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RESONANCE CO_2 ABSORPTION (10.6 μ) BEHIND A SHOCK FRONT

R. I. Soloukhin and N. A. Fomin

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Carbon dioxide is widely used in various molecular laser systems, and therefore there is interest in measurements of the coefficient of absorption of CO_2 at the frequency of the usually used laser transition (100, J) → (001, J + 1) at the wavelength of 10.6 μ in a wide range of temperatures and densities of the gas. For this purpose it is helpful to use the method of compression and heating of the gas being studied in shock waves [1, 2]. The dependence of the coefficient of absorption α of CO_2 at the 10.6-μ wavelength on the pressure of the gas at room temperature has been studied in sufficient detail at present [3, 4], although even in this region of variation of the parameters attempts at comparing the calculated values of α with experiment (at $p \geq 1$ atm) have revealed considerable discrepancies, connected primarily with the imperfection of the approximation of the shape of the lines of the vibrational–rotational spectrum of the CO_2 molecule by a Lorentzian contour.

It is interesting to trace the behavior of the coefficient of absorption at high gas temperatures, where the values of α can be especially large. The temperature dependence of α at $p \simeq 1$ atm in the temperature range of 300-420°K was determined in [5]. It was discovered that the measured values of the coefficient of absorption are considerably higher than the calculated values, and therefore to explain the dependence α(T) it was assumed in [5] that the cross section for collisional broadening of the line contour of CO_2 molecules varies as a function of the temperature in the form $\sigma_c \sim T^{-1/2}$. In [6] similar measurements were made up to temperatures of ~615°K. On the basis of the calculations of [7] the authors explained the high values of α which they obtained by the contribution of transitions of "mixed" states ($11^10 \rightarrow 01^11$, $12^00 \rightarrow 02^01$) without resorting to the temperature dependence of the broadening cross section. Measurements of α up to T ~ 1600°K were made in [8] using the shock-tube technique. In the calculations of the coefficient of absorption the authors of this report allowed for the contribution of "mixed" transitions and they also assumed a dependence $\sigma_c \sim T^{-1/2}$.

In the present work the temperature dependence was determined both for CO_2 and for a mixture of CO_2 and nitrogen in a wide range of temperatures (500-2100°K). Measurements of α were also made as a function of the pressure at high gas temperatures (1100 and 1600°K) behind both the incident and the reflected shock waves. The purpose of the tests described

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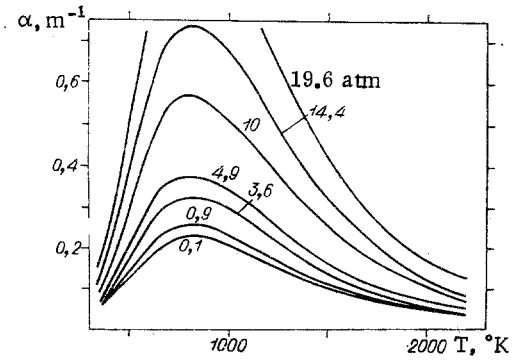

Fig. 1

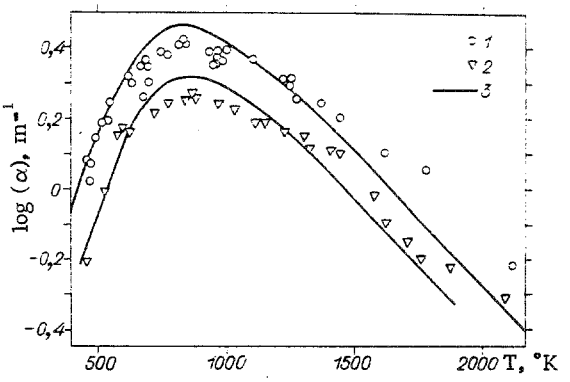

Fig. 2

below is the determination of the limits of applicability of the simplest methods of theoretical description of resonance absorption in CO_2 at moderate and elevated gas pressures.

