

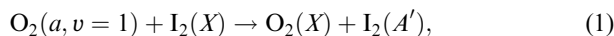
Distribution of O₂ molecules over vibrational levels at the output of a singlet-oxygen generator

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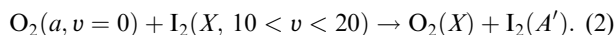
Abstract. Simple formulas are obtained for determining the population of the vibrational levels of singlet oxygen generated chemically in a singlet-oxygen generator. The rate of decrease in the vibrational energy of oxygen is limited by the exchange between its first vibrational level and the bending mode of the water molecule. It is shown that the populations of singlet oxygen molecules at the second and third vibrational levels are comparable with the population of oxygen in the excited electronic state $b^1\Sigma_g^+$. The possibility of formation of electronically excited iodine in the reaction $O_2(a^1A_g, v=2) + I_2(X) \rightarrow O_2(X^3\Sigma_g^-) + O_2(A^3\Pi_{1u})$, which may be the intermediate state in the process of dissociation of iodine in singlet-oxygen medium, is substantiated.

Keywords: singlet oxygen, vibrational kinetics, iodine dissociation.

The emission spectra of an oxygen dimole at the output of a chemical singlet-oxygen generator (SOG) were recorded in Ref. [1]. A comparison of the intensities of the emission bands at 579 and 634 nm showed a considerable deviation of the population of oxygen at the first vibrational level from equilibrium. The vibrationally excited singlet oxygen (SO) may play a significant role in the formation of the active medium of an oxygen–iodine laser, especially during the dissociation of I₂ [2, 3], whose mechanism has not been established unequivocally so far. In subsequent publications [3, 4], a number of arguments were put forth suggesting that the intermediate state during the dissociation of iodine is its excited electronic state I₂(A' ³Π_{2u}). It was assumed in Ref. [3] that this state is populated through the reaction



while the following reaction was proposed in Ref. [4]:



Hereafter, O₂(X), O₂(a), O₂(b) are the oxygen molecules in the electronic states $X^3\Sigma_g^-$, a^1A_g , and $b^1\Sigma_g^+$, respectively,

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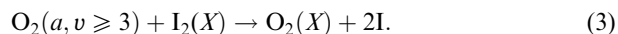
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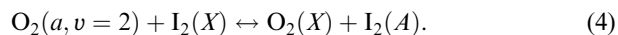
Translated by Ram Wadhwa

I₂(X), I₂(A'), and I₂(A) are the iodine molecules in the electronic states $X^1\Sigma_g^+$, $A'^3\Pi_{2u}$, $A^3\Pi_{1u}$, respectively.

Vibrationally excited SO with $v > 1$ can also be involved in the dissociation of I₂. For example, it was assumed in Ref. [5] that iodine dissociates during the process



Note that the excitation energies of the states of O₂(a, $v=2$) and I₂(A) are almost identical (equal to 10823 and 10847 cm⁻¹, respectively). It can be assumed that the resonance energy exchange between these states occurs in the process

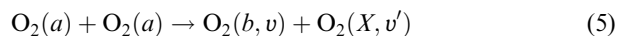


The I₂(A) state was observed during the dissociation of I₂ in Ref. [6]. This excited electronic state can also be the intermediate state in the dissociation of iodine in the SO medium.

Thus, the processes involving the vibrationally excited SO may dominate during the dissociation of molecular iodine, and hence it should be certainly interesting to determine the population of vibrational states of O₂(a, v) during the formation of the active medium of the oxygen–iodine laser. This work is devoted to the study of the distribution of oxygen molecules over the vibrational levels at the output of a SOG.

