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***A. V. Turin*[1], *G. P. Prepelitsa*[2], *V. P. Kozlovskaya*[3]**

[1] *I. I. Mechnikov Odessa National University,*
[2] *Odessa National Polytechnical University,*
[3] *Odessa State Environmental University*

Kinetics of energy and heat exchange in mixture CO_2-N_2-N_2O of atmospheric gases interacting with ir laser radiation: Precise 3-mode kinetical model

A kinetics of energy and heat exchange in the mixture CO_2-N_2-H_20 gases in atmosphere under passing the powerful CO_2 laser radiation pulses is studied. More precise three-mode model of kinetical processes is formulated. The estimate for realization of the kinetical cooling effect by CO_2 for different atmosphere conditions and laser pulses parameters (pulse form, duration etc.) is given.

Studying interaction of the powerful laser radiation with aerosol atmosphere and search of new non-linear optical effects is related to class of actual serious problems of modern aerosol laser physics (c.f.[1-6]). It is well known that in the resonant absorption of IR laser radiation by the atmospheric molecular gases a redistribution of molecules on the energy levels of internal degree of freedom occurs and the saturation of absorption results in the changes of the absorption coefficient of gas [1]. The change of level population for gas composition leads to the disturbance of thermodynamic equilibrium between the oscillations of molecules and its translation. Because of this circumstance an effect of the kinetic cooling of environment may take a place, as it was at first predicted in ref. [2,5]. It should be noted that a new effect of kinetical cooling CO_2 in a process of absorption of the laser energy by molecular gas was considered for the middle latitude atmosphere and for special form of a laser pulse. Besides, there were used approximate values for constants of collisional deactivation and resonant transfer in reaction CO_2-N_2. At the same time using more precise values for all constants may lead to quantitative changing temporary dependence of the resonant absorption coefficient by CO_2. This is accompanied by additional important effects. The formation and accumulation of the excited molecules of nitrogen owing to the resonant transfer of excitation from the molecules CO_2 results in the change of environment polarizability. Perturbing the complex conductivity of environment, all these effects are able to transform significantly the impulse energetics of IR lasers in an atmosphere and significantly change realization of different non-linear laser-aerosol effects. An effective example is the pulsed enlightenment of artificial water aerosol by the CO_2 laser radiation [2]. In this paper we study a kinetics of energy and heat exchange in the mixture CO_2-N_2-H_20 gases in atmosphere under passing the powerful CO_2 laser radiation pulses and given the estimate for realization of the kinetical cooling effect by CO_2 for different atmosphere conditions and laser pulses parameters (pulse form, duration etc.).

To describe the energy exchange and relaxation processes in the CO_2.– N_2 — H_2O mixture, which interacts with laser radiation, we start from the modified three-mode model of kinetic processes [5,6]. We consider a kinetics of three levels: 10°0, 00°1 (CO_2) and v = 1 (N_2). Availability of atmospheric constituents O_2 and H_2O is allowed for the definition of the rate of vibrating-transitional relaxation of N_2. The system of balance equations for relative populations is written in a standard form as follows:

$$\frac{dx_1}{dt} = -\beta(\omega + 2gP_{10})x_1 + \beta\omega x_2 + 2\beta gP_{10})x_1^0 ,$$

$$\frac{dx_2}{dt} = \omega x_1 - (\omega + Q + P_{20})x_2 + Qx_3 + P_{20}x_2^0 , \tag{1}$$

$$\frac{dx_3}{dt} = \delta Q x_2 - (\delta Q + P_{30})x_3 + P_{30}x_3^0 .$$

Here, $x_1 = N_{100}/ N_{CO_2}$, $x_2 = N_{001}/ N_{CO_2}$, $x_3 = \delta N_{N_2} / N_{CO_2}$; $N_{100,}$ N_{001} are the level populations 10°0, 00°1 (CO_2); N_{N_2} is the level population v = 1 (N_2); N_{CO_2} is the concentration of CO_2 molecules; δ is the ratio of the common concentrations of CO_2 and N_2 in the atmosphere ($\delta = 3.85 \cdot 10^{-4}$); x_1^0 , x_2^0 and x_3^0 are the equilibrium relative values of populations under gas temperature T:

$$x_1^0 = \exp\left(-E_1/T\right), \tag{2}$$

$$x_2^0 = x_3^0 = \exp\left(E_2/T\right);$$

Values E_1 and E_2 in (1) are the energies (K) of levels 10°0, 00°1 (consider the energy of quantum N_2 equal to E_2); P_{10}, P_{20} and P_{30} are the probabilities (s^{-1}) of the collisional deactivation of levels 10°0, 00°1 (CO_2) and v = 1 (N_2), Q is the probability (s^{-1}) of resonant transfer in the reaction $CO_2 \rightarrow N_2$, ω is the probability (s^{-1}) of CO_2 light excitation, $g = 3$ is the statistical weight of level 02°0, в = $(1+g)^{-1} = 1/4$.