The following circumstances have been taken into account in calculations of α. An important contribution to the value of α for the main transition (100, J) → (001, J + 1) is also made by transitions of the type (1n0, J) → (0n1, J ± 1) for which the frequencies lie close enough to the frequency of the main transition and the populations of the levels (n = 1, 2) become significant at T ≳ 500°K. According to the estimate of [7] such transitions at T = 620°K and p = 1 atm already increase α at the center of the P 20 line by 42.1%. Moreover, the overlapping of neighboring rotational lines, which must lead to an increase in α by several times [4], becomes important at a pressure p ≳ 1 atm. Therefore, one must use the following expression for the coefficient of absorption:

$$\alpha(J) = \sum_{(J',n)} \{\alpha^{100\to001}(J')\Phi(J,J') + \alpha^{1n0\to0n1}(J')\Phi_n(J,J')\},$$

where Φ is the form factor. In the case of a collisionally broadened contour the quantity α for the center of an individual line can be found from the equation [9]

$$\alpha = (\lambda^2 A_{21}/4\pi^2\Delta\nu_c)(N_2 - (g_2/g_1)N_1)$$

(the notation is standard).

For a collisional contour

$$\Phi(J,J') = \frac{1}{1+\left[\frac{2(\lambda(J)-\lambda(J'))}{\Delta\nu_c}\right]^2}.$$

The populations of the vibrational–rotational levels were calculated on the assumption of complete thermodynamic equilibrium behind the shock front:

$$N_i = (Nr_i/Q_v Q_R)\exp\{-\Theta_{vi}/T - \Theta_{Ri}J_i(J_i+1)/T\},$$

where Q are statistical sums; Θ are the characteristic temperatures of the i-th level (the indices v and R pertain to vibrational and rotational degrees of freedom, respectively); r_i is the statistical weight of the i-th level. Equations for calculating the transition frequencies, as well as the principal vibrational–rotational constants, are presented in [7]. The choice of the temperature dependence of the half-width of the contour is important in the performance of the calculations:

$$\Delta\nu_c = Cp(300/T)^k. \qquad (1)$$

The best agreement between the calculated and experimental functions is obtained if, by analogy with [5, 8], one sets the exponent k = 1 in Eq. (1), i.e., one takes $\sigma_c \sim T^{-1/2}$.

The constant C depends weakly on the rotational number J. From the approximation of the data of [11] which was made in [10] this dependence has the form

$$C = 0.118 - 0.0008(J \pm 1),$$

where plus pertains to the P branch and minus to the R branch of the laser transition in CO_2. The calculations show that: a) At room temperature in the pressure range of 0.1-10 mm Hg the gain increases linearly with pressure from $2\cdot10^{-5}$ to $1.6\cdot10^{-3}$ cm^{-1} (Doppler broadening) and then does not vary with an increase in pressure to p ≈ 0.2 atm; b) in the pressure range

