Quantum Electronics **40** (7) 629 – 632 (2010)

ACTIVE MEDIA

PACS numbers: 33.20.Ea; 42.55.Lt
DOI: 10.1070/QE2010v040n07ABEH014305

On the temperature dependence of collisional linewidths of the $10^00 - 00^01$ laser transition in the CO_2 molecule

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***Abstract*. Unsaturated absorption coefficients in pure carbon dioxide and $CO_2 - N_2$ and $CO_2 - He$ binary mixtures are measured at a pressure of 100 Torr in the temperature range of 300 – 700 K using a frequency-stabilised tunable CO_2 laser. The relative coefficients of collisional broadening caused by N_2 and He buffer gases and their temperature dependence are determined for the R(22) absorption line ($10^00 - 00^01$ transition) of the CO_2 molecule.**

Keywords: *absorption coefficient, collisional linewidth, carbon dioxide, nitrogen, helium.*

1. Introduction

Almost all technological CO_2 lasers of any excitation type are known to operate at active-medium pressures providing homogeneous broadening of lasing lines due to the collisions of working-gas (CO_2) molecules with other atoms and molecules of the active medium [1 – 4].

The dependence of the collisional width $\Delta\nu_{CO_2}$ (FWHM) of a spectral line of the CO_2 molecule on the gas temperature T is generally described by the formula [5]

$$\Delta\nu_{CO_2} = \gamma_{CO_2-CO_2} p_{CO_2} (300\ \mathrm{K}/T)^n, \qquad (1)$$

where $\gamma_{CO_2-CO_2}$ is the collisional linewidth related to the collisions between CO_2 molecules at a pressure of 1 Torr and temperature of 300 K (collisional self-broadening coefficient for the CO_2 molecule) and p_{CO_2} is the carbon dioxide pressure. The exponent n in (1) depends on the mechanism of interaction between colliding molecules; for the $10^00 - 00^01$ transition lines it ranges from 0.5 to 1 [5 – 12].

It is also known that the main components of active media in highly efficient technological CO_2 lasers are carbon dioxide, nitrogen, and helium in different ratios, at pressures corresponding to the dominance of binary collisions between atoms and molecules [1]. Therefore, the total collisional width $\Delta\nu_g$ of the gain line profile under these conditions can be presented as the sum of contributions to broadening from pair collisions between the major-component atoms and molecules, i.e., in the form

$$\Delta\nu_g \approx \Delta\nu_{CO_2} + \Delta\nu_{N_2} + \Delta\nu_{He}, \qquad (2)$$

where $\Delta\nu_{N_2}$ and $\Delta\nu_{He}$ are the widths related to the collisions of CO_2 molecules with buffer N_2 molecules and He atoms, respectively.

Numerical estimates of the gain linewidth in these CO_2 lasers for the strongest $00^01 - 10^00$ lasing transition are generally based on the formula obtained in [13] for the P(20) line of this transition:

$$\Delta\nu_g = \gamma_{CO_2-CO_2}(\xi_{CO_2} + b_{N_2}\xi_{N_2} + b_{He}\xi_{He}) p_\Sigma \sqrt{300/T}, \qquad (3)$$

where ξ_{CO_2}, ξ_{N_2}, ξ_{He} are the CO_2, N_2, and He fractions in the mixture; $b_{N_2} = \gamma_{CO_2-N_2}/\gamma_{CO_2-CO_2} = 0.73$ and $b_{He} = \gamma_{CO_2-He}/\gamma_{CO_2-CO_2} = 0.64$ are the relative collisional broadening coefficients of CO_2 lines for buffer gas molecules (N_2) and atoms (He), respectively; and p_Σ is the pressure of $CO_2 - N_2 - He$ gas mixture.

The fact that the coefficients b_{N_2} and b_{He} enter formula (3) as constants means in essence that the temperature dependences of $\Delta\nu_{CO_2}$, $\Delta\nu_{N_2}$ and $\Delta\nu_{He}$ values from (2) are assumed to be identical, i.e., the character of spectral line broadening upon interaction of CO_2 molecules with collisional partners having different properties is assumed to be the same; however, this assumption contradicts the existing representations about the mechanisms of collisional line broadening [5, 6].

