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CO_2 Lasing on Non-Traditional Bands

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1. Introduction

Construction of powerful and efficient laser sources, lasing in various IR ranges, is of importance for further development of a number of trends, e.g., spectroscopy, laser chemistry, sounding of the atmosphere, and metrology. The most natural way to solve this problem is to use unconventional (nontraditional) transitions to produce lasing in commonly used CO_2 lasers. The spectral range of CO_2 lasers is greatly increased in lasing on transitions of the so-called "hot" band 01^11-11^10, whose P-branch is in the range of 10.9-11.3 μm. Thorough investigations of gain, vibrational temperatures (T_1, T_2, T_3), and output parameters on lines of the hot band made it possible to achieve efficient lasing both for pulse TEA and for cw longitudinal-discharge CO_2 lasers.

In studying the lasing spectrum of hot transitions in TEA CO_2 lasers some lines not belonging to the 01^11-11^10 band. We suggested, that these lasing lines belong to higher level transitions, e.g., 10^01-20^00 (04^00), which were called "doubly hot," i.e., transitions in which compared to hot transitions two deformation quanta or one symmetric quantum rather than one deformation quantum is added both to the upper and to the lower energy level.

In the present work lasing in both a TEA laser and a low-pressure laser with longitudinal discharge on some transitions of the CO_2 molecule in the range of 11.0-11.6 μm is reported. The rather high resolution of the spectral equipment used and calculation of transition frequencies on the basis of recent spectroscopic constants made it possible to identify definitively the lasing lines obtained as belonging to the doubly hot bands 02^21-12^20 and 10^0l-20^00 and the sequence hot band 01^12-11^11. To find optimum conditions for lasing on the aforementioned bands experimental studies of vibrational temperatures in active media of a TEA CO_2 laser and a low-pressure laser with longitudinal discharge were carried out.

Earlier the lasing on the $02^00(10^00)$-01^10 band of the CO_2 molecule has been obtained in the specific systems at cryogenic temperatures under the lowest efficiency. The optimization of the active medium and its electrical discharge pumping conditions based on the original technique of the temperature model allowed to obtain in the simple TE CO_2 laser with UV preionization the powerful lasing on the 02^00-01^10 band at room temperature. The dependencies of the output and spectral performances of the 16 (14) micrometers lasing vs. a content of the active medium, pumping parameters and cavity characteristics have been carried out.

To increase the power performances of the 16 (14) microns CO_2 laser the possibility of lasing on the $02^01(10^01)$-01^11 band have been experimentally and theoretically investigated under the combined (electrical + optical) excitation of the active medium. The conditions for

obtaining effective lasing at the rotational-vibrational transitions of the 02^01-01^11 (λ = 16.4 μm) and 10^01-01^11 (λ = 14.1 μm) bands of the CO_2 molecule are examined. To obtain population inversion in the indicated channels one should initially populate the 00^02 vibrational level, considerable population of which can be accomplished comparatively simply, for example, in an electric discharge. Then a powerful two-frequency radiation resonant with the 00^02-02^01 (10^01) and 01^11-11^10(03^10) transitions acts on the medium excited in such a way. We will discuss by what means such a scheme of lasing in one active medium can be accomplished.

The lidar complex of equipment based on CO_2 laser specially designed for atmospheric sensing, with tuning on generation lines in the spectral ranges 9-11.3 and 4.5-5.6 μm will be described. Considerable extension of the spectral range to the short-wave region is attained due to effective CO_2 laser second harmonic generation in nonlinear crystals. Taking into the real potentialities of the lidar complex in hand, using a package of spectroscopic data HITRAN, computer simulation of atmospheric transmission has been made. On this basis, by the method of differential absorption a method has been elaborated for measuring of small concentrations of a number of gases.

2. Effective oscillation of a cw CO_2 laser in the range of 11 μm (01^11-11^10 band)

The CO_2 laser oscillation spectrum expansion to the long-wave region is of interest for various scientific and practical applications, for example, for spectroscopy, atmosphere monitoring, etc. From this point of view, the use of the P-branch of the hot 01^11- 11^10 band (10.9—11.4 μm) (see Fig. 1) has considerable promise. Weak hot band lasing was registered in the middle of the 60s in specific long tube (2—4 m) laser systems. The problem of obtaining the hot band oscillation in commercially available cw CO_2 lasers is associated with the low gain realized under conventional conditions. Therefore, the effective hot band cw CO_2 laser oscillation demands, first of all, as in the pulsed TEA CO_2 system, comprehensive study of excitation and active medium composition effects on the hot band gain.

In this work we present experimental results of searching for optimal conditions of the hot band line lasing in a cw CO_2 laser with a commercial 1.2 m sealed-off tube. The hot band gain and output optimization was carried out depending on the active medium composition as well as on the discharge current.

The gain in the active medium was measured by small signal probing using the compensation method. We used as a probing laser a specially developed cw stabilized CO_2 laser tunable over many of the hot band, sequence 00^02-10^01(02^01) band and regular 00^01-10^00(02^00) band lines.

Analysis of the results obtained was carried out on the basis of the universally accepted CO_2-molecule vibrational temperature model (Petukhov et. al.., 1985). The vibrational temperatures of the asymmetric (ν_3) and bound symmetric-bend ($2\nu_2 \approx \nu_1$) mode, T_3 and T_2, respectively, have been determined from the following expressions (Petukhov et. al.., 1985):

$$T_3 = -\frac{3380}{\ln\frac{K_s}{2.1 \cdot K_t} - \frac{36}{T}}, T_2 = -\frac{960}{\ln\frac{K_h}{K_t} - \frac{18}{T}}, \quad (1)$$

where K_r , K_s, K_h are measured small signal gains of the corresponding regular, sequence and hot band lines; T is the translational temperature determined from the gain distribution over the regular band lines (Petukhov et. al.., 1985). The lock-in amplifier and box-car integrator used in the recording system allowed us to achieve a better than 2% measurement gain accuracy for all bands.

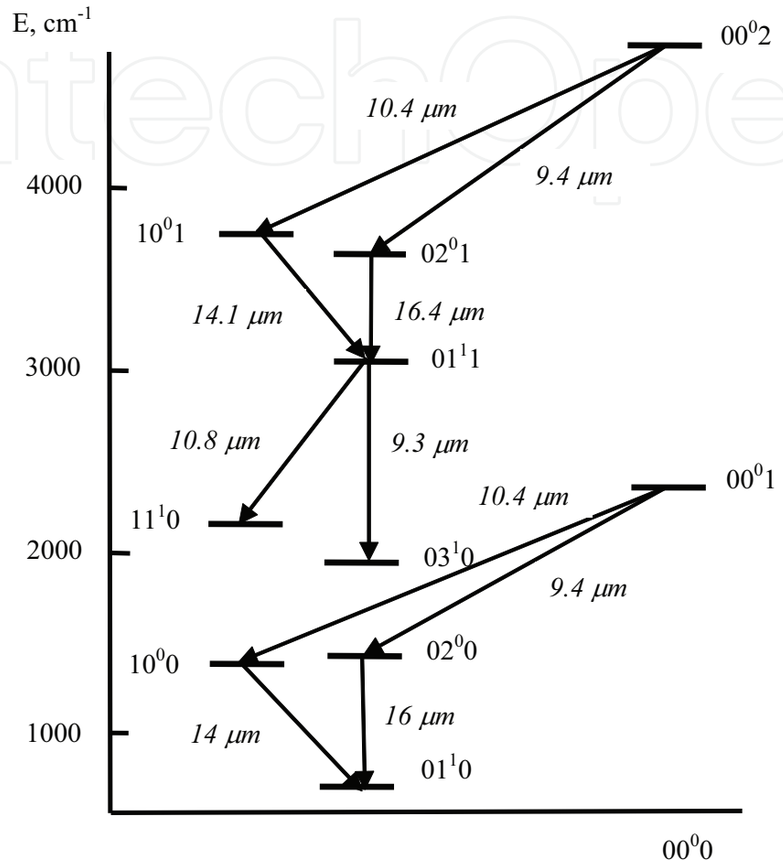


Fig. 1. Simplified diagram of lower vibrational levels of the CO_2 molecule.

The first step was optimization of active medium composition and pressure P and of discharge current. The measurement of the small signal gain K_h has shown that the optimum mixture for the hot band is CO_2:N_2:He = 1:1.4:3.5 at a total pressure P = 11 Torr *(I* = 15 mA) in which on the strong lines K_h = 0.08m^{-1}. It is important that this mixture contains less He and has a large partial content of CO_2 as compared with the mixture 1:1.6:6.5 (P = 15 Torr, I = 10 mA) optimum for the regular band 00^01-$10^00(02^00)$. The theoretical and experimental investigations of vibrational temperatures of a CO_2 molecule has shown that to obtain considerable hot band gain K_h of a CO_2 molecule it is necessary to heat up the $v_2(v_1)$ mode characterized by vibrational temperature T_2 along with the excitation of the v_3 mode (temperature T_3). For the conventional values of T_3 ≈1600 – 2200 K realized in an electric discharge the K_h gain is shown to achieve its maximum if T_2~l/3T_3 (Bertel et. al.., 1983). Such a relationship between T_2 and T_3 can be reached in gas mixtures with greater CO_2 and lesser He contents, as compared to those optimal for the regular band oscillation. In addition, an increased specific energy input is also required.

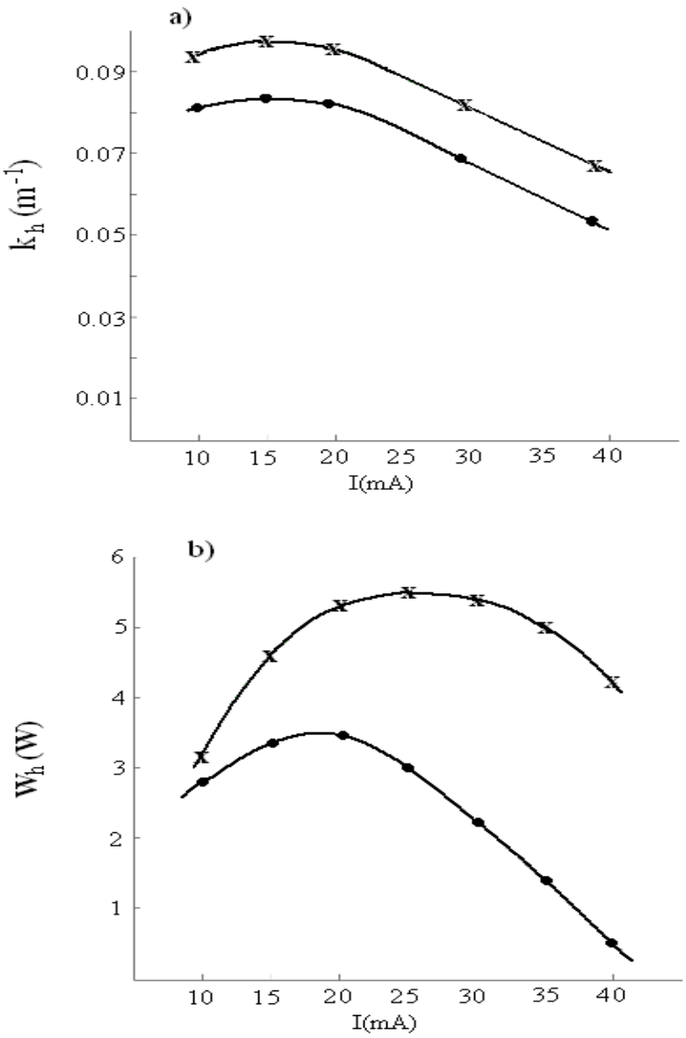


Fig. 2. A typical pattern of the hot band gain K_h (a) and output power W_h (b) as functions of the disharge current I for the CO_2:N_2:He – 1:1.4:3.5 (P – 11 Torr) mixture without Xe (•) and with optimal Xe content (0.3 Torr) (×).

It is known for the CO_2 laser regular band that addition of Xe to the active medium sometimes results in an output power increase (Gorobets et al., 1990). We have also investigated the influence of Xe on the characteristics of the active medium and the lasing parameters (Fig. 2). It has been found that small additions of Xe to the mixture (~ 30% of the CO_2 content) increase K_h by 25% and the lasing power in the hot band by a factor of 1.5. The analysis of vibrational temperatures shows that this is due to the increase in excitation efficiency of vibrations of N_2 and the v_3 asymmetric mode of CO_2 in electric discharge [Fig. 3(a)]. Besides, using the reconstruction method of K_h, K_s and K_r gains we have found from the experimental values of T_3, T_2 and T that addition of Xe reduces the CO_2 molecule dissociation in the discharge [Fig. 3(b)]. This also results in an output increase. It is noteworthy that addition of Xe considerably improves the output parameters only for the low gain transitions. The study of the Xe effect on the laser output for the regular band

where gain is fairly high (K_r≈0.6 m^{-1}) has shown that in this case an output power increase is not large (~15%).

After optimization of the gas content, pressure and discharge current we optimized the laser resonator. In the optimal case, the laser resonator was formed by a flat 100 lines/mm^{-1} grating and a totally reflecting concave mirror (R = 3m). The resonator length was 1.5 m. About 6% of the radiation was extracted through the grating zeroth order.