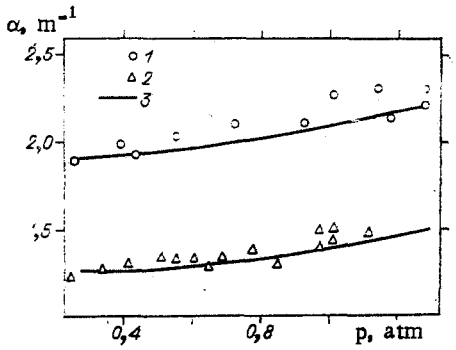

Fig. 3

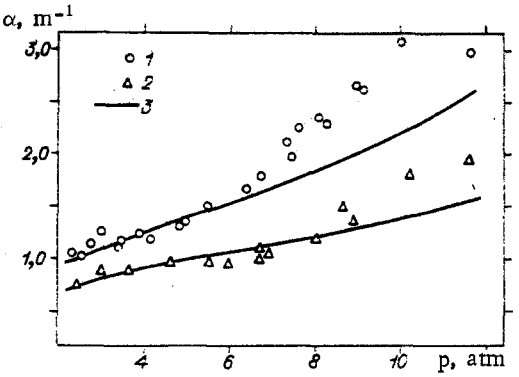

Fig. 4

from 0.2 to 1 atm with T ≳ 500°K an increase occurs in the coefficient of absorption of CO_2 due to the contribution of bound states; c) at pressures of more than 1 atm one must allow for the contribution of neighboring lines to the absorption [4], and this effect at pressures on the order of 10 atm more than doubles the coefficient of absorption; d) the temperature dependence of the coefficient of absorption is very strong [5, 6, 8]: at pressures on the order of 0.01 atm and above α increases by an order of magnitude with a change in temperature from 300 to 600°K, it reaches a maximum at T = 800°K, and then declines relatively slowly. The degree of sensitivity of the coefficient of absorption to the temperature and pressure is well illustrated by the calculating functions α(p, T) for the P 18 line of the 100–001 transition of CO_2 presented in Fig. 1 (the numbers above the curve correspond to the pressure in atm).

The heating of the CO_2 was accomplished in a shock tube with a square cross section of 5 × 5 cm^2. The method of measuring the coefficient of absorption is analogous to the system for measuring the amplification in a gasdynamic laser [12]: The light beam of an electric-discharge CO_2 laser with a density of ~10 W/cm^2 is limited by two light filters to a wavelength of 10.6 μ, passes through the channel of the shock tube parallel to the wave front, and is recorded by a germanium receiver. Since the pressure in the test gas exceeded the pressure in the resonator of the probing laser by more than an order of magnitude, the contour of the absorption line considerably exceeded the contour of the laser emission line, which practically eliminated the error in measuring the coefficient of absorption due to mismatch of the spectra [13]. With a delay of 50–200 μsec after recording the signal of absorption in the shock wave the laser beam was "cut off" with an electrooptical gate to determine its intensity during the recording period.

In measurements behind the fronts of reflected shock waves one must take into account the effects of the interaction of the reflected wave with the boundary layer which are manifested especially strongly in CO_2. Therefore, by analogy with [14], in work with reflected waves we used the method of partial reflection from the end of a cylinder 30 mm in diameter mounted in the central part of the shock tube channel. The results of the experiment are compared with calculations by the method described above in Fig. 2 (here and in Figs. 3 and 4, curve 1 is measurements in pure CO_2, curve 2 is measurements in a mixture of $CO_2 + N_2$, and curve 3 is the calculation). The initial pressure was chosen so that the pressure behind the reflected shock wave was about 3 atm. The density of the compressed gas in these tests varied in the range of $(1\text{-}3)\cdot 10^{19}$ cm^{-3}. Calculations of shock adiabats presented in [15] were used in analyzing the data; for the $CO_2 + N_2$ mixture the adiabat was calculated by the method described in [16]. From the data obtained it is seen that the temperature dependence of the coefficient of absorption of CO_2 at pressures p ≳ 1 atm is well described by the simplest theory of absorption with collisional broadening on the assumption that the broadening cross section depends on the temperature ($\sigma_c \sim T^{-1/2}$). At temperatures T ≳ 1500°K the experimental points lie somewhat higher than the calculated points. It is possible that transitions between high-lying levels, of the type $00^0 2 \rightarrow 10^0 1$, for example [7], become important at such temperatures.

The temperature dependence α(T), having a transition through a maximum, illustrates the existence of the effect of "clearing" of the medium upon an increase in the temperature T > 800°K. Such an effect was actually observed in the experiments: The absorption was lower behind the reflected shock wave than in the incident wave when the gas temperature in the incident shock wave was 800–1000°K.

