Explosive absorption of CO_2-laser radiation in the mixture of atmospheric air with carbon dioxide

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Absorption of 35 W CO_2 laser radiation is studied for the case of the laser beam focusing in an optical cell filled with a mixture of atmospheric air of room temperature and carbon dioxide of varying concentration (from 1 to 100%). The temperature of the gas mixture in a focal zone was controlled by the shift of interference fringes in a Mach–Zehnder interferometer based on a He–Ne laser. The interferometer was designed so that one its arm crossed the focus of the CO_2-laser beam. At the carbon dioxide concentration higher than 20%, the mixture warms significantly (up to 600 K) in the focal zone, what can be explained by the effect of explosive absorption as the concentration of carbon dioxide achieves some critical value.

Introduction

Explosive absorption of IR laser radiation in molecular gases has been thoroughly studied theoretically (Refs. 1–8). Experimentally it was studied only partially (Refs. 9–14). The mechanism of explosive absorption falls in the category of chain reactions, which occur in the presence of a positive feedback between absorption and growing population of the lower absorbing level, which corresponds to an excited vibrational state. The effect of explosive absorption is characterized by the exponential growth of the population of absorbing levels as the temperature increases and sharp increase of the absorption coefficient due to heating the gas in the laser beam channel. Thanks to the positive feedback, this phenomenon has a threshold character. The threshold condition depends on the radiation intensity, concentration of the absorbing gas, and the mode of gas cooling in the laser beam channel. It can manifest itself in laser media as instabilities of different kind leading to fluctuations of the laser radiation power, bistability, hysteresis effects, etc.[9] In Ref. 8, the threshold conditions were estimated for the case of propagation through the atmosphere of high-power large-diameter laser beams of the 10.6 μm wavelength.

The goal of this work is direct observation of the explosive absorption of the CO_2-laser radiation in the mixture of atmospheric air and carbon dioxide, as well as estimation of the carbon dioxide threshold concentration and maximum air temperature in the laser beam channel.

Experiment

The optical layout of the experiment is shown in Fig. 1. We used the standard interferometric method for measuring the gas temperature in a cell.[15] This method employs a single-pass Mach–Zehnder interferometer (mirrors M_3 and M_4 and beam-splitting plates P_1 and P_2). A tight cell containing the gas mixture under study was set in one arm of the interferometer.

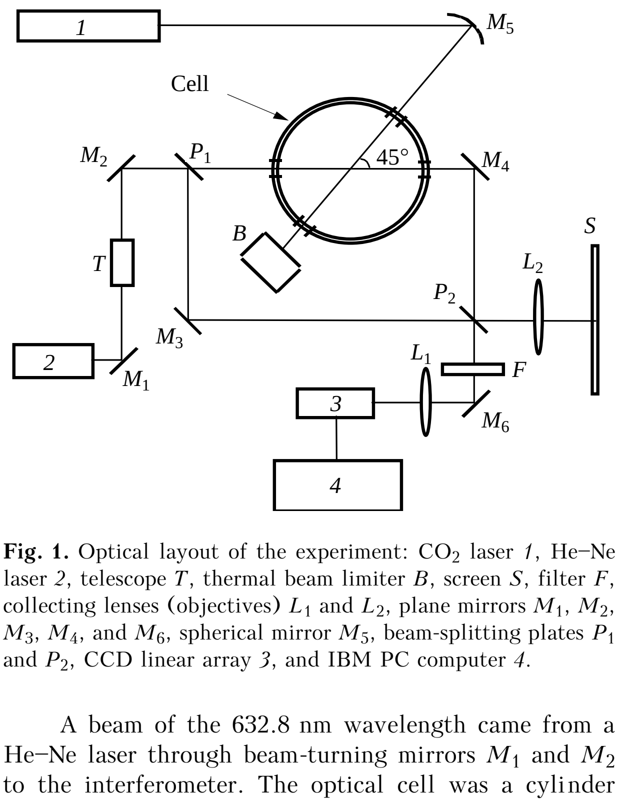

Fig. 1. Optical layout of the experiment: CO_2 laser *1*, He–Ne laser *2*, telescope *T*, thermal beam limiter *B*, screen *S*, filter *F*, collecting lenses (objectives) L_1 and L_2, plane mirrors M_1, M_2, M_3, M_4, and M_6, spherical mirror M_5, beam-splitting plates P_1 and P_2, CCD linear array *3*, and IBM PC computer *4*.

A beam of the 632.8 nm wavelength came from a He–Ne laser through beam-turning mirrors M_1 and M_2 to the interferometer. The optical cell was a cylinder made of stainless steel with windows for entrance and exit of IR and visible radiation. The inner diameter of the cell was 120 mm, and the inner height was 100 mm. The centers of the windows were at the height of

80 mm above the cell base. The diameters of all windows were equal to 20 mm. They were arranged so that the axes of the visible and thermal beams crossed at the center of the cell at an angle of 45°. The input windows for the CO_2-laser radiation were made of ZnSe in the form of 4-mm thick plane-parallel plates. The cell was equipped with pipes for filling and pumping out and dosage of the gas under study.

