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MODELING OF NONLINEAR OPTICAL EFFECTS IN THE INTERACTION OF LASER RADIATION WITH ATMOSPHERE AND SENSING FOR ENERGY EXCHANGE IN A MIXTURE ATMOSPHERIC GASES

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Abstract. It is presented an advanced quantum-kinetic model to describe the nonlinear-optical (spectroscopic) effect caused by the interaction of infrared laser radiation with a gas atmosphere. We determine the quantitative features of energy exchange in a mixture of CO_2-N_2-H_2O atmospheric gases of atmospheric gases, which can be used in the development of new sensory spectroscopic technologies for observing the state of the atmosphere.

Keywords: kinetics of energy exchange, gases in atmosphere, laser radiation, sensing

МОДЕЛЮВАННЯ НЕЛІНІЧНИХ ОПТИЧНИХ ЕФЕКТІВ ВЗАЄМОДІЇ ЛАЗЕРНОГО ВИПРОМІНЮВАННЯ З АТМОСФЕРОЮ І ДЕТЕКТУВАННЯ ЕНЕРГООБМІННИХ ПРОЦЕСІВ В СУМІШУ АТМОСФЕРНИХ ГАЗІВ

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Анотація. Розроблено вдосконалену квантово-кінетичну модель для опису нелінійно-оптичного (спектроскопічного) ефекту, спричиненого взаємодією інфрачервоного лазерного випромінювання з атмосферою. Визначені кількісні особливості обміну енергією в суміші CO_2-N_2-H_2O атмосферних газів, які можуть бути використані при розробці нових сенсорних спектроскопічних технологій спостереження за станом атмосфери.

Ключові слова: кінетика енергообміну, атмосферні гази, випромінювання лазера, детектування

МОДЕЛИРОВАНИЕ НЕЛИНЕЙНЫХ ОПТИЧЕСКИХ ЭФФЕКТОВ ВЗАИМОДЕЙСТВИЯ ЛАЗЕРНОГО ИЗЛУЧЕНИЯ С АТМОСФЕРОЙ И ДЕТЕКТИРОВАНИЕ ЭНЕРГООБМЕННЫХ ПРОЦЕССОВ В СМЕСИ АТМОСФЕРНЫХ ГАЗОВ

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Аннотация. Разработана усовершенствованная квантово-кинетическая модель для описания нелинейно-оптического (спектроскопического) эффекта, вызванного взаимодействием инфракрасного лазерного излучения с атмосферой. Определены количественные особенности обмена энергией в смеси CO_2-N_2-H_2O атмосферных газов, которые могут быть использованы при разработке новых сенсорных спектроскопических технологий наблюдения за состоянием атмосферы.

Ключевые слова: кинетика энергообмена, атмосферные газы, излучение лазера, детектирование

1. One of the most important problems in the modern sensor electronics, molecular and environmental physics is connected with a searching new physical effects and construction of new sensors (e.g. [1-7]). In this paper we present theoretical fundamentals a new, improved quantum-kinetic atomic-molecular approach to theoretical modeling of nonlinear optical (spectroscopic) effects in the interaction of electromagnetic (laser) radiation with the gas atmosphere of an industrial city and quantitatively elucidate the features of energy CO_2-N_2 in the energy-N_2 exchange of atmospheric gases during the passage of powerful laser radiation pulses. This topic is of a great importance for further solving the problems of laser (lidar) sounding of atmosphere and creation new sensor devices on the laser system basis [1-21]. The required theoretical modeling is based on the numerical solution of the differential equation system, which describes the time evolution of

the relative populations of levels of atmospheric gas molecules.

First, let us consider qualitatively some fundamental aspects of the interaction of electromagnetic radiation with atoms and molecules of the atmospheric environment. Indeed, in the case of an intense external field, a nonlinear response of atoms and molecules will obviously occur. It should be noted that cases of both resonant and non-resonant interaction of electromagnetic radiation with atoms and molecules of atmospheric gases can be considered separately.

The obvious consequence of resonant interaction (in particular, absorption) of electromagnetic radiation (hereinafter, as a rule, will be coherent, that is, laser radiation) by molecular gases of the atmosphere is the quantitative redistribution of molecules by the energy levels of internal degrees of freedom. In turn, this will change the so-called gas absorption coefficient. Changing the population levels of the mixture of gases causes a disturbance of thermodynamic equilibrium between the vibrations of molecules and their translational motion, resulting in kinetic cooling of the environment.

At the same time, as shown in [2,5], it is very important to use more realistic and accurate values of constant constants in the corresponding quantum-kinetic models. For example, we are talking about realistic data regarding the dependence of the resonance absorption coefficient of CO_2 (and other atmospheric gases too) over time.