The collisional linewidth is generally determined either directly (by measuring the absorption line profile using a tunable radiation source) or by measuring the unsaturated absorption coefficient at one (for example, central) frequency within the absorption line at a pressure providing a collisionally broadened profile. The precise measurement of the entire line profile of the CO_2 molecule is a more complex experimental problem than the single-frequency measurement of the absorption coefficient. For the lines of the $[10^00, 02^00]_{I,II} - 00^01$ transitions [3] this problem is somewhat facilitated, because one can use the resonant radiation of a frequency-stabilised CO_2 laser, tunable to the lines of these transitions, as the probe radiation.

The purpose of this study was to determine the relative collisional broadening coefficients b_{N_2} and b_{He} of the R(22) absorption line ($10^00 - 00^01$ transition) of the CO_2 molecule for N_2 molecules and He atoms, respectively, in the temper-

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Received 11 March 2010
Kvantovaya Elektronika **40** (7) 629 – 632 (2010)
Translated by Yu.P. Sin'kov

ature range of 300 – 700 K, which is characteristic of the active media used in electric-discharge technological CO_2 lasers.

2. Measurement technique and discussion of results

The absorption coefficients were measured using a frequency-stabilised tunable CO_2 laser at the central frequency of the R(22) line of the $10^00 - 00^01$ transition in pure CO_2 (α_{CO_2}) and $CO_2 - N_2$ and $CO_2 - He$ binary mixtures (α_{CO_2-M}) with the component ratios $CO_2{:}M = 1{:}Y$ at pure CO_2 and binary mixture pressures (p_Σ) of 100 Torr, which provide Lorentzian profiles of absorption lines.

The relative collisional broadening coefficients b_M for N_2 and He buffer gases were determined from the expression

$$\frac{\alpha_{CO_2}}{\alpha_{CO_2-M}} = 1 + Yb_M. \qquad (4)$$

The R(22) line was chosen because the contribution to the absorption coefficient at its central frequency from the absorption line of the other overlying transitions in the CO_2 molecule can be neglected for the pressures and temperatures under consideration.

Note also that the use of this technique excludes the errors caused by uncertainties in the spectroscopic parameters determining the measured values of absorption coefficients [14].

The experimental setup for measuring the absorption coefficients in gases according to the double-beam compensation scheme on the lasing lines of a frequency-stabilised CO_2 laser, tunable to the lines of the main $00^01 - [10^00, 02^00]_{I,II}$ laser transitions, was described in detail in [15, 16]. The long-term instability of the lasing frequency with respect to the lasing line central frequency, which is resonant to the central frequency of the absorption line, did not exceed ±0.5 MHz. This allowed us to assume with high accuracy that, for the experimentally realised absorption linewidths, the absorption coefficients were measured at the central frequencies of absorption lines.

The error in determining the gas pressure, Δp, was ±0.5 Torr. The gas temperature in the measuring cell was maintained with errors of ±0.4 K (in the range of 293 K $\leqslant T \leqslant$ 420 K) and ±0.9 K (in the range of 470 K $\leqslant$ $T \leqslant$ 700 *K*).

The results of measuring α_{CO_2} in pure carbon dioxide at the pressure $p_{CO_2} = 100$ Torr and temperatures from 300 to 700 K are shown in Fig. 1. The measured coefficients α_{CO_2-M} in $CO_2 - N_2$ and $CO_2 - He$ binary mixtures with different component ratios at the mixture pressure $p_\Sigma = 100$ Torr and in the same temperature range are shown in Figs 2 and 3, respectively.

For each measurement temperature, according to formula (4), we plotted the dependence of the $\alpha_{CO_2}/\alpha_{CO_2-M}$ ratio on Y. The coefficients b_{N_2} and b_{He} were determined from the slopes of the corresponding straight lines.

Figure 4 shows as an example the dependences for the $CO_2 - N_2$ mixture at the limiting temperatures (300 and 700 K). These dependences gave $b_{N_2} = 0.74 \pm 0.02$ at $T = 300$ K and 0.99 ± 0.04 at $T = 700$ K. Similar dependences for the $CO_2 - He$ mixture are shown in Fig. 4b. According to these data, $b_{He} = 0.58 \pm 0.03$ at $T = 300$ K and 0.88 ± 0.04 at $T = 700$ K.

