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Three-block model of the kinetics of vibrationally excited $I_2(X)$ molecules in the active media of oxygen-iodine lasers

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Abstract. A three-block model of the kinetics of vibrationally excited $I_2(X)$ molecules in the active media of chemical oxygen-iodine lasers is developed. Instead of the system of equations describing a change in the concentrations of $I_2(X, v)$ (v = 0 - 47) molecules, this model uses equations for the total concentrations of iodine molecules belonging to the blocks of vibrational levels with $v \le 10$, v = 11 - 24, and $v \ge 25$. Effective deactivation rate constants of $I_2(X, 11 \le v \le 24)$ molecules are found for laser media of different compositions. The results of calculations performed using the proposed model agree with experimental data and are close to the parameters calculated previously by using the total system of equations for populations of individual vibrational levels of $I_2(X, v)$.

Keywords: chemical oxygen-iodine laser, vibrationally excited iodine molecules, effective deactivation rate constants.

1. Introduction

Oxygen-iodine lasers (OILs) are promising sources of coherent radiation with a power up to several megawatt [1]. At present, OILs are extensively studied for different purposes: development of chemical [2–8] and gas-discharge [9–14] generators of singlet oxygen $O_2(a^1\Delta)$, investigation of kinetic processes in the active media of OILs [15–30], improvement of energy characteristics [31–41], etc. Numerical simulation of kinetic processes allows one to considerably decrease the cost of optimisation of OIL characteristics [25, 26, 42, 43]. However, simulation of three-dimensional gas-dynamic reacting flows is complicated by a variety of chemical and energy-exchange processes occurring in the active media of lasers.

Figure 1 shows the energy levels of I, O₂, I₂, and H₂O molecules that are most important in the kinetics of processes occurring in the active media of chemical OILs (COILs). Hereinafter, the atoms and molecules $I(^{2}P_{3/2})$, $I(^{2}P_{1/2})$, $O_{2}(X^{3}\Sigma)$, $O_{2}(a^{1}\Delta)$, $O_{2}(b^{1}\Sigma)$, $I_{2}(X^{1}\Sigma)$, $I_{2}(A'^{3}\Pi_{2u})$, and $I_{2}(A^{3}\Pi_{1u})$ are denoted as I, I^{*}, $O_{2}(X)$, $O_{2}(a)$, $O_{2}(b)$, $I_{2}(X)$, $I_{2}(X)$, $I_{2}(A')$, and $I_{2}(A)$, respectively. About 50 vibrational levels of $I_{2}(X)$ are taken into account in calculations using a multichannel model of dissociation of iodine molecules [21, 44]. More than 200 reactions for more than 70 components describe the kinetics of processes in the active media of COILs, and the complete

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Figure 1. Energy levels of the I atom and O₂, H₂O, and I₂ molecules.

account of them is a difficult problem in the numerical calculation of laser characteristics, especially in the case of threedimensional simulation [25, 26, 42, 43].

The main goal of this work is to simplify the kinetic model of COILs for convenience of calculations. In the proposed model, the system of equations for populations in vibrationally excited molecules $I_2(X, v)$ with v > 10 is reduced to two equations for the total populations of iodine molecules at vibrational levels with v = 11-24 and $v \ge 25$. The effective deactivation rate constants of $I_2(X,11 \le v \le 24)$ molecules are calculated taking into account conditions in different active media of COILs. The applicability of this model for calculating the rates of I_2 dissociation observed in recent experiments was verified.

