Linear and nonlinear absorption of the overtone CO laser radiation in the atmosphere

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Received March 6, 2001

Linear and nonlinear absorption of the overtone CO laser radiation by different atmospheric gases is considered. Emission lines are selected for using in spectroscopic absorption diagnostics of particular molecules. Emission and absorption spectral characteristics of the overtone CO laser are analyzed in comparison with the chemical HF and DF lasers. The information on the 50 most weakly and 50 most strongly absorbed in the atmosphere lines of the overtone CO laser is presented. The absorption is measured at different frequencies of the overtone CO laser in the N_2O/N_2 mixture. It is shown that using the dependence of N_2O absorption coefficient on the foreign gas pressure it is possible to identify spectral lines of the overtone CO laser and determine its frequency. Nonlinear absorption spectra are calculated for high-intensity multifrequency and monochromatic radiation of the overtone CO laser in the atmosphere to demonstrate the feasibility of using this laser in new promising schemes of diagnostics.

Introduction

The laser operated on the first vibrational overtone of the CO molecule[1,2] is an efficient source of coherent IR radiation in the spectral region from 2.5 to 4.2 μm (Refs. 3–11). The experimentally observed and theoretically calculated efficiency of lasing of the pulsed multifrequency first-overtone CO laser reaches 11 and 20%, respectively.[6,9–11] The frequency selective mode of the pulsed first-overtone CO laser was obtained experimentally on more than 400 rotational-vibrational (RV) transitions with the maximum efficiency of 0.6% (Refs. 7, 9, 11). The spectral region of the first-overtone CO laser radiation is very attractive, because it overlaps the atmospheric window of 3 to 5 μm and the absorption bands of numerous natural atmospheric constituents and gaseous pollutants, for example, H_2O, CO_2, O_3, CH_4, N_2O, NO_2, NO, SO_2, HCN, NH_3, H_2CO, HOCl, CH_3Cl, CO, HCl, HF, HBr, HI, OH, C_2H_2, C_2H_4, C_2H_6, C_3H_6, C_3H_8, C_6H_6, C_7H_8, CH_3OH, C_2H_5OH, $C_2H_8N_2$, etc. Thus, the first-overtone CO laser is a promising source for various applications in remote sensing, spectroscopic gas analysis, and medicine. It is highly competitive with the well-known chemical HF (DF) lasers[12,13] and tunable differential frequency generators[14,15] operating in the same spectral region.

This paper considers linear and nonlinear absorption of the first-overtone CO laser radiation by various atmospheric gases. Frequencies are selected for spectroscopic absorption diagnostics of particular gases. The spectral characteristics of the absorption of the first-overtone CO laser radiation and radiation of HF (DF) lasers by atmospheric gases are compared. The practically important data on the 50 most weakly and 50 most strongly absorbed in the atmosphere lines of the overtone CO laser are presented. The spectra of nonlinear absorption of the high-intensity multifrequency radiation of the first-overtone CO laser in the atmosphere are modeled to demonstrate the capabilities of this laser in new promising schemes of laser gas diagnostics.

1. Linear absorption by atmospheric gases in the spectral region of the first-overtone CO laser

The laser operating on the first vibrational overtone of the $^{12}C^{16}O$ molecule has the lasing lines in the region from 2.5 to 4.2 μm corresponding to the RV transitions $(V + 2, J - 1) - (V, J)$, where $V = 4 - 36$, $J = 4 - 25$ (Refs. 5–11).

Table 1 allows one to compare the spectral densities of lines of the first-overtone CO laser and HF and DF lasers. The positions of the lines of the first-overtone CO laser with $V = 4 - 36$, $J = 4 - 25$ (a total of 726 lines; lasing is now obtained at not all of these lines) were calculated with the use of the $^{12}C^{16}O$ spectroscopic constants.[16] The positions of the $H^{19}F$ lines (a total of 58) and $D^{19}F$ lines (a total of 63) were borrowed from Ref. 17. Absorption was calculated with the use of the HITRAN-96 database[18] for the conditions of the standard USA atmosphere of 1976 [model No. 6 (Ref. 19)] at the height $H = 0$ km. The $H^{19}F$ frequencies corresponded to the transitions $(V + 1, J - 1) - (V, J)$ with $V = 0 - 5$, $J = 2 - 15$, and the $D^{19}F$ frequencies – to the transitions $(1, J + 1) - (0, J)$ with $J = 0 - 14$ and $(V + 1, J - 1) - (V, J)$ with $V = 0 - 3$, $J = 1 - 17$ (for every vibrational band we used its own range of the rotational quantum numbers J).