The gaseous mixture at the output of the SOG consists of oxygen, chlorine Cl₂ unreacted in the generator, a buffer gas (N₂, Ar or He), water (H₂O) and hydrogen peroxide (H₂O₂) vapours. The vibrational kinetics in this gaseous mixture is mainly determined by the processes involving O₂ and H₂O molecules [3]. We will assume that the mixture emerging from the SOG consists of only two gases – oxygen and water vapour, with characteristic concentrations $N_O \approx 10^{17} - 10^{18} \text{ cm}^{-3}$ and $N_w \approx 10^{15} - 3 \times 10^{16} \text{ cm}^{-3}$, respectively. Oxygen may be in one of the electronic states $X^3\Sigma_g^-$, a^1A_g and $b^1\Sigma_g^+$ with concentrations N_X, N_a, N_b , and the fraction of the electronically excited singlet oxygen O₂(a) in the entire mass of oxygen may be significant: $\eta_A = N_a/N_O \approx 0.4 - 1$, where $N_O = N_X + N_a + N_b$.

The relaxation of SO in the pooling reaction



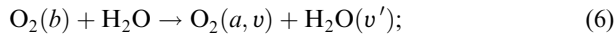
occurs with a characteristic time $\tau_A = (K_5 N_a)^{-1} \approx 0.01 - 0.1 \text{ s}$, where K_5 is the rate constant for reaction (5). Such a lifetime makes it possible to transport SO over relatively

Table 1. Rate constants for the reactions in the O₂ – H₂O medium

Rate constant $K_i/\text{cm}^3 \text{ s}^{-1}$	References
$K_5 = 2 \times 10^{-17}$	[7]
$K_6 = 6.7 \times 10^{-12}$	[8]
$K_7 = 10^{-12}$	[9, 10]
$K_8 = 1.7 \times 10^{-11}$	[11]
$K_9^O = 2 \times 10^{-18}$	[12]
$K_9^w = 10^{-16}$	[13]
$K_{10} = 3 \times 10^{-13}$	[3, 14]
$K_{11} = 5 \times 10^{-11}$	[15]

large distances with insignificant losses. Table 1 contains the numerical values of the rate constants of reactions employed by us.

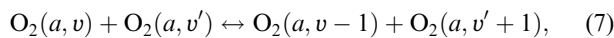
The electronically excited oxygen O₂(*b*) formed in reaction (5) is quenched upon collisions with water molecules:



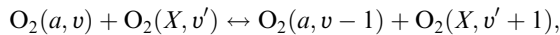
the characteristic quenching time being $\tau_6 = (K_6 N_w)^{-1} \approx 10^{-5} - 10^{-4}$ s. The steady-state concentration N_b of the electronically excited oxygen in the gas flow is established during the same time. The fraction of O₂(*b*) in the total mass of oxygen $\eta_b = N_b/N_O$ is insignificant and can be determined to a high degree of accuracy from the balance of the rates of its production in reaction (5) and its reduction in reaction (6): $\eta_b = K_5 \eta_A^2 / (K_6 \eta_w) \approx 10^{-5} - 10^{-4}$ (see Table 1 and Fig. 1), where $\eta_w = N_w/N_O$.

The vibrational levels of oxygen are populated at the output of SOG during electronic energy deactivation processes (5) and (6). Oxygen is produced in reaction (5) mainly at the second vibrational level [16]. The information about the distribution of vibrational energy in the products of reaction (6) is scarce. The probability of the vibrational excitation of H₂O(001) was estimated as 0.1 in Ref. [17]. Apparently, the main part of the energy released in the reaction is spent for exciting the vibrational levels of oxygen up to $v = 3$.

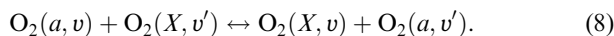
The redistribution of the vibrational quanta between the vibrational levels of SO occurs during the VV exchange:



while the redistribution between the electronic states occurs during the VV' exchange



and the EE exchange



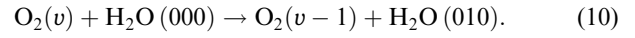
The characteristic time of the vibrational-distribution establishment in oxygen is $\tau_{VV} = (K_7 N_O)^{-1} \approx 10^{-6} - 10^{-5}$ s, which is much smaller than the characteristic relaxation time of SO.