At the same time, in the interaction of laser radiation with a mixture of atmospheric gases, relatively complex processes of resonant excitation transfer, in particular, from CO_2 molecules to nitrogen molecules, will take place. As a result, a quantitative change in the polarizability of the atmosphere will be observed. As a result, the complex dielectric constant of the atmospheric medium will change, which will lead to a significant transformation of the energy of laser pulses in the gas atmosphere [1-4].

Indeed, in a nonlinear medium, the dielectric constant depends on the intensity of the electromagnetic wave *I*:

$$\varepsilon = \varepsilon(I) = \bar{\varepsilon}_0 + \varepsilon_N(I) \qquad (1)$$

$$I = \frac{c\sqrt{\varepsilon_0}}{8\pi}\left|\vec{E}\right|^2$$

where *c* is the speed of light, *E* is the electric field strength of the wave.

Obviously, expression (1) defines a specific type of nonlinear interaction - nonlinear response of the medium. In (1) the index “0” indicates the undisturbed value of the dielectric constant:

$$\bar{\varepsilon}_0 = \varepsilon_0 + i\alpha_0 / k_0\,, \qquad (2)$$

and the index “N” - the corresponding increase due to nonlinear interaction. It should be noted that a generalization of equation (1) in the case of propagation of radiation in an aerosol medium leads to introduction of the corresponding additive [1]:

$$\bar{\varepsilon}_a = \sum_{\nu=1}^{N} \varepsilon_{a\nu}(\vec{r} - \vec{r}_\nu)\,, \qquad (3)$$

which is the sum of the perturbations of the complex dielectric constant from the individual centers. In (3), the vector $\vec{r}_\nu$ determines the position of the particles in space, N is the total number of particles. The value $\bar{\varepsilon}_a$ is equal to the value of the complex dielectric constant of the particle and its halo, when the observation point is inside the localized inhomogeneity, and is equal to zero otherwise. The halos around the aerosol particles are due to the perturbation of the dielectric constant due to temperature, vapor, or plasma inhomogeneities (see more details in Refs. [1-6]). The latter result from the nonlinear interaction of laser radiation with the substance of particles.

When laser radiation interacts with atoms and molecules of atmospheric gases, there is also the so-called Kerr electronic effect, which arises due to the deformation of the electron density distributed by the field, almost immediately following the change of field, as well as the orientation effect of Kerr [1] . The relaxation time of this effect for atmospheric air under normal conditions is 10^{-13} s. This effect leads to the dependence of the dielectric constant on the field of the electromagnetic wave in the formula (1) of the form

$$\varepsilon_N = \varepsilon_2 \left|E\right|^2 . \qquad (4)$$

Based on the measurement of nonlinear air polarization, it was shown [3] that the contribution of the electronic mechanism to nonlinear air polarization is very small, and the value of the constant for air is $5 \cdot 10^{-16}$ units SGSE. For Gaussian beams and plateau beams, the Kerr effect leads to the self-focusing of light, described in detail, for example, in [1-4]. If the length of the nonlinear interaction (self-focusing) is a Gaussian beam with radius R_0

$$L_N = \frac{R_0}{\sqrt{\varepsilon_2 \left|E\right|^2}} = R_0 \left(\frac{8\pi\varepsilon_2}{c\sqrt{\varepsilon_0}} I \right)^{-1/2} , \qquad (5)$$

then the realization of the effect on distance $L_{||}$ is possible if the threshold intensity is defined [1]:

$$I_{ПОР} \geq \frac{c\sqrt{\varepsilon_0}}{8\pi} \frac{R_0^2}{\varepsilon_2 L_{||}^2} . \qquad (6)$$

$I_{ПОР} \approx 10^{10}$ W·cm^{-2} for $R_0 = 0{,}1$

and $L_{||} = 10^3$ m. If $L_{||} = 10^5$ m, then $I_{ПОР} \approx 10^8$ W·cm^{-2}.

For infrared laser wavelength λ =10.6 μm, the critical autofocus ($L_{||} = L_d$) power is as follows:

$$P_{кр} = \pi R_0^2 I_{ПОР} = \frac{c\sqrt{\varepsilon_0}}{8k^2\varepsilon_2} = 1{,}7 \cdot 10^{11} \text{ W}. \qquad (7)$$

Correspondingly, one has $P_{кр} = 1{,}7 \cdot 10^9$ W for λ =1,06 μm.

2. Here we construct an improved quantum-kinetic model to describe the nonlinear-optical (spectroscopic) effect caused by the interaction of infrared laser radiation with a gas atmosphere and consider the quantitative features of energy exchange in a mixture of CO_2-N_2-H_20 atmospheric gases of atmospheric gases [2,5].