The theory of explosive absorption at vibrational transitions in molecular gases is expounded in detail in the references mentioned above and in review 16. Figure 2 illustrates the mechanisms of excitation and relaxation of energy levels of carbon dioxide and nitrogen molecules taking part in the mixture heating. According to Ref. 8, the thermal regime of the atmospheric air interaction with high-power CO_2-laser radiation of 10.6 μm is described by the equation of thermal explosion

$$d\eta/dt = \exp \eta/\tau_1 - \eta/\tau_0, \quad (1)$$

where $\eta = (T - T_0)\theta_{(100)}/T_0^2$ is the dimensionless temperature; $\tau_1 = \exp(\theta_{(100)}/T_0)(\tau_I + \tau_\Sigma)T_0^2/A\theta_{(100)}$ is the characteristic time of heat release in the laser channel; τ_0 is the characteristic time of heat removal from the channel; T and T_0, respectively, are the current and initial temperatures in the laser channel; $\theta_{(100)}$ is the characteristic temperature of the vibrational level (100) of the CO_2 molecule [where the resonance absorption of the 10.6 μm radiation occurs and the CO_2 molecule transits to the level (001)]; $\tau_I = h\nu/\kappa\sigma I$ is the pumping time for this resonance transition, σ is the transition cross section, I is the laser radiation intensity, κ is the ratio of concentrations of carbon dioxide and nitrogen in air; $\tau_\Sigma = \kappa/\tau_{CO_2-H_2O} + 1/\tau_{N_2-H_2O} + 1/\tau_{N_2-O_2} + \kappa/\tau_{CO_2-N_2}$ is the generalized time of relaxation of a generalized excited level of the system CO_2–N_2.

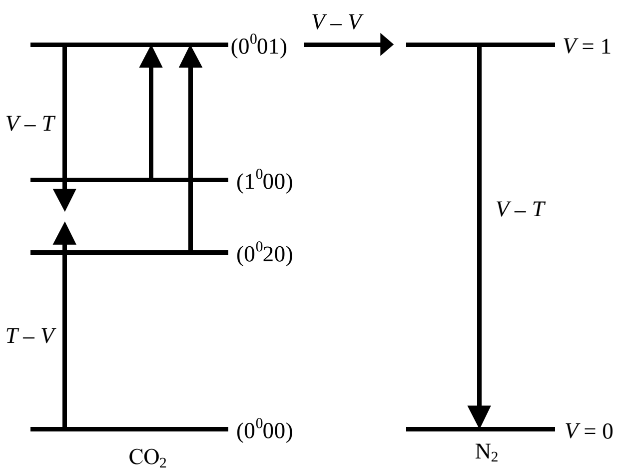

Fig. 2. Mechanism of positive feedback at the resonance radiation absorption (9.6 and 10.6 μm wavelengths) by molecules of CO_2 in air.

The thermal explosion occurs when the parameter τ_0/τ_1 exceeds some critical value.

For atmospheric air (A = 1000 K, $\theta_{(100)}$ = 2000 K, T_0 = 300 K), the threshold condition can be expressed as

$$\tau_0/(\tau_I + \tau_\Sigma) > 10. \quad (2)$$

In our experiment, the conditions $\tau_I = 10^{-2}$–10^{-4} s and $\tau_\Sigma = 10^{-4}$–10^{-6} s hold, respectively, in the focal zone of the 35-W CO_2-laser radiation ($I \sim 10^4$ W/cm^2 at the focus) and at the carbon dioxide concentration in the cell from 1 to 100% (κ = 0.01–1). In this case, the threshold condition for occurrence of the explosive absorption is determined by the established regime of heat removal from the focal zone. That is, under the condition $\tau_0/\tau_I > 10$, the parameter $\tau_0 = D/V$ is determined by cooling of the heated focal zone by convective flows formed in this process. Here D is the characteristic cross size of the focal zone, and V is the convection rate. Since possible values of D and V in our case are within 0.01–0.1 cm and 1–10 cm/s, respectively, then τ_0 falls within 10^{-3}–10^{-1} s range. Obviously, the considered conditions of our experiment allow the threshold conditions of explosive absorption to be realized, what just has been observed by us at saturating the atmospheric air in the cell with carbon dioxide.

In the experiment, we used gaseous mixtures with the following content of carbon dioxide in air: natural concentration of CO_2 in the atmosphere (0.035%), 1, 5, 10, 20, 30, 50, 80, and 100%. The power of the CO_2-laser radiation was kept at the level of 35 W. Shifts of the interference pattern in response to the variation of the carbon dioxide concentration in the cell were recorded on the computerized CCD linear array.

Figure 3 shows a characteristic interference pattern observed on the CCD linear array in two cases: when the CO_2-laser radiation is focused into the cell and when the CO_2-laser beam is cut off.

Figure 4 demonstrates the obtained dependence of the temperature in the focal zone on the carbon dioxide concentration. As is seen, the threshold concentration of carbon dioxide in the cell is about 15% under our experimental conditions.