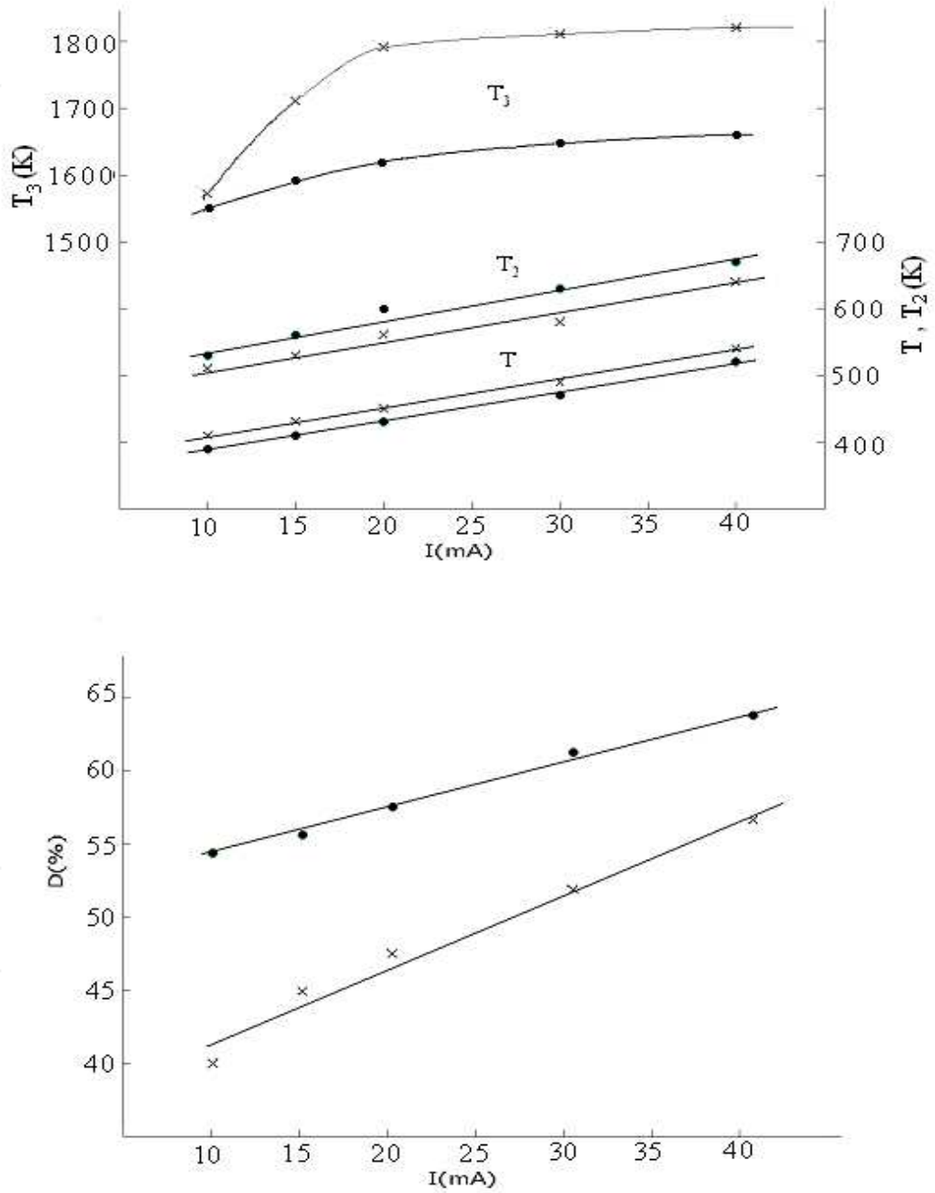


Fig. 3. Vibrational T_3, T_2 and transitional T temperatures (a) and CO_2 molecules dissociation degree D (b) vs the discharge current I for the CO_2:N_2:He – 1:1.4:3.5 (P – 11 Torr) mixture without Xe (•) and with optimal Xe content (0.3 Torr) (×).

Thus, after the above improvements the commercially available sealed-off laser (LG-22) (FSUE RPC "Istok", Fryasino, Russia) oscillates on more than 30 lines of the P-branch of the 01^11- 11^10 band in the 10.9—11.3 μm range with output power no less than 0.5 W. On strong lines (P(16)—P(26)) output power was ~ 6W at efficiency ~3% which makes up ~40% of analogous laser parameters in the case of oscillation on the lines of regular bands 00^01-10^00 (02^00) under optimum conditions.

3. New laser transitions of the CO_2 molecule in the wavelength range of 11.0-11.6 μm

Construction of powerful and efficient laser sources, generating in various IR ranges, is of importance for further development of a number of trends, e.g., spectroscopy, laser chemistry, isotope separation, sounding of the atmosphere, and metrology. The easiest and most natural way to solve this problem is to use unconventional transitions to produce lasing in commonly used CO_2 lasers (Churakov et al., 1987). The spectral range of CO_2 lasers is greatly increased in lasing on transitions of the so-called "hot" band 01^11-11^10, whose P-branch is in the range of 10.9-11.3 μm. Thorough investigations of gain, vibrational temperatures, and output parameters on lines of the hot band made it possible to achieve efficient lasing both for pulse TEA and for cw longitudinal-discharge CO_2 lasers.

In studying the lasing spectrum of hot transitions in TEA CO_2 lasers (Bertel at al., 1983). some lines not belonging to the 01^11-11^10 band occurred in the spectral range of 875-882 cm^1 that were not identified due to the poor resolution of the monochromator used and the lack of reliable spectroscopic data in the literature at that time. It was suggested, that these lasing lines belong to higher level transitions, e.g., 10°l-20°0(04^00), which were called 'doubly hot," i.e., transitions in which compared to hot transitions two deformation quanta or one symmetric quantum rather than one deformation quantum is added both to the upper and to the lower energy level.

In the present work lasing in both a TEA laser and a low-pressure laser with longitudinal discharge on some transitions of the CO_2 molecule in the range of 11.0-11.6 μm is reported for the first time. The rather high resolution of the spectral equipment used and calculation of transition frequencies on the basis of recent spectroscopic constants made it possible to identify definitively the lasing lines obtained as belonging to the doubly hot bands 02^2l-12^20 and 10°1-20^00 and the sequence hot band 01^12-11^11 (Fig. 1).

Within the scope of a commonly used model of vibrational temperatures (Gordiets at al. 1980, Smith and Thompson, 1981) let us analyze what gain coefficients for a weak signal can be realized for the aforementioned bands in electrical-discharge lasers. Estimates showed that to achieve a suitable gain at the new transitions it is necessary, together with the heating up of the asymmetric type of oscillations, characterized by the vibrational temperature T_3, to strongly excite the connected deformation and symmetric modes (T_2). Moreover for the sequence hot band 01^12-11^11 there is one additional condition – the excitation of the asymmetric mode must be at the same high level as for the sequence bands 00^02-10^01 (02°1) (Petukhov et. al.., 1985).

To find optimum conditions for lasing on the aforementioned bands experimental studies of vibrational temperatures in active media of a TEA CO_2 laser and a low-pressure laser with longitudinal discharge were carried out using to the techniques described in (Bertel at al.,

1983, Gorobets et al., 1990). According to the measurements, mixtures of the composition CO_2:N_2:Ne = 1:1:1, in which with an increased specific energy contribution the gain coefficients for a weak signal are — 0.2 m^{-1} for doubly hot transitions and —0.3 m^{-1} for sequence hot ones, are optimum for TEA CO_2 lasers. The vibrational temperatures T_3 and T_2 must have values of —2000 K and —650 K, respectively. For low-pressure lasers with longitudinal discharge mixtures of the compositions CO_2:N_2:He:Xe = 1:1.2:2.5:0.4 (doubly hot bands) and 1:1.5:2.5:0.4 (sequence hot) are optimum. The gain coefficient in such mixtures for the aforementioned transitions can reach —0.04 m^{-1} (T_3—1800 K, T_2—600 K). It should be noted that at these transitions the gain is considerably lower than that at the ordinary (~20 times) and hot (~4 times) bands, and consequently a high-Q cavity, lack of harmful losses, and careful selection of the active medium and the conditions of its excitation are necessary to obtain lasing on transitions with such a low gain.

The lasing mode on the new transitions was studied first on a TEA CO_2 laser with UV preionization. The distance between electrodes that were 4 cm wide was 2 cm. The length of the discharge gap was 70 cm. The main charge and the UV preionization were energized from a battery of low-inductance capacitors with a total capacitance of 0.25 μF, charged to a voltage of 30 kV. The design and of the laser and its performance are described in detail in (Gorobets et al., 1995). A two-transmission three-mirror resonator was used to increase the length of the active medium to 140 cm. A planar grating with 150 lines/mm working in the first order according to an autocollimation scheme with a reflection coefficient not less than —90%, was one of the end mirrors of the resonator. Radiation from the resonator (~5%) was extracted through the zero order of the grating. The other two mirrors were spherical (R = 10 m) with a highly reflective coating. The active medium of the laser was a mixture of gases of the composition CO_2:N_2:He = 0.8:1.0:1.2 with a total pressure of 200 Torr, which is close to the optimum found from experimental investigations of vibrational temperatures. For this mixture lasing was achieved at more than 10 lines in new bands of the 11.3-11.6 μm range. At the strongest lines the energy in the pulse exceeded 150 mJ. The peak power with a pulse length at half-height of ~0.5 μs attained ~0.3 MW. More thorough investigations of the lasing spectrum of the new transitions were done in the present work for a low-pressure CO_2 laser.

Experiments were performed with a GL-501 production-type gas-discharge tube of an LG-22 commercial laser (FSUE RPC "Istok", Fryasino, Russia). The inner diameter of the tube— 15 mm, length of the discharge gap —1.2 m. The tube, which worked in the sealed-off mode, was filled with a gas mixture of the composition CO_2:N_2:He:Xe = 1:1.2:2.5:0.4 under the total pressure of 13.5 Torr. The total reflection spherical mirror (R = 3 m) of the commercial laser was not replaced, and a diffraction grating, which worked according to an autocollimation scheme in the first order, was used instead of the output mirror. The emission was extracted through the zero order. The cavity base was 1.5 m. Most of the new lines were obtained with a grating with 100 lines/mm (reflection coefficient – 95%, extraction of emission – 3%). A number of lines in the range of 11.0-11.4 μm, where comparatively strong hot transition are located, were successfully obtained with a more selective grating with 150 lines/mm (93 and 3%, respectively). In addition, to increase the Q-factor of the cavity the germanium etalon was placed before the grating (perpendicular to the output radiation), which not only increased the Q-factor of the grating, returning 75% of the radiation back to the cavity, but also increased its selectivity substantially. As a whole, this device, consisting of a grating and an etalon, was a highly selective output mirror with a reflection coefficient of 97.5% for a grating with 100 lines/mm and 95.5% for a grating with 150 lines/mm. It should be noted,

that this original technique made it possible to separate weak lines of the new transitions from closely positioned ones in some regions of the spectrum of stronger hot lines.

Line	λ_{meas}, μm	λ_{cal}, μm	Intensity, W	Line	λ_{meas}, μm	λ_{cal}, μm	Intensity, W
02^21-12^20				10^01-20^00			
P(12)	11.2513	11.251536	10	*P(14)*	11.0358	11.036728	10
*P(14)**	11.2729	11.273239	25	*P(16)*	11.0590	11.058304	10
P(15)	11.2834	11.284243	15	*P(18)*	11.0794	11.080312	15
P(16)	11.2944	11.295321	16	*P(20)*	11.1027	11.102758	20
P(17)	11.3064	11.306522	24	*P(22)*	11.1260	11.125648	20
*P(18)**	11.3173	11.317785	35	*P(24)*	11.1488	11.148987	16
P(19)	11.3300	11.329186	30	*P(28)*	11.1961	11.197036	20
P(20)	11.3410	11.340633	28	*P(30)*	11.2223	11.221757	17
P(21)	11.3517	11.352241	25	*P(32)*	11.2467	11.246953	20
P(22)	11.3634	11.363870	20	*P(34)*	11.2729	11.272628	25
P(23)	11.3754	11.375690	20	*P(36)*	11.2978	11.298791	20
P(24)	11.3872	11.387499	30	*P(38)*	11.3251	11.325449	10
P(25)	11.3991	11.399538	25	01^12-11^11			
P(26)	11.4117	11.411523	18				
P(27)	11.4241	11.423790	16	*P(23)*	11.0408	11.041722	25
P(28)	11.4357	11.435948	15	*P(24)*	11.0512	11.050460	18
P(29)	11.4494	11.448451	15	*P(25)*	11.0650	11.064261	25
P(30)	11.4596	11.460775	15	*P(26)*	11.0733	11.072789	15
P(31)	11.4730	11.473525	9	*P(28)*	11.0953	11.095487	30
P(32)	11.4855	11.486009	10	*P(29)*	11.1096	11.110543	30
P(33)	11.4981	11.499017	8	*P(30)*	11.1176	11.118556	25
P(34)	11.5112	11.511655	12	*P(32)*	11.1425	11.142003	28
P(35)	*11.5245*	11.524934	4	*P(33)*	11.1591	11.158465	20
P(36)	11.5375	11.537715	4	*P(34)*	11.1667	11.165831	30
				P(36)	11.1908	11.190045	30
				*P(46)**	11.3173	11.317088	35
01^11-11^10							
P(47)	11.3075	11.307393	40				
P(49)	11.3352	11.334278	35				
P(50)	11.3601	11.359850	40				
P(51)	11.3610	11.361589	18				
P(53)	11.3907	11.389331	20				

*The lines are not identified ambiguously (they may belong to the both lines).

Table 1. Measured and calculated values of wavelengths and experimental values of intensities for new transitions

The lasing spectrum in the range of 11.0-11.6 μm was studied in detail with the gas-discharge tube being energized from a pulsed source. It was found experimentally that the following pumping parameters are optimum for lasing at the new transitions: a pulse rate of

375 Hz, a length of the excitation pulse of ~5O μs, an average current of 8.5 mA. Under these conditions with careful adjustment of the diffraction grating and the etalon we managed to obtain more than *50* new lasing lines (see Table 1). Lasing wavelengths were measured with an SPM-2 monochromator (Carl Zeiss Jena, Germany) with a highly selective diffraction grating, whose resolution was not worse than 0.0005 μm. Absolute calibration of the monochromator was done using the technique described in (Gorobets et al., 1992), which is based on a search for a line with an anomalously high gain, e.g., the line *P(23)* of the hot band. In addition, correction calibration against known wavelengths of hot transitions was done on virtually the entire investigated spectrum. New lines were identified by comparing measured and calculated values of transition wavelengths. Calculations were done using standard methods. Values of the constants G, B, D, H were well known (Witteman, 1987).

The peak power (intensity) on the strongest lines of the new bands with a lasing pulse length at half-height of -50 ns was $-$ 30 W. The average output power reached -0.2 W. Lasing was achieved at a number of new transitions and in the continuous mode with the discharge tube being energized from a dc power supply. More than 25 new lasing lines with λ = 11.1-11.4 μm, belonging to all the aforementioned bands, were observed in this mode in the spectral range studied. The output power on strong lines attained 0.25 W.

The characteristics of the output radiation given in the present work are not the best attainable. Optimization of the active medium composition, the conditions of its excitation, and the cavity parameters will make it possible to increase the efficiency of lasing at the new transitions. However at present the large number of new lasing lines obtained substantially broadens the potentialities of simple laser systems on CO_2 for various applications.

4. Optimisation of a cw CO_2 laser output

4.1 Optimisation technique

To the present time the number of optimization methods of CO_2 laser power parameters is developed. However, the known methods are either complex, since they are based on the calculations calling for a knowledge of a great number of parameters or by virtue of sufficiently rough approximations, not always provide the necessary accuracy, as in the development of laser systems generating on the nonregular transitions – 00^02-10^01, 02^01 (sequence bands); 01^11- 11^10 (hot band); 02^21-12^20, 02^01-12^00 (double hot bands). The gain on these transitions is much weaker than on the regular transitions 00^01-10^00, 02^00 and hence a careful optimization of the active medium composition and of the resonator and pumping parameters is required to provide the lasing on them. Therefore until now remains to actual search new and perfecting of known methods of optimization CO_2 lasers.