A distinguishing feature of the oxygen medium is the low rate of VT relaxation (see Table 1)

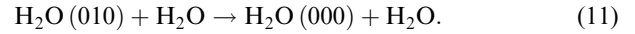


with a characteristic time $\tau_{VT} = (K_9^w N_w + K_9^O N_O)^{-1} \approx 0.1 -$

1 s. The conversion of the vibrational energy of O₂ into thermal energy mainly occurs as follows [3]. At the first stage, oxygen transfers vibrational quanta to water molecules during the process



At the second stage, the vibrational energy is transferred to the translational degrees of freedom during the process



It has been established experimentally [1] that the fraction of vibrationally excited oxygen at the first vibrational level at the SOG output does not exceed a few percent. The population of oxygen at the levels $v \geq 2$ is even smaller than at the first level, i.e., $\eta_i = N_i/N_O \ll 1$ ($i = 1, 2, \dots$), where N_i is the oxygen concentration at the i th vibrational level. Note that $\tau_{VT} \gg \tau_{VV}$, $\eta_w \ll \eta_O$. In this case, the vibrational quanta are transferred to the first vibrational level of oxygen in fact without any losses during the VV and VV' exchange processes. Because the concentration N_1 of oxygen at the first vibrational level is much higher than at the higher vibrational levels, the process (10) makes a significant contribution to a decrease in the number of vibrational quanta of oxygen for the first level only.

Under quasi-equilibrium conditions, the concentration of oxygen molecules at the first vibrational level and of water molecules H₂O(010) can be determined from the balance between the rates of formation and disappearance of these molecules under the assumption that the distribution of vibrational energy is established rapidly for all three electronic states of oxygen:

$$nK_5 N_a^2 - K_{10} N_1 N_w + K_{10} N_{w1} N_O \exp(\Delta E/kT) - N_1 (K_9^O N_O + K_9^w N_w) + (K_9^O N_O^2 + K_9^w N_O N_w) \exp(-E_1/kT) = 0, \quad (12)$$

$$K_{10} N_1 N_w - K_{10} N_{w1} N_O \exp(\Delta E/kT) + mK_5 N_a^2 - K_{11} N_w N_{w1} + K_{11} N_w^2 \exp(-E_w/kT) = 0. \quad (13)$$

Here, N_{w1} is the concentration of H₂O(010) molecules; $\Delta E = E_w - E_1$; E_w and E_1 are the energies of the vibrational level of H₂O(010) and of the first level of O₂; n is the total number of vibrational quanta of O₂ formed in reactions (5) and (6); m is the average number of vibrational quanta for the bending mode of the water molecule, which are excited in reaction (6); K_9^M and K_{10} are the rate constants of processes (9) and (10) for $v = 1$. Because $\tau_A \gg \tau_6$, the deactivation rate of the electron energy is limited by process (5). Eqn (13) takes into account the fact that the VV exchange between the valence and bending modes of H₂O occurs much faster than the VT relaxation of valence modes [3, 15].

The deactivation of the electronic energy in reaction (6) may be accompanied by the excitation of SO molecules to vibrational levels up to $v = 3$. Estimates show that vibrationally excited oxygen with $v = 2, 3$ may play an active role in the formation of the active medium of the oxygen – iodine laser if its fraction in the total mass of oxygen is $\eta_i \geq 10^{-5}$. The concentration of oxygen molecules at these levels can be

determined from the balance equations analogous to (12) and (13):

$$\begin{aligned} \gamma_2 K_5 N_a^2 + K_{1,1} N_1^2 + K_{3,0} N_3 N_O \\ - K_{2,0} N_2 N_O - K_{2,1} N_2 N_1 = 0, \end{aligned} \quad (14)$$

