Effect of NO_2-gas mantle around the optical breakdown cells in air on the main parameters of plasma luminescence

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The concentration of nitrogen dioxide produced under the effect of the ionizing radiation from an optical-breakdown plasma cell in air is calculated. Basic parameters of an NO_2 gas mantle are obtained as functions of the laser pulse duration and intensity. It is noted that the mantle with an enhanced concentration of nitrogen dioxide and oxygen ions around a plasma cell significantly affects the time parameters of plasma radiation, because it screens the plasma luminescent radiation in the visible spectral region.

Introduction

Modern high-power lasers make it possible to generate plasma formations with parameters, close to those in a lightning or nuclear explosion, in various media. At a high pressure the optical-breakdown plasma forms a shock wave and emits energy in a wide spectral range, which leads to photoionization of the ambient medium.

The development of methods of laser spectral analysis made it possible to reveal regularities in the plasma spread dynamics and the plasma interaction with the ambient medium, as well as to consider the processes of plasma recombination. However, the complete pattern of phenomena occurring during the optical breakdown in a wide range of the laser radiation intensity is still to be understood.

This paper considers the dynamics of emission of plasma formations in air in a wide range of the intensity of laser radiation with the wavelength of 10.6 μm: from 10^6 to 10^9 W/cm^2.

Measurement procedure and results

A single-pulse CO_2 laser was used as a source of radiation. The laser pulse was shaped as the main peak with a half-width of 1.5 μs followed by a long 10-μs-duration tail with a slow falloff of the intensity. Seventy five percent of the total pulse energy is concentrated in the leading peak. The beam structure on the path is formed with a Cassegrainian telescope. The radiation energy density in the focal plane varied within 0.2 to $2 \cdot 10^3$ J/cm^2. The laser pulse energy was measured with a bolometric detector. The spectral-energy measurements were carried out with two photomultiplier tubes (PMTs), detecting the radiation from plasma formations in the spectral ranges of 0.4, 0.63, or 0.51 μm. These ranges were separated by interference filters. The signals from PMT were recorded with a dual channel memory oscilloscope. To synchronize the detection of luminescence of plasma formations with the CO_2 laser pulse, signals from PMTs and a FSG-223 detector of the scattered laser radiation were recorded using another one oscilloscope. Photos of plasma formations were taken with the use of a Zenit-E camera.

Figure 1*a* shows the typical oscillograms for the luminescence in the propagation channel of the CO_2 laser pulse of microsecond duration with the power density of $5 \cdot 10^6$ W/cm^2, averaged over the beam cross section, in the blue (0.4 ± 0.02) μm and green (0.51 ± 0.02) μm spectral ranges along with the time behavior of the laser pulse power density.

The luminescence in the channel starts 0.2–1 μs after the arrival of the leading front of the pulse. The maximum values of the luminescence power in the spectral ranges mentioned above coincide in time, but come after the maximum value of the laser pulse power. The strongest fluctuations of the luminescence power are observed by the end of the laser pulse. The duration of luminescence in the visible region is comparable with the laser pulse duration.

Figure 1*b* depicts the signals of luminescence from the propagation channel of the CO_2 laser radiation with the power density of $5 \cdot 10^7$ W/cm^2 in the blue (0.4 ± 0.02) μm and red (0.63 ± 0.02) μm spectral ranges. A plasma formation begins to luminesce some time later after the beginning of a CO_2 laser pulse. The lag (~ 30 ns) corresponds to the finite time of generation of a plasma formation near an aerosol particle. The luminescence in different spectral ranges appears simultaneously, while the maximum values of the signals, not coinciding with the maximum value of the laser pulse, are spaced in time. The maximum value of luminescence is first achieved by the radiation with shorter wavelengths. The duration of the luminescence increases with the increase of the laser radiation intensity, as can be seen from the comparison of Figs. 1*a* and 1*b*. The time mismatch of the maximum luminescence power for different wavelengths is caused by the competition of two processes: the increase of the luminescence power due to the growth of a plasma formation at its adiabatic expansion and the decrease of the spectral power density of the radiation from a plasma cell with the decrease of temperature. The longer-wave radiation is less dependent on temperature, which leads to the lag of its maximum.[1]

