

# Pulsed electron-beam-sustained discharge in oxygen-containing gas mixtures: electrical characteristics, spectroscopy, and singlet oxygen yield\*

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**Abstract.** The electrical and spectroscopic characteristics of electron-beam-sustained discharge (EBSD) in oxygen and oxygen-containing gas mixtures are studied experimentally under gas pressures up to 100 Torr in a large excitation volume ( $\sim 18$  L). It is shown that the EBSD in pure oxygen and its mixtures with inert gases is unstable and is characterised by a small specific energy contribution. The addition of small amounts ( $\sim 1\% - 10\%$ ) of carbon monoxide or hydrogen to oxygen or its mixtures with inert gases considerably improves the stability of the discharge, while the specific energy contribution  $W$  increases by more than an order of magnitude, achieving  $\sim 6.5$  kJ L $^{-1}$ atm $^{-1}$  per molecular component of the gas mixture. A part of the energy supplied to the EBSD is spent to excite vibrational levels of molecular additives. This was demonstrated experimentally by the initiation of a CO laser based on the O<sub>2</sub> : Ar : CO = 1 : 1 : 0.1 mixture. Experimental results on spectroscopy of the excited electronic states O<sub>2</sub>( $a^1\Delta_g$ ) and O<sub>2</sub>( $b^1\Sigma_g^+$ ), of oxygen formed in the EBSD are presented. A technique was worked out for measuring the concentration of singlet oxygen in the O<sub>2</sub>( $a^1\Delta_g$ ) state in the afterglow of the pulsed EBSD by comparing with the radiation intensity of singlet oxygen of a given concentration produced in a chemical generator. Preliminary measurements of the singlet-oxygen yield in the EBSD show that its value  $\sim 3\%$  for  $W \sim 1.0$  kJ L $^{-1}$ atm $^{-1}$  is in agreement with the theoretical estimate. Theoretical calculations performed for  $W \sim 6.5$  kJ L $^{-1}$ atm $^{-1}$  at a fixed temperature show that the singlet-oxygen yield may be  $\sim 20\%$ , which is higher than

the value required to achieve the lasing threshold in an oxygen–iodine laser at room temperature.

**Keywords:** singlet oxygen, non-self-sustained discharge, CO laser, oxygen–iodine laser.

## 1. Introduction

A singlet-oxygen molecule in the excited electronic O<sub>2</sub>( $a^1\Delta_g$ ) state serves as an energy donor for the emitting atomic iodine in chemical oxygen–iodine lasers (COILs). The generation of singlet oxygen in an electric discharge (see reviews [1–3]) makes it possible to avoid the use of toxic materials required for obtaining singlet oxygen in a chemical generator. The possibility of developing an electric-discharge oxygen–iodine laser (OIL) was pointed out in a large number of publications (see, for example, [4–19]). It was emphasised that to obtain a positive gain for atomic iodine, the singlet-oxygen yield

$$Y = \frac{[\text{O}_2(a^1\Delta_g)]}{[\text{O}_2(a^1\Delta_g)] + [\text{O}_2(X^3\Sigma_g^-)]},$$

where  $[\text{O}_2(a^1\Delta_g)]$  and  $[\text{O}_2(X^3\Sigma_g^-)]$  are the concentrations of singlet oxygen and oxygen in the ground state, must exceed the temperature-dependent threshold yield

$$Y_{\text{th}} = [1 + 1.5 \exp(402/T)]^{-1},$$

which is equal to 15% for the gas at room temperature. The attainment of such (and even higher) values of the singlet-oxygen yield in a self-sustained discharge was reported in papers [7, 12–14]. However, the low partial pressures of oxygen (less than 1 Torr) in experiments on the formation of singlet oxygen in a self-sustained discharge do not correspond to the present-day requirements imposed on OILs, since the partial pressure of oxygen must exceed 10 Torr.

It was mentioned in [16–18] that the efficiency of oxygen excitation from the ground state to the singlet O<sub>2</sub>( $a^1\Delta_g$ ) state depends on the reduced field strength  $E/N$ . This dependence has a distinct peak for the field strength  $E/N \sim 10^{-16}$  V cm<sup>2</sup> characteristic of a non-self-sustained discharge. The EBSD in mixtures of oxygen with inert gases was studied experimentally as a non-self-sustained discharge for generating singlet oxygen in [5, 6, 11], while a mixture of oxygen with nitrogen was used for this purpose in Ref. [10]. In these

\*This paper is based on the materials of the report TuR2-11 presented at the 'XI Conference on Laser Optics', 30 June–4 July 2003, St. Petersburg, Russia.