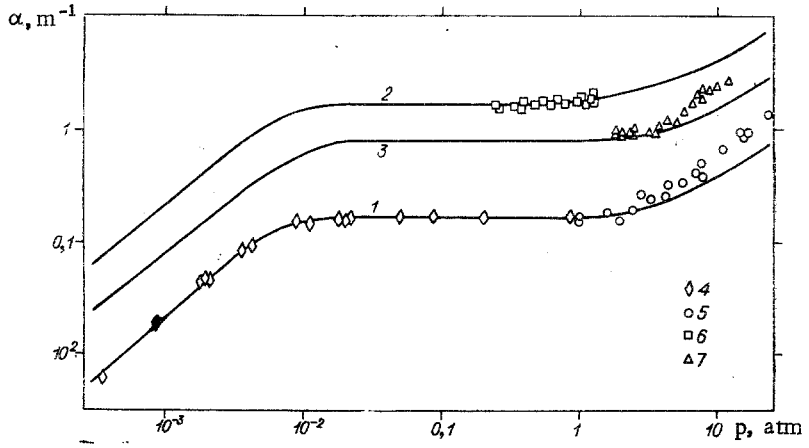

Fig. 5

Since carbon dioxide is usually used in a mixture with N_2 in the active media of lasers, we also measured the coefficients of absorption of 10.6-μ radiation for a mixture of CO_2 and N_2. Nitrogen acts mainly as an inert diluent, although because CO_2 has a somewhat smaller (by about 25%) broadening cross section in collisions with N_2 than with CO_2 this leads directly to an increase in the coefficient of absorption, since $\alpha \sim 1/\Delta\nu_c$, on the one hand, and somewhat decreases the contribution of neighboring lines to α because of their smaller overlapping, on the other. Considering this, one can be confident that the dependence α(T) for the mixture $CO_2 + N_2$ is also described sufficiently well by the simplest theory.

The results of measurements of the dependence of α on the gas pressure are presented in Figs. 3 and 4 [measurements behind incident shock waves (T = 1100°K) and measurements behind reflected shock waves (T = 1600°K), respectively]. It is interesting to note that the observed growth of α with variation in the pressure in the range of 0.2-1.2 atm (Fig. 3) is mainly due to an increase in the contribution of the transitions of "bound" states. According to the data of [7], the variation in α in this range is even more considerable for the P 20 line than for P 18 because of the closer resonance of the P 20 line of the transition (100 → 001) and the R 23 line of the transition ($01^11 \rightarrow 11^10$). And the continued increase in α (see Fig. 4) is connected mainly with the effect of overlapping of the neighboring rotational lines [4], with the differences between the experimental and calculated values of the coefficient of absorption becoming considerable at pressures $p \gtrsim 6$ atm. Evidently this disagreement can be partly explained by the difference of the absorption contour from a Lorentzian contour [17]. Moreover, considerable irregularities and turbulization of the medium [14], which can introduce additional losses in measurements of the coefficient of absorption, are possible behind a reflected shock wave in CO_2.

A summary of the data on the dependence of α on pressure within wide limits (from $0.3 \cdot 10^{-3}$ to 20 atm) and for different gas temperatures is presented in Fig. 5: curves 1-3) 300, 1100, and 1600°K, respectively; curve 4) data of measurements from [3]; curve 5) data from [4], temperature 300°K; curves 6 and 7) measurements of present work, 1100 and 1600°K, respectively. As seen from these data, the values of α can be considerable, and therefore the absorption of radiation at 10.6 μ (for example, in "pockets" of gasdynamic streams in the resonators of circulating CO_2 lasers) can have a considerable effect on the modes of generation.

Summing up, we note that systematic measurements of the coefficient of absorption of the resonance radiation (10.6 μ) in CO_2 and in the mixture $CO_2 + N_2$ in a wide range of variations of the temperature and pressure of the gas were conducted in the work. The functions α(p, T) at pressures $p \lesssim 6$ atm are described accurately enough by the simplest theory with allowance for the contribution of the transitions of "bound" states and on the assumption that the broadening cross section depends on the temperature in the form $\sigma_c \sim T^{-1/2}$. At pressures $p > 6$ atm more accurate allowance for the difference of the absorption contour from a Lorentzian contour is evidently needed.

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