The difference ΔT in the temperature of the focal zone with respect to the rest of the cell was determined by the well-known equation[15]:

$$\Delta T = \lambda/L\ (dn/dT)\ \Delta S, \quad (3)$$

where L is the length of the cell zone heated by the CO_2-laser beam (in our case, it is the projection of the heated zone to the interferometer ray); λ is the interferometer operating wavelength (in the experiment, we used a He–Ne laser with the wavelength λ = 632.8 nm); dn/dT is the derivative of the refractive index with respect to temperature; ΔS is the shift of peaks on the interference pattern in relative units of the distance between the neighboring peaks.

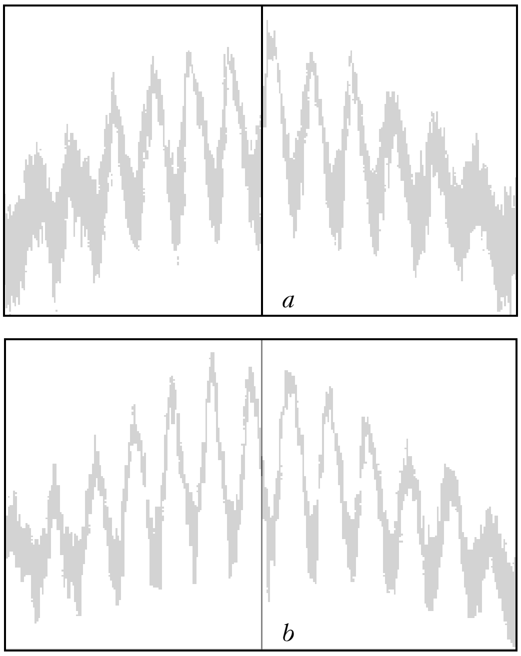

Fig. 3. Position of interference fringes at carbon dioxide concentration of 50% in air: laser beam is focused in the cell (*a*); laser beam is cut off before the entrance to the cell (*b*).

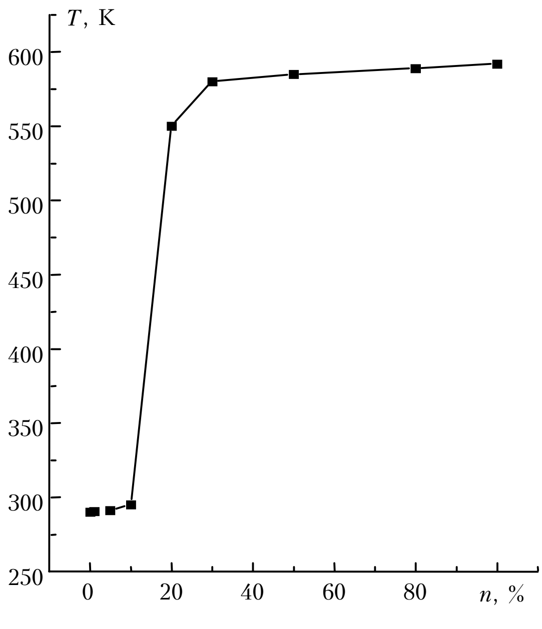

Fig. 4. Temperature of the gas mixture heated by CO_2-laser beam vs. carbon dioxide concentration *n* in air.

In our experiments, the maximum shift ΔS of the peaks of the interference pattern (100-% concentration of carbon dioxide) was equal to 3/4 distance between the neighboring peaks and the projection of the heated gas zone to the interferometer ray $L = 1.2$ mm. The value of dn/dT was borrowed from the tables for air.[15] The maximum air temperature in the focal zone under the after-threshold conditions (100% carbon dioxide) was estimated as 595 ± 60 K. This value far exceeds the temperature under the before-threshold conditions (0.035, 1, 5, and 10% carbon dioxide), when the gas temperature is almost equal to the room one (accurate to several degrees), i.e., about 290 K. In spite of high sensitivity of the interferometric method, the error in determination of the temperature change ΔT in the focal zone was rather high (about 20%) in our case. In an interference pattern displayed from the CCD linear array, the shift of the peak can be determined rather accurately (to 5–10% error). Therefore, the error in determination of the temperature by this method is mostly due to the error in measuring L. In our case, the value of L was found directly from the size of the shifted interference pattern relative to the full interference pattern on the screen S. The measuring error was about 20%. Just this value determined the characteristic error in the calculation of ΔT by Eq. (3).

Conclusion

In this experiment, we used a relatively simple and efficient procedure to observe the explosive absorption in gaseous media. The effect of explosive absorption of the CO_2-laser radiation was first observed in the mixture of atmospheric air with carbon dioxide. This work can be considered as one of the first steps in experimental research into a wide variety of critical phenomena occurring at interaction of resonance IR radiation with molecular gases.[16] It can be also useful in interpreting possible manifestation of similar thermal effects in the atmosphere of Venus[17] and in the case of future accumulation of greenhouse gases, mostly carbon dioxide, in the Earth's atmosphere.[18,19]

Acknowledgments

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