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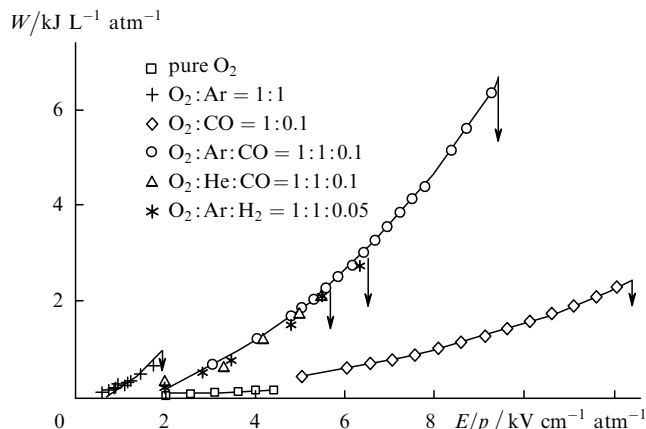
experiments, the specific energy contribution  $W$  corresponding to the partial pressure of oxygen was  $100\text{--}200\text{ J L}^{-1}\text{ atm}^{-1}$ . For such values of  $W$ , the singlet-oxygen yield does not exceed a few percent. Such low values of  $W$  are due to the discharge instability in oxygen-containing gas mixtures even in small discharge volumes ( $\sim 0.1\text{ L}$ ).

The addition of molecular components to gas mixtures in Ref. [20] increased the free electron concentration in the discharge and, hence, the stability of the EBSD. In this paper, we continue the investigations of the electrical characteristics of the EBSD in oxygen-containing gas mixtures, which was started in Ref. [20]. The aim of our work is to estimate the singlet-oxygen yield in the EBSD with and without molecular additives. Spectroscopic analysis of glow and afterglow of pulsed EBSD in the visible and near-IR spectral regions is carried out. A technique is worked out for measuring the singlet-oxygen concentration from its luminescence in the afterglow of a pulsed EBSD by comparing with the radiation intensity of singlet oxygen of a given concentration produced in a chemical singlet-oxygen generator.

## 2. EBSD in oxygen-containing gas mixtures

The experiments on measuring the characteristics of electric discharge in oxygen-containing gas mixtures were carried out on a pulsed electroionisation setup with an excited volume of  $\sim 18\text{ L}$  [20]. The electrical conductivity of the gas in a non-self-sustained discharge was created by an external ionisation source in the form of an electron beam with an electron energy  $\sim 150\text{ keV}$ . The voltage was supplied to the EBSD from a capacitor bank with total capacitance  $C$  which could be varied in different experiments from  $45$  to  $370\text{ }\mu\text{F}$ . The energy  $Q$  supplied to the EBSD was determined by measuring the initial voltage  $U_0$  and the final voltage (after the termination of the pulse)  $U_1$  across the electric discharge gap:  $Q = C(U_0^2 - U_1^2)/2$ . The EBSD pulse duration was determined by the duration of the electron beam, which was  $\sim 100\text{ }\mu\text{s}$  in our experiments.

We studied in our experiments the properties of the pulsed EBSD in oxygen-containing gas mixtures. The purity of oxygen was  $99.999\%$ . Figure 1 shows the dependences of  $W$  on the initial reduced field intensity  $E/p$  of the EBSD in various oxygen-containing mixtures. The maximum value of the specific energy contribution to the EBSD was limited by the electric breakdown in the gap between the electrodes (vertical arrows in Fig. 1). For pure oxygen under pressures between  $3$  and  $100\text{ Torr}$ ,  $W \leq 0.1\text{ kJ L}^{-1}\text{ atm}^{-1}$ . For a given initial value of parameter  $E/p$ , dilution of oxygen by argon increased the value of  $W$  reduced to the partial pressure of molecular components of the gas mixture. The stability of the EBSD decreased with increasing field strength. This was manifested in the emergence of pulsations in the temporal dynamics of the current pulses. The mechanism of such instability of argon-containing mixtures was discussed in Ref. [21]. Small (up to  $10\%$ ) additives of CO or  $\text{H}_2$  into the gas mixture increased the stability of the EBSD and led to a higher peak value of  $W$  [22]. Earlier, CO additives were used for stabilising the EBSD for developing an effective pulsed  $\text{N}_2\text{O}$  laser [23, 24]. The addition of  $\sim 10\%$  carbon monoxide to pure oxygen in our experiments increased the maximum value of  $W$  to  $\sim 2.3\text{ kJ L}^{-1}\text{ atm}^{-1}$  (see Fig. 1). The maximum value of  $W$  reduced to the molecular components of the mixture ( $\text{O}_2$ , CO or  $\text{H}_2$ ) in