The solution of system (1) allows defining a coefficient of absorption of the radiation by the CO_2 molecules according to the formula:

$$\alpha_{CO_2} = \sigma(x_1 - x_2)N_{CO_2} . \tag{3}$$

The у in (3) is dependent upon the thermodynamical medium parameters as follows [2]:

$$\sigma = \sigma_0 \frac{P}{P_0} \left(\frac{T}{T_0} \right)^{1/2}, \quad (4)$$

Here T and p are the air temperature and pressure, y_0 is the cross-section of resonant absorption under $T = T_0$, $p = p_0$.

It is well known that the absorption coefficient for carbon dioxide and water vapour is dependent upon the thermodynamical parameters of aerosol atmosphere. In particular, for radiation of CO_2-laser the coefficient of absorption by atmosphere $\alpha_g = \alpha_{CO_2} + \alpha_{H_2O}$ is equal in conditions, which are typical for summer mid-latitudes, $б_g(H=0) = 2.4 \cdot 10^6$ cm^{-1}, from which $0.8 \cdot 10^6$ cm^{-1} accounts for CO_2 and the rest — for water vapour (data are from ref. [2]) . On the large heights the sharp decrease of air moisture occurs and absorption coefficient is mainly defined by the carbon dioxide. The physics of resonant absorption process is defined by changing the population of low level 10°0 (CO_2), population of level 00°1 and the vibrating-transitional relaxation (VT-relaxation) and the inter modal vibrating-vibrating relaxation (VV'-relaxation), which redistribute the energy between the vibrating and transitional freedom of the molecules. According to ref.[1], the threshold value, which corresponds to the decrease of absorption coefficient in two times, for the strength of saturation of absorption in vibrating-rotary conversion give $I_{sat} = (2 \div 5)\ 10^5$ W cm^{-2} for atmospheric CO_2. In this case the pulse duration t_i must satisfy the condition $t_R << t_i < t_{VT}$, where t_R and t_{VT} are the times of rotary and vibrating-transitional relaxation's. by The fast exchange of level 10°0 with basic state, and by the relatively slow relaxation of high level 00°1 defines a renewal process of thermodynamic equilibrium is characterized. This can results in an energy outflow from the transitional degree of freedom onto vibrating ones and in the cooling of environment.

In table 1 we present the data for the relative coefficient of absorption $\bar{\alpha}_{CO_2}$, which is normalized on the linear coefficient of absorption, calculated using (1) on corresponding height H. All data for $\bar{\alpha}_{CO_2}$ are obtained for the height distribution of pressure and temperature according to the mid-latitude atmospheric model [2] (A-data from ref.[2]; B — data of present paper).

In table 2 there are presented analogous our data for the relative coefficient of absorption $\bar{\alpha}_{CO_2}$ and the height distribution of pressure and temperature are chosen for the Odessa-like atmospheric conditions according to atmospheric model [7,8]. It is clear that the temporary dependence of resonant absorption relative coefficient $\bar{\alpha}_{CO_2}$ of laser radiation by CO_2 molecules for rectangular and gauss laser pulses differs.

A significant aspect of modelling is connected with the correct choice of probabilities P_{10}, P_{20} and P_{30} of the collisional deactivation of levels 10°0, 00°1 (CO_2) and v = 1 (N_2), probability Q of resonant transfer in the reaction $CO_2 \rightarrow N_2$, probability ω of CO_2 light excitation and other constants. We have used the calculated and compiled data for all constants, which are, in our opinion, to be more correct ones at present time (c.f. [9-11] and refs in it). A quality of choice of the molecular constants may be significant under modelling the effect of kinetic cooling of the CO_2 under propagation of the laser radiation in atmosphere. Let us note further that an effect of kinetic cooling of the CO_2 is defined by the following condition [1]:

$$\alpha^0_{\mathrm{H_2O}} < (E_1/(E_2 - E_1))\alpha^0_{\mathrm{CO_2}} = 1.44\alpha^0_{\mathrm{CO_2}} \tag{5}$$

Table 1

Temporary dependence of resonant absorption relative coefficient $\bar{\alpha}_{\mathrm{CO_2}}$ (sm^{-1}) of laser radiation (λ=10,6μm) by CO_2 for rectangular (R) laser pulses (intensity I, 10^5 W/sm^2) on the height (H, km) for the mid-latitude atmospheric model [1]: A- data of modelling [2]; B- (present paper).

T μs	A [2] I; R H=0	A [2] 10·I; R H=0	A[2] I;R H=10	A[2] 10·I;R H=10	B I;R H=0	B 10·I; R H=0	B I; R H=10	B 10·I; R H=10	B I; G H=0	B 10·I; G H=0	B I; G H=10	B 10·I; G H=10
0	1,0	1,0	1,0	1,0	1,0	1,0	1,0	1,0	1,0	1,0	1,0	1,0
1	0,94	0,60	0,48	0,12	0,93	0,57	0,45	0,11	0,91	0,55	0,40	0,10
2	0,88	0,52	0,34	0,08	0,85	0,48	0,31	0,05	0,82	0,43	0,25	0,03
3	0,90	0,63	0,41	0,27	0,88	0.60	0,36	0,19	0,85	0.56	0,30	0,17
4	0,91	0,67	0,48	0,35	0,90	0,65	0,43	0,28	0,89	0,61	0,36	0,25