Table 1. Comparison of the peculiarities in the absorption and emission spectra of CO, HF, and DF laser radiation in the region from 2.5 to 4.2 μm

Laser and spectral region, cm^{-1}	N	ρ, lines/cm^{-1}	η_{an}, %	Detectable molecules (as it follows from HITRAN-96)	η_w, %	η_s, %	η_{ss}, %
CO 2364.93–4034.06	726	0.43	19	H_2O, CO_2 , N_2O, CH_4, NO_2, NH_3, OH, HF, HCl, HBr, H_2CO, HOCl, HCN, C_2H_2	41	13	28
HF 2749.49–3693.41	58	0.06	33	N_2O, NO_2, NH_3, OH, HF, HCl, H_2CO, HOCl, HCN, C_2H_2	26	16	42
DF 2750.08–2463.33	63	0.22	17	N_2O, CH_4, HBr, H_2CO	91	–	–

Note. N is the total number of the considered laser lines; ρ is the spectral density (or the number of laser lines per unit spectral interval) in lines/cm^{-1}; η_{an} is the percentage of analytical lines among the considered laser lines; η_w, η_s, and η_{ss} are the percentages of lines weakly, strongly, and very strongly absorbed in the atmosphere among the analytical lines, in %.

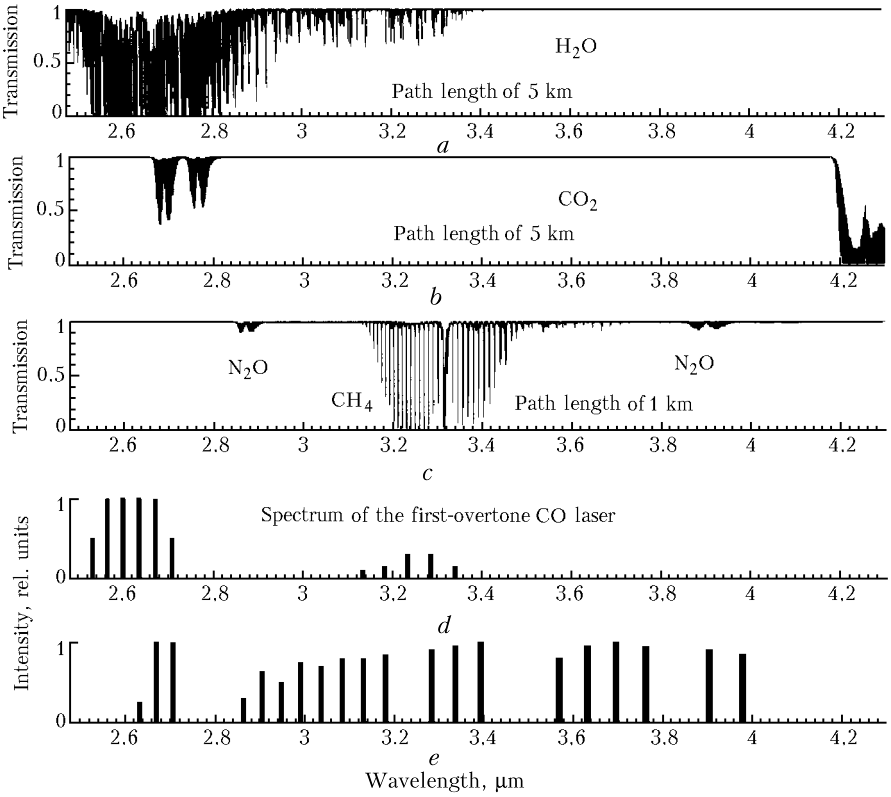


Fig. 1. Linear transmission spectra of water vapor (*a*), carbon dioxide gas (*b*), methane and nitrous oxide (*c*) in the region from 2.5 to 4.3 μm. Experimentally obtained multifrequency spectra of the first-overtone CO laser without rotational structure[5,6,9,10] (*d*, *e*). Standard atmospheric conditions at the height $H = 0$ km: pressure of 1 atm, temperature of 288.2 K; gas concentrations: H_2O – 7750, CO_2 – 330, CH_4 – 1.7, and N_2O – 0.32 ppmV.