**Figure 1.** Dependences of specific energy contribution  $W$  corresponding to the partial pressure of the molecular components ( $\text{O}_2$ , CO, or  $\text{H}_2$ ) on parameter  $E/p$  for the total gas pressure  $p = 30\text{ Torr}$ ;  $C = 370\text{ }\mu\text{F}$ .

$\text{O}_2 : \text{He} : \text{CO} = 1 : 1 : 0.1$  and  $\text{O}_2 : \text{Ar} : \text{H}_2 = 1 : 1 : 0.05$  gas mixtures was  $\sim 2\text{ kJ L}^{-1}\text{ atm}^{-1}$ . In order to increase the maximum value of  $W$ , it is apparently expedient to use the  $\text{O}_2 : \text{Ar} : \text{CO} = 1 : 1 : 0.1$  gas mixture for which the maximum energy contribution ( $\sim 6.5\text{ kJ L}^{-1}\text{ atm}^{-1}$ ) was attained.

## 3. Theoretical estimates for the singlet-oxygen yield in EBSD

The results of our experimental investigations show that the addition of CO or  $\text{H}_2$  makes it possible to increase substantially the stability of EBSD in oxygen-containing mixtures and attain a high specific energy contribution ( $\sim 6.5\text{ kJ L}^{-1}\text{ atm}^{-1}$ ) corresponding to the molecular components of the mixture in a large ( $\sim 18\text{ L}$ ) volume being excited. For such an energy contribution, it can be expected that the singlet-oxygen yield would exceed  $Y_{\text{th}}$  for OILs. However, when molecular gases are added, a part of the energy supplied to the discharge is spent on their excitation. In order to estimate the role of inelastic processes of electron interaction with CO,  $\text{H}_2$  or  $\text{D}_2$  additives and to evaluate the singlet-oxygen yield in the EBSD, we simulated the kinetic processes occurring in the discharge plasma.

Our investigations were based on the kinetic model described in detail in Ref. [25] where it was used for calculating the reduced electric field strength  $E/N$  in the positive column of a self-sustained discharge in the singlet oxygen:  $\text{O}_2 = 1 : 1$  gas mixture.

Complication of the model due to allowance for the molecular components CO,  $\text{H}_2$  and  $\text{D}_2$  is described briefly in Ref. [20]. In the present work, the processes of excitation of vibrational levels of molecular components, the processes of detachment of electrons from  $\text{O}^-$  ions during collisions with CO,  $\text{H}_2$  and  $\text{D}_2$  molecules, processes of ionisation and recombination, as well as excitation of atoms and molecules under the action of an electron beam were included additionally. For large values of  $W$  attained in the experiments, the distribution function of molecules over vibrational levels is found to be considerably nonequilibrium. An analogous model [26] was used to describe a strong departure from equilibrium and the effects of molecular anharmonicity in vibration–vibration exchange processes.

The excitation of vibrational levels of molecular additivs may lead to a decrease in the efficiency of the singlet-oxygen excitation. For example, an addition of 5% CO to the  $O_2 : Ar = 1 : 1$  mixture leads to a decrease in the maximum efficiency of the electron energy fraction (56%) spent on exciting the state  $O_2(a^1A_g)$  to 18% (with allowance for cascade excitation). The optimal value of  $E/N$  corresponding to the maximal excitation of singlet oxygen increased in this case from  $0.69 \times 10^{-16}$  to  $2.2 \times 10^{-16}$  V cm<sup>2</sup>. This is due to a large cross section of excitation of CO molecular vibrational levels. Since the cross section of vibrational excitation of  $H_2$  and  $D_2$  molecules is considerably lower, an addition of 2.5%  $H_2$  or  $D_2$  to the  $O_2 : Ar = 1 : 1$  mixture does not lead a noticeable change in the electron energy balance. In particular, the maximum effective electron energy fraction spent on the excitation of singlet oxygen decreases to 43% and 49%, respectively upon an addition of 2.5%  $H_2$  or  $D_2$ . The optimal value of  $E/N$  changes only slightly in this case.