We have developed and experimentally tested the method of optimizations of the cw CO_2 lasers energy parameters. To realize it, it is necessary to known the vibrational temperatures of the symmetrical (T_1), bending (T_2) and asymmetrical (T_3) modes of the CO_2 molecule vibrations. At the present time, the generally recognized fact is that the knowledge of these temperatures as well of the gas temperature (T) of the gas mixture makes it possible to determine all the most important characteristics of the active medium (population of the energy levels, the energy accumulated in different modes of CO_2, the efficiency of excitation, and so on). Next, the main energy characteristics of the laser system can be calculated based on the information about the temperatures of the medium.

In our works we used the method of determination of vibrational temperatures, which is based on the measurements of the gain on separate vibrational lines of regular and nonregular CO_2 bands (Petukhov et. al.., 1985). The advantages of this method are the possibility of determination of all vibrational and translational temperatures at once, a relative simplicity and sufficiently high accuracy as compared with other known methods. Besides, a knowledge of the absolute values of the gain factors and of the active medium composition is not needed here, which is sometimes very important.

It would appear reasonable that within the limits of the model of vibrational temperatures the output power (P) for every above-indicated bands is dependent only on temperatures. The experimental investigations performed by us show that for a typical low-pressure CO_2 laser with a longitudinal continuous discharge the active medium in the lasing regime differs significantly from that in the absence of lasing in only the value of the asymmetric vibration temperatures T_3, while for the other temperatures T_2, T_1 and T the difference is insignificant (less than 10 %). Such a temperature approximation is predominantly due to fact that the energy capacitance for vibrations of symmetric and bending modes of CO_2 is much greater than that for vibrations of the asymmetric mode as well as due to the constant effective heat abstraction from the low laser levels. This approximation may be thought of as by true for laser system with on efficiency of transformation of the energy contributed to the discharge to the lasing energy of -10 % or less percent, which is characteristic of all real continuous CO_2 lasers. In this case, using the ratio between the temperature of the asymmetric mode and average number of vibrational quanta accumulated in this mode we can write the following simple expression for the output power in every above-indicated band:

$$\varepsilon_3 = \exp\left(-\frac{h\nu_3}{kT_3}\right) \Big/ \left[1-\exp\left(\frac{h\nu_3}{kT_3}\right)\right], \tag{2}$$

$$P = A \cdot \frac{K_{loss}^{us}}{K_{loss}^{h} - K_{loss}^{us}} \cdot \left(\frac{\exp\left(-\frac{h\nu_3}{kT_3}\right)}{\left[1-\exp\left(-\frac{h\nu_3}{kT_3}\right)\right]} - \frac{\exp\left(-\frac{h\nu_3}{kT_3^*}\right)}{\left[1-\exp\left(-\frac{h\nu_3}{kT_3^*}\right)\right]} \right), \tag{3}$$

where A is the proportionality factor dependent on the CO_2 content and independent on the lasing band; K_{loss}^{us} is the useful loss factor; K_{loss}^{h} is the harmful loss factor; T_3 and T_3^* are the vibrational temperatures of the asymmetric mode of the CO_2 molecule in the regime of amplification and lasing, respectively.

The temperature T_3 as well as T_2, T_1 and T can be found if the gain factors of the weak signal in different bands are known (Petukhov et. al.., 1985). To determine the temperature T_3^* we will draw on the fact that in the regime of lasing (continuous) the gain factor is equal to the total loss factor:

$$K_g^*\left(T_3^*, T_2, T_1, T\right) = K_{loss}^{h} - K_{loss}^{us}, \tag{4}$$

Then, using the dependence of the gain factor on the difference in the population of the upper and low laser levels, expressed through vibrational temperatures. We can easily

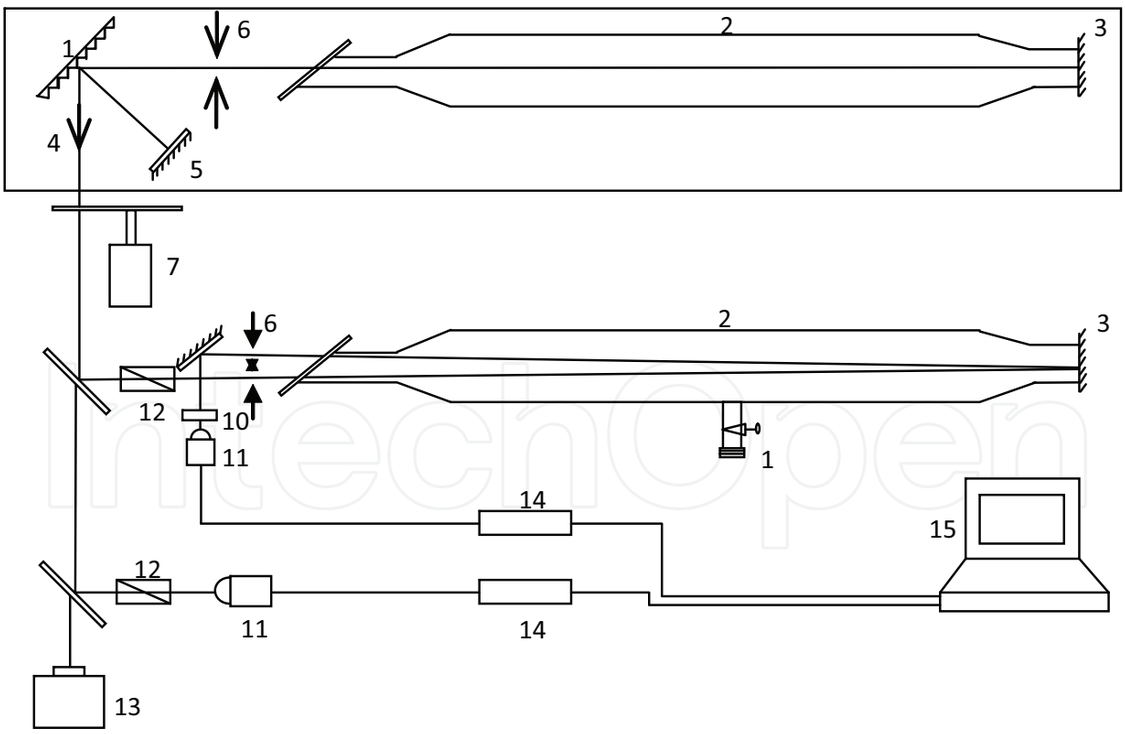
obtain expressions for determining T_3 for the above-indicated bands. For example, for the 00^02-10^01 band it has the form

$$\left[1-\exp\left(-\frac{h\nu_3}{kT_3^*}\right)\right]\cdot\left\{\exp\left(-\frac{h\nu_3}{kT_3^*}\right)-\exp\left(-\frac{h\nu 1}{kT_1}\right)\exp\left(-\frac{h\nu_3}{kT_3^*}\right)\right\}= \\ =\frac{K_{loss}^h-K_{loss}^{us}}{K_g^s}\cdot\left[1-\exp\left(-\frac{h\nu_3}{kT_3^*}\right)\right]\cdot\left\{\exp\left(-\frac{h\nu_3}{kT_3^*}\right)-\exp\left(-\frac{h\nu 1}{kT_1}\right)\exp\left(-\frac{h\nu_3}{kT_3^*}\right)\right\}, \quad (5)$$

Similar expressions are also true for other bands. Thus, if the all temperatures in the regime of amplification and the loss factors are known, it is an easy matter to calculate the output power for different bands. The temperatures T_3, T_2, T_1 and T can be determined from the measurement of the gain factor of a weak signal on the lines of different bands by the method described in (Petukhov et. al.., 1985).

4.2 Experimental setup

Figure 4 shows a block diagram of the experimental setup for CO_2 laser optimization. A sealed off a cw CO_2 laser was the source of probe radiation. It could be tuned over the vibrational-rotational lines of the regular (00^01-10^00, 02^00) bands, the sequence (00^02-10^01, 02^01) bands, the hot (01^11- 11^10) band or the new (02^21-12^20, 02^01-12^00….) bands.



1 – probing laser; 2 – discharge tube; 3 – 100% reflection mirror; 4 – grating; 5 – additional mirror; 6 – iris diaphragm; 7 – chopper; 8 – ZnSe plane-parallel plate; 9 – mirror; 10 – interference filter; 11 – photo detector; 12 – polarizer; 13 – spectrum analyzer; 14 – ADC; 15 – computer; 16 gas valve.

Fig. 4. Experimental setup for CO_2 laser optimization

A production-type water-cooled sealed-off gas-discharge tube of GL-501 type (FSUE RPC "Istok", Fryasino, Russia) was used as an active element of the probe laser. It has discharge-gap length of ~1 .2 m, and the inside diameter is of 15 mm. The tube was filled with a CO : N_2 : He :Xe mixture in a proportion of 1.0:1.6:4.0:0.6 at a total pressure of *13.5* Torr. The laser cavity was formed by ~100% reflecting mirror with a curvature radius of 3 m built in the tube, a plane diffraction grating and an additional mirror with a large curvature radius.

We used a nonconventional scheme of the laser cavity. The diffraction grating operated in the first diffraction order in the nonLittrow scheme. Laser radiation was extracted from the cavity through the zero order. Our studies have shown that the diffraction grating with 150 lines/mm and a reflectance of >95%, combined with the additional mirror with a curvature radius of 10 m, are optimal for obtaining the necessary high spectral resolution with a sufficiently high output power. A more detailed description of the construction of the probe laser is given in the next part.

A signal from the probe laser passed a two times through the active medium under study in the discharge tube and was recorded by a liquid nitrogen cooled photo detector. This discharge tube was similar to one used as the active element of the probe laser. In addition to the measuring signal, we used a reference signal that does not pass along the investigated active medium and appears as a result of reflection of a portion of radiation from the ZnSe plane-parallel plate (see Fig. 4). This portion of radiation was directed to the another liquid nitrogen cooled Ge:Au photodetector of the reference channel, which makes automatically possible to account for the possible instability of the output laser radiation by way of normalization of the measuring signal to reference one.

The electric signals from two photodetectors were fed into an two-channel digital registration system on the base of PC. Lasing wavelengths were measured with SPM-2 spectrum analyzer (Carl Zeiss Jena, Germany) with a highly selective diffraction grating, whose resolution was not worse than 0.0005 μm.

4.3 Results and discussions

To test the method proposed we have performed experimental investigations and calculations of the output power (*P*) dependence on the discharge current (*I*) for a cw CO_2 laser, operated on four different bands (00^01-10^00, 00^02-10^01, 1^11- 11^10 and 02^21-12^20). The cw CO_2 laser was similar to one used as the probe laser. The only distinction is the using of the appropriate diffraction grating with optimum Q-factor for each band.

The temperatures T_3, T_2 and T (see Fig. 5a), used in the calculations, were determined from the measurement of the gain factor of a weak signal by the method described in (Petukhov et. al.., 1985). For our experiments T_1 is approximately equal T_2. According to our calculations the loss factors for different bands have the following values:

$$for\ P(18)\ \ 00^01-10^00-K_{loss}^{h}=4.8\times10^{-4}cm^{-1};K_{loss}^{us}=5.3\times10^{-4}cm^{-1},$$

$$for\ P(19)\ \ 00^02-10^01-K_{loss}^{h}=4.8\times10^{-4}cm^{-1};K_{loss}^{us}=5.3\times10^{-4}cm^{-1},$$

$$for\ P(19)\ \ 01^11-11^10-K_{loss}^{h}=2.3\times10^{-4}cm^{-1};K_{loss}^{us}=1.9\times10^{-4}cm^{-1},$$

$$for\ P(19)\ \ 02^21-12^20-K_{loss}^{h}=2.1\times10^{-4}cm^{-1};K_{loss}^{us}=1.1\times10^{-4}cm^{-1},$$

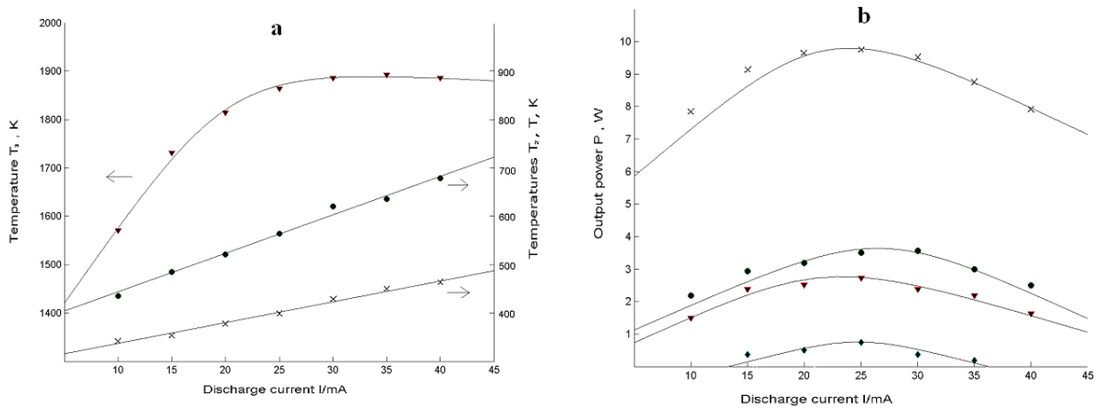


Fig. 5. Dependencies of temperatures T_3 (∇), T_2 (O), and T (×) (a) and output power P (b) from a discharge current (× - for P(18) 00^01-10^00, O - for P(19) 00^02-10^01, ∇ - for P(19) 01^11-11^10, ◊ - for P(19) 02^21-12^20)

Figure 5b shows the calculated curves and experimental values of the output power for the considered bands. At first we calculated the value of P/A for every band in accordance with discharge current. Then, we determined the proportionality coefficient A from the experimental data for the 00^01-10^01 band at I=25 mA. As this takes place, the coefficient A has a common value for all lasing bands. Next, the dependence P on I was constructed.

The method of output optimization of cw CO_2 lasers has been developed. The method is based on vibrational and translational temperatures determination by gain measurements on the ro-vibrational lines of regular (00^01-10^00, 02^00) and nonregular (00^02-10^01,02^01; 01^11-11^10; (02^21-12^20, 02^01-12^00…) bands of CO_2 molecule. To test the validity of the method, the experiment realization has been done for a low pressure CO_2 laser with the cw longitudinal discharge, that can oscillate on the lines of regular and nonregular lines. The good agreement between calculation and experiment data has been observed. Thus, a good agreement between the calculated and experimental data, which is observed, as a whole, for all the investigated bands, is demonstration that this method can be applied to the optimization of the output power of cw CO_2 lasers. This method can be also successfully used for the optimization of the output parameters depending on the pumping and Q-factor of the resonator of the lasers generating only on the regular transitions 00^01-10^00 and 00^01-02^00.