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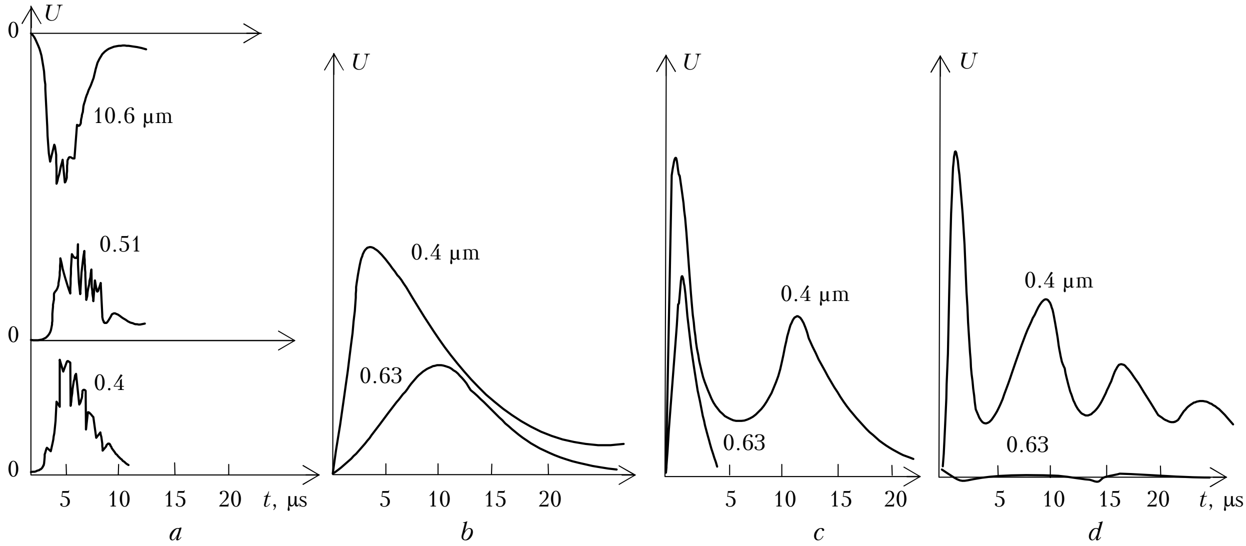

Fig. 1. Typical oscillograms of luminescence of optical breakdown cells in the visible spectral ranges: $I = 5 \cdot 10^6$ (*a*), $5 \cdot 10^7$ (*b*), $3 \cdot 10^8$ (*c*), and 10^9 W/cm^2 (*d*).

No fluctuations of the luminescence brightness were observed when the plasma passed into an equilibrium state after the termination of the high-power laser pulse, and this determined the main role of the continuum spectrum in the energy of luminescence of a plasma formation.

As the power density increases higher than 10^8 W/cm^2, the decrease in the duration of the plasma luminescence is observed along with the appearance of an additional lagged pulse, and restriction on the luminescence duration is the most tough for radiation of the red spectral range, which is not observed (does not exceed the detector noise) at the power density of laser radiation higher than $7 \cdot 10^8$ W/cm^2 (Figs. 1*c* and *d*). The power of luminescence in the blue spectral range fluctuates in time and decays. With the increase of the laser pulse energy, the number of periods of luminescence fluctuations in the blue spectral range increases, while the maximum power of luminescence changes only slightly.

The restriction on the duration and the appearance of fluctuations in the luminescence power of the optical breakdown plasma in different spectral ranges with the increasing energy of laser radiation are caused by the change of the optical properties of the air around plasma formations due to the photochemical reactions under the effect of the ionizing radiation from the plasma and the shock wave from a breakdown center.