For analysing the peculiarities of the singlet-oxygen excitation in a non-self-sustained discharge, the dependences of the singlet-oxygen yield on the energy supplied to the discharge are presented in Fig. 2 for various mixtures under a pressure of 30 Torr and at a constant gas temperature of 300 K, i.e., under the conditions of the effective gas cooling. The current pulse of the electron beam was assumed to be rectangular and had a duration of 100  $\mu$ s. The reduced electric field was assumed to be constant in our calculations and corresponded to the maximum effective excitation of singlet oxygen for each mixture of gases. The theoretical value of the energy supplied to the discharge was varied in the numerical model by changing the electron beam current.

The results of these calculations show that the recombination mechanism is predominantly responsible for perishing of electrons. This points to a high rate of detachment of electrons in oxygen-containing gas mixtures with an addition of molecular gases. The concentration of singlet oxygen was calculated for the instant of time 200  $\mu$ s beginning 100  $\mu$ s after termination of the EBSD pulse. The vertical lines in Fig. 2 show the maximum energy contribution corresponding to the partial pressure of molecular components of the mixture, attained in experi-

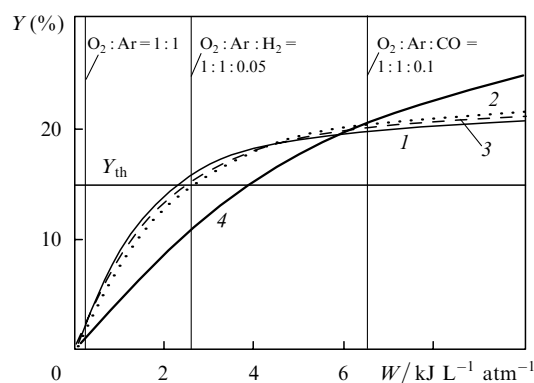
ments for various gas mixtures. The horizontal lines show the threshold value of the singlet-oxygen yield ( $Y_{th} = 15\%$  at  $T = 300$  K). For small energy contributions, the singlet-oxygen concentration increases linearly and the slope of the straight line is determined by the effective fraction of the singlet-oxygen excitation. For the  $O_2 : Ar = 1 : 1$  mixture, the maximum energy contribution obtained in the experiment for a preserved stability of the EBSD was  $\sim 2$  kJ L<sup>-1</sup> atm<sup>-1</sup>. The singlet-oxygen yield for such an energy contribution is much lower than  $Y_{th}$ . The addition of  $H_2$  to the gas mixture slightly lowers the effective fraction of singlet-oxygen excitation, while the experimentally attained energy contribution increases considerably to 2.6 kJ L<sup>-1</sup> atm<sup>-1</sup> and the singlet-oxygen yield approaches  $Y_{th}$ . When CO was added to the gas mixture, the stability of the EBSD increased considerably, which led to the value  $W = 6.5$  kJ L<sup>-1</sup> atm<sup>-1</sup> corresponding to  $\sim 20\%$  yield of singlet oxygen, i.e., higher than  $Y_{th}$ .

#### 4. Spectroscopy of EBSD and luminescence of excited oxygen

The concentration of singlet oxygen is determined most frequently (see for example, [1–3]) by measuring the singlet-oxygen luminescence intensity near the wavelength  $\lambda = 1.27$   $\mu$ m of the  $O_2(a^1A_g) \rightarrow O_2(X^3\Sigma_g^-)$  transition. However, the calculation of the singlet-oxygen concentration from these data requires the knowledge of the Einstein coefficient whose value may vary by a factor of two in different works [27]. The application of such a measuring technique requires very precise calibration of the IR detector using the blackbody surface radiation. In real experiments, however, the intensity of the bulk radiation source is measured, which casts a doubt on the precision of such a calibration (see also [28]).