5. 16(14) microns TE CO_2 laser working on the $02^00(10^00)$-01^10 and $02^01(10^01)$-01^11 bands

Earlier the lasing on the $02^00(10^00)$-01^10 band of the CO_2 molecule (see Fig. 1) has been obtained in the specific systems at cryogenic temperatures under the lowest efficiency (Wexler, 1987). The optimization of the active medium and its electrical discharge pumping conditions based on the original technique of the temperature model (gain measurements on the several bands: 00^01-10^00, 00^02-10^01, 01^11- 11^10 of CO_2 molecule) allowed to obtain in the simple TE CO_2 laser with UV preionization (the active media length is 65 cm the width of electrodes is 2.5 cm, the interval between electrodes is 1.8 cm, (the voltage on the 0.2 micro Farad capacitor is 6.5 kV) the powerful lasing on the $02^00(10^00)$-01^10 bands at the room temperature. The output pulse energy of 57 mJ and the peak power of some tens kWatt have

been reached. The dependencies of the output and spectral performances of the 16 (14) micrometers lasing vs. a content of the active medium, pumping parameters and cavity characteristics have been carried out.

To increase the power performances of the 16 (14) microns CO_2 laser the possibility of lasing on the $02^01(10^01)$-01^11 band have been experimentally and theoretically investigated under the combined (electrical + optical) excitation of the active medium. The conditions for obtaining effective lasing at the rotational-vibrational transitions of the 02^01-01^11 (λ = 16.4 μm) and 10^01-01^11 (λ= 14.1 μm) bands of the CO_2 molecule are examined. To obtain population inversion in the indicated channels one should initially populate the 00^02 vibrational level, considerable population of which can he accomplished comparatively simply, for example. in an electric discharge (Petukhov et. al.., 1985). Then a powerful two-frequency radiation resonant with the 00^02-$02^01(10^01)$ and 01^11-11^10 (03^10) transitions, saturating an individual rotational-vibrational transition, acts on the medium excited in such a way. As a result of this the first electromagnetic field, resonant with the 00^02-$02^01(10^01)$ transition, populates the upper laser level $02^01(10^01)$, while simultaneously the second field, resonant with the 01^11-11^10 (03^10) transitions, depopulates the lower level 01^11 which also leads to inversion of the populations in the 02^01 (10^01)-01^11 16(14) μm channel. In (Churakov et al., 1987). we have discussed by what means such a scheme of lasing in one active medium can be accomplished.

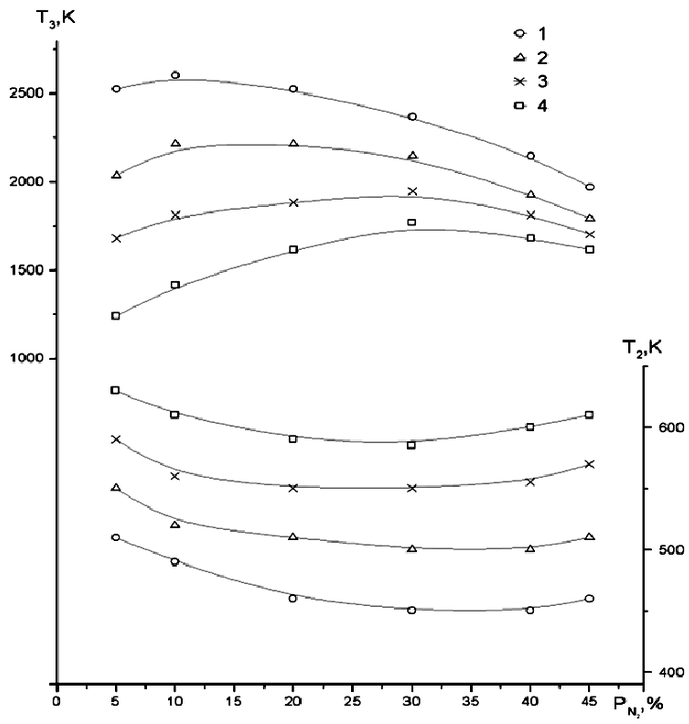


Fig. 6. Vibrational temperatures T_2 and T_3 vs N_2 content at fixed values of P_{CO2} 5(1), 10 (2), 20 (3), and 30% (4) in the mixture CO_2:N_2:He (P = 200 Torr, U= 30 kV, C = 0.25 μF)

Let us examine formation of inversion on the $02^01(10^01)$-01^11 transition using the example of an ordinary pulsed TEA CO_2 laser. For this we employ a temperature model which at the

present time is widely used to describe processes occurring in the active media of CO_2 lasers and amplifiers. According to this model, population of the vibrational levels is unambiguously connected with the vibrational temperature of the symmetric (T_1), bending (T_2). and asymmetric (T_3) modes of the CO_2 molecule. We performed experimental investigations of the vibrational temperatures in the active medium of the TEA CO_2 laser, directed toward a search for the optimum conditions for lasing in the 16(14) μm channel. The vibrational temperatures T_3 and T_2 (T_1=T_2 for conditions examined) were determined from the ratios of the measured amplification coefficients of a weak signal at the individual rotational-vibrational lines of the 00^01-10^00, 00^02-10^01 and 1^11-11^10 bands by the procedure described early.

Let us examine what kind of the small gain and the output energy can be attained in the TEA CO_2 laser on the $02^01(10^01)$-01^11 transitions. On the basis of the experimentally determined vibrational temperatures T_3 and T_2 (see Fig. 6) using the well-known expression (Gordiets et al., 1980) we calculated the small gain. The calculations shown that the small gain in the $02^01(10^01)$- 01^11 band can attain a significant value (>1m^{-1}). The necessary conditions for the effective lasing have been determined. It is shown that in optimum conditions the output energy can reach *1.3* J/l at the peak power 5 MW and at the full efficiency of 2 %.

6. A stabilized cw CO_2 laser automatically switched between generations lines

This part describes a cw CO_2 (CO) laser with stabilized output parameters that can be automatically switched from line to line. The laser generates 115 vibration-rotation CO_2 lines between 9.15 and 11.3 μm and 100 CO lines between 5.3 and 6.4 μm. The laser is switched from CO_2 operation to CO operation by replacing a sealed laser tube. Then computerized control of the laser spectrum is described.

Although there are many publications on tunable lasers (Gorobets et al., 1992) it is premature to think that all design and operation problems of tunable CO_2, and especially of CO lasers, have been resolved. Computer control over the tuning of the generation wavelength is required (Gorobets et al., 1992). Fully computerized CO_2, and CO lasers could be extensively used to monitor active media to improve lidar systems, in the spectroscopy and analysis of gases.

We have described the design of a laser head with a sealed tube and separate units (a high-voltage power supply, unit for tuning the lasing wavelength, an AFT unit, and a modulator) of an actively stabilized a cw CO_2 (CO laser) that can he automatically switched between generation lines. The laser is switched from CO_2 to CO operation by replacing the discharge tube.

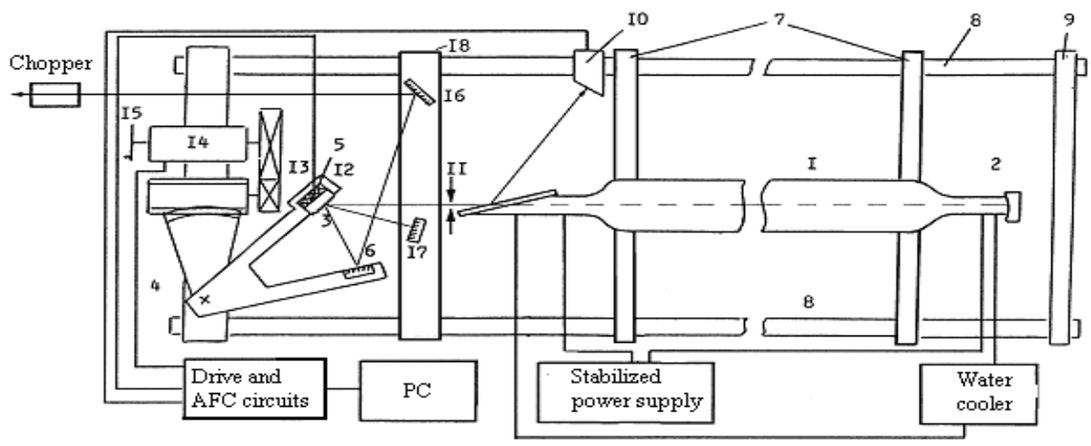
Further improvements include an electro-mechanical drive for the diffraction grating, an electronic control unit compatible with various computers, interfaces, and a control algorithm to link the laser to the computer.

6.1 Laser structure

The laser structure is shown in Fig. 7. The GL 501 (CO_2) or GL-509 (CO) (FSUE RPC "Istok", Fryasino, Russia) commercial discharge tubes 1 are used because they have similar

structures and the same discharge distance – 1.2 m. The laser cavity is formed by a 100% end mirror *2* with a curvature radius of ~3 m that is integrated in the tube and a flat diffraction grating *3* with 100 lines/mm set on a rotating arm 4 and a PZT drive *5*. The grating reflects in the first diffraction order when operating with CO_2 and in the second order with CO (the reflectivity is about 90% in both cases). There is an additional mirror 6 on the rotating arm which forms a corner reflector (Gorobets et al., 1992) to keep the direction of the output beam unchanged when the laser is switched from line to line. This optic layout is particularly suited to the laser tubes we were employing. Besides, when the output beam has zero-order reflection at the grating, the optic losses are much lower, and more laser lines can he generated, which is essential for CO tubes whose gains are relatively small.

The diffraction-grating rotating arm and the tube braces 7 are fixed on three invar rods *8*. The end plate *9* is rigidly fixed to the laser frame, and unit *4* is attached to the lower rod via two bearings. As a result, thermal expansion in the rods does not lead to any misalignment of the grating. The use of invar rods and the high rigidity of the structure leads to a good passive stability of the cavity length.



1 – laser tube; 2 – end mirror; 3 - diffraction grating; 4 – rotating arm stage; 5 – piezo-electric drive; 6 – turning mirror; 7 – laser tube braces; 8 – invar rods; 9 – end plate; 10 – pyro-electric detector; 11 – iris diaphragm; 12 – holder of the diffraction grating; 13 – gear-box with a worm wheel; 14 – stepper motor; 15 – graduated wheel; 16 – turning mirror

Fig. 7. Diagram of the laser structure

The radiation frequency is stabilized by coupling the output power to the wavelength and an appropriate curve is used in the stabilization system. The laser is in fact stabilized by automatically tuning the cavity length with the PZT drive *5* to which the diffraction grating *3* is fixed. The AFC circuit is similar to that of the Edinburgh instruments lasers. The signal for feedback loop is taken from a pyro-electric detector 10 which is exposed to the radiation reflected from the GaAs Brewster window of the discharge tube (see Fig. 7). The laser only generates the fundamental transverse mode because of the iris diaphragm 11 and the AFC system.

The key element in the laser is the system for tuning the lasing wavelength. Structurally, it is the rotating arm *4* of the diffraction grating and driven by the electronics driving the AFC system. The laser is switched between the lines by turning the grating with respect to the cavity axis.

The rotating arm *4* comprises the grating holder *12* which aligns the grating in vertical and horizontal planes, a two-stage reduction gear-box *13* with a worm-wheel (the transfer ratio is 1/1620), and a small stepper motor *14*. The precise reduction box, which has split gears, can rotate the grating and the corner mirror 6 through 40° and can set the grating angle to within 10″. The laser spectrum can be also tuned manually with a calibrated wheel 15. When active, the control unit feeds pulses to the stepper motor, and the counter displays the number of motor steps on a front panel. The turning rate can be set to between 50—500 step/sec (up to 5 lines/sec). It can be also made to go in single steps. The motion of the arm is limited in both directions by limit switches.

An initial-state indicator with a low-voltage spark discharger acts as one limit switch and was designed to set the rotation arm in the starting position. At a constant voltage of several volts, the spark discharge in air occurs at a very small gap width (~1.5 μm) and, hence, the discharger generates a signal. The uncertainty the gap width at which the discharger generates a signal is less than 1 μm, with corresponds to a grating turn of ±4″. Thus the reproducibi1ity and accuracy of the initial position is good and corresponds to one step of the system. Note that an error of one step is not significant when switching the laser to a particular line since the distance between neighboring lines in CO_2 and CO lasers in terms of the grating turning angle is ~ 240″, which is equivalent to 60 steps of the motor.

The laser tuning system can be linked to computers through appropriate interfaces and software. The turning angles of the diffraction grating with respect to the cavity axis corresponding to each laser line are leaded into the computer. The turning angles in terms of motor steps are derived from the grating pitch. When the laser is operating, the calculated angles may differ from the real values by some constant. This difference may be due to composition variations of the gas in the tube as a result of the electric discharge. Even so the intervals between neighboring spectral lines, and hence the distance between the grating positions in terms of the turning angle remain unchanged. The correction to the calculated positions should been found experimentally for all lines in terms of motor steps. This experimental correction also takes into account the uncertainty of the position of the zero-angle discharger.

The correction can be determined in two ways using the laser tuning software. The first method is semiautomatic and requires an external spectral device (a monochromator or a gas cell with a known absorption spectrum, e.g., NH_3). The position of the grating for the selected reference line is determined, and the difference between this value and the calculated posit ion is fed into computer as the correction. In the second method, the correction is determined automatically by finding a reference line without an external spectral reference (Gorobets et al., 1992). The correction is determined using bright lines which can be easily identified in the output spectrum. In the case of the CO_2 laser a good line is P(56) in the 00^01-10^00 band, which coincides with the P(23) line of the 01^11-11^10 band. The algorithm for finding this line was described in (Gorobets et al., 1992) and it reliably determines the correction. A similar method is possible for the CO laser.

Once the experimental correction is found, the laser is tuned to the selected line. The correction is added to the angle corresponding to the selected line, and the computer moves the grating to the correct angle with respect to the previous position, then it activates the AFC system.

The temperature of the liquid cooling the laser tube should be kept constant. This is particularly important for the CO laser since the number of lasing lines, especially in the

short-wave band, depends on the gas temperature in the tube (Aleinikov and Masychev, 1990). We used a standard water cooler with a closed cycle to remove the heat from the laser tube. It cools the tube with distilled water at a temperature between 2 and 10 °C and keeps it constant to within 1 °C.

The stabilized power unit is standard for CO_2 lasers and an additional current stabilizer built around a vacuum tube. The current stabilizer suppresses current oscillations by several orders of magnitude, especially those at the mains frequency of 50 Hz. The current through the tube can be tuned between 10 and 40 mA.

To modulate the laser power, we used a electromechanical chopper. It was built around a electric motor. A thin precisely made disk with sixteen slits made from titanium foil 0.1 mm thick was mounted on the motor axis. The signal for the feedback of the active frequency control was taken from an optic couple. The electronics drive the modulator at 125, 250, 500, and 1000 Hz, and it can be detuned by ±5.9% from these frequencies. With a crystal oscillator and automatic control of the modulation frequency its very stable (the frequency usually differs from the preset value by less than 0.01%).

6.2 Output laser parameters

The laser characteristics have been measured on an optic bench using traditional techniques (Gorobets et al., 1995). We first consider the spectral and energy parameters.