2. Three-block model of the kinetics of vibrationally excited I₂(X) molecules in the active medium of COILs

It is well known that excited iodine atoms I^\ast are rapidly quenched by $I_2(X)$ molecules in the electronic-vibrational process

$$I^* + I_2(X) \to I + I_2(X, v > 10)$$
. (1)

A high relative concentration of $I_2(X,v)$ molecules excited to vibrational levels with $v \le 45$ was observed in [45–47]. It was experimentally shown that reaction (1) leads to the formation of vibrationally excited $I_2(X, 25 \le v \le 43)$ molecules [45]. In [45], excited iodine atoms were obtained as a result of photolysis either of the I₂-Ar mixture by radiation with λ = 475 nm or of the CF₃I – I₂ – Ar mixture by radiation with λ = 266 nm. $I_2(X,v)$ molecules were also recorded in the $I_2-O_2(a)-O_2$ mixture by laser-induced fluorescence (LIF) [46, 47]. In experiments of [46, 47], $O_2(a)$ molecules were obtained using a gas-discharge generator, and I₂ vapours were injected into the oxygen flow. In the presence of singlet oxygen, iodine molecules dissociated and excited I* atoms were formed as a result of the EE-exchange process $I + O_2(a)$ \rightarrow I^{*} + O₂(X). In [46], I₂(X,v) molecules with vibrational quantum numbers $33 \le v \le 44$ were detected. A distribution of the absolute concentration of iodine molecules over vibrational levels with $5 \le v \le 45$ was experimentally found in [47].

To describe the mechanism of vibrational excitation of iddine molecules, it is necessary to know the probabilities $\gamma_{\rm m}$ of the formation of $I_2(X)$ molecules at the *v*th vibrational level in reaction (1). Paper [48] presents the distribution of γ_{ν} (in arbitrary units) over vibrational levels with $24 \le v \le 47$, which was obtained from the analysis of measured relative populations of $I_2(X, v)$. This distribution has a maximum at v = 40. Experiments performed in [47] revealed that the concentration of molecules $I_2(X, v = 11-23)$ formed immediately after injection of molecular iodine into the flow with singlet oxygen significantly exceeds the concentration of vibrationally excited iodine molecules with $v \ge 30$. So high populations of vibrational levels of $I_2(X)$ molecules with $10 \le v \le 23$ can be explained only by their direct pumping in the course of reaction (1) [21, 49, 50]. Analysis of the concentrations of $I_2(X, v)$ molecules measured in [47] shows that the distribution of γ_v over vibrational levels has two maxima, at v = 40and v = 20-22 [21, 49, 50]. The unusual double-humped distribution of γ_v can be explained by the existence of two different mechanisms of quenching of I^* by $I_2(X)$ molecules [49]. The probabilities of excitation of the vth vibrational level of $I_2(X)$ molecules in reaction (1) were estimated in [49] by comparing the calculated populations of $I_2(X, v)$ with experimental values. A satisfactory agreement with experimental data was achieved when the total probabilities of vibrational exci-tation of I₂(X) molecules were $\Gamma_{25 \le v \le 47} = \sum_{v=25}^{47} \gamma_v \approx 0.1$ and $\Gamma_{15 \le v \le 24} = \sum_{v=15}^{24} \gamma_v \approx 0.9$. Relaxation of I₂(X, v) molecules was studied in [45–48].

Relaxation of $I_2(X, v)$ molecules was studied in [45–48]. The rate constants of vibrational–rotational relaxation of $I_2(X, v, J)$ molecules are given in [48] for vibrational–rotational levels with v = 23, J = 57; v = 38, J = 49; and v = 42, J = 17. The rate constants $K_{v,v-1}^M$ of the VT relaxation processes $I_2(X, v) + M \rightarrow I_2(X, v - 1) + M$ were determined for v = 23 and 38 and M = He, Ar, N₂, O₂, Cl₂, I₂, H₂O from the numerical analysis of LIF spectra of iodine molecules. No dependence of $K_{v,v-1}^M$ on the vibrational level number was found. In [48], it was supposed that the VT relaxation rate constants of $I_2(X)$ molecules are determined by the expressions $K_{v,v-1}^M = v^s K_{1,0}^M$, where the value of *s* lies within the range of 0-1.