$$\gamma_3 K_5 N_a^2 + K_{2,1} N_2 N_1 - K_{3,0} N_3 N_O = 0, \quad (15)$$

where γ_i is the sum of the excitation probabilities of the i th vibrational level of O_2 in processes (5) and (6), and $K_{i,j}$ are the rate constants of the VV exchange [18]. The first terms in Eqns (14) and (15) describe the pumping rate for the corresponding level in processes (5) and (6), while the remaining terms describe the single-quantum exchanges between the vibrational levels of oxygen. Only the most significant processes have been taken into account in Eqns (14) and (15). If the pumping rate for the i th level in processes (5) and (6) is much lower than the VV exchange rate (i.e., the first term in Eqns (14) and (15) is much smaller than the remaining terms), the well-known Treanor distribution of oxygen molecules over vibrational levels is established [18]:

$$N_n = N_O \exp(-\theta n) \exp(-E_n/kT), \quad (16)$$

where E_n is the energy of the n th vibrational level of the O_2 molecule; a parameter θ does not depend on the level number and can be expressed in terms of the population of the first vibrational level: $\theta = \ln(N_O/N_1) - E_1/kT$. The population N_1 of the first level is determined from Eqns (12) and (13).

Consider the characteristic parameters of a gaseous medium, which are realised at the output of a SOG, namely, $\eta_A = 0.4 - 1$ and $\eta_w = 0.01 - 0.1$. The case $\eta_w \ll 0.01$ was considered in Ref. [5]. The rate constants of process (10) were not measured directly. According to the estimates obtained in Ref. [3], $K_{10} \approx 3 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$, which coincides with the value calculated in Ref. [14]. For $K_{10} \geq 10^{-14} \text{ cm}^3 \text{ s}^{-1}$, the last and the next to last terms in Eqn (12) can be neglected. Going over to the fractions of the components, we obtain from Eqns (12) and (13)

$$\eta_1 = n \frac{K_5 \eta_A^2}{K_{10} \eta_w} + (n+m) \frac{K_5 \eta_A^2}{K_{11} \eta_w^2} \exp(\Delta E/kT) + \exp(-E_1/kT), \quad (17)$$

$$\eta_{w1} = (n+m) \frac{K_5 \eta_A^2}{K_{11} \eta_w} + \eta_w \exp(-E_w/kT), \quad (18)$$

where $\eta_{w1} = N_{w1}/N_O$.

Considering that $K_{1,1} \cong K_{2,0}$ and $K_{2,1} \cong K_{3,0}$, we obtain from (14) and (15) the relative population of oxygen molecules at the second and third vibrational levels:

$$\eta_2 = \eta_1^2 + (\gamma_2 + \gamma_3) K_5 \eta_A^2 / K_{1,1}, \quad \eta_3 = \eta_1 \eta_2 + \gamma_3 K_5 \eta_A^2 / K_{2,1}. \quad (19)$$

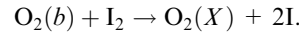
We are mainly interested in the vibrational population of SO. The relative population $\eta_{Ai} = N_{Ai}/N_O$ of SO at the i th vibrational level, can be found from the total relative population η_i assuming a rapid EE exchange in process (8):

$$\eta_{Ai} = \frac{\eta_i}{(1/\eta_A - 1) \exp(-\Delta E_i/kT) + 1}, \quad (20)$$

where ΔE_i is the difference in the energies of the i th vibrational level of the O_2 molecule in the states $^3\Sigma$ and $^1\Delta$.

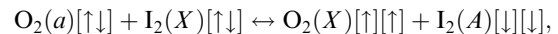
It follows from expression (17) that for the composition of the medium considered here and for $K_{10} \gg 3 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$, the energy transfer from a vibrational to a thermal reservoir will be limited by process (11), and the vibrational population of the oxygen will slightly decrease. On the other hand, the deactivation of vibrationally excited molecules for $K_{10} \leq 3 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$ is limited by process (10). It follows from (17)–(20) that the relative population of the vibrational levels of oxygen is determined by the medium composition and the rate constants of the process only and does not depend on the pressure of the medium. The fraction of vibrationally excited oxygen at the first level at the output of the SOG is several percent. For example, for $\eta_A = 0.8$, $\eta_w = 0.01$, $K_{10} = 3 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$ and $T = 300 \text{ K}$ ($n = 4$, $m = 0.2$), the fraction of vibrationally excited oxygen is $\eta_1 = 4.5\%$, while for $K_{10} = 3 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$, we have $\eta_1 = 2.6\%$. Although the rate constant K_{10} is higher by an order of magnitude in the latter case, the value of η_1 decreases only by a factor of 1.7.