Figure 2 shows the photos of the optical breakdown in the air under the exposure to the CO_2 laser radiation focused with a mirror of 6-m focal length. The mean power density of the laser radiation at the caustic varied from $5 \cdot 10^7$ W/cm^2 (*a*) to 10^9 W/cm^2 (*e*) for different pulses. The scale of the photos is shown overhead, and the direction of laser radiation propagation is indicated by an arrow on the top photo.

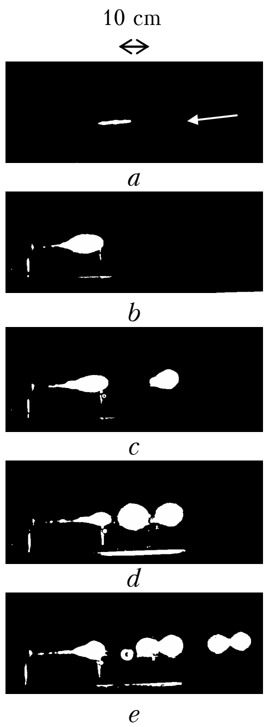

Fig. 2. Photos of optical breakdown cells.

The photo (*a*) corresponds to the time dependence of the luminescence power shown in Fig. 1*b*, and the following photos of plasma cells correspond to the time dependence of luminescence similar to that shown in Fig. 1*d*.

Discussion

As is seen in Fig. 2, as the laser pulse energy increases, the plasma formation shape changes from cylindrical to ellipsoidal, and further is takes a form of a sphere exceeding in size the laser beam diameter.

The spectral and temporal characteristics of the plasma luminescence in the ranges of 0.4 and 0.63 μm (see Fig. 1) change simultaneously with the change in the shape of a plasma cell.

These changes are caused by the formation of a large amount of nitrogen dioxide around a plasma cell. Nitrogen dioxide screens the plasma luminescent radiation and thus restricts the brightness of plasma luminescence in the visible region. The production of NO_2 is attributed to the ionization of the air around a plasma cell by the high-power radiation from plasma.

A quickly formed mantle of the ionized air around a laser spark was discovered in Ref. 2. The observations of the source of continuum radiation in a laser spark have shown that it consists of two components. The first component is the radiation from the plasma center, and the second one is the radiation from the zone near the cell, which spreads at a speed higher than 10^8 cm/s. The instrumentation used did not permit detailed records of temporal changes in the front of this radiation. A possible cause of the appearance of the fast component of continuous radiation may be the ionization of N_2 and O_2 in the air in the front of a shock wave by the electron and short-wave radiation, arising at the initial stage of the expansion of laser-induced plasma due to the emission of multicharged ions and vortex structure of the fields in plasma.

The fast signal (luminescence of the air before the arrival of the shock wave during the optical breakdown), whose origin is attributed to the photoionization of the air by the radiation from a laser plume was observed in Refs. 3 to 5.

Upon focusing of a high-intensity laser pulse ($I > 10^9$ W/cm^2), the plasma, intensely emitting in the soft-X-ray and UV regions, is produced in the zone of optical breakdown. The spectral composition and the intensity of the ionizing radiation from the plasma depend on its optical thickness. For the optically thick plasma, the spectral composition is described by the Plank's function[6]:

$$f(\nu) = 15\nu^3/\pi^4 T^4{}_{,\mathrm{br}}\,[\exp(\nu/T_{\mathrm{br}} - 1)]^{-1}, \qquad (1)$$

where ν is the energy of a quantum.

With the Plank spectral distribution of the radiation from the plasma cell, the brightness temperature of the plasma is[5]:

$$T_{\mathrm{br}} = 6.3 \cdot 10^{-4}\, I^{4/9}, \qquad (2)$$

where I is the intensity of laser radiation, in W/cm^2; T_{br} is the temperature, in eV.