It is reasonable to divide the vibrational levels of $I_2(X, v)$ molecules into three blocks: levels with $0 \le v \le 10$, $11 \le v \le 24$, and $v \ge 25$. Let us assume that $I_2(X, v)$ molecules dissociate upon collisions with $O_2(a)$ molecules if $v \ge 25$,

$$I_2(X, v \ge 25) + O_2(a) \to 2I + O_2(X).$$
 (2)

In this case, we achieve good agreement between the distribution of concentrations of $I_2(X, v)$ molecules over vibrational levels calculated in [21 49, 50] and the experimental distribution found in [47], which demonstrates a steep fall of the $I_2(X, v)$ concentration at v > 24. The $I_2(X, v)$ molecules at the vibrational levels with $11 \le v \le 24$ can participate in the formation of electronically excited molecules $I_2(A')$:

$$I_2(X, 11 \le v \le 24) + O_2(a) \to I_2(A') + O_2(X).$$
 (3)

Different estimates of the rate constant of process (3) give $10^{-12}-2 \times 10^{-11}$ cm³ s⁻¹ [21, 25]. The vibrationally excited iodine molecules with $v \le 10$ make no considerable contribution to the dissociation of I₂ molecules because their energy is not high enough to form any intermediate state upon collisions with $O_2(a)$ molecules. In the proposed three-block model of vibrational excitation and relaxation of I₂(X, v) molecules, the I₂(X, v ≤ 10) molecules will be considered as molecules at the effective zero vibrational level. A temporal evolution in the concentration $N_{I_2}(v)$ of I₂(X, v) molecules with $v \ge 11$ in the active medium of COILs is described by the equations

$$\frac{\mathrm{d}N_{\mathrm{I}_{2}}(v)}{\mathrm{d}t} = \gamma_{v}K_{\mathrm{I}}N_{\mathrm{I}^{*}}N_{\mathrm{I}_{2}} - K_{3}(v)N_{\mathrm{a}}N_{\mathrm{I}_{2}}(v)$$

$$+ \sum_{\mathrm{M}} [K_{v-1,v}^{\mathrm{M}}N_{\mathrm{I}_{2}}(v-1) + K_{v+1,v}^{\mathrm{M}}N_{\mathrm{I}_{2}}(v+1)$$

$$- K_{v,v-1}^{\mathrm{M}}N_{\mathrm{I}_{2}}(v) - K_{v,v+1}^{\mathrm{M}}N_{\mathrm{I}_{2}}(v)]N_{\mathrm{M}}$$
(4)

for v = 11 - 24 and

$$\frac{dN_{I_2}(v)}{dt} = \gamma_v K_I N_{I^*} N_{I_2} - K_2(v) N_a N_{I_2}(v)$$

$$+ \sum_{M} [K_{v-1,v}^M N_{I_2}(v-1) + K_{v+1,v}^M N_{I_2}(v+1) - K_{v,v-1}^M N_{I_2}(v) - K_{v,v+1}^M N_{I_2}(v)] N_M$$
(5)

for v = 25, 26, ... Here, N_{I^*}, N_a , and N_M are the concentrations of I^{*} atoms, $O_2(a)$ molecules, and M component (M = O_2 , N_2 , He, CO_2), respectively; K_1 is the rate constant of reaction (1); $K_{v,v-1}^{M}$ and $K_{v-1,v}^{M}$ are the rate constants of the direct and reverse processes $I_2(X,v) + M \leftrightarrow I_2(X,v-1) + M$; $K_3(v)$ and $K_2(v)$ are the rate constants of the processes $O_2(a)$ + $I_2(X,v) \rightarrow O_2(X) + I_2(A')$ and $O_2(a) + I_2(X,v) \rightarrow O_2(X) + 2I$, respectively. A decrease in the number density of $I_2(X, v)$ molecules in these processes is described by the second terms in Eqns (4) and (5). The first terms in these equations are the rates of pumping of the vth vibrational level of $I_2(X,v)$ in reaction (1). The sum of terms in square brackets is the rate of direct and reverse cascade VT processes on the assumption that the change in the level number in these processes is $\Delta v =$ ±1. In the calculations, it is assumed that $K_{v,v-1}^{M} = vK_{1,0}^{M}$, and the values of $K_{v-1,v}^{M}$ are calculated using the detailed balance principle. The $K_{1,0}^{M}$ constants are given in [48]. In addition, it is supposed that the rate constants $K_2(v)$ and $K_3(v)$ do not depend on the vibrational level number, i.e., $K_2(v) = K_2$ and $K_3(v) = K_3$. Then, summing Eqns (4) over $11 \le v \le 24$ and Eqns (5) over $25 \le v \le 47$ and assuming that $N_{\rm I_2}(v) = 0$ for v > 047, we obtain