To increase the reliability of measurement of the absolute concentration and the yield of singlet oxygen, we preliminarily compared in our experiments the radiation intensity of singlet oxygen obtained in an EBSD and in a chemical generator with a controllable concentration of singlet oxygen. The geometry of the excited oxygen volume and the optical scheme of radiation recording were identical in both cases. It should be noted that, for such a calibration, it is not necessary to use the Einstein coefficient or absolute calibration of the IR detector. A similar technique can be used for determining the oxygen concentration in the  $O_2(b^1\Sigma_g^+)$  state. In a chemical generator, the concentration is determined by comparing the radiation intensity of oxygen in this state ( $\lambda \approx 762$  nm) and at the  $O_2(a^1A_g) + O_2(a^1A_g) \rightarrow O_2(X^3\Sigma_g^-) + O_2(X^3\Sigma_g^-)$  transition with a wavelength  $\lambda \approx 634$  nm. We used this method to determine, for example, the concentration of oxygen in the  $O_2(b^1\Sigma_g^+)$  state obtained in a self-sustained pulsed discharge in oxygen ( $p = 2.8$  Torr,  $U = 15$  kV,  $C = 10$  nF,  $Q_{disch} = 2$  J,  $V = 0.8$  L). The concentration in these experiments was  $\sim 10^{14}$  cm<sup>-3</sup>.

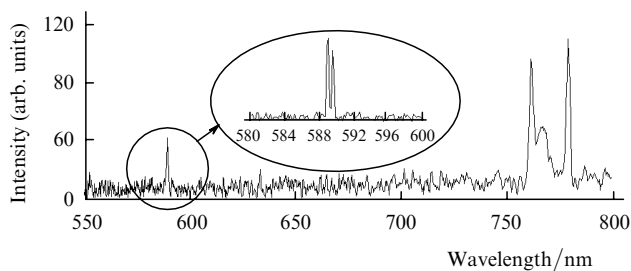
The oxygen state  $O_2(b^1\Sigma_g^+)$  plays an important role in the formation of singlet oxygen in an electric discharge; consequently, luminescence in the region of 762 nm provides information on the singlet-oxygen concentration [17]. The luminescence of the EBSD in the visible spectral range was recorded with an ISP-51 prism spectrometer equipped with a CCD array. The emission spectrum of the EBSD and its afterglow recorded with a spectral resolution of  $\sim 2$  nm are



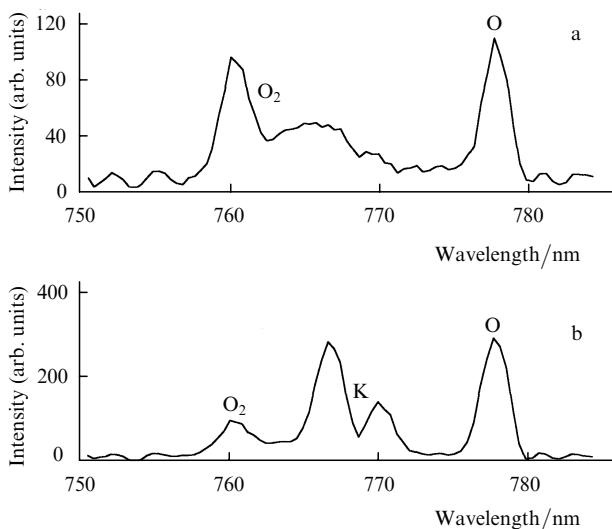
**Figure 2.** Theoretical dependences of the singlet-oxygen yield  $Y$  on the specific energy contribution corresponding to the molecular components for the mixtures  $O_2 : Ar = 1 : 1$  (1),  $O_2 : Ar : H_2 = 0.95 : 1 : 0.05$  (2),  $O_2 : Ar : D_2 = 0.95 : 1 : 0.05$  (3), and  $O_2 : Ar : CO = 0.9 : 1 : 0.1$  (4). Vertical lines show the maximum energy contribution attained experimentally for the mixtures  $O_2$ -Ar,  $O_2$ -Ar-CO, and  $O_2$ -Ar- $H_2$ . The horizontal line corresponds to the threshold value  $Y_{th}$  at  $T = 300$  K.

presented in Fig. 3. The spectrogram shows the  $P$  and  $R$  branches of the  $O_2(b^1\Sigma_g^+) \rightarrow O_2(X^3\Sigma_g^-)$  transition ( $\lambda \approx 762$  nm), the luminescence of atomic oxygen ( $\lambda \approx 778$  nm), and the spectral line at the wavelength  $\sim 589$  nm. It was proposed in Ref. [29] that this line is the luminescence line of gold oxide. Experiments carried out with a high spectral resolution show that this is a doublet of atomic lines identified by us as the luminescence lines of sodium (589.0 and 589.6 nm). The appearance of sodium in the discharge may be due to the fact that this electro-ionisation setup was earlier used as a  $CO_2$  laser whose optical elements were prepared from NaCl and KCl. Indeed, the EBSD spectrum for high-energy contributions (higher than  $1 \text{ kJ L}^{-1} \text{ atm}^{-1}$ ) also contains the potassium doublet (766.5 and 769.9 nm) (Fig. 4b). This doublet overlaps with the  $P$  branch of the  $O_2(b^1\Sigma_g^+) \rightarrow O_2(X^3\Sigma_g^-)$  transition (Fig. 4a), which may lead to errors in the measurement of luminescence intensity of the  $O_2(b^1\Sigma_g^+)$  state, especially when spectral filters are used. However, the potassium lines do not prevent the recording of luminescence on the  $R$  branch of the  $O_2(b^1\Sigma_g^+) \rightarrow O_2(X^3\Sigma_g^-)$  transition and of the atomic oxygen line ( $\lambda \approx 778$  nm).