Figure 1 shows the spectra of linear transmission of the major atmospheric gaseous absorbers in the region from 2.5 to 4.3 μm. Water vapor plays the main role in absorption of radiation of the first-overtone CO laser radiation in the short-wave spectral region. But the laser radiation is absorbed very weakly at the wavelengths from 3.4 to 4.2 μm, and therefore it is suitable for efficient transfer of laser energy through the atmosphere. It should be noted that the spectrum of CO laser depends strongly on the spectral characteristics of an optical cavity. For this reason, the output spectrum can be controlled in a rather wide region.

1.1. Analysis of diagnostic capabilities of the first-overtone CO laser

For spectroscopic diagnostics, it is necessary for laser frequencies to be in resonance with the strong lines of substances to be detected (such laser frequencies are called analytical). Using the HITRAN-96 database,[18] we sought analytical frequencies of the first-overtone CO laser and HF (DF) lasers for detection of various atmospheric molecules. We used the conditions of the standard USA atmosphere of 1976 [model No. 6 (Refs. 19, 20)] at the height $H = 0$ km: pressure of 1 atm, temperature of 288.2 K. The relative concentrations of the major absorbing gases were taken to be the following: H_2O – 7750 (relative humidity of 46%), CO_2 – 330, CH_4 – 1.7, and N_2O – 0.32 ppmV.

The spectral profile of the absorption lines was assumed to be Lorentzian. All isotopic modifications of the molecules were taken into account. For analytical frequencies, we used rather rigorous selection criterion: (1) almost exact resonance of the laser frequency with a molecular absorption line (within the halfwidth of the absorption line) and (2) contribution of the considered molecular line to the total absorption cross section at the given frequency larger than 10^{-20} cm^2. Atmospheric absorption coefficients at the selected analytical frequencies were calculated as well. For detection of pollutants in the atmosphere, these coefficients must be small. All analytical frequencies were divided into four categories: frequencies with weak (absorption coefficient $\alpha_{air} < 0.1$ km^{-1}), medium ($0.1 < \alpha_{air} < 1$ km^{-1}), strong ($1 < \alpha_{air} < 10$ km^{-1}), and very strong ($\alpha_{air} > 10$ km^{-1}) absorption. The absorption was calculated neglecting the contribution coming from H_2O and CO_2 continuum, as well as from aerosol.

Table 1 summarizes the obtained results. One can see from this table that (1) the spectral region of the first-overtone CO laser is much wider than those of HF and DF lasers; (2) the spectral density of lines of the first-overtone CO laser is higher than those for the HF and DF lasers; (3) the number of analytical frequencies ($N\,\eta_{an}$) of the first-overtone CO laser radiation (138) is markedly larger than the number of analytical frequencies of the emission of HF (19) and DF (11) lasers; (4) the percentage of the first-overtone CO laser lines weakly absorbed in the atmosphere is rather high, but it occupies the intermediate position between the HF and DF lasers (DF laser takes the first place); (5) the total fraction of strongly and very strongly absorbed analytical lines is smaller for the first-overtone CO laser than for the HF laser (DF laser has no such lines); (6) the first-overtone CO laser undoubtedly is in the lead as concerning the variety of detectable gases. Thus, the first-overtone CO laser has considerable capabilities for application to spectroscopic gas analysis, and these capabilities are, on the whole, wider than those of the HF and DF lasers.

For practical use of lasers in atmospheric optics, it is very important to determine the frequencies of lines, whose absorption in air is the weakest or the strongest. The results for 50 such frequencies of the first-overtone CO laser are given on Tables 2 and 3. The conditions of calculations were the same as in Table 1, but the contribution coming from the H_2O and CO_2 continuums was taken into account. It follows from the tabulated data that the contribution of the continuum is quite significant for weakly absorbed laser lines.