In the $^1\Sigma$ state, the fraction $\eta_b = N_b/N_O$ of oxygen in the gaseous medium is insignificant and is determined to a high degree of accuracy from the condition that processes (5) and (6) are stationary (see Fig. 1). It was assumed for a long time that I_2 dissociates in collisions with the electronically excited oxygen $O_2(^1\Sigma)$ in a process with a rate constant close to the constant for the gas-kinetic process ($2 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$) [19]:



Direct measurements of the rate constant showed that it is much smaller ($4 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$) [20]. However, the process with the gas-kinetic rate constant provides a fairly correct description of certain observed dissociation rates in the oxygen–iodine laser. The authors of Ref. [6] treat this as an accidental coincidence.

Several mechanisms of dissociation of I_2 are being considered at present [3, 4]. Each channel considered in the literature makes its own contribution to the process of dissociation. The main problem lies in determining the dominating channel among these. We propose another possible channel for dissociation of iodine and will try to substantiate it. It is assumed that the electronically excited state $I_2(A)$, which is populated as a result of collisions with the vibrationally excited singlet oxygen $O_2(a, v = 2)$ [process (4)], is the intermediate state in the process of dissociation. The rate of EE exchange depends strongly on the defect in the excitation energy of the colliding partners [21, 22]. In this case, the energy gap is only 24 cm^{-1} , and hence the process of population of $I_2(A)$ may be quite rapid. The transfer of the electronic energy between the molecules considered here is allowed only under the condition of conservation of total spin of the system (the Wigner rule). Indeed, process (4) occurs with the conservation of total spin according to the scheme [22]



where the brackets denote the orbits: paired electrons are shown by two oppositely directed arrows, while unpaired electrons are shown by single arrows. The transfer of the excitation energy in this process occurs through the electron-exchange transport. Therefore, the slight difference

in the excitation energies of oxygen and iodine molecules and the conservation of total spin of the electrons may be responsible for a rapid energy exchange in process (4). The results of experimental investigations [4, 6, 23] also suggest a rapid electron energy exchange.

Using expressions (16)–(20), we can determine the populations of SO at the second and third levels and compare them with the population η_b of the electronically excited oxygen O₂(*b*) for $\eta_A=0.75$, $\gamma_2 + \gamma_3 = 2$ and $T = 300$ K. Fig. 1 shows the dependence of the fractions of vibrationally excited oxygen O₂(*a*, $v = 2$) and the electronically excited oxygen O₂(*b*) on η_w . One can see that the populations of these two components are of the same order at the output of the SOG. For such concentrations of O₂(*a*, $v = 2$), process (4) with the rate constant $K_4 > 10^{-11}$ cm³ s⁻¹ can ensure the experimentally observed dissociation rates for iodine.