The ratio S of the black body radiation density W_{ion} in the spectral range from $\lambda = 0$ to $\lambda_1 = 0.11$ μm to the total radiation density W_{o} significantly depends on temperature. At high temperatures, the main part of the plasma radiation falls in the range of gas absorption ($T_{\mathrm{br}} = 80 \cdot 10^3$ K, $S = 0.9$).

The X-ray and UV radiation ionizes the ambient medium by breaking loose electrons from the inner (K) and outer (L) electron shells of atoms and molecules. In addition, molecules dissociate and their electronic levels are excited. In the case of ionization from the K-shell by the radiation with the characteristic temperature of 100 eV, besides a photoelectron, an additional high-energy electron is produced due to the Auger effect (transition of an electron from an outer shell to the K-vacation produced). For N_2 and O_2 molecules of the air, the Auger effect occurs with the probability of 99%, while the probability of spontaneous luminescence is only 1%. It can be believed that, during the photoionization, Auger electrons are additionally produced from the K-shell. These electrons are uniformly distributed in the energy ranges $\Delta E = 315$–385 eV for nitrogen and 456–520 eV for oxygen.[6] The calculations show[6] that, at short distances $x \sim 1$–3 mm from a plasma cell and at low air pressures $p < 0.1$ mm Hg, the photoionization by quanta plays the major role in the production of electrons. With the increase of x and p, the contribution of the photoionization decreases sharply and the ionization by fast electrons becomes the main mechanism.

The largest fraction of secondary electrons produced has the energy lower than 20 eV. They lose their energy and become thermal electrons for the time about $2 \cdot 10^9/n$ [s/cm^3], where n is the total concentration of air molecules.

The parametric effects in plasma (aperiodic instability, decay into two plasmoids, decay into the ion-acoustic and plasma waves, stimulated Brillouin scattering, stimulated Raman scattering) are known to lead to generation of the significant ionizing radiation of plasma with the penetration power up to several centimeters in the air.[7]

The plasma exposed to the high-power electromagnetic wave is unstable with respect to the build-up of high-frequency plasma electron waves (plasmons), caused by the absorption of the wave by two plasmons. The threshold value of the light flux needed for the development of the decay instability $\omega \to 2\omega_{\mathrm{Le}}$ in a homogeneous plasma is equal to 10^{12} W/cm^2, if the plasma is produced by the Nd laser radiation with a wavelength of 1.06 μm, and 10^{10} W/cm^2, if the plasma is exposed to the CO_2 laser radiation with a wavelength of 10.6 μm [Ref. 8]. Other parametric effects have close threshold values of the radiation power density, which are comparable with the possible local peak values of the radiation intensity of a multimode laser used in the experiment.

The energy distribution of electrons, obtained from the simulation of the processes of high-intensity radiation interaction with plasma, has spatial modulation in the phase space with a scale of $\lambda/2$ due to the presence of a standing electromagnetic wave, playing an important role in the heating of electrons.[9] The electron motion becomes stochastic for several periods of the laser field.

Thus, it can be concluded that for the extended plasma the stochastic electron heating is the main process providing for the energy accumulation by electrons. The idea of the considered model assumes nonequilibrium plasma with two electron temperatures, and the temperature ratio of the hot and cold electrons is 10^2–10^3 at the ion temperature an order of magnitude lower than the temperature of cold electrons. The number of hot electrons is small as compared to the cold ones, but they carry quite comparable energy.[10]

The results of experimental investigations of the air ionization at the atmospheric pressure by the radiation from plasma produced by a pulse of TEA CO_2 laser radiation are presented in Ref. 11. It is shown that the electron concentration decreases slowly with the distance from the plasma boundary (in the inverse proportion to the distance).

The slow decrease of the electron concentration with the distance from the source of ionization was observed not only in the air, but also in other gases[11] at the distances of 1 to 10 cm.

Thus, on the basis of numerous observations of the high air ionization at the distances up to 10 cm from the front of the shock wave of optical breakdown,[2–5] detection of high-energy electrons,[12] and theoretical models of electron acceleration in the laser plasma,[9,13] we can speak about the presence of the ionizing radiation in front of the shock wave of a plasma formation. This radiation is attenuated in the air in the inverse proportion to the distance x from the spark.