Table 2. Fifty most strongly absorbed lines of the first-overtone CO laser

Laser line	ν, cm^{-1}	α_{air} (km^{-1})	
		With continuum	Without continuum
8–6 $P(21)$	3853.96327	10043	10036
9–7 $P(6)$	3870.07913	7503	7497
12–10 $P(25)$	3629.55301	4155	4151
12–10 $P(17)$	3668.72055	3075	3069
38–36 $P(24)$	2369.18124	2924	2924
8–6 $P(23)$	3843.75541	2797	2790
9–7 $P(10)$	3853.59929	2038	2032
9–7 $P(16)$	3826.82768	1405	1397
13–11 $P(8)$	3656.48902	1378	1372
7–5 $P(17)$	3925.17210	1150	1148
14–12 $P(7)$	3609.43154	1125	1121
10–8 $P(14)$	3784.62486	717	710
9–7 $P(15)$	3831.45951	690	683
12–10 $P(21)$	3649.67172	581	575
9–7 $P(8)$	3861.97677	554	547
12–10 $P(12)$	3691.01304	553	547
10–8 $P(11)$	3797.80367	546	539
10–8 $P(20)$	3756.43786	524	517
10–8 $P(21)$	3751.50457	514	508
9–7 $P(5)$	3874.02676	508	502
10–8 $P(23)$	3741.43748	506	499
38–36 $P(25)$	2364.92918	483	483
13–11 $P(17)$	3617.90477	472	467
9–7 $P(17)$	3822.12824	464	456
11–9 $P(23)$	3690.51185	406	399
15–13 $P(5)$	3566.13858	398	396
10–8 $P(10)$	3802.06014	388	381
12–10 $P(7)$	3711.60138	377	370
8–6 $P(24)$	3838.55148	334	327
16–14 $P(12)$	3488.03866	312	312
10–8 $P(24)$	3736.30397	283	277
9–7 $P(20)$	3807.62563	274	267
11–9 $P(8)$	3758.91458	268	262
7–5 $P(21)$	3905.41978	261	258
38–36 $P(23)$	2373.36532	258	258
11–9 $P(16)$	3724.32725	244	237
14–12 $P(11)$	3593.37840	240	238
10–8 $P(18)$	3766.10319	239	232
8–6 $P(17)$	3873.57501	235	229
10–8 $P(5)$	3822.31232	232	224
9–7 $P(21)$	3802.65716	226	219
9–7 $P(7)$	3866.06242	224	218
15–13 $P(15)$	3525.67663	221	221
8–6 $P(13)$	3892.10702	210	205
11–9 $P(10)$	3750.67740	208	202
11–9 $P(11)$	3746.45604	202	195
8–6 $P(10)$	3905.29186	199	196
10–8 $P(9)$	3806.24812	189	182
12–10 $P(13)$	3686.69029	178	171
13–11 $P(10)$	3648.39215	174	168

Note. A total of 726 RV transitions of the $^{12}C^{16}O$ molecule $(V + 2,\ J - 1) \to (V,\ J)$ with $J = 4 - 25$ and $V = 4 - 36$ were considered. Standard USA atmosphere of 1976 [model No. 6 (Ref. 19)] at the height $H = 0$ km.