$$\frac{\mathrm{d}N^*}{\mathrm{d}t} = \Gamma_{11 \le v \le 24} K_1 N_1^* N_{\mathrm{I}_2} - K_3 N_a N_1^*$$

$$-\sum_{M} K_{1 \to 0}^{M} N^{*} N_{M} + \sum_{M} K_{2 \to 1}^{M} N^{**} N_{M}, \qquad (6)$$

$$\frac{\mathrm{d}N^{**}}{\mathrm{d}t} = \Gamma_{25 \leqslant v \leqslant 47} K_1 N_{\mathrm{I}^*} N_{\mathrm{I}_2} - K_2 N_{\mathrm{a}} N^{**} - \sum_{\mathrm{M}} K_{2 \to 1}^{\mathrm{M}} N^{**} N_{\mathrm{M}}.$$
 (7)

Here, N^* and N^{**} are the total concentrations of $I_2(X, v)$ molecules at the levels with v = 11-24 (effective vibrational level 1) and with v = 25-47 (effective vibrational level 2), respectively;

$$K_{1 \to 0}^{M} = \frac{K_{11,10}^{M} \{ N_{I_{2}}(11) - \exp[-\Delta E_{11}^{I_{2}}/(kT)] N_{I_{2}}(10) \}}{N^{*}},$$

$$K_{2 \to 1}^{M} = \frac{K_{25,24}^{M} \{ N_{I_{2}}(25) - \exp[-\Delta E_{25}^{I_{2}}/(kT)] N_{I_{2}}(24) \}}{N^{**}}$$

are the effective deactivation rate constants of $I_2(X, 11 \le v)$ \leq 24) and I₂(X, 25 $\leq v \leq$ 47) molecules due to collisions with molecules or atoms of M component [51]; $\Delta E_v^{I_2}$ is the difference in the energies of $I_2(X, v)$ molecules at the levels with the numbers v and v - 1. Note that the probability $\Gamma_{11 \le v \le 24} = \sum_{v=11}^{24} \gamma_v$ in (6) is equal to the above-mentioned total probability $\Gamma_{15 \le v \le 24}$ if $\gamma_v = 0$ for v = 11 - 14. It is obvious that the terms describing the changes in the concentrations of $O_2(a)$, $I_2(X)$, and $I_2(A')$ in reactions (2), (3) will include the variables N^* and N^{**} introduced into consideration and will not include the populations of individual vibrational levels of $I_2(X)$ molecules. It is reasonable to assume that, in contrast to reactions (2) and (3), the reactions in which the degree of vibrational excitation of $I_2(X)$ is unspecified involve only the $I_2(X, 0 \le v \le 10)$ molecules. The concentrations of these molecules only slightly differs from the total $I_2(X)$ concentration. Thus, instead of the system with a large number of equations for populations of $I_2(X, v)$ ($0 \le v \le 47$) molecules [44], one can use in calculation only the equation for the concentration of $I_2(X)$ molecules and Eqns (6), (7) for the total populations N^* and N^{**} of iodine molecules at vibrational levels with $11 \le v \le 24$ and $25 \le v \le 47$. It is important that this does not require knowledge of the probabilities γ_v of formation of $I_2(X, v)$ molecules at each vibrational level, since Eqns (6), (7) contain only the total probabilities $\Gamma_{11\leq}$ $_{v \leq 24}$ and $\Gamma_{25 \leq v \leq 47}$.