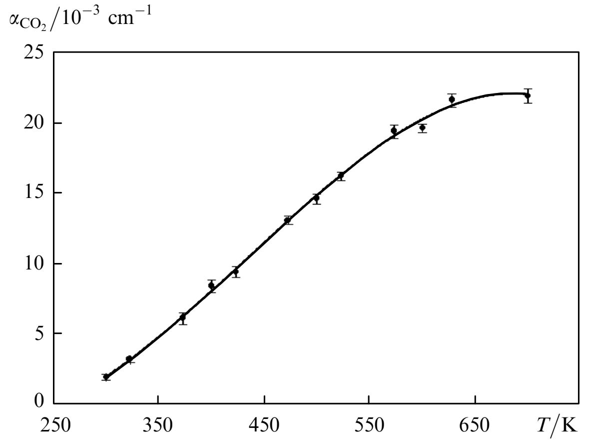

Figure 1. Temperature dependence of the absorption coefficient of pure CO_2 at a pressure of 100 Torr on the 10R(22) line.

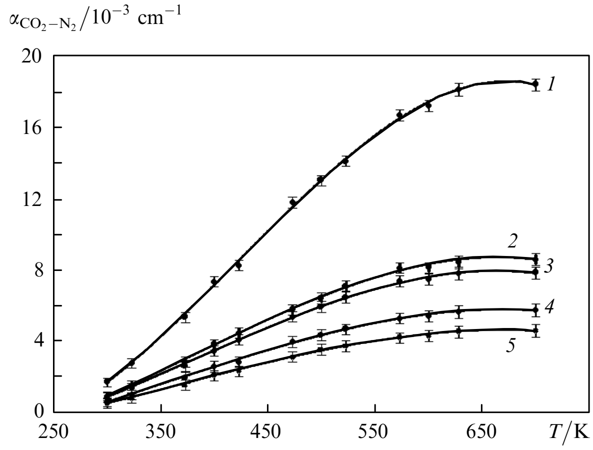

Figure 2. Temperature dependences of the absorption coefficient of a $CO_2{:}N_2 = 1{:}Y$ gas mixture at a pressure of 100 Torr on the 10R(22) line for $Y = 0.19$ (*1*), 1.5 (*2*), 1.78 (*3*), 3 (*4*), and 4 (*5*).

Thus, the experimental data in Fig. 4 clearly indicate that in the temperature range under study (300 – 700 K) the $\alpha_{CO_2}/\alpha_{CO_2-N_2}$ and $\alpha_{CO_2}/\alpha_{CO_2-He}$ ratios, and, therefore, the

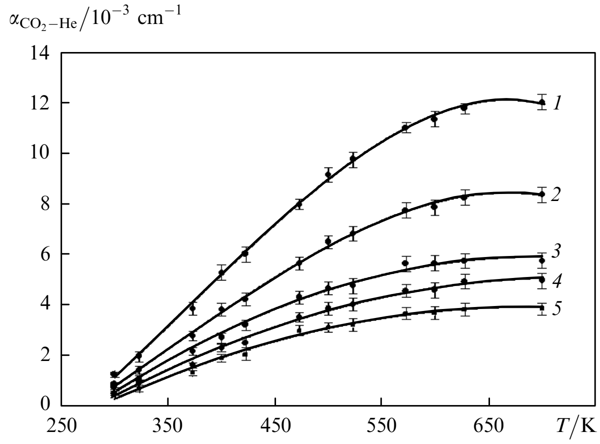

Figure 3. Temperature dependences of the absorption coefficient of a $CO_2{:}He = 1{:}Y$ gas mixture at a pressure of 100 Torr on the 10R(22) line for $Y = 1$ (*1*), 2 (*2*), 3 (*3*), 4 (*4*), and 5 (*5*).