Typically, for the quantitative description of energy exchange and the corresponding relaxation processes in a mixture of CO_2-N_2-H_20 gases in the laser radiation field, one should first consider the kinetics of three levels: 10°0, 00°1 (CO_2) і v = 1 (N_2). The system of differential equations of balance for relative populations is written in the following form:

$$\begin{aligned} \frac{dx_1}{dt} &= -\beta(\omega + 2gP_{10})x_1 + \beta\omega x_2 + 2\beta g P_{10})x_1^0 + F_N(x_1) , \\ \frac{dx_2}{dt} &= \omega x_1 - (\omega + Q + P_{20})x_2 + Qx_3 + P_{20}x_2^0 + F_N(x_2), \\ \frac{dx_3}{dt} &= \delta Q x_2 - (\delta Q + P_{30})x_3 + P_{30}x_3^0 + F_N(x_3) . \end{aligned} \qquad (8)$$

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_{CO_2} is concentration of CO_2 molecules; N_{N_2} is the level population v=1(N_2); Q is the probability (s^{-1}) of resonant transfer in the reaction $CO_2 \rightarrow N_2$,ω is a probability (s^{-1}) of CO_2 light excitation, g = 3 is statistical weight of level 02°0, $\beta=(1+g)^{-1}$= ¼; δ is ratio of common concentrations of CO_2 and N_2 in atmosphere ($\delta = 3.85 \cdot 10^{-4}$); $F_N(x)$ – additional nonlinear term; x_1^0, x_2^0 and x_3^0 are the equilibrium relative values of populations under gas temperature T:

$$\begin{aligned} x_1^0 &= \exp(-E_1/T), \\ x_2^0 = x_3^0 &= \exp(E_2/T) \end{aligned} \qquad (9)$$

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).

Note that having obtained the solution of the differential equation system (8), one can further calculate the absorption coefficient of radiation by CO_2 molecules:

$$\alpha_{CO_2} = \sigma(x_1 - x_2) N_{CO_2} . \qquad (10)$$

The σ in (10) is dependent upon the thermodynamical medium parameters according to [1]. The different estimates (c.g.[1-5]) show that for emission of the CO_2-laser the absorption coefficient:

$$\alpha_g = \alpha_{CO_2} + \alpha_{H_2O}\,. \qquad (11)$$

is equal in conditions, which are typical for summer mid-latitudes $\alpha_g(H=0) = (1.1\text{-}2.6)\cdot 10^6$ 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. T

It is known [2-5] that the resonance absorption by the molecules of the atmospheric mixture of laser radiation is determined by the change in the population of the low-lying level 10°0 (CO_2), the population of the level 00°1 and vibration-translational relaxation (VT-relaxation), as well as intergenerational vibration relaxation (VV'-relaxation). For the wavelength of infrared laser radiation (eg, CO_2 laser of 10.6μm), the duration of the corresponding pulse will satisfy the inequality $t_R \ll t_i < t_{VT}$", where t_R , t_{VT} are the values of time, respectively, of rotational and oscillatory relaxation. For accurate numerical calculations, it is important to accurately determine the probabilities of P_{10}, P_{20} , P_{30} deactivation due to the levels of 10°0, 00°1 (CO_2) and v = 1 (N_2), the probability of Q resonance energy transfer $CO_2 \rightarrow N_2$, the excitation probability ω pulse of CO_2 laser and other constants.

computing was performed with using the PC code Superatom [24-28]). It is clear that the time dependence of the relative resonance absorption coefficient of laser radiation by CO_2 molecules for different laser pulses differs. In Table 1 we list the Temporary dependence of resonant absorption relative coefficient $\overline{\alpha}_{CO_2}$ (sm^{-1}) for rectangular (R), gaussian (G) and soliton-like (S) laser pulses (intensity I, 10^5 W/sm^2) on the height H=10km : A- data of modelling [1,2]; B and C- our data.

The effect of kinetic cooling of the CO_2 is determined by the condition (for Odessa region):

$$\alpha^0_{H_2O} < (E_1/(E_2 - E_1))\alpha^0_{CO_2} = 1.51\alpha^0_{CO_2}\,. \quad (12)$$

Note that expression (12) is significantly different from early qualitative estimates [1,2,5]. The numerical parameters obtained allow us to further quantify the effects of the kinetic cooling of CO_2, depending on the parameters of the model of the atmosphere and the parameters of laser radiation. The analysis shows that the energy flux that causes the gas to heat through the absorption of water vapor radiation is proportional to the intensity of the laser radiation.

Table 1.