Since the diffraction grating has a reflectivity of 90% the laser with a CO_2 tube, generates about 90 lines between 9.15 and 10. 95 μm (00^01-10^00, 02°0 bands) The output power in the fundamental mode reaches 10 W for strong lines and is over 1 W side lines. Using the same grating, the laser generates 25 lines in the P-branch of he hot 01^11–11^10 band of the CO_2 molecule. In this case the spectrum is shifted to the red to 10.94 –11.25 μm. The output power at strong lines was 3-4 W and 0.5 W at the band edge. However the conditions needed for the hot baud, where the gain is considerably smaller, are not optimum. When the laser tube is filled with CO_2, N_2, He and Xe, output was considerably higher and, the number of lines was larger (see 1-3 parts).

When the CL-509 tube is inserted, the laser efficiently generates about 100 vibration-rotation lines of the CO molecule between 5.28 and 6.43 μm. Output powers at the strongest lines were ~1 W in the fundamental mode at the optimum discharge current. The lines were identified using the data in (Aleinikov and Masychev, 1990).

Note that the parameters of the grating are better when generating CO and hot of CO_2 lines, where the gain is smaller than in the more conventional 00^01-10^00,02^00 bands. The lines in the conventional bands will clearly be stronger in a cavity with a lower Q factor.

6.3 Instability of the output laser power

The long-term instability of the output power was checked using a laser calorimeter whose signal was fed to a chart-recorder. Figure 8 shows a typical plot of the laser output power over one hour. The instability in the laser output power on the P(16) line of the 00°1-02°0 band over one hour was ±1.1%. Similar measurements with other lines of CO and CO_2 molecules demonstrated that the long-term instability of the laser power is less than ±1.25%. However the time constant of the calorimeter is long and the short-term instability could not

be monitored. The instability over the measurement time needs to be known in many applications. For example, when determining contaminants by the differential method (at the absorption line and off the line), the measurement may last from several seconds to a minute. The output power instability in this time interval was measured as follows.

The laser beam was modulated by the chapter and fed to a light detector cooled by liquid nitrogen (Gorobets et al., 1995). The detector's electric signal was processed by a lock-in amplifier, digitized and sent to a computer. One measurement, including the signal processing in the ADC took about 0.7 s. Measurements lasting over 70 s demonstrated that the output power of the laser generating at the P(24) line of the $00^{0}1\text{-}10^{0}0$ band of the CO_2 molecule varied by ±1.6% around the mean with the AFC system on and by 12.5% with the AFC system off. The larger instability measured by the second method may be due to longer time constant of the calorimeter. The short-term power instability was also measured using an digital oscilloscope (band-width of about 1MHz) connected to the Ge:Au detector. The instability over times of the order of microseconds was estimated to be several times smaller than the long-term instability quoted above. Several lasers have been used to monitor the atmosphere and to obtain spectroscopic measurements over a long time. They have proven to the reliable devices with a long service life.

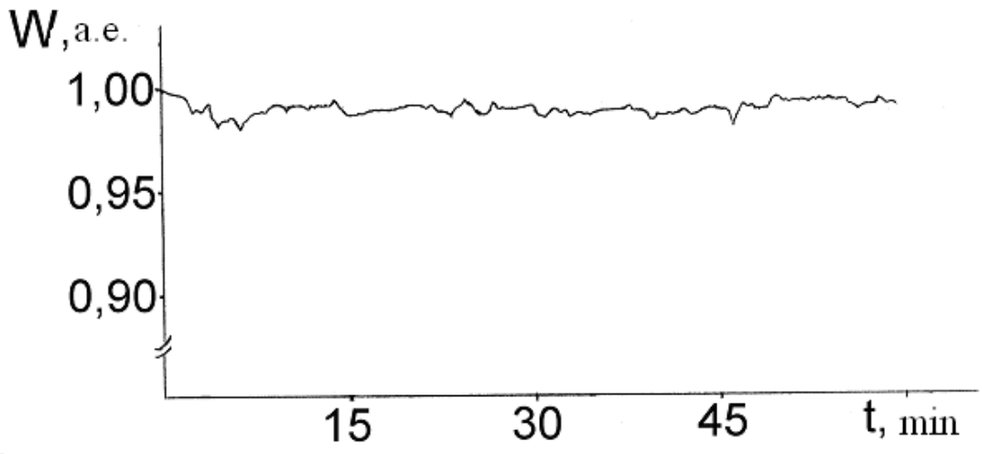


Fig. 8. Trace of the output power over 1 h at the P(16) line of the CO_2 molecule's $00^{0}1\text{-}02^{0}0$ band

7. Detection of small N_2O concentrations using a frequency doubling $^{12}C^{18}O_2$ laser

The destruction of the protective ozone layer of the Earth (so called "ozone holes") can result in a global environmental and climatic catastrophe showing for many years a continuous unflagging. It is well known that the products of human activity such as freons and nitrous oxide (N_2O) are responsible for "ozone holes". Freons appear as a result of manufacturing some kinds of plastics and using refrigerators. Nitrous oxides penetrate into the atmosphere primarily due to microbiological changes in soil caused by agricultural human activity. Moreover, (Crutzen, 1996) determined that there is a direct coupling between the life of microorganisms in soil and the ozone layer thickness.

The conservation of the ozone envelope depends on many factors. However it is beyond doubt that modern reliable techniques monitoring the atmosphere for the presence of freons and nitrous oxide would assist greatly in a solution of this serious global problem. Laser gas-analysis methods are well suited to this task. They are capable of working with high speed, i.e. practically in real time mode. The ability to determine extremely low gas concentrations (for laser photoacoustics on the level of 0.1 ppb) and to cover extensive areas of the earth from a single point of observation (for lidars – about 10 km) give them unquestionable advantages as compared to other known diagnostic methods. There is reliable and effective laser procedure based on CO_2 laser for the detection of prevailing freons, the strong absorption bands of which overlap with emission lines of the laser.

Spectral analysis of N_2O has shown that the characteristic feature of this molecule consists of the absorption now low in the ranges where known effective lasers can operate. There are only the complex and (or) inefficient multitasked systems with nonlinear frequency conversation (generation of harmonics with the subsequent frequency summation), parametric oscillators, and tunable diode lasers. Therefore, the development of reliable and efficient laser methods for N_2O sensing remains a topical problem. An additional difficulty arising with the development of such methods applies to the necessity to detect low concentrations of nitrous oxide (background content of this gas in the atmosphere is 0.2—0.4 ppm).

The main goal of the present investigation is the development of a reliable and high-efficiency laser method for detecting low concentrations of N_2O. The other goal of the work is the test of this method as a remote gas analyzer. The procedure is based on the idea of using a nonlinear frequency-doubled CO_2 laser operating on the isotopic carbon dioxide modification $^{12}C^{18}O_2$. Such a powerful laser system would emit neighboring lines both coinciding well and adjacent to N_2O absorption lines. This fact allows one to apply the highly accurate technique which uses corresponding on/off line pairs for the differential absorption. The high efficiency of the system and strong absorption of N_2O molecules (we use the strongest band in the range of $\lambda \sim 4.5$ μm) would give a possibility to measure low gas concentrations both in short and long (~10 km) measurement paths.

7.1 Active medium optimization of the $^{12}C^{18}O_2$ laser

It is known that the use isotopically substituted carbon dioxide molecules make it possible to increase substantially the number of lines and to extend the spectral range of CO_2 lasers. That is important for different applications, in particular for atmospheric gas detection. The use of $^{12}C^{18}O_2$ as molecules of the active medium of lasers is of special interest, since for this molecule the maximum gain lies at wavelength ~9.4 μm and not ~10.6 μm, as for $^{12}C^{16}O_2$, and, consequently. there is a possibility for efficient lasing in a shorter wavelength range down to 8.9 μm. The doubled emission frequencies of $^{12}C^{18}O_2$ laser well coincide with absorption lines of many molecules including nitrous oxide.

However, for a number of reasons, and particularly because of the much higher price, CO_2-lasers based on isotopically substituted carbon dioxide molecules are not in wide use. For $^{12}C^{18}O_2$, molecules there is also the problem of the isotoporeplacement of $^{18}O_2$ with $^{16}O_2$ as these molecules are active in discharge plasma. The electrode surface, discharge chamber and tubes walls accumulate with time ordinary oxygen $^{16}O_2$. Then, under the discharge conditions, $^{16}O_2$ replaces (isotopically) $^{18}O_2$, in the active medium. This results in rapid degradation of the $^{12}C^{18}O_2$ active medium. This fact is especially important for TEA CO_2-

lasers the discharge chambers of which are much more difficult to pump out as compared to low-pressure sealed-off lasers. Another problem is the necessity of proper choice of materials which accumulate less ordinary oxygen. Therefore, we have performed detailed investigations aimed at active medium optimization for high-energy parameters with a simultaneous decrease in the price of the active medium based on isotopically substituted molecules $^{12}C^{18}O_2$ at the expense of its dilution with inexpensive carbon dioxide $^{12}C^{16}O_2$.

This section gives the results of our spectral investigations of the gain and the lasing for the TEA CO_2 laser operating both on $^{12}C^{18}O_2$ and, just for comparison, on ordinary $^{12}C^{16}O_2$. The analysis of the conditions required for efficient lasing in the range of 9 μm is given too.

Experiments were performed with a UV-preionized TEA module specialty developed for lidar systems (Gorobets et al., 1995). The module had a working volume of 70 × 2.5 × 2 cm. The distance between electrodes was 2 cm. Both main and auxiliary discharges were fed from low-inductance capacitors having a total capacity of 0.2 μF charged up to the 25 kV voltage. The discharge duration was ~500 ns.

The isotopically substituted form of carbon dioxide $^{12}C^{18}O_2$ with an ^{18}O enrichment factor of 80% obtained as a gas mixture containing 4% $^{12}C^{16}O_2$, 32% $^{12}C^{16}O^{18}O$, and 64% $^{12}C^{18}0_2$ was used in the experiments. It is much more expensive to prepare a mixture with a higher factor of enrichment for $^{12}C^{18}O_2$. The first measurements and calculations concerned the gain. For example, Fig. 9 shows respective gain for some lines of the P-branch of 00°1-10°0 band of the $^{12}C^{18}O_2$ molecule (λ~9.4 μm) and of the $^{12}C^{16}0_2$ molecule (λ~10.6 μm) for the mixture $^{12}C^{16}O_2$: $^{12}C^{16}O^{18}O$: $^{12}C^{18}O_2$: N_2 : He = 10:3:7:20:60 (the manufactured mixture was diluted with $^{12}C^{16}O_2$) with a total pressure of 500 Torr. Measurements carried out at t = 4 μs after the start of the discharge when the highest gains were realized. Gain measurements were performed by probing the active medium with a cw laser, lasing on the corresponding lines.

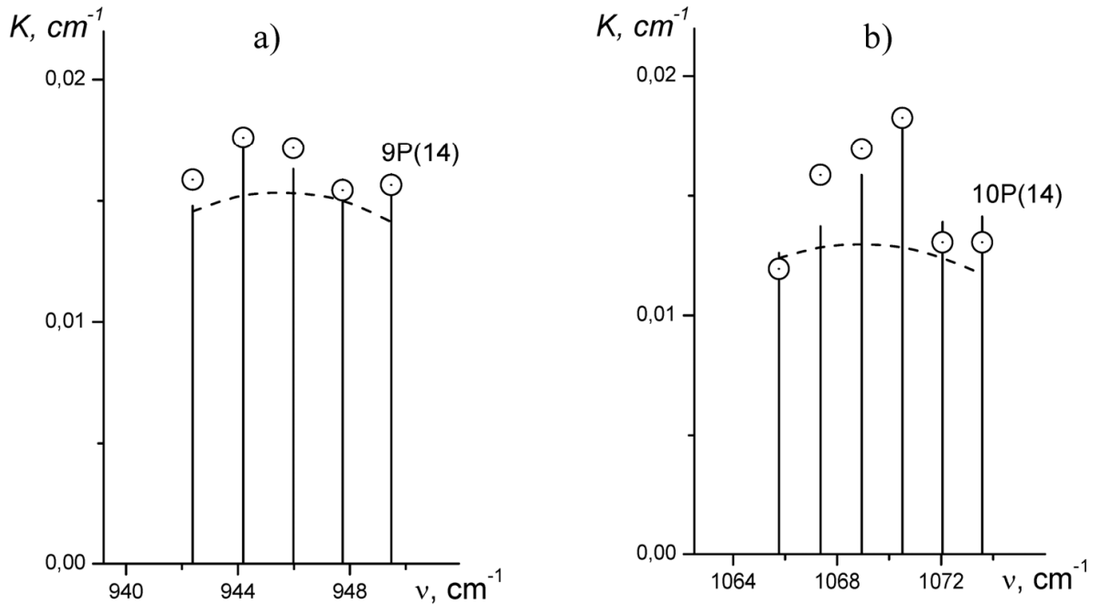


Fig. 9. Measured (O) and calculated (|) values of the gain at the lines of the 10P($^{12}C^{16}O_2$) (a) and 9P($^{12}C^{18}O_2$) (b). Dashed lines – the gain calculated without lines overlapping.

After passing the medium in question, the probing emission came to a photodetector producing a signal to a digital oscilloscope, connected to a personal computer where the measurements were stored and averaged. The calculations were performed based on a vibrational temperature model (Petukhov et. al.., 1985) that is in wide use for simulations of the active medium of CO_2, lasers. The measurements have shown that under the above conditions: T= 350 ± 10 K, $T_1 = T_2$ = 420±20 K, and T_3=1475, 1510 and 1550, ±15 for molecules $^{12}C^{16}O_2$, $^{12}C^{16}O^{18}O$, and $^{12}C^{18}O_2$, accordingly. T designates the gas temperature. T_1 and T_2 are the temperatures of symmetric and deformation vibrations that are virtually the same for the whole scope of carbon dioxide variations in question. T_3 is the temperature of asymmetric vibrations. The technique used and apparatus to measure gains and to determine vibrational temperatures were described early. The gain spectrum was calculated using the scheme described in (Gordiets et al., 1980). Additionally the overlap of individual vibration— rotation lines in bands $00^01\text{-}10^00$, $00^01—02^00$, $00^02\text{-}10^01$, $00^02\text{-}02^01$, $01^11\text{-}11^10$, $01^11\text{-}03^10$ for molecules $^{12}C^{16}O_2$ and $^{12}C^{18}O_2$ and bands $00^01\text{-}10^00$, $00^01—02^00$ for molecule $^{12}C^{16}O^{18}O$ and spectroscopic data given in (Witteman,.1987) were allowed for.