Table 3. Fifty most weakly absorbed lines of the first-overtone CO laser

Laser line	ν, cm^{-1}	α_{air} (km^{-1})	
		With continuum	Without continuum
31–29 *P*(9)	2760.68731	0.0106	0.0026
34–32 *P*(9)	2616.37580	0.0107	0.0014
31–29 *P*(13)	2746.45306	0.0113	0.0034
33–31 *P*(22)	2615.02977	0.0118	0.0024
31–29 *P*(16)	2735.06054	0.0120	0.0040
31–29 *P*(24)	2701.70687	0.0131	0.0050
30–28 *P*(18)	2775.11382	0.0131	0.0051
34–32 *P*(13)	2602.54833	0.0136	0.0035
28–26 *P*(23)	2849.91696	0.0137	0.0052
29–27 *P*(22)	2806.26756	0.0138	0.0056
34–32 *P*(4)	2632.10366	0.0138	0.0048
32–30 *P*(16)	2687.10766	0.0141	0.0057
33–31 *P*(19)	2627.44142	0.0142	0.0051
32–30 *P*(4)	2728.56865	0.0144	0.0064
32–30 *P*(15)	2690.93954	0.0147	0.0064
33–31 *P*(23)	2610.75792	0.0149	0.0054
33–31 *P*(18)	2631.44358	0.0151	0.0062
32–30 *P*(11)	2705.58525	0.0153	0.0072
33–31 *P*(14)	2646.77331	0.0153	0.0064
33–31 *P*(8)	2667.71626	0.0157	0.0069
32–30 *P*(24)	2654.01934	0.0163	0.0074
33–31 *P*(12)	2654.02896	0.0165	0.0076
31–29 *P*(25)	2697.23596	0.0166	0.0084
34–32 *P*(15)	2595.22263	0.0172	0.0068
33–31 *P*(25)	2602.01305	0.0174	0.0073
29–27 *P*(25)	2792.85529	0.0177	0.0096
33–31 *P*(13)	2650.43535	0.0177	0.0088
32–30 *P*(9)	2712.49707	0.0178	0.0097
27–25 *P*(23)	2898.10094	0.0178	0.0081
34–32 *P*(11)	2609.59976	0.0181	0.0086
29–27 *P*(8)	2860.84429	0.0184	0.0099
34–32 *P*(14)	2598.91970	0.0186	0.0084
32–30 *P*(18)	2679.24032	0.0187	0.0102
29–27 *P*(14)	2839.08263	0.0190	0.0106
34–32 *P*(23)	2563.19871	0.0193	0.0063
33–31 *P*(7)	2670.96560	0.0193	0.0107
34–32 *P*(10)	2613.02227	0.0194	0.0099
34–32 *P*(5)	2629.09721	0.0195	0.0105
31–29 *P*(12)	2750.11431	0.0198	0.0118
33–31 *P*(6)	2674.14565	0.0204	0.0118
27–25 *P*(25)	2888.95738	0.0204	0.0111
33–31 *P*(21)	2619.23439	0.0204	0.0112
29–27 *P*(12)	2846.60978	0.0205	0.0121
29–27 *P*(6)	2867.54936	0.0205	0.0118
34–32 *P*(17)	2587.62375	0.0207	0.0098
34–32 *P*(18)	2583.72221	0.0207	0.0094
32–30 *P*(23)	2658.39071	0.0207	0.0119
35–33 *P*(17)	2539.92998	0.0208	0.0045
32–30 *P*(17)	2683.20788	0.0209	0.0125
34–32 *P*(6)	2626.02105	0.0213	0.0122

Note. A total of 726 RV transitions of the $^{12}C^{16}O$ molecule $(V+2,\ J-1) \to (V,\ J)$ with $J = 4 - 25$ and $V = 4 - 36$ were considered. Standard USA atmosphere of 1976 [model No. 6 (Ref. 19)] at the height $H = 0$ km.

1.2. Absorption of the first-overtone CO laser radiation by nitrous oxide

The high density of spectral lines of the first-overtone CO laser complicates identification of laser RV transitions. Therefore, in the experiments on studying the lasing characteristics of the pulsed first-overtone CO laser, the linear absorption of radiation by the nitrous oxide was measured for the purpose of identification of the laser transitions. The N_2O molecule is an asymmetric triatomic molecule with the well-known spectral characteristics.[18] A 2-m long absorption cell was filled with the mixture of N_2O and N_2. The radiation of the first-overtone CO laser operating in the single-frequency mode was directed onto the cell. The absorption coefficient was determined from the energy ratio of the incident pulse and the pulse passed through the cell with calibration against pure nitrogen and vacuum.

The laser radiation with the wavelength of 3.9 μm is absorbed by nitrous oxide with the excitation of the symmetric vibrational mode 20^00. The central frequency of the CO overtone transition $35 \to 33$ *P*(13) (2554.7228 cm^{-1}) is spaced from the central frequency of the closest strong absorption line of the basic isotopic species $^{14}N_2{}^{16}O$ (*P*10E, $\nu_0 = 2554.6535$ cm^{-1}) by ~0.07 cm^{-1} (Ref. 18). The halfwidth of this line is roughly 0.08 cm^{-1} · atm^{-1}. Therefore, the line broadening with the increasing nitrogen pressure (at constant nitrous oxide partial pressure of 0.01 atm) can cause the increase of the absorption coefficient up to 1 m^{-1} (Fig. 2*a*). This value agrees well with the calculated absorption coefficient.