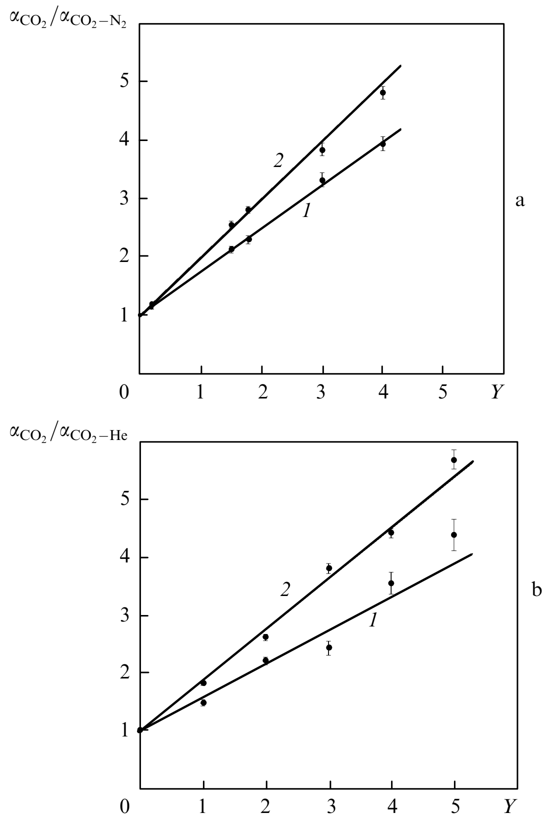

Figure 4. Dependences of the ratio of absorption coefficients, $\alpha_{CO_2}/\alpha_{CO_2-M}$, on the ratio $Y = [M]/[CO_2]$ of the concentrations of binary mixture components for M = N_2 (a) and He (b) at temperatures of 300 (*1*) and 700 K (*2*).

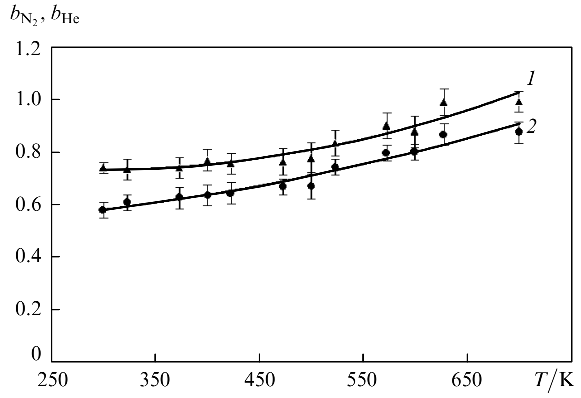

Figure 5. Temperature dependences of the relative collisional broadening coefficients of the 10R(22) line of the CO_2 molecule for N_2 (*1*) molecules and He (*2*) atoms.

coefficients b_{N_2} and b_{He} increase with increasing gas temperature.

Note that Abrams [13], who proposed formula (3), measured the linewidth P(20) of the $10^00 - 00^01$ transition in pure CO_2 and in $CO_2{:}N_2 = 1{:}1$ and $CO_2{:}He = 1{:}1$ binary mixtures by optoacoustic spectroscopy only at one temperature: $T = 298$ K. The coefficients b_{N_2} and b_{He} for several lines, including R(22), were measured later in the temperature range of 300–650 K [17] using the technique similar to that applied in the present study. At $T = 300$ K the coefficients $b_{N_2} = 0.81 \pm 0.04$ and $b_{He} = 0.70 \pm 0.04$ were measured in [17]. Although these values somewhat exceed those obtained in this study, the quantitative agreement can be considered as fairly good.

At the same time, a qualitative comparison of our results and the data of [17], i.e., a comparative analysis of the behaviour of the coefficients b_{N_2} and b_{He} with a change in temperature leads to radically different conclusions. In particular, only small variations in b_{N_2} and b_{He} were observed [17] in the temperature range studied (300–650 K); thus, it was concluded that these coefficients hardly changed. The character of the change in the coefficients b_{N_2} and b_{He} with temperature, observed by us, is shown in Fig. 5. It can be seen that b_{N_2} barely changes up to $T \sim 550$ K, which is in agreement with the results of [17], but noticeably increases at $T > 550$ K. The coefficient b_{He} weakly depends on temperature in the range of $T = 300 - 550$ K.

Thus, our measurements unambiguously indicate that the coefficients b_{N_2} and b_{He} depend (and rather differently) on temperature. This means that the widely used formula (3) is incorrect at $T > 550$ K. According to the physical meaning of expression (2), each term in the correct formula should depend on temperature in its own way, just as the rate constants of collision-induced vibrational relaxation of the 00^01 upper lasing level of the CO_2 molecule in pure CO_2 and in $CO_2 - N_2$ and $CO_2 - He$ binary mixtures are characterised by their own temperature dependences (see, e.g., [18]).

Active media with a content of CO_2 molecules much lower than that of N_2 molecules and He atoms are optimal for high-power technological electric-discharge CO_2 lasers, operating with fast flow of active medium. These are $CO_2{:}N_2{:}He \approx 1{:}(5-22){:}(5-22)$ mixtures or mixtures with even a lower content of working molecules (see, for example, [19–22]). In these lasers the active-medium temperature corresponding to the maximum lasing power reaches 600–700 K [23–25]. Simple estimates show that the collisional gain linewidths for such CO_2 lasers, obtained from formula (3) and using our results, may differ by more than 100 %.