As is evident in Fig. 9, the gain maximum in the P-branch of the band $00^01\text{-}10^00$ of the $^{12}C^{16}O_2$ molecule in the mixture of isotopic forms of carbon dioxide $^{12}C^{16}O_2$, $^{12}C^{16}O^{18}O$ and $^{12}C^{18}O_2$, occurred at lines P(18) and P(20). These lines are especially affected by the overlap with lines from other bands. The P(18) line of the 00°1-10°0 band for the $^{12}C^{18}O_2$, molecule is shifted from lines P(3) 00°1-02°0 $^{12}C^{16}O^{18}O$, R(8) 00°1-02°0 $^{12}C^{16}O_2$ and P(26) $01^11\text{-}11^10$ $^{12}C^{18}O_2$, at 0.046, 0.045 and 0.021 cm^{-1}, and line P(20) $00^01\text{-}10^00$ $^{12}C^{18}O_2$-from lines P(5) 00°1-02°0 $^{12}C^{16}O^{18}O$, R(6) 00°1-02°0 $^{12}C^{16}O_2$, and P(28) $01^11\text{-}11^10$ $^{12}C^{18}O_2$ at 0.007, 0.072, 0.085 cm^{-1}, respectively. The homogeneous width of the line at half-height under our conditions is ~0.05 cm^{-1}. We note that the calculation and experimental data are in a good agreement. For instance, the gain calculated for line P(18) $00^01\text{-}10^00$ $^{12}C^{18}O_2$ is 1.83×10^{-2} cm^{-1} while the measured value is $(1.81 \pm 0.06) \times 10^{-2}$ cm^{-1}

It's also worth noting that higher gains are realized for the $^{12}C^{18}O_2$ molecule than for the $^{12}C^{16}O_2$ molecule. As seen from Fig. 9, the gains on the strongest lines of the P-branches for both isotopes are approximately equal, though this mixture contains 1.8 times less $^{12}C^{18}O_2$ than $^{12}C^{16}O_2$. There are three reasons for this. First, the limiting gain for the lines of the P-branch of the band $00^01\text{-}10^00$ of $^{12}C^{18}O_2$ is approximately 1.5 times higher than that for the P-branch of the band $00^01\text{-}10^00$ of $^{12}C^{16}O_2$ (Witteman,.1987); second, for the lines of the P-branch of this band of the $^{12}C^{18}O_2$ molecule in the $^{12}C^{16}O_2$+$^{12}C^{16}O^{18}O$ mixture the effect of the overlap with other bands is essential; and, finally, under the conditions of dynamic equilibrium between the v_3 modes of both isotopic forms the temperature T_3 for $^{12}C^{18}O_2$, is higher than for $^{12}C^{16}O_2$.

We also measured the effect of the dilution of molecules $^{12}C^{18}O_2$ by the usual form of carbon dioxide $^{12}C^{16}O_2$, on the output energy. The lasing pulse energies measured in the ranges of 9.4 μm ($^{12}C^{18}O_2$) (1) and 10.4 μm ($^{12}C^{16}O_2$) (2) are given in Fig. 10. These data were acquired for the TEA-module described above using a non-selective cavity composed of a non-transmitting and germanium output (R=~50%) mirrors. The energies emitted in the ranges 10.4 and 9.4 μm are approximately equal to each other at the 30 Torr content of $^{12}C^{18}O_2$, approaching to the content of $^{12}C^{18}O_2$ at which the corresponding gains get equal. Also note that the total emission energy ($E_{9.4}$ + $E_{10.6}$) is independent in this case of isotoporeplacement of carbon dioxide and reaches the value of 5 J.

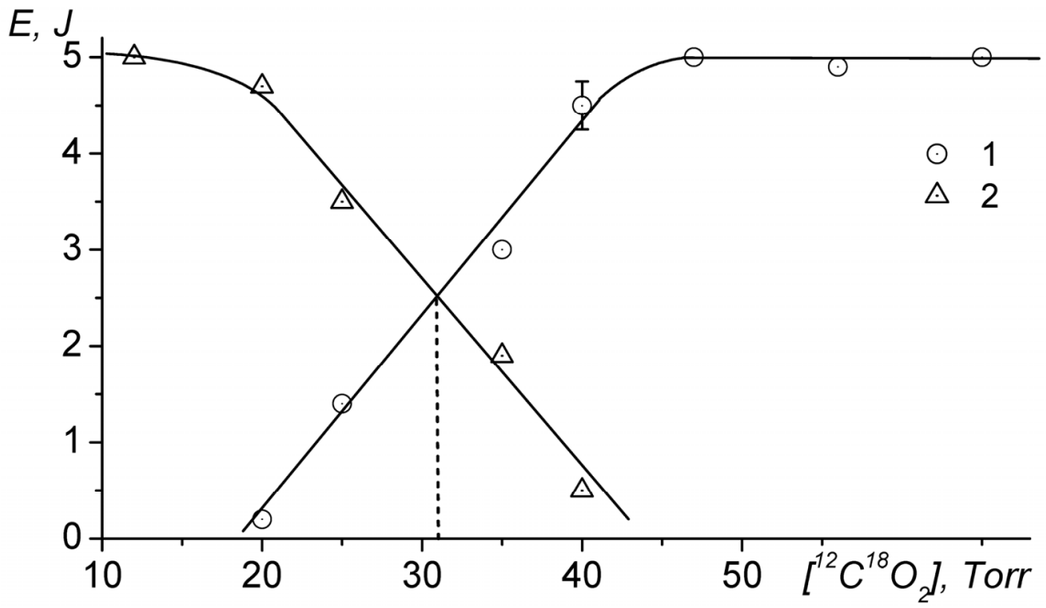


Fig. 10. Lasing energy measured in the ranges of 9.4 μm for $^{12}C^{18}O_2$ (O) and 10.4 μm for $^{12}C^{16}O_2$ (Δ) versus $^{12}C^{18}O_2$ content

Thus, the results of the investigations demonstrate that efficient lasing of the CO_2 laser even with non-selective cavity is possible in the range of 9.4 μm when the $^{12}C^{18}O_2$, content is 30% of total carbon dioxide amount. When the laser operates with selective cavity such as that based on a diffraction grating, the percentage of $^{12}C^{18}O_2$, can go down to ~20%. This fact is key for reduction of the price of the active medium.

An experimental series performed as well by us was oriented toward providing long-duration autonomous laser operation without active mixture replacement and noticeable degradation of its composition. To this end, the discharge chamber was well evacuated (no more than 0.2 Torr/day inleakage) and the material it was made of was properly selected. The best results were achieved with a glass-epoxy cylinder when the operation was virtually quasi-sealed-off, i.e. without replacement of the active medium on $^{12}C^{18}O_2$ during 1—2 months (more than 10^5 pulses) without noticeable decrease of the laser energy. In addition, after long operation and before working gases replacement. the old mixture was pumped through liquid nitrogen traps to recover carbon dioxide for repeated use.

7.2 Using a nonlinear crystal etalon for second harmonic generation from CO_2 lasers

The poor efficiency of the frequency conversion attributable on the whole to mid IR lasers can be compensated to a large degree by application of non-traditional optical schemes. Therefore, the problem remaining topical in the mid IR range along with creation of higher-quality crystals applies to development of novel nonlinear conversion schemes and search for the laser operation modes which are optimal for frequency conversion. To this end and with the aim of reaching high conversion efficiency, we have performed some investigations on the basis of which it was possible to realize original optical frequency conversion schemes including intracavity versions.

In this work we used an original high-efficiency optical nonlinear conversion scheme without focusing optics developed by ourselves. An $AgGaSe_2$ nonlinear crystal was acting as an Fabry-Perot etalon. In this case it could be placed in a laser cavity without reflection-reducing coating. However sonic problems connected with the spacing of its Fabry-Perot transmittance bandwidths and laser cavity modes can arise from it. To clarify these problems some calculations have been made (see Fig. 11).

The line shape function in Figs. 11a and 11b were calculated using the Foight and Lorentz expression, correspondingly. The intervals between the longitudinal modes for the lasers were calculated according with the condition $c/2L$, where L is the cavity length and c is the velocity of light. The curves in Fig. 11c are the Fabry-Perot etalon transmittance bandwidths calculated for different temperatures according with Airy's Formula for the etalon made from the $AgGaSe_2$, crystal with the length of 17 mm.

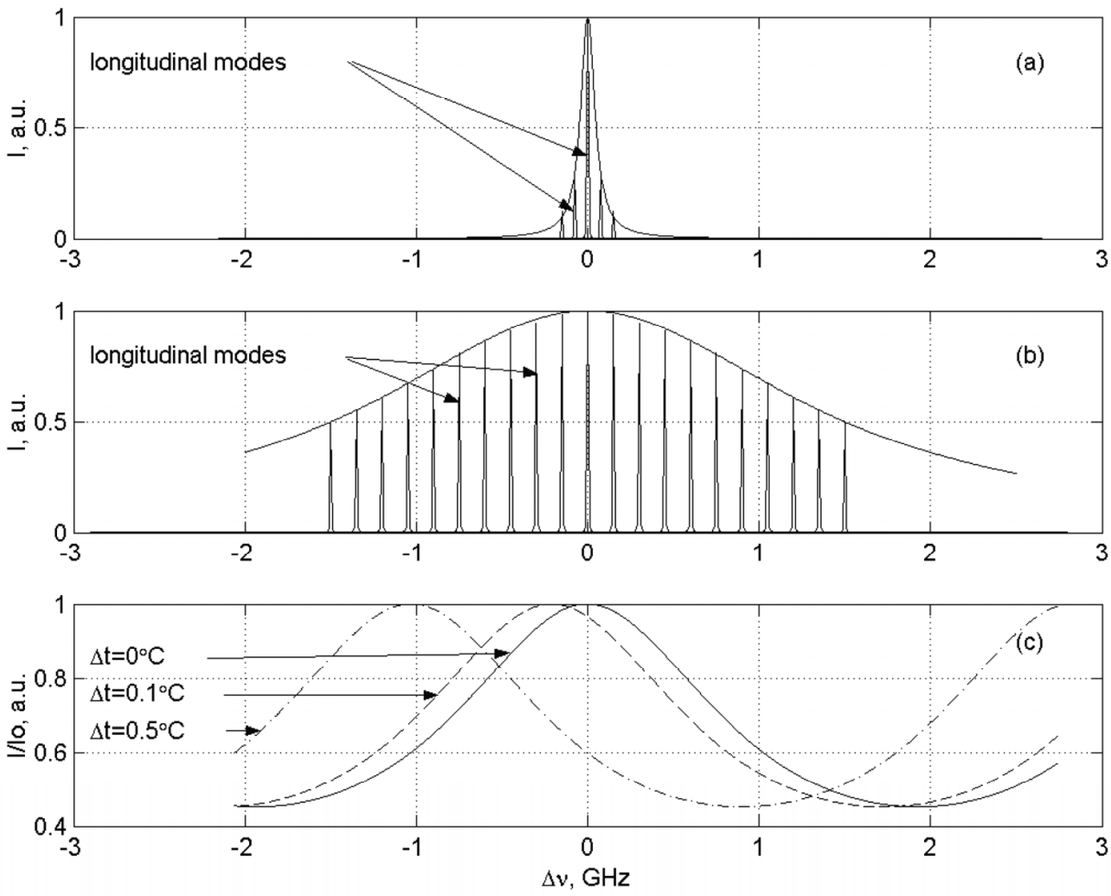


Fig. 11. Line shape function for a low-pressure longitudinal-discharge CO_2 laser (a) and for a TEA CO_2 laser (b) and a transmittance bandwidth of the $AgGaSe_2$ crystal acting as an Fabri-Perot etalon for different temperature variations (c).

In spite of the fact that the pressure broadened line width strongly differs for the TEA CO_2-laser and the low-pressure longitudinal-discharge CO_2 laser. For the first the full width at half maximum of the line shape function is about 3 GHz, and for the second ~0.l GHz. At

the fixed temperature the etalon practically does not influence on oscillations of both lasers, as its bandwidth is much wider than longitudinal modes and comparable to the pressure broadened line width for the TEA laser. Therefore, it is easy to achieve a concurrence between maxima of the line shape function (for TEA and low-pressure longitudinal-discharge lasers) and transmittance bandwidth of the Fabry-Perot etalon by a small angular tuning of the crystal.

However in real conditions a nonlinear crystal has a temperature drift due to the d*(nl)*/d*t* thermal expansion. For the used $AgGaSe_2$ crystal according with the dates of Clevelend Crystals Inc. the factor of linear expansion (α) is $15x10^6$ °C^{-1} and the thermo-optical factors (dn_0/dt and dn_e/dt) are ~$50x10^{-6}$ °C^{-1}. Our calculations (see Fig.11c) demonstrate the rather strong temperature drift of the crystal. It is especially important in the case of crystal operating for a low-pressure longitudinal-discharge CO_2 laser. In our experiments the crystal was supported by a massive metal holder heated by a thermoelement and it was stabilized with accuracy <0.1°C. Besides it should be noted, that in our experiments the crystals having, the very small absorption (~0.01 cm^{-1}) were used. This fact essentially simplifies process of temperature stabilization. Possible changes of the cavity losses because of the crystal temperature drift for a low-pressure longitudinal-discharge laser did not influence strongly on its output from the fact that we used a pulse-periodic regime of the lasing.

7.3 Second harmonic generation from a TEA $^{12}C^{18}O_2$ laser

The optical crystal was used as a nonlinear output mirror of the TEA $^{12}C^{18}O_2$, laser. The cavity of the laser was formed by a 150 line/mm grating and an $AgGaSe_2$ nonlinear crystal. A plate made of LiF was used to select second harmonic emission generated in the nonlinear crystal. A high-quality monocrystal sample made of $AgGaSe_2$, with a 12 x 10-mm section *(L* = 19 mm) was used as a nonlinear output mirror of the laser. The working faces of the crystal were mechanically polished and were not coated. The highly parallel faces (better than 10″) caused the sample to operate as a Fabri-Perot etalon. The angle Θ (phase matching angle) was adjusted near 46°, and φ = 45°. The angle Θ is such that the highest efficiency of the second harmonic oscillation is observed at a normal incidence at line 9P(32) *(*λ = 9.06 μm) and at the neighboring line 9P(34) (λ = 9.05 μm) of the isotopic modifications $^{12}C^{18}O_2$ of carbon dioxide.

The cavity length was 1.1 m. Before the output mirror ($AgGaSe_2$ crystal) there was in the cavity an iris diaphragm (diameter ~8 mm). In the case when the TEA module was filled with the mixture $^{12}C^{16}O_2$: $^{12}C^{16}O^{18}O$: $^{12}C^{18}O_2$: N_2 : He = 104 : 32 : 64 : 200: 600 at a total pressure 500 Torr and a nonlinear crystal was used as an output mirror (~60% reflection), the output energy at lines 9P(32) and 9P(34) of carbon dioxide isotope $^{12}C^{18}O_2$, was ~0.8 J while the peak power was ~4MW. The diameter of the output beam was ~7 mm. The lasing spot had a good spatial distribution. The energy density of the output emission was ~2 J/cm^2 (~10 MW/cm^2).