Such an emission spectrum of an EBSD and its afterglow in oxygen-containing mixtures was also observed at cryogenic temperatures ( $\sim 100$ – $150$  K). Low temperatures of oxygen-containing gas mixtures are interesting from the point of view of lowering the threshold value of the singlet-



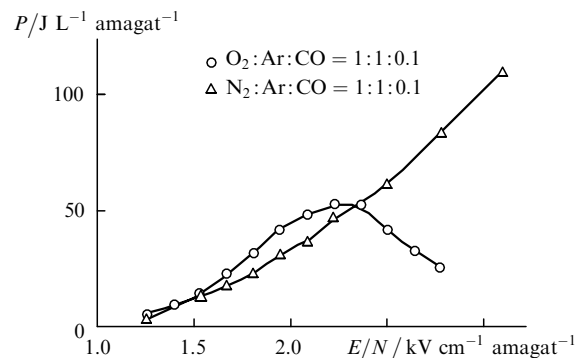
**Figure 3.** Spectrum of the EBSD and its afterglow in the visible region. Gas mixture  $O_2 : CO = 1 : 0.1$ ,  $p = 30$  Torr,  $W \sim 1 \text{ kJ L}^{-1} \text{ atm}^{-1}$ .



**Figure 4.** Spectrum of the EBSD and its afterglow near the wavelength 760 nm. Gas mixture  $O_2 : CO = 1 : 0.1$ ,  $p = 30$  Torr,  $W \sim 1 \text{ kJ L}^{-1} \text{ atm}^{-1}$  (a) and  $2.3 \text{ kJ L}^{-1} \text{ atm}^{-1}$  (b).

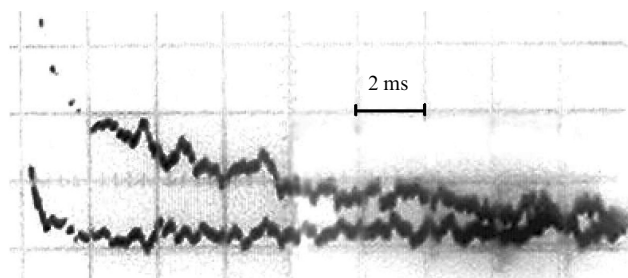
oxygen yield ( $Y_{th} \sim 1\%$  at  $T = 100$  K). It should be noted that the role of vibration–vibration VV-exchange between CO molecules contained in the gas mixtures increases at cryogenic temperatures. Such an exchange leads to the population of a large number (right up to 40) of vibrational levels  $V$  of the CO molecule, on which luminescence appears; in the case of feedback, lasing may occur in the intermediate IR region ( $\lambda \sim 5$ – $6 \mu\text{m}$ ) (see, for example, [30]).

Indeed, we observed in our experiments generation by a  $CO_2$  laser operating on the gas mixture  $O_2 : Ar : CO = 1 : 1 : 0.1$  at a low temperature  $T \sim 100$  K. The optical cavity of this laser is formed by a highly reflecting spherical copper mirror and a partially reflecting mirror (reflection coefficient  $\sim 50\%$ ). Figure 5 shows the dependences of the specific energy removal  $P$  on the parameter  $E/N$  obtained for the oxygen-containing  $O_2 : Ar : CO = 1 : 1 : 0.1$  gas mixture and the nitrogen-containing  $N_2 : Ar : CO = 1 : 1 : 0.1$  mixture which is normally used in a CO laser for a density  $N = 0.08$  amagat (the relative density of 1 amagat corresponds to a pressure of 1 atm under normal conditions). One can see that unlike a CO laser with a typical nitrogen-containing mixture, the CO laser with an oxygen-containing mixture exhibits a distinct peak of specific energy removal for  $E/N \approx 2.2 \text{ kV cm}^{-1} \text{ amagat}^{-1}$ . A decrease in  $P$  upon a further increase in the parameter  $E/N$  in a laser with an oxygen-containing gas mixture may be associated with an increase in the pump efficiency of electronic levels of the oxygen molecule and with a decrease in the pump efficiency of vibrational levels of the CO molecule. Thus, investigation of the generation parameters of such a laser provides information on processes of oxygen excitation in an electric discharge.