For a fraction $\eta_w < 0.04$ of water in oxygen, the

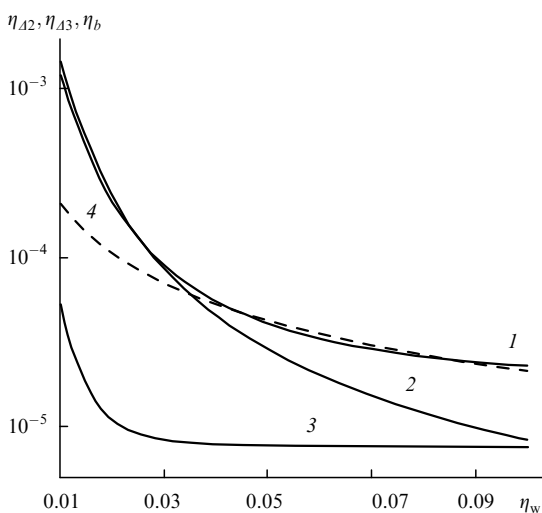


Figure 1. Fractions η_{A2} of the vibrationally excited SO at the second level (curves 1, 2) and η_{A3} at the third level (curve 3), as well as the fraction O₂(*b*) η_b of the electronically excited oxygen O₂(*b*) (curve 4) as functions of the water content η_w ; curves 1, 3 were calculated by expressions (17)–(20) for $\eta_A=0.75$, $\gamma_2 + \gamma_3 = 2$ and $T = 300$ K, while curve 2 was calculated using the Treanor distribution.

fractions of vibrationally excited SO at $v = 2$ obtained from the Treanor distribution (curve 2 in Fig. 1) are identical to those obtained from formulas (17)–(20) (curve 1). Thus, for the above-mentioned compositions of the medium, the pumping does not affect the form of the steady-state vibrational distribution, which can be described with a fairly high degree of precision by the Treanor distribution.

If the excitation to the third vibrational level of oxygen molecules in processes (5) and (6) occurs with a probability 0 and 1 ($\gamma_2 = 1$, $\gamma_3 = 1$) respectively, pumping always dominates over the arrival of vibrational quanta from lower levels for the medium compositions considered here. It can be seen from Fig. 1 that in this case, the fraction of SO molecules at the level $v = 3$ (curve 3) is much smaller than at the level $v = 2$, and is practically independent of the fraction of water in oxygen for $\eta_w > 0.03$. However, if the rate constant of process (3) is close to the gas-kinetic rate constant, the effect of this process on the dissociation of

iodine may turn out to be significant, especially for $\eta_w \leq 0.01$ [5]. Note that the concentration of oxygen at the levels $v > 3$ is much lower than at the levels considered here; hence, their contribution to the formation of the active medium can be neglected.

Thus, the population of oxygen at the output of SOG deviates considerably from the equilibrium population. The decrease in energy from the vibrational reservoir occurs during the VV' exchange with water molecules. Since the concentration of oxygen at the first vibrational level is considerably higher than the concentration at higher levels, process (10) accounts for a significant decrease in the number of vibrational quanta from oxygen for the first level only.

The rate of vibrationally translational VT exchange with the medium components is much lower than the characteristic rates of the remaining processes and does not affect the steady-state distribution of the vibrational energy over the levels. In some cases, the pumping rate of vibrational levels of oxygen during the deactivation of the electron energy is higher than the rate of their population from lower levels. In this case, the relative population η_{Ai} of the lower vibrational levels of SO is described by the formula

$$\eta_{Ai} = \frac{\eta_i}{(1/\eta_A - 1) \exp(-\Delta E_i/kT) + 1}, \quad i = 1, 2, 3,$$

where

$$\eta_1 = n \frac{K_5 \eta_A^2}{K_{10} \eta_w} + (n + m) \frac{K_5 \eta_A^2}{K_{11} \eta_w^2} \exp \frac{\Delta E}{kT} + \exp \left(-\frac{E_1}{kT} \right);$$

$$\eta_2 = \eta_1^2 + \frac{(\gamma_2 + \gamma_3) K_5 \eta_A^2}{K_{1,1}};$$

$$\eta_3 = \eta_1 \eta_2 + \frac{\gamma_3 K_5 \eta_A^2}{K_{2,1}}.$$

If the pumping is carried out to the levels with $v \leq 2$, the population of levels for a water fraction $\eta_w < 0.04$ in the gas flow is described to a fairly high degree of precision by the Treanor distribution.

The concentration of SO with $v = 2$ is comparable with the concentration of the electronically excited oxygen O₂(*b*), which was treated for a long time as a potential 'partner' causing the dissociation of iodine. The electronically excited state $A^3\Pi_{lu}$ of iodine may be an intermediate state during the dissociation of I₂. The process described by (4) may ensure the experimentally observed iodine dissociation rates if its rate constant $K_4 > 10^{-11}$ cm³ s⁻¹.

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