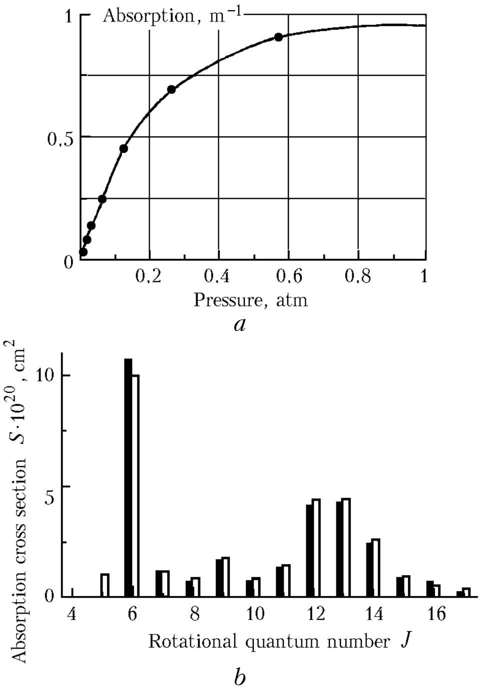


Fig. 2. Absorption coefficient for the first-overtone CO laser radiation at the transition $35 \to 33$ *P*(13) vs. the pressure of the N_2O–N_2 mixture at the constant nitrous oxide partial pressure of 0.01 atm (*a*) and N_2O absorption cross section for the first-overtone CO laser radiation at rotational components of the vibrational transition $35 \to 33$ *P*(*J*) (*b*): experiment ( ) and theory ( ).

The absorption coefficient was measured for eleven rotational components of the vibrational band 35 → 33 (from $J = 6$ to $J = 17$). Based on these measurements, absorption cross sections were determined. They turned out to be in close agreement with the calculated data (Fig. 2*b*). These experiments showed that the frequency of the first-overtone CO laser even at such highly excited transition as 35 → 33 agrees with the calculated value accurate to, at least, 10^{-5}, i.e., within the accuracy about several hundredths reciprocal centimeter.

2. Nonlinear absorption of the first-overtone CO laser radiation

The consistent theoretical model and computer program for modeling of the atmospheric absorption spectra under the exposure to high-intensity IR radiation were described earlier in Refs. 21–24. The model and program consist of two related parts: spectroscopic and kinetic. The spectroscopic part involves calculation of the absorption cross section and the absorption coefficient of the mixture of H_2O, CO_2, N_2, and O_2 with the use of the data on population of vibrational levels of the considered molecules, as well as gas temperature and pressure, which depend on the kinetics of laser excitation and relaxation. In their turn, the data on the absorption cross section for the high-intensity laser radiation serve as input information for the kinetic part.

Nonlinear absorption of the multifrequency radiation of the first-overtone CO laser was calculated for the spectrum of laser radiation shown in Fig. 1*d* assuming conditions of the tropical atmosphere [model No. 1 (Refs. 19 and 20)] at the heights of 0 and 10 km. For $H = 0$ km, the pressure is 1 atm, the temperature is 299.7 K, and the concentrations of H_2O and CO_2 are 25900 and 330 ppmV, respectively. For $H = 10$ km, the pressure is 0.282 atm, the temperature is 237 K, and the H_2O and CO_2 concentrations are 191 and 330 ppmV. Only the basic isotopic modifications of the molecules were taken into account.

Figure 3 illustrates the evolution of the absorption coefficients for individual laser lines of the first-overtone CO laser radiation and shows that absorption changes considerably in the process of laser pumping of the air. For some lines, the absorption coefficient increases, whereas it decreases for the other lines. Analysis has shown that this effect is caused by laser heating of air (in the considered case at the pulse duration ~3 ms, the temperature can reach 548 K). That noticeable heating leads to a marked deformation of the spectrum in the process of laser pulse propagation, as well as to the corresponding change of the refractive index and to thermal blooming of the laser beam.

The results of modeling of nonlinear absorption of the first-overtone CO laser radiation in the atmosphere are shown in Figs. 4 and 5. In calculations, the laser spectrum overlapped the ν_3 band of H_2O. Note that only the high-frequency group of laser lines (3680–3960 cm^{-1}) efficiently interacts with this band. From Fig. 4 it is seen that the laser effect on water vapor manifests itself not only in a decrease of the absorption (laser clearing-up), but also in its increase (laser-induced absorption). Since the nature of absorption variation is purely thermal in this case, it is natural that variations in air absorption are observed not only in the region of pump frequencies, but also in most remote spectral regions. The marked decrease (down to one third) in absorption occurs in the central regions of the ν_2 and ν_3 bands of H_2O (≈ 1600 and 3800 cm^{-1}), as well as in the ν_3 band of CO_2. The significant increase in absorption (up to tens times for weak lines) due to laser heating is observed for many lines of water vapor. The complex changes in absorption occur in the region of 1200–2200 cm^{-1} (decrease and violet shift of absorption in the region of the ν_2 band of H_2O, increase and red shift in the region of the $2\nu_2$–ν_2 band of H_2O). The calculations showed that the laser-induced changes in atmospheric absorption depend strongly on the spectral composition of the radiation and the height. For example, already at the height $H = 10$ km the changes in the absorption spectrum are insignificant (change of the temperature for the same parameters of radiation as in Fig. 4 is insignificant because of low absorption at the given height).

