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PHOTOKINETICS OF THE INFRARED LASER RADIATION EFFECT ON MIXTURE OF THE CO_2-N_2-H_2O ATMOSPHERIC GASES: ADVANCED MODEL

Within the refined 3-modal model of kinetic processes there is examined a photokinetics of the energy exchange in a mixture of CO_2-N_2-H_2O atmospheric gases while passing powerful laser CO_2 radiation through an atmosphere.
Key words*: photokinetics, laser field, mixture of gases, refined 3-model model*

1. Introduction

At present time the environmental physics has a great progress, provided by implementation of the modern quantum electronics and laser physics methods and technologies in order to study unusual features of the "laser radiation- substance (gases, solids etc.) interaction. A special interest attracts a problem of interaction of the powerful laser radiation with an aerosol ensemble and search of new non-linear optical effects. The latter is directly related with problems of modern aerosol laser physics (c.f.[1-15]). One could remind that there is a redistribution of molecules on the energy levels of internal degree of freedom in the resonant absorption of IR laser radiation by the atmospheric molecular gases. As a result of quite complicated processes one could define an essential changing of the gases absorption coefficient due to the saturation of absorption [1]. One interesting effect else to be mentioned is an effect of the kinetic cooling of environment (mixture of gases), as it was at first predicted in ref. [2,5]. Usually the effect of kinetical cooling (CO_2) in a process of absorption of the laser pulse energy by molecular gas is considered for the middle latitude atmosphere and for special form of a laser pulse. Besides, the approximate values for constants of collisional deactivation and resonant transfer in reaction CO_2-N_2 are usually used.

In series of papers (see, for example, [8-11]) computational modelling energy and heat exchange kinetics in the mixture of the CO_2-N_2-H_20 atmospheric gases interacting with IR laser radiation has been carried out within master three-mode kinetical model. It is obvious that using more precise values for all model constants and generally speaking the more advanced atmospheric model parameters may lead to quantitative changing in the temporary dependence of the resonant absorption coefficient by CO_2. So, in the last there are developed more refined, advanced models [12-15] regarding determination of the atomic and molecular constants and correspondingly interaction of the atoms and molecules of the atmosphere gases with a powerful laser radiation. Let us remind that the creation 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. This paper, which goes on our work on atmospheric optins and photochemistry [8-15], is devoted to receiving more refined data on 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. The master model is the refined atmospheric model and generalized three-mode kinetical model with using refined values for all molecular, kinetical and photochemical constants.

2. Advanced three-mode kinetical model for the "laser pulse – medium" interaction

As usually, we start from the modified three-mode model of kinetic processes (see, for example, [11]) in order to take into consideration the energy exchange and relaxation processes in the CO_2.– N_2 – H_2O mixture interacting with a laser radiation. As in ref. [11-13] 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:

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

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

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

Here the following notations are used:

$$x_1 = N_{100}/N_{CO_2},$$
$$x_2 = N_{001}/N_{CO_2}, \qquad (2)$$
$$x_3 = \delta N_{N_2}/N_{CO_2}.$$

where 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 ($\delta = 3.85\cdot10^{-4}$); x_1^0, x_2^0 and x_3^0 are the equilibrium relative values of populations under gas temperature T:

$$x_1^0 = \exp(-E_1/T), \qquad (3)$$
$$x_2^0 = x_3^0 = \exp(E_2/T).$$

The 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, $\beta = (1+g)^{-1} = 1/4$.

As usually, the solution of the differential equations system (1) allows defining a coefficient of absorption of the radiation by the CO_2 molecules according to the formula:

$$\alpha_{CO_2} = \sigma(x_1 - x_2)N_{CO_2}. \qquad (4)$$

The σ in (4) is dependent upon the thermodynamical medium parameters as follows [2]

$$\sigma = \sigma_0 \frac{P}{P_0}\left(\frac{T}{T_0}\right)^{1/2}. \qquad (5)$$

Here T and p are the air temperature and pressure, σ_0 is the cross-section of resonant absorption under $T = T_0$, $p = p_0$.