Under above conditions the second harmonic generation energy *($E_{2\omega}$)* was 52 mJ and the peak power *($P_{2\omega}$)* was ~2 MW. The conversion efficiency reached almost 15%, and at the peak power it was ~50%. The efficiency was calculated by the standard method ($\eta = E_{2\omega} / E_{\omega}$ – *for* the per pulse energy and $\eta = P_{2\omega} / P_{\omega}$ – for peak power). E_{ω}, P_{ω} and $E_{2\omega}$, $P_{2\omega}$ – are energy parameters of the laser emission in the ranges 9 and 4.5 μm, respectively.

7.4 Low-pressure longitudinal-discharge $^{12}C^{18}O_2$ laser with frequency doubling in $AgGaSe_2$ crystal

There are two types of electric-discharge CO_2, lasers which are promising to detect N_2O content along a measurement path. First of them — low-pressure longitudinal-discharge excitation — is more efficient for small and average paths *(L* = 0.1 – 2.0 km). As a rule, it must operate with the laser beam reflection by a so-called corner reflector . The second — TEA — is suitable for long paths (*L* > 2 km) when the lidar operates either using the backscattering signal or pulses reflected by a topographic target]. This section considers the CO_2 laser intended for gas analysis in small and average paths.

The laser the optical system of which is shown in Fig. 12 is automatic tunable and output stabilized as described early. The active element was a sealed-off gas-discharge tube like the industrial GL-50l (see Fig. 7) with the discharge gap of 1.2 m. Our experiments were performed with the $^{12}C^{18}O_2$ isotopic forms of carbon dioxide with the low enrichment factor with respect to $^{18}O_2$ described earlier. It is known that when a gas-discharge tube is fed by a pulsed power supplier, the peak power in optimal regime may go up more than by one order as compared to cw electric pumping. This is of especial importance for lasers used in lidar systems. First, the length of the probing path increases up; secondly, pulse-periodic lasing at an optimal repetition rate (~1 kHz) are suitable for receiving and processing of optical and electrical signals and do not require additional devices for modulation.

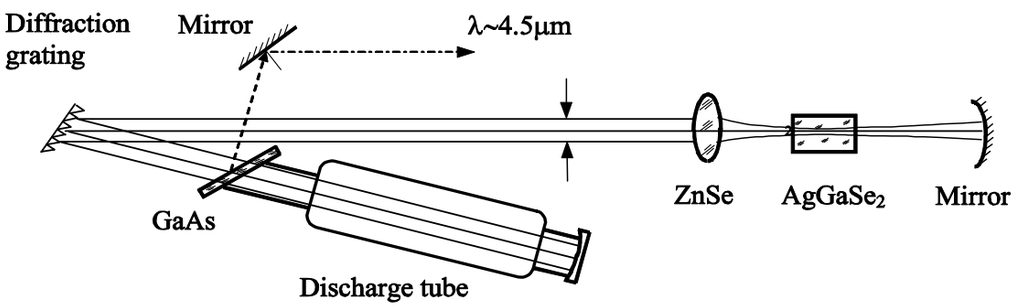


Fig. 12. Optical scheme of the $^{12}C^{18}O_2$ laser with intracavity frequency doubling by non-linear crystal.

Application of the pulse-periodic regime is of especial importance for second harmonic generation in nonlinear crystals. In this case the benefit in the conversion efficiency considerable at the peak power. Our experiments showed the output peak power of the laser to go up to ~l00 W (almost by one order as compared with cw lasing) at lines 9P(32) and 9P(34) of the $^{12}C^{18}O_2$ molecule (at each individual line in single-mode operation) when a pulsed power supply is applied.

It is very difficult for longitudinally excited CO_2 lasers to obtain efficient frequency conversion in nonlinear crystals, as their output power is several orders lower than the peak value attainable in pulsed TEA CO_2-systems. Thus, for the $AgGaSe_2$ crystal, for instance, with a mean length *(l*~ 2 cm) the second harmonic conversion efficiency attained by us with cw discharge was a little more than a tenth of a percent (in case of pulse-periodic discharge it was ~1%), which was a record-breaking value for such laser sources. Therefore, it is even

more attractive for such lasers than for powerful TEA systems to place a nonlinear crystal into the cavity.

In our experiments a monocrystal sample made of $AgGaSe_2$ and having a high optical quality (absorption factor ~0.0l cm^{-1}) was used. The crystal was uncoated and had the rectangular 3.5 x 8.5-mm^2 section. The length of the crystal *(l)* was 17mm. Highly parallel (~.4") working faces provided for a possibility to use the crystal as a Fabri-Perot etalon. The phase-matching angle was ~46°. When the incident pumping emission was normal to the crystal, the highest efficiency of second harmonic generation occurred at lines 9P(32) and 9P(34) which were emitted by the laser.

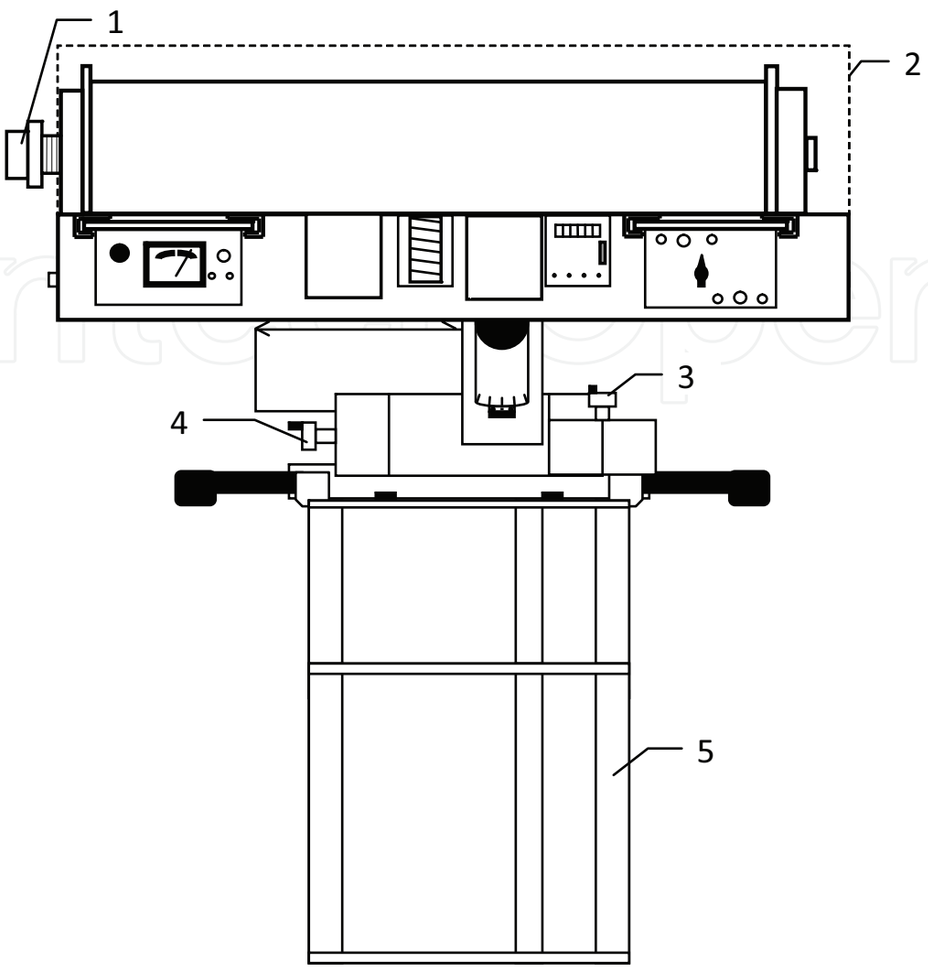
With the optimal radius of the spherical mirror and the focus of the coated lens it was possible to decrease the diameter of the laser beam passing through the crystal by more than one order and, therefore, to increase considerably the pumping density. Along with this we have provided for the pumping beam to be quasi-parallel in the nonlinear crystal. To couple out second harmonic (~75%), we used a Brewster window (GaAs). As the second harmonic polarization was orthogonal to pumping emission, its output was much more than that attainable in a typical conversion system.

A characteristic feature of the proposed system consists of application of the absolutely reflecting (with no out coupling) cavity for pumping emission, which even more increases its intensity. Our experiments showed the highest second harmonic peak power output of the cavity had been equal to 2 W. This is more than one order higher than the analogous parameters reached with the same laser operating with typical optical systems. It is important here to use high quality lens coating with a high damage threshold. This intracavity optical system is simply adjusted and provides with high stable output.

7.5 Laser detection of N_2O

The lidar apparatus complex is shown in Fig. 13. The CO_2 laser was either TEA or low pressure as described previously. All optical elements CO_2 laser, receiving telescope with the objective, photo-detectors, beam-splitting plates, etc. were fixed on a massive metal base to provide good repeatability of the experimental results. The load carrying base of the lidar complex was placed on a construction equipped with mechanisms rotating the system in horizontal (360°) and vertical (45°) directions for immediate and reliable targeting. The target is made visible using an optical sight (12 x 50). Comparatively low dimensions (1.4 x 0.7 x 1.2 m) and mass (~200 kg) give a possibility for development of a mobile version.

A schematic drawing of the receiver/transmitter is given in Fig. 14. The CO_2 laser pulses are output through the beam splitter and are directed into the atmosphere using a transmitting telescope. The emission passed through the atmosphere and trapped by the telescope of the Cassegrain type and with a 250 mm aperture. It consists of two mirrors: front and rear made of sittal with a reflecting aluminum coating protected by corrosion-preventive film. The system uses additionally a ZnSe coated multi-lens objective. The optical configuration of the objective telescope provides for focusing in the focal plane of the detected emission with a 250-mm cross-section down to a diameter ~0.5 mm. After that the detected emission gets on the photodetector which has the sensitive area with a diameter ~1 mm. Optical signals are measured in two channels. Besides the measurement signal, there is a reference one not transmitted through the atmosphere.

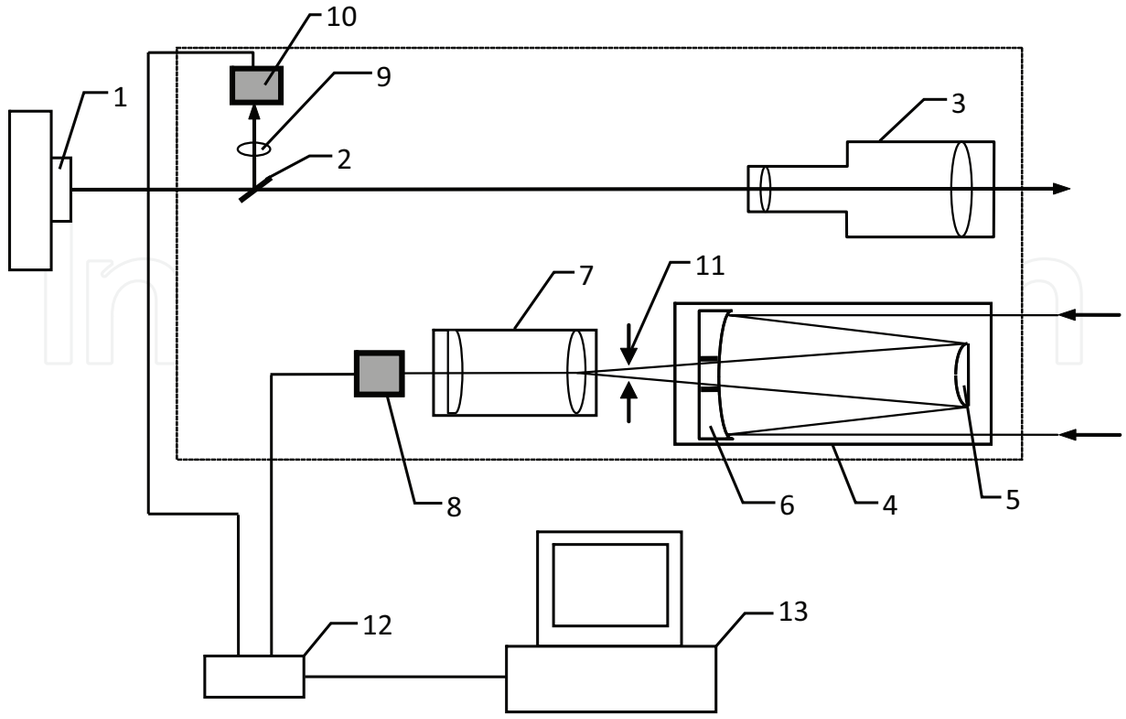


1 — output mirror of CO_2-laser; 2 — CO_2-laser; 3 — horizontal rotation unit; 4 — vertical rotation unit; 5 — metallic base; 6 — receiver/transmitter

Fig. 13. Lidar complex.

The reference signal is produced using a 1-mm-thickness beam-splitter made of BaF_2. Such a thin plate located at an angle ~50° introduces minimum loss for polarized radiation of the TEA CO_2 laser. The loss in the reflection by both faces was no more than 2%. At the same time, only small portion of the emission would be reflected to provide the reference channel. Passing the focusing lens, the reflected portion comes to the sensitive area of the photodetector of the reference channel so organized that this allows automatic account of possible instability of the laser output by normalizing the measurement signal on the reference one, which essentially increases the measurement accuracy and reliability.

As photodetectors nitrogen-cooled photoresistors based on CdHgTe, InSb or germanium doped with gold (Ge:Au) were used. To increase the sensitivity in the measurement channel we used an amplifier. The photodetectors were placed on alignment units. This provided accurate adjustment of the sensitive area of the detector with respect to incident emission. Photodetector signals either arrive to a two-channel ADC or digital oscilloscope and then, via an interface unit, into a computer.



1 — output mirror of TEA CO_2 laser; 2 — BaF_2 beam-splitter; 3 — transmitting telescope; 4 — receiving telescope; 5, 6 — mirrors; 7 — receiving objective (ZnSe); 8 — photodetector (CdHgTe) of measurement channel; 9 — lens (ZnSe) of reference channel; 10 — photodetector (Ge:Au) of reference channel; 11 — iris; 12 — ADC; 13 — personal computer

Fig. 14. Schematic drawing of the receiver/transmitter of the lidar complex.

For monochromatic radiation propagating in homogeneous medium containing several absorbing gases, the transmission T_λ is described by the Buger law :

$$T_\lambda = I_\lambda / I_0 \times exp(-\tau_\lambda) , \quad (6)$$

where I_0 and I_λ, are the intensities of the emission with the wavelength λ before and after its passage through a gas layer with length L, $\tau_\lambda = L\sum_i k_{\lambda i} c_i$ is the optical thickness, $k_{\lambda i}$ and c_i are accordingly the absorption coefficient at the wavelength λ_i and the concentration of the ith absorbing gas.