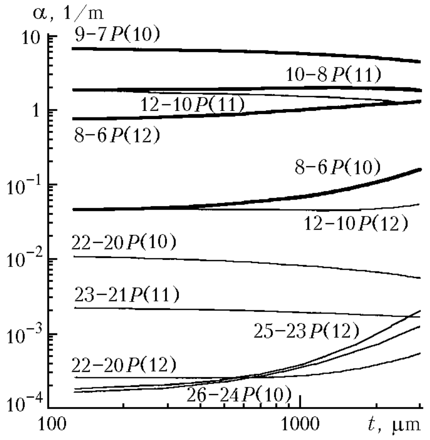


Fig. 3. Evolution of the absorption coefficients in the atmosphere for individual rotational-vibrational lines of the first-overtone CO laser. The radiation intensity is 10 kW/cm^2 for the typical laser spectrum (see Fig. 1*d*). The conditions of the tropical atmosphere at the height $H = 0$ km. Continuum absorption is neglected. The bold curves present the case of laser lines of the first-overtone CO laser having high relative intensity in the lasing spectrum.

The joint effect of laser heating and saturation of absorption manifests itself at short pulse duration and high intensity of radiation. The high peak intensity in combination with the short duration of a laser pulse can be obtained in a Q-switched mode. Figure 5 shows the height dependence of the absorption coefficient in the atmosphere for two frequencies. Laser heating due to absorption of the pump frequency decreases with height (at the height $H = 0$ km the temperature achieves 490 K, and heating becomes insignificant already at $H > 7$ km). As to the effect of absorption

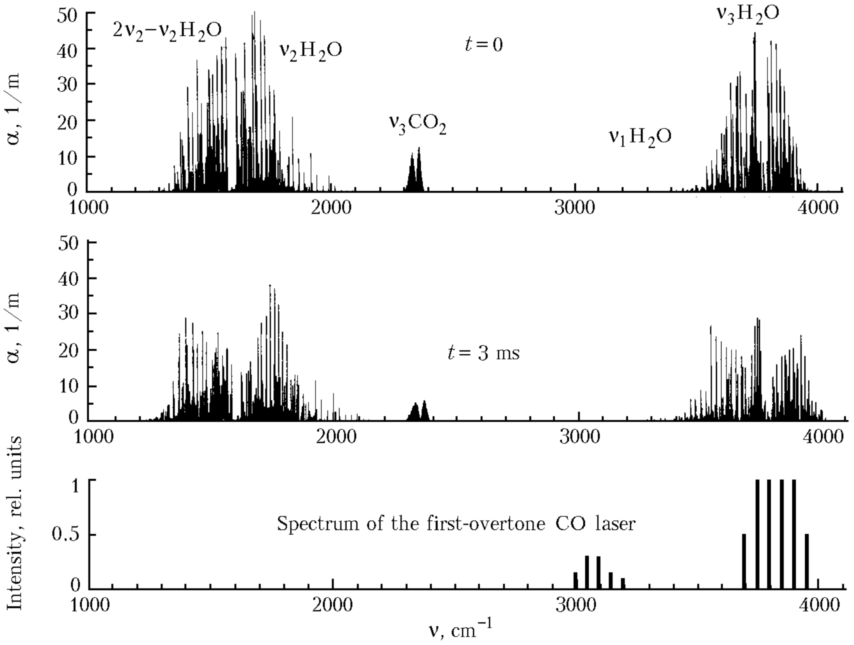
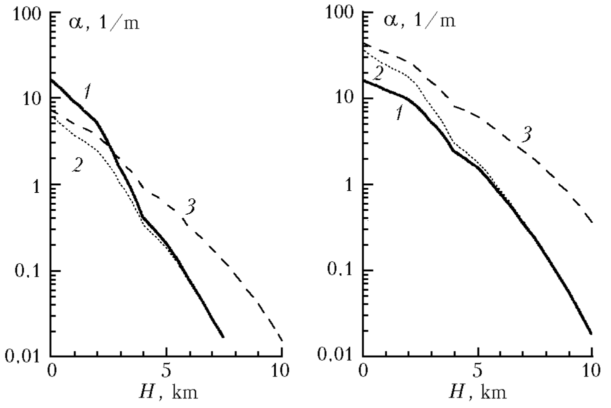