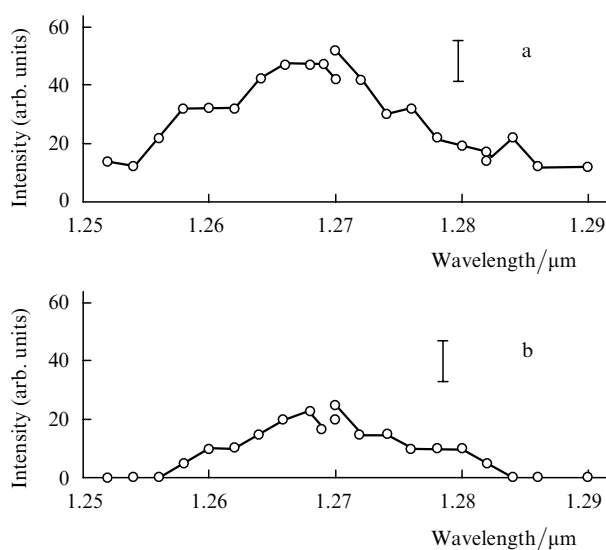


**Figure 5.** Dependence of the specific energy removal  $P$  of an electro-ionisation CO laser on parameter  $E/N$  for  $N = 0.08$  amagat,  $T = 100$  K for the  $O_2 : Ar : CO = 1 : 1 : 0.1$  and  $N_2 : Ar : CO = 1 : 1 : 0.1$  mixtures.

The singlet-oxygen luminescence in an EBSD and its afterglow was recorded with an IKS-31 diffraction monochromator (spectral resolution in our experiments was  $\sim 10$  nm) equipped with an uncooled germanium photodiode with a response time of  $\sim 200 \mu\text{s}$ . The luminescence signal at a wavelength of  $1.27 \mu\text{m}$  decayed exponentially for  $\sim 10$  ms (Fig. 6). To analyse the spectral-temporal characteristics of luminescence of the EBSD, we measured the signal amplitude at a certain instant at different wavelengths. Figure 7 shows the spectral distribution of luminescence at the second and sixth milliseconds from



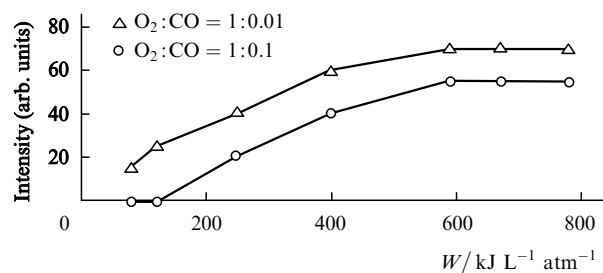
**Figure 6.** Time dependence of the singlet-oxygen luminescence signal at the wavelength  $1.27 \mu\text{m}$  (top) and the signal from the photodetector without an EBSD in the presence of an electron beam (bottom). Gas mixture  $\text{O}_2 : \text{He} : \text{CO} = 1 : 2 : 0.01$ ,  $p = 90 \text{ Torr}$ ,  $T = 300 \text{ K}$ ,  $W \sim 0.5 \text{ kJ L}^{-1} \text{ atm}^{-1}$ .



**Figure 7.** Spectral characteristics of singlet-oxygen luminescence at the second (a) and sixth (b) milliseconds after the onset of the EBSD. Here and in Fig. 8, the vertical segments show the measuring error.

the beginning of the EBSD. In both cases, the FWHM of the spectral line was  $\sim 20 \text{ nm}$ .