The analysis of the optical characteristics of the detected gases was performed using the differential absorption technique that is in wide use now for laser atmospheric probing. The probing is made at an on/off pair of laser emission lines. "On" line has the maximally possible resonance absorption, and "off"—minimal. The two-frequency differential absorption technique takes useful information only in the resonance absorption by a gas in question. The effects of such factors as water vapour continuum, non-resonance molecular and aerosol absorption, dust, smog, etc. scattering, atmospheric turbulence will be virtually absent due to the comparatively weak monotonic spectral dependence when "on" and "oil" lines are located near each other.

Prior to measuring N_2O, the lidar complex was tested and calibrated for CO and H_2O measurements using CO_2 lasers both TEA and low pressure operating on the ordinary

$^{12}C^{16}O_2$ with frequency doubling by a nonlinear $AgGaSe_2$ crystal. Measurements of the CO and H_2O concentrations also allowed us to account these gases as background for the N_2O measurements.

The theoretical analysis of the absorption lines of CO and background gases (particularly, H_2O) for a path with 2L = 200 m has shown that it is reasonable to select "on" line among the doubled frequencies of the $^{12}C^{16}O_2$, laser such as 9R(30) (at λ = 4.6099 μm the absorption is 50%/ppm). 9R(18) (λ= 4.6412 μm—45%/ppm) and 9P(24) (λ= 4.7931 μm—37%/ppm). Accordingly. the most suitable "off" line belongs to the same laser and are 9R(28) (λ= 4.6148 μm). 9R(20) (λ = 4.6357 μm) and 9P(26) (λ = 4.8018 μm) at which the absorption by carbon dioxide and background gases is virtually absent.

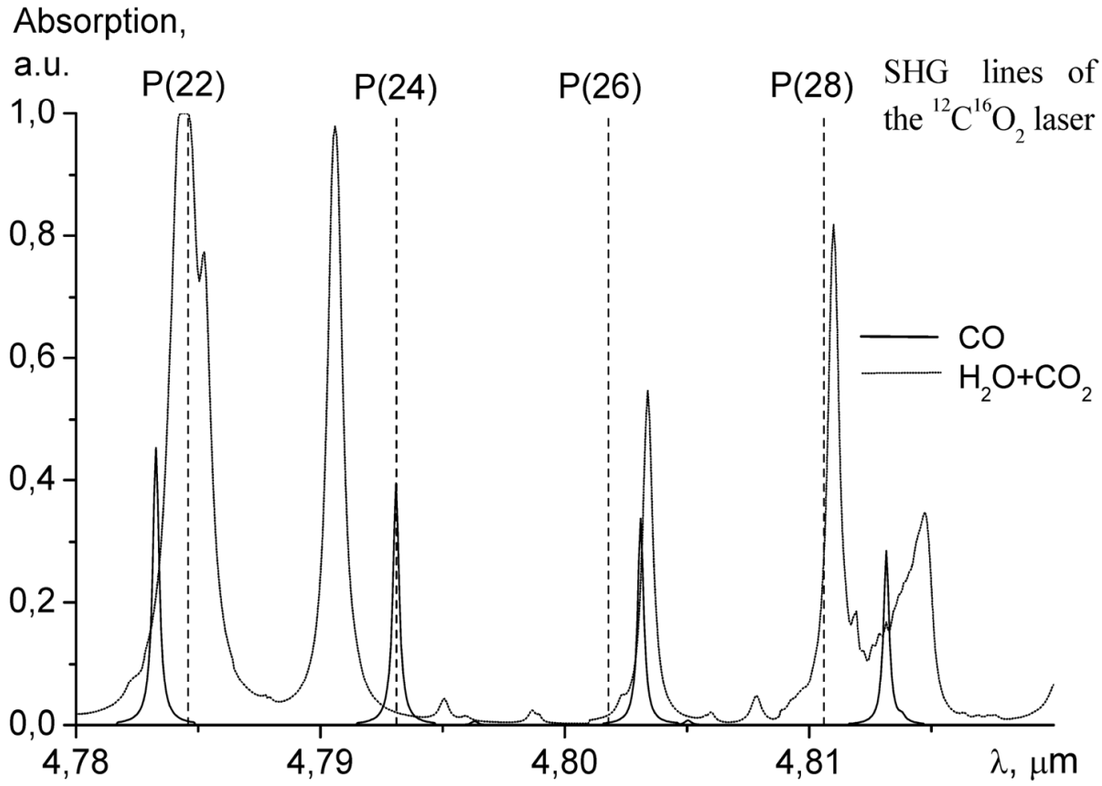


Fig. 15. Absorption spectrum of CO and background gases (H_2O and CO_2) in the range of 4.78 - 4.82 μm. The conditions: path length (2L)=200 m, P=1 atm, T=287 K, gas contents: CO – 1 ppm, H_2O – 10000 ppm, CO_2 – 330 ppm.

Fig. 15 shows a calculated absorption spectrum of the atmospheric gases in the range of our investigations. We select this spectral range due to the following advantages. One of the four selected laser lines (9P(22)— 9P(28)), namely 9P(24) ("on line"), coincides well with the absorption peak of CO. Two of them (9P(22) and 9P(28)) coincides sufficiently well with the absorption lines of H_2O, while 9P(26) demonstrating no absorption of both CO and H_2O is quite suitable as the "off" line. In addition, there is no noticeable absorption by other atmospheric gases (CO_2, for instance) at these lines. Then, carrying out consecutive measurements at these lines, it will be possible to measure concentrations of CO and H_2O.

Then, from the known concentration of H_2O determined independently (for instance, using psychometric devices), it will be possible to calibrate the technique, i.e. obtain the evidence that the results of the laser atmospheric probing are reliable. We used the line 9P(22) that was almost fully absorbed by H_2O as a reference one to check laser tuning at the selected lines. Based on such an original technique we have measured carbon dioxide and water vapour near a highway at a height about 10 m over the cart!, surface. The laser emission was reflected by a plywood sheet painted with a metallic color. The length of probing was 2L = 0.2 km.

The carbon monoxide concentration measured in autumn (5^{00}-6^{00} p.m.) has varied from 0.8 to 1.2 ppm. The measured mean concentration of CO was ~1 ppm. The measurement accuracy determined from H_2O calibrations was ~5%.

N_2O measurements were performed with the same path *(2L* = 0.2 km) using tile low-pressure $^{12}C^{18}O_2$ laser with frequency doubling by a nonlinear crystal. As in the previous case, the emission was reflected by a metallized plywood sheet. Fig. 16 shows a calculated spectrum of the atmospheric gases absorption in the range of 4.5—4.55 μm. We select this spectral range as there are some doubled frequencies of efficient tines of the $^{12}C^{18}O_2$ laser. It is reasonable to select frequency doubled R(32) or R(40) as "on' line, and doubled frequencies of the neighboring R(34) or R(38)—as "off" line. It is important that the indicated lines do not coincide with the absorption lines of background gases H_2O and CO which are always present in the atmosphere. In this way we carried out a number of measurements of N_2O concentration along a researched path at various seasons and times of day. The analysis of the received data has shown that N_2O content in the atmosphere varied considerably, and it is mainly caused by intensity of the transport movement. For example, our experiments performed in autumn in different times during a few days have shown that the N_2O concentration in the path was from 0.35 to 0.5 ppm. The measurement accuracy is estimated to be better than 15%.

We also have measured N_2O for a longer path *(2L* = 1.4 km) using the frequency-doubling TEA $^{12}C^{18}O_2$ laser described earlier. In this case, the laser beam was reflected by a building wall. The averaged content of N_2O was in a good agreement with the value obtained for the shorter path.

The experimental investigations and the calculations carried out have proved conclusively the promising character of the technique developed for the determination of low nitrous oxide concentrations. The technique is based on the use of $^{12}C^{18}O_2$, lasers with effective frequency doubling in nonlinear crystals.

The research carried out has given a reliable technique for laser atmospheric probing of nitrous oxide and effective laser systems to implement this procedure. It is of importance that the path probing is made with a powerful molecular gas laser. Such lasers have narrow emission lines and high stability of spectral and energy output. These characteristics are achieved, as distinct from semiconductor and solid state lasers, naturally without any additional devices. Thus the laser system is simplified and the measurement accuracy increases. The $^{12}C^{18}O_2$ laser system with effective nonlinear frequency-doubling is much promising for global network of lidar stations for atmosphere monitoring.

A reliable procedure or remote high-accuracy laser detection of N_2O as one of the principal destroyers of the protective ozone layer of the Earth has been developed. The procedure is based on using a CO_2 laser system emitting efficiently in the ~4.5 μm range. In this case

lasing from isotopic modification $^{12}C^{18}O_2$ of carbon dioxide with its subsequent frequency doubling by a nonlinear crystal is used. With the object of reducing the price the composition of the active medium (both for TEA laser and low-pressure longitudinal-discharge excitation laser) has been optimized. New high-efficiency intracavity frequency doubling schemes based on nonlinear $AgGaSe_2$ crystals have been developed for CO_2 lasers of both types. Low concentrations of N_2O and concentrations of the principal background gases CO and N_2O have been measured under real atmosphere conditions with the aid of the lidar complex built around these lasers.

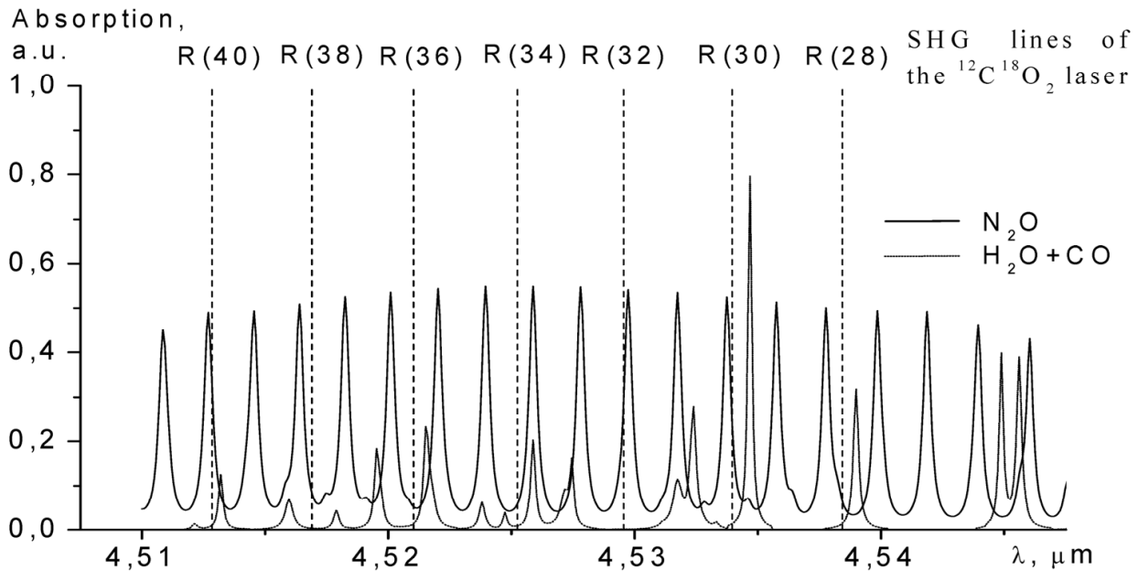


Fig. 16. Absorption spectrum of N_2O and background gases (CO and H_2O). The conditions: path length (2L)=0.2 km, P=1 atm, T=287 K, gas contens: N_2O — 0.4 ppm, H_2O — 10 000 ppm, CO — 1 ppm

8. Conclusion

Optimization of the gas content, pressure, discharge current and the cavity of a low-pressure laser with longitudinal discharge were carried out. Thus, after the above improvements the commercially available sealed-off laser oscillates on more than 30 lines of the P-branch of the 01^11- 11^10 band in the 10.9—11.3 μm range with output power no less than 0.5 W. On strong lines [P(16)—P(26)] output power was ~6W at efficiency ~3% which makes up ~ 40% of analogous laser parameters in the case of oscillation on the lines of regular bands 00^01-10^00 (02^00) under optimum conditions.

The peak power on the strongest lines of the new bands (10^01-20^00(04^00) with a lasing pulse was 30 W. The average output power reached 0.2 W. Lasing was achieved at a number of new transitions. More than 25 new lasing lines with λ = 11.1–11.4 μm, belonging to all the aforementioned bands, were observed in the spectral range studied.

The method of output optimization of cw CO_2 lasers has been developed. The method is based on vibrational and translational temperatures determination by gain measurements on the ro-vibrational lines of regular (00^01-10^00, 02^00) and nonregular (00^02-10^01, 02^01; 01^11-

11^10; (02^21-12^20, 02^01-12^00…) bands of CO_2 molecule. To test the validity of the method, the experiment realization has been done for a low pressure CO_2 laser with cw longitudinal discharge, that can oscillate on the lines of regular and nonregular lines. The good agreement between calculation and experiment data has been observed

We examined what kind of the small gain and the output energy can be attained in the TEA CO_2 laser on the 16(14) μm $02^01(10^01)$-01^11 transitions. On tile basis of the experimentally determined vibrational temperatures T_3 and T_2 we calculated the small gain. The calculations shown that the small gain in the $02^01(10^01)$-01^11 band can attain a significant value (>1 m^{-1}). The necessary conditions for the effective lasing have been determined. It is shown that in optimum conditions the output energy can reach *1.3* J/l at the peak power 5 MW and at the full efficiency of 2 %.

The experimental investigations and the calculations carried out have proved conclusively the promising character of the technique developed for the determination of low nitrous oxide concentrations. The technique is based on the use of $^{12}C^{18}O_2$, lasers with effective frequency doubling in nonlinear crystals. The research carried out has given a reliable technique for laser atmospheric probing of nitrous oxide and effective laser systems to implement this procedure. It is of importance that the path probing is made with a powerful molecular gas laser. Such lasers have narrow emission lines and high stability of spectral and energy output. They were much promising for global network of lidar stations for atmosphere monitoring.

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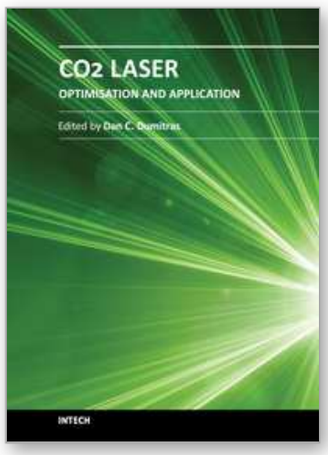

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The present book includes several contributions aiming a deeper understanding of the basic processes in the operation of CO2 lasers (lasing on non-traditional bands, frequency stabilization, photoacoustic spectroscopy) and achievement of new systems (CO2 lasers generating ultrashort pulses or high average power, lasers based on diffusion cooled V-fold geometry, transmission of IR radiation through hollow core microstructured fibers). The second part of the book is dedicated to applications in material processing (heat treatment, welding, synthesis of new materials, micro fluidics) and in medicine (clinical applications, dentistry, non-ablative therapy, acceleration of protons for cancer treatment).

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