3. Conclusions

Unsaturated absorption coefficients in pure carbon dioxide and in $CO_2 - N_2$ and $CO_2 - He$ binary mixtures for the R(22) line of the $10^00 - 00^01$ transition in CO_2 molecules were measured at a total pressure of 100 Torr in the temperature range of 300–700 K using a frequency-stabilised tunable CO_2 laser. The relative collisional-broadening coefficients b_{N_2} and b_{He} for N_2 and He buffer gases were determined. These parameters were found to depend on gas temperature.

References

1. Siegman A.E. *Lasers* (Mill Valey, Cal.: University Science Book, 1986).
2. Hall D.R., Hill C.A., in *Handbook of Molecular Lasers* (New York: Marcell Dekker, 1987) Ch. 3, p. 328.
3. Witteman W.J. *The CO_2 Laser* (Berlin: Springer, 1987; Moscow: Mir, 1990).
4. Plinski E.F., Abramski K.M. *DC-excited Continuous-wave Conventional and RF-excited Waveguide CO_2 Lasers, Gas Lasers* (New York: CRC Press, 2007) p. 239.

5. Achasov O.V., Kudryavtsev N.N., Novikov S.S., Soloukhin R.I., Fomin N.A. *Diagnostika neravnovesnykh sostoyanii v molekulyarnykh lazerakh* (Diagnostics of Nonequilibrium States in Molecular Lasers) (Minsk: Nauka i tekhnika, 1985).
6. Starikov V.I., Lavrent'eva N.N. *Stolknovitel'noe ushirenie spektral'nykh linii pogloshcheniya molekul atmosfernykh gazov* (Collisional Broadening of Absorption Lines of Atmospheric Gas Molecules) (Tomsk: Izd-vo Inst. Optiki Atmosfery SO RAN, 2006).
7. Bulanin M.O., Bulychev V.P., Khodos E.B. *Opt. Spektrosk.*, **48**, 732 (1980).
8. Osipov V.M. *Opt. Spektrosk.*, **63**, 1013 (1987).
9. Arie E., Lacome N., Levy A. *Appl. Opt.*, **26**, 1636 (1987).
10. Gross L.A., Griffiths P.R. *Appl. Opt.*, **26**, 2250 (1987).
11. Rosenmann L., Perrin M.Y., Taine J. *J. Chem. Phys.*, **88**, 2995 (1988).
12. Arshinov K.I., Leshenyuk N.S. *Kvantovaya Elektron.*, **24**, 517 (1997) [*Quantum Electron.*, **27**, 497 (1997)].
13. Abrams R.L. *Appl. Phys. Lett.*, **25**, 609 (1974).
14. El'yashevich M.A. *Atomnaya i molekulyarnaya spektroskopiya* (Atomic and Molecular Spectroscopy) (Moscow: Fizmatlit, 1962).
15. Arshinov K.I., Kablukov N.G., Leshenyuk N.S. *Prib. Tekh. Eksper.*, **1**, 237 (1991).
16. Arshinov K.I., Kablukov N.G., Tikhonov F.V. *Prib. Tekh. Eksper.*, **1**, 103 (1996).
17. Robinson A.M., Weiss J.S. *Can. J. Phys.*, **60**, 1656 (1982).
18. Nevdakh V.V., Orlov L.N., Leshenyuk N.S. *Zh. Prikl. Spektrosk.*, **70**, 246 (2003).
19. Nath A.K., Kumar M. *IEEE J. Quantum Electron.*, **29**, 1199 (1993).
20. Markillie G.A.J., Baker H.J., Betterton J.G., Hall D.R. *IEEE J. Quantum Electron.*, **35**, 1134 (1993).
21. Chang S.-H., Cheng C.-C., Chang M.-F., et al. *Jpn. J. Appl. Phys. Pt.1*, **39**, 82 (2000).
22. Nevdakh V.V., Gandzhali Monire. *Zh. Prikl. Spektrosk.*, **71**, 490 (2004).
23. Harry J.E., Evans D.R. *IEEE J. Quantum Electron.*, **24**, 503 (1988).
24. Abramski K.M., Colly A.D., Baker H.J., Hall D.R. *IEEE J. Quantum Electron.*, **32**, 340 (1996).
25. Nevdakh V.V. *Kvantovaya Elektron.*, **31**, 525 (2001) [*Quantum Electron.*, **31**, 525 (2001)].