Figure 8 shows the dependences of the singlet-oxygen luminescence intensity at the second millisecond on the value of  $W$  for  $\text{O}_2 : \text{CO} = 1 : 0.1$  and  $\text{O}_2 : \text{CO} = 1 : 0.01$  gas mixtures with approximately 10% and 1% carbon monoxide content. When a mixture with a lower CO concentration was used, the singlet-oxygen luminescence intensity increased. This is apparently associated with an increase in the fraction of energy spent on exciting oxygen. A comparison of the helium- and argon-containing gas mixtures ( $\text{O}_2 : \text{Ar} : \text{CO} = 1 : 1 : 0.01$  and  $\text{O}_2 : \text{He} : \text{CO} = 1 : 1 : 0.01$ ) shows that the use of helium makes it possible to almost double the singlet-oxygen yield. Such an effect may be associated with an increase in the role of diffusion in thermal conduction in the helium-containing gas and with smoothing of spatial gradients. The argon-containing mixture exhibits intense luminescence during the first one or two milliseconds, which was identified by us as the glow of excited argon ( $\lambda = 1.2702 \mu\text{m}$ ); this hampers the observation of singlet-oxygen luminescence.



**Figure 8.** Amplitudes of the singlet-oxygen luminescence signal at the second millisecond after the onset of the EBSD as functions of the specific energy contribution for mixtures  $\text{O}_2 : \text{CO} = 1 : 0.01$  and  $\text{O}_2 : \text{CO} = 1 : 0.1$ .

Thus, our experiments show that the choice of the gas mixture plays an important role in increasing the energy contribution and the singlet-oxygen yield. Preliminary measurements of the singlet-oxygen yield in the EBSD show that its value for  $W \sim 1.0 \text{ kJ L}^{-1} \text{ atm}^{-1}$  is  $\sim 3\%$ . The theoretical model in which the temperature variation of the gas upon its excitation in the EBSD was taken into account predicted a singlet-oxygen yield of 4%–6% under these experimental conditions, which is in good agreement with the results of measurement of the singlet-oxygen concentration.

## 5. Conclusions

The results of experimental investigations show that molecular additives (CO or  $\text{H}_2$ ) in pure oxygen or oxygen diluted with inert gases lead to a considerable increase in the EBSD stability and to a high specific energy contribution for a large excitation volume ( $\sim 18 \text{ L}$ ). The highest value of  $W$  recalculated for the molecular components of the mixture ( $\sim 6.5 \text{ kJ L}^{-1} \text{ atm}^{-1}$ ) was obtained experimentally for the  $\text{O}_2 : \text{Ar} : \text{CO} = 1 : 1 : 0.1$  gas mixture for a total gas pressure of 30 Torr. Part of the energy supplied to the EBSD is spent on exciting vibrational levels of molecular additives. This was demonstrated experimentally by the initiation of a CO laser based on the  $\text{O}_2 : \text{Ar} : \text{CO} = 1 : 1 : 0.1$  mixture.

An analysis of experimental results on spectroscopy of luminescence and afterglow of the EBSD in oxygen-containing gas mixtures in the visible and near-IR spectral regions shows that the composition of the gas mixture determines the singlet-oxygen yield to a considerable extent. The optical scheme for recording the singlet-oxygen luminescence was calibrated by comparing the luminescence intensities of excited oxygen obtained in chemical and electrical singlet oxygen generators. Preliminary measurements of the singlet-oxygen yield in the EBSD show that its value for  $W \sim 1.0 \text{ kJ L}^{-1} \text{ atm}^{-1}$  is  $\sim 3\%$ , which is in good agreement with the theoretical estimate. Theoretical analysis carried out for  $W \sim 6.5 \text{ kJ L}^{-1} \text{ atm}^{-1}$  under the assumption of a constant gas temperature shows that the singlet-oxygen yield in the first electron excited state  $\text{O}_2(a^1\Delta_g)$  may amount to  $\sim 20\%$ , i.e., exceed the value required for attaining the generation threshold in the oxygen-iodine laser at room temperature. A large singlet-oxygen yield with a high efficiency of its production should be expected for other types of non-self-sustained discharge, e.g., for the discharge described in Ref. [16]. It should be noted that experimental results were obtained for the pulsed discharge without

cooling a gas, which is required for developing an OIL with an electric-discharge source of singlet oxygen [18]. A lowering of gas temperature simplifies the conditions for developing an OIL based on a singlet-oxygen generator using a non-self-sustained discharge.

**Acknowledgements.** This work was supported by the Russian Foundation for Basic Research (Grant No. 02-02-17452), AFRL, EOARD, International Centre for Science and Technology (Project No. 2415P), the Russian Science Support Foundation, and Grant Nos MK-930.2004.2 and NSh-794.2003.2 from the President of the Russian Federation.

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