Table 2

Temporary dependence of the resonant absorption relative coefficient $\bar{\alpha}_{\mathrm{CO_2}}$ (sm^{-1}) of laser radiation (λ=10,6μm) by CO_2 for gauss (G) laser pulse (intensity *I*, 10^5 W/sm^2) on the height (*H*, km):

T μs	B I; G H=0	B 10·I; G H=0	B I; G H=10	B 10·I; G H=10
0	1,0	1,0	1,0	1,0
1	0,91	0,55	0,40	0,10
2	0,82	0,43	0,25	0,03
3	0,85	0.56	0,30	0,17
4	0,89	0,61	0,36	0,25

Taking into account the data of the tables 1 and 2 one can conclude and wait for the changing CO_2 kinetic cooling effect realization in dependence upon the atmospheric model conditions and parameters of laser radiation. The effect of kinetic cooling vanishes under some critical intensity. According to [2], under large intensities of radiation the energy flux, which is responsible for an existence of effect of kinetic cooling, from translational degrees of freedom into vibrating ones reaches the maximal magnitude and not depends on the intensity of incident radiation. But, the energy flux, which results in the heating of gas, onto translational degrees of freedom due to the absorption of radiation by the water vapor remains proportional to the intensity of radiation. Consequently, starting with some critical intensity, the gas heating prevails over its cooling for any time instant. The more less exact estimate of the correspondent values must be based on using correct data for the atmospheric conditions, laser radiation and molecular parameters.

References

1. Zuev V.E., Zemlyanov A.A., Kopytin Y.D. Nonlinear Optics of Atmosphere.- L.:Hidrometeoizd., 1991. — 256 p.
2. Zuev V.E., Zemlyanov A.A., Kopytin Y.D., Kuzikovsky A.V. Powerful laser radiation in an atmospheric aerosol. — Novosybirsk, 1984. — 224 p.
3. Bagratashvili V.N., Letokhov V.S., Makarov A.A., Ryabov E.A. Multi-Photon Processes in Molecules in Infrared laser field. — M.: Nauka, 1991 — 180 p.
4. Glushkov A.V., Prepelitsa G.P. Polischuk et al QED Theory of non-linear interaction of the complex atomic systems with laser field. Multiphoton Resonances // J. Techn. Phys. — 1997. — Vol. 38. — P. 219-224.
5. Gordiets B.F., Osipov A.I., Khokhlov R.V. About cooling the gas under powerful CO_2 laser radiation passing in atmosphere// J.Techn.Phys. — 1974. — Vol. 14. — P. 1063-1069.
6. Ambrosov S.V., Prepelitsa G.P. Spectroscopy of Atmosphere gases atoms and molecules: Nonlinear Spectroscopic Effects// Proc.32nd EPS Conf. EGAS. — Vilnius. — 2000. — P. 30.
7. Glushkov A.V., Mahmud A.A. Calculation of turbulent ventilation of industrial city// Meteorology and hydrology. — 1999. — №38. — P. 77-61.
8. Mahmud A.A. Calculation of turbulent regime of industrial city in a zone of dry subtropics// Meteorology and hydrology. — 1999. — №38. — P. 182-186.
9. Glushkov A.V., Ambrosov S.V., Malinovskaya S.V. et al. Spectroscopy of carbon dioxide: Oscillator strengths and energies of transitions in spectra of CO_2 // Optics and Spectroscopy. — 1996. — T. 80, № 1. — C. 60-65.

А. В. Тюрин , Г. П. Препелица, В. П. Козловская

Кинетика энерго и тепло-обмена в смеси CO_2-N_2-H_20 атмосферных газов, взаимодействующей с ИК лазерным излучением: Уточненная 3-модовая модель кинетических процессов

АННОТАЦИЯ

Рассмотрена кинетика энерго- и тепло-обмена в смеси CO_2-N_2-H_20 атмосферных газов при прохождении через атмосферу мощного излучения CO_2 лазера в рамках уточненной 3-модовой модели кинетических процессов. Дана оценка условий реализации эффекта кинетического охлаждения углекислого газа в процессе поглощения лазерной энергии молекулярным газом для различных атмосферных условий и параметров лазерного импульса.

А. В. Тюрін , Г. П. Препелица, В. П. Козловська

Кінетика енерго-і тепло-обміну у сумішу CO_2-N_2-H_20 атмосферних газів, що взаємодіють з ІЧ лазерним випромінюванням: Уточнена 3-модова модель кінетичних процесів

АНОТАЦІЯ

Розглянуто кінетику енерго- й тепло-обміну у сумішу CO_2-N_2-H_20 атмосферних газів при проходженні скрізь атмосферу міцного випромінювання CO_2 лазера у межах уточненої 3-модової моделі кінетичних процесів. Наведено оцінку умов реалізації ефекта кінетичного охолодження вулекислого газу в процесі поглинення лазерної енергії молекулярним газом для різних атмосферних умов й параметрів лазерного імпульсу