For calculating $K_{1 \rightarrow 0}^{M}$ and $K_{2 \rightarrow 1}^{M}$, the concentrations $N_{I_2}(10), N_{I_2}(11), N_{I_2}(24), N_{I_2}(25), N^*$, and N^{**} can be found from the numerical solution of Eqns (4), (5) together with the equations of chemical and vibrational kinetics of processes in the active medium of COILs [44]. In this work, we calculated N^* , N^{**} , $K_{1 \rightarrow 0}^{M}$, and $K_{2 \rightarrow 1}^{M}$ for typical conditions in the COIL active media of the composition $O_2(X)$: $O_2(a)$: N_2 (He, CO₂) = 0.5:0.5: ε (ε = 1-3). The calculated values of $K_{1\to 0}^{\rm M}$ and $K_{2\rightarrow 1}^{M}$ depend on the used value of K_{3} and on the relative content ε of the buffer gas in the mixture. In all the cases, K_{1-0}^{M} , $K_{2\rightarrow 1}^{\text{M}} \ll K_2$ and $N^*/N^{**} \ge 100$. This means that the rate of the transition from the effective vibrational level 2 to the effective level 1 is much lower than the rate of process (2) and the rate of pumping of the effective vibrational level 1 in process (1). The calculation results show that the last terms in Eqns (6) and (7) are significantly smaller than the other terms and can be neglected for the sake of simplicity. It is important to note that the calculated rate constants $K_{1 \to 0}^{M}$ are almost independent of time, which proves that they can be used as effective constants. The calculated effective deactivation rate constants of $I_2(X, 11 \le v \le 24)$ molecules in the active media of COILs at different rate constants of reaction (3) are listed in Table 1.

Table 1. Effective deactivation rate constants of $I_2(X, 11 \le v \le 24)$ molecules in the $O_2(X): O_2(a): N_2$ (He, $CO_2) = 0.5: 0.5: \varepsilon$ mixture.

$\frac{K_3}{\mathrm{cm}^3 \mathrm{s}^{-1}}$	ε	$\frac{K^{O_2}_{1 \to 0}}{10^{-12} \text{ cm}^3 \text{ s}^{-1}}$	$\frac{K_{1 \to 0}^{N_2}}{10^{-12} \text{ cm}^3 \text{ s}^{-1}}$	$\frac{K_{1 \to 0}^{\text{Ne}}}{10^{-12} \text{ cm}^3 \text{ s}^{-1}}$	$\frac{K_{1 \to 0}^{\rm CO_2}}{10^{-12} \rm \ cm^3 \ s^{-1}}$
10-12	1	1.8	2.3	2.7	4.1
	2	1.9	2.4	2.7	4.3
	3	2.0	2.5	2.8	4.3
10-11	1	1.3	1.7	1.9	2.9
	2	1.5	1.9	2.2	3.4
	3	1.6	2.1	2.4	3.7
2×10^{-11}	1	0.9	1.1	1.3	2.0
	2	1.2	1.5	1.7	2.6
	3	1.3	1.7	2.0	3.0