Fig. 4. Atmospheric absorption in the region of 1000–4000 cm^{-1} before (t = 0) and after (t = 3 ms) CO laser pulse. The radiation intensity is 10 kW/cm^2. The conditions of tropical atmosphere at the height H = 0 km. Continuum absorption is neglected. The spectrum of laser radiation without rotational structure is shown below.



a *b*

Fig. 5. Altitude profiles of the water vapor absorption coefficient for the tropical atmosphere at the frequencies of 3546.9 cm^{-1} (*a*) and 3752.2 cm^{-1} (*b*) under the exposure to radiation of the first-overtone CO laser (line 8–6 P(21), ν = 3853.9633 cm^{-1}). Radiation intensity I = 1 MW/cm^2, pulse duration of 1 μs. Continuum absorption is taken into account; allowance for the absorption saturation and heating (*1*), allowance only for the absorption saturation, air temperature in the beam is constant and equal to the atmospheric temperature at a given height (*2*); linear absorption (I = 0) (*3*).

saturation, it becomes stronger with height for the considered pump frequency because of the increase of the absorption cross section and the time of collisional relaxation of H_2O energy levels. At the sensing frequency of 3546.9 cm^{-1}, absorption increases with the increasing temperature, and at 3752.2 cm^{-1} it decreases, while the saturation of absorption of the pump radiation decreases absorption at both frequencies. The above-said explains the behavior of the altitude profiles α(H) shown in Fig. 5. In particular, it becomes clear why the dependence α(H) for the case (*b*) proves to be generally smoother than that for the case (*a*).

Conclusions

1. The comparative analysis of spectral characteristics of radiation of the first-overtone CO laser and the HF and DF lasers has shown that the spectral region of the first-overtone CO laser (2.5–4.2 μm) is wider than the regions of the HF and DF laser emissions, and the spectral density of the CO laser lines is much higher. The capabilities of the first-overtone CO laser are very promising for various applications to remote sensing, spectroscopic gas analysis, and medicine, and these capabilities are wider than those of the HF and DF lasers.

2. Water vapor plays the major role in extinction of radiation of the first-overtone CO laser in the atmosphere. Strong absorption is characteristic of many frequencies (for some of them the absorption coefficient exceeds 1 m^{-1}). Such absorption is now unachievable for the HF and DF lasers. The lines of the first-overtone CO laser with strong absorption by H_2O can be used for laser heating of water-containing media, for example, in medicine. On the other hand, the lines of this laser with $\lambda \geq 3.4$ μm are convenient for efficient energy transfer through a medium.

3. The absorption coefficient of nitrous oxide has been measured for eleven rotational components of the CO vibrational band 35 → 33 (J = 6 ... 17). The absorption cross sections have been determined; they are in a good agreement with the calculated data. The experiments have shown that the frequency of the first-overtone CO laser even at such highly excited

transition as 35 → 33 agrees with the calculated value accurately to, at least, about 10^{-5}, i.e., with the accuracy up to several hundredths reciprocal centimeter.

4. The character of variation of the absorption coefficients for different lines in the process of absorption of the high-power multifrequency radiation of the first-overtone CO laser in the atmosphere may be different: increase or decrease (for some frequencies, variations achieve several times). The high-intensity radiation of the first-overtone CO laser can change markedly the absorption spectrum of the atmospheric air. The calculations have shown that laser-induced changes in the atmospheric absorption depend strongly on the intensity and spectral composition of radiation, height in the atmosphere, absorbing substance, and its concentration. The interaction was discovered between the effects of air heating by the pump radiation and saturation of absorption of the pump radiation, which occurs at short pulse durations and high radiation intensity. This interaction manifests itself differently at different frequencies of sounding radiation. This fact has a marked effect on the corresponding altitude profiles of the absorption coefficient.

Nonlinear laser-induced transformations of the absorption spectrum of air can be used in the future for the development of new methods for spectroscopic diagnostics of molecular gases.

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