Temporary dependence of resonant absorption relative coefficient $\overline{\alpha}_{CO_2}$ (sm^{-1}) for rectangular (R), gaussian (G) and soliton-like (S) laser pulses (intensity I, 10^5 W/sm^2) on the height H=10km : A- data of modelling [2]; B and C –our data.

t μs	A I=10^5 R	A I=10^6 R	B I=10^5 R	B I=10^6 R	B I=10^5 G	B I=10^6 G	C I=10^5 S	C I=10^6 S
0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
1	0.48	0.12	0.46	0.12	0.41	0.11	0.43	0.10
2	0.34	0.08	0.32	0.07	0.26	0.04	0.29	0.05
3	0.41	0.27	0.37	0.19	0.31	0.18	0.34	0.19
4	0.48	0.35	0.44	0.29	0.37	0.26	0.39	0.27

Below we present the results of computing the relative absorption coefficient $\overline{\alpha}_{CO_2}$ (normalized to linear absorption coefficient) based on the solutions of the system (8). All data obtained for the distribution of pressure altitude and temperature are taken from the model of the atmosphere of the middle latitudes (Odessa) [22, 23]. The

At the same time, when the critical value of the critical parameter is reached, the heating of the steam will prevail over its cooling for any moment of time. In such a physical situation, the effect of kinetic cooling will cease to exist. In any case, the quantitative manifestation of the kinetic effect may vary for different atmospheric conditions, laser radiation parameters, and different values of atomic-molecular parameters (set of energy, spectroscopic and radiation characteristics). Obviously, this will significantly influence and appropriately determine the energy conditions of the laser sounding of the atmosphere of an industrial city, and the latter, in turn, will redefine the quantitative possibilities of finding quantitative characteristics of spatio-temporal fields of concentrations of substances in the atmosphere of an industrial city.

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Summary

The aim of the work is to develop and present a new approach for advanced analysis, modeling of nonlinear optical effects in the interaction of laser radiation with atmosphere and studying the quantitative features of the energy exchange in a mixture atmospheric gases. The obvious consequence of resonant interaction (in particular, absorption) of electromagnetic radiation (a rule, coherent, that is, laser radiation) by molecular gases of the atmosphere is the quantitative redistribution of molecules by the energy levels of internal degrees of freedom. In turn, this will change the so-called gas absorption coefficient. Changing the population levels of the mixture of gases causes a disturbance of thermodynamic equilibrium between the vibrations of molecules and their translational motion, resulting in kinetic cooling of the environment. It is presented an advanced quantum-kinetic model to describe the nonlinear-optical (spectroscopic) effect caused by the interaction of infrared laser radiation with a gas atmosphere. We determine the quantitative features of energy exchange in a mixture of CO_2-N_2-H_2O atmospheric gases of atmospheric gases, which can be used in the development of new sensory spectroscopic technologies for observing the state of the atmosphere. The results of computing the relative absorption coefficient (normalized to linear absorption coefficient) are presented.

Keywords: kinetics of energy exchange, gases in atmosphere, laser radiation, sensing

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МОДЕЛЮВАННЯ НЕЛІНІЧНИХ ОПТИЧНИХ ЕФЕКТІВ ВЗАЄМОДІЇ ЛАЗЕРНОГО ВИПРОМІНЮВАННЯ З АТМОСФЕРОЮ І ДЕТЕКТУВАННЯ ЕНЕРГООБМІННИХ ПРОЦЕСІВ В СУМІШУ АТМОСФЕРНИХ ГАЗІВ

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Реферат

Метою роботи є розробка нового підходу до вдосконаленого аналізу, моделювання нелінійних оптичних ефектів при взаємодії лазерного випромінювання з атмосферою та вивчення кількісних особливостей обміну енергією в сумішу атмосферних газів. Внаслідок резонансної взаємодії (зокрема, поглинання) електромагнітного випромінювання молекулярними газами атмосфери має місце кількісний перерозподіл молекул за енергетичними рівнями внутрішніх ступенів свободи. У свою чергу, це змінює званий коефіцієнт поглинання відповідного газу. Зміна рівня населеностей суміші газів спричиняє порушення термодинамічної рівноваги між коливаннями молекул та їх поступальним рухом, що призводить до кінетичного охолодження середовища. Представлена вдосконалена квантово-кінетична модель для опису нелінійно-оптичного (спектроскопічного) ефекту, спричиненого взаємодією інфрачервоного лазерного випромінювання з атмосферою газу. Представлена вдосконалена квантово-кінетична модель для опису нелінійно-оптичного (спектроскопічного) ефекту, спричиненого взаємодією інфрачервоного лазерного випромінювання з атмосферою газу. Визначені кількісні особливості обміну енергією в суміші атмосферних газів CO_2-N_2-H_2O атмосферних газів, які можуть бути використані при розробці нових сенсорних спектроскопічних технологій спостереження за станом атмосфери. Представлені результати обчислення відносного коефіцієнта поглинання (нормованого до лінійного коефіцієнта поглинання).

Ключові слова: кінетика енергообміну, атмосферні гази, випромінювання лазера, детектування