3. Calculation results

We numerically solved the system of differential equations describing changes in the relative concentrations η_i (concentrations divided to the total concentration of oxygen) of atoms I^{*}, molecules $O_2(a)$, $O_2(b)$, $I_2(X)$, $I_2(A')$, $I_2(A)$, and vibrationally excited molecules $O_2(v)$ (v = 1, 2, 3, 4) and $H_2O(010)$, as well as changes in the gas temperature, on the distance x = Vt (V is the gas flow rate) [44]. In these calculations, instead of equations for the relative concentrations of $I_2(X, v)$ molecules, we used equations describing a change in the relative total concentrations $\eta^* = N^*/N_{\rm O_2}$ and $\eta^{**} =$ N^{**}/N_{O_2} , with distance x, which are analogous to Eqns (6), (7). The last terms in these equations, which contain the constants $K_{2 \to 1}^{M}$, were assumed to be zero, while the constants $K_{1 \to 0}^{M}$ for $M = O_2$ and N_2 were taken from Table 1. The initial values of the component concentrations, temperature, and gas flow rates corresponded to the experimental data of [21]. Figure 2 shows the concentrations of I* atoms measured in [21] for the $O_2: N_2=1:1$ mixture with the water vapour concentration



Figure 2. Dependences of the concentrations of I^{*} atoms on the distance *x* along the flow for the O₂: N₂ = 1:1 mixture with the water vapour concentration $\eta_w = 3\%$ and the initial relative concentration of singlet oxygen $\eta_a = 50\%$ at the initial iodine vapour concentrations $\eta_{12} = 1.23\%$ (mixture pressure p = 2.2 Torr) [\circ , (1)], 0.94% (p = 2.1 Torr) [\Box , (2)], 0.52% (p = 2 Torr) [\triangle , (3)], and 0.29% (p = 1.9 Torr) [\blacksquare , (4)]. Symbols show the experimental data of [21]. Solid curves are the dependences calculated in this paper, and dashed curves show the results calculated in [21, 44] using equations for the populations of individual vibrational levels of I₂(X).

 $\eta_{\rm w}$ = 3% and the initial relative concentration of singlet oxygen $\eta_a = 50\%$ at the initial iodine vapour concentrations changing from 0.29% to 1.23%. Figure 2 also presents the dependences of I_2 concentrations on the distance x along the flow calculated in the present work and the results of [21, 44] calculated using equations for populations of individual vibrational levels of $I_2(X)$. In our calculations, the rate constant of reaction (3) was taken to be 10^{-11} cm³ s⁻¹, and the total probabilities of vibrational excitation of $I_2(X)$ molecules in process (1) were $\Gamma_{15 \le v \le 24} = 0.9$ and $\Gamma_{25 \le v \le 47} = 0.1$. Figure 3 presents the distributions of the concentration of excited iodine atoms along the flow at the initial relative concentration of iodine vapours $\eta_{I_2} = 0.94\%$ for different concentrations of the buffer gas N_2 in the mixture. Figures 2 and 3 demonstrate that the calculated dependences well agree with experimental data. The calculation results obtained in [21, 44] only slightly differ from the results of calculations performed in this work.



Figure 3. Dependences of the concentrations of I^{*} atoms on the distance *x* at the initial relative concentration of iodine vapour $\eta_{1_2} = 0.94\%$ and different concentrations of nitrogen buffer gas in the mixture: N₂: O₂ = 1:1 (*p* = 2.1 Torr) [\Box , (1)], 2:1 (*p* = 3 Torr) [\Box , (2)], and 3:1 (*p* = 3.8 Torr) [Δ , (3)]. Symbols are the experimental data of [21]. Solid curves show the dependences calculated in this work.

4. Conclusions

Thus, in this work we developed a three-block model of the kinetics of vibrationally excited iodine molecules $I_2(X, v)$ in the active medium of a COIL. Instead of a system with a large number of equations describing the change in the concentration of $I_2(X, v)$ molecules, this model uses equations for total concentrations of iodine molecules within blocks of vibrational levels with $v \le 10$, v = 11-24, and $v \ge 25$. Taking into account the conditions typical for the active media of COILs, we found effective deactivation rate constants of $I_2(X, 11 \le v \le 24)$ molecules upon collisions with O_2 , N_2 , CO_2 molecules and He atoms. The results of calculations performed using the three-block model equations well agree with experimental data and are close to the data calculated previously using the system of equations for the populations of individual vibrational levels of $I_2(X)$ molecules.

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