One could remind 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 defined as

$$\alpha_g = \alpha_{CO_2} + \alpha_{H_2O}$$

is equal in conditions, which are typical for summer mid-latitudes, $\alpha_g(H=0) = 2.4 \cdot 10^6$ 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 changing population of the low level 10°0 (CO_2), population of the level 00°1, the vibrating-transitional relaxation (VT-relaxation) and the inter modal vibrating-vibrating relaxation (VV'-relaxation) processes define the physics of resonant absorption processes. Moreover, the above indicated processes result in a redistribution of 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 $t_R \ll t_i < t_{VT}$, 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°1define a renewal process of thermodynamic equilibrium is characterized. The latter provides an energy outflow from the transitional degree of freedom onto vibrating ones and in the cooling of environment. It is easily understand that using more powerful laser radiation sources can lead to a strong non-linear interaction phenomena and, as result, significantly change a photo-kinetics of the corresponding processes.

3. The results and conclusion

In table 1 we present mode accurate our data (column C) for the relative coefficient of absorption $\overline{\alpha}_{CO_2}$, which is normalized on the linear coefficient of absorption, calculated using (1) on corresponding height H. All data for $\overline{\alpha}_{CO_2}$ are obtained for the height distribution of the pressure and temperature according to the advanced mid-latitude atmospheric model (all data are presented in series of refs. [1,16-19]). In table 1 there are presented also the analogous data from ref. [2] (column A) , from ref. [9-11] (column B).

In ref.[,2 11-13] the analogous data for the relative coefficient of absorption $\overline{\alpha}_{CO_2}$ and the height distribution of pressure and temperature are presented and obtained in a case of using the Odessa-latitude atmospheric conditions according to atmospheric model [7,8]. Here we use the world atmospheric model conditions. In is important to note that preliminary more refined values of the 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 have been determined (in comparison with results of refs. [9-11] and further used in the model.

Table 1 -Temporary dependence of resonant absorption relative coefficient

T μs	A [2] 10·I; R H=0	A[2] 10·I;R H=10	B [13] 10·I; G H=0	B [13] 10·I; G H=10	C 10·I; G H=0	C 10·I; G H=10
0	1,0	1,0	1,0	1,0	1,0	1,0
1	0,60	0,12	0,57	0,13	0,52	0,10
2	0,52	0,08	0,46	0,05	0,40	0,04
3	0,63	0,27	0.59	0,19	0.56	0,14
4	0,67	0,35	0,64	0,28	0,59	0,23

$\overline{\alpha}_{CO_2}$ (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- data of modelling [13], C- this work.

Let us in conclusion to note that obviously a quality of choice of the corresponding molecular constants and the corresponding atmospheric model parameters is of a great importance in modelling the effect of kinetic cooling of the CO_2 under propagation of the laser radiation in atmosphere. In any case it provides a variance of the final results within 5-10% interval. From the other, side, we did not consider more exotic situation with using super intense laser pulses (for example, in the radio frequency range). In the last case it is necessary to say about "climatic weapon" and this problem requires using another approaches. Obviously, another intensity of the kinetical processes will be observed here.

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Фотокінетіка суміші CO2-N2-H20 атмосферних газів під дією ІК лазерного випромінювання: Узагальнена модель

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Розглянута фотокінетіка процесів обміну енергії в суміші CO2-N2-H20 атмосферних газів при проходінні через атмосферу потужного випромінювання CO2-лазера в рамках узагальненої 3-модовой моделі.

Ключові слова: *фотокінетіка, лазерне поле, суміш газів, поліпшена 3-модовая модель*

Фотокинетика смеси CO_2-N_2-H_20 атмосферных газов под действием ИК лазерного излучения: Обобщенная модель

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Рассмотрена фотокинетика процессов обмена энергии в смеси CO_2-N_2-H_20 атмосферных газов при прохождении через атмосферу мощного излучения CO_2-лазера в рамках обобщенной 3-модовой модели.

Ключевые слова: *фотокинетика, лазерное поле, смесь газов, улучшенная 3-модовая модель*