With the allowance for the divergence, the intensity of ionizing radiation W in front of the shock wave of the plasma formation can be presented in the form

$$W = W_{\text{ion}} \frac{R_0^2}{(R_0 + x)^2} \frac{1}{\alpha x + 1}, \tag{3}$$

where R_0 is the radius of a plasma cell; x is the distance from the plasma; W_{ion} is the intensity of the ionizing radiation of the plasma; α is the extinction coefficient. The intensity of the ionizing radiation achieves the threshold value W_{th} by the end of the path and can be determined from the degree of air ionization at the distance of propagation of the radiation from the front of the shock wave.

The relation between the power density of the plasma radiation, the mean free path Z of the ionizing particles, and the radiation extinction coefficient α is described by the equation, which allows α to be determined from the measurements of the air ionization as a function of the distance to the plasma cell:

$$Z \approx R_0 \left(\frac{W_{\text{ion}}}{W_{\text{th}} \alpha R_0} \right)^{1/3}. \tag{4}$$

The irradiation intensity J of air is characterized by the energy of the ionizing radiation per unit volume of the medium per unit time:

$$J = \frac{W}{Z} \approx \frac{(W_{\text{ion}}^2 W_n \alpha R_0)^{1/3} R_0}{(R_0 + x)^2 (\alpha x + 1)}. \tag{5}$$

The Table summarizes the main parameters of the laser radiation and the plasma of optical breakdown of air at the time t measured from the beginning of the laser pulse.

I, W/cm^2	T_{br}, K	W_0, W/cm^2	S	t, s	R_0, cm	Z, cm	$J(x = 0)$, eV/(cm^3·s)
10^8	$26 \cdot 10^3$	$2.7 \cdot 10^6$	0.2	10^{-7}	0.06	0.6	$4.8 \cdot 10^{24}$
				$5 \cdot 10^{-7}$	0.3	2.1	$1.6 \cdot 10^{24}$
10^9	$73 \cdot 10^3$	$1.6 \cdot 10^8$	0.85	10^{-7}	0.125	5.94	$1.4 \cdot 10^{26}$
				$5 \cdot 10^{-7}$	0.625	21.6	$4 \cdot 10^{25}$

Excited molecules, atoms, and ions, produced under the effect of ionizing radiation from the plasma formation on the air, undergo chemical reactions with each other and with N_2 and O_2 molecules. The investigations show[14] that these reactions yield nitrogen oxides NO, N_2O, NO_2, and ozone O_3 in the air. Soon after the formation, NO reacts with O_2, thus transforming NO_2:

$$2NO + O_2 \rightarrow 2NO_2. \tag{6}$$

At a weak irradiation, O_3 is the third main component of the air after N_2 and O_2. At the strong irradiation, the air becomes a mixture of nitrogen and nitrogen oxides.[14]

The investigations of the dependence of the rate of NO_2 production in the air on the electron energy have shown that nitrogen oxides are formed due to the ionization of nitrogen molecules and atoms. This is proved by the correlation between the nitrogen ionization function and the rate of formation of the nitrogen dioxide.[14]

The rate of formation of the nitrogen dioxide V for any ionizing radiation (except for neutrons) can be expressed by the following equation[15]:

$$V = 0.7 \cdot 10^{-7} \frac{Jp}{\varepsilon_0} \sqrt{\frac{RT(273 + T)}{\sigma M}}\, e^{\frac{-E}{R(273+T)}} \times$$

$$\times \int_0^{\varepsilon_{\max}} F(\varepsilon) g(\varepsilon) \mathrm{d}\varepsilon \ \frac{\text{mol}}{\text{cm}^3 \cdot \text{s}}, \tag{7}$$

where p is pressure, in mbar; J is the irradiation intensity, eV/(cm^3·s); R is the gas constant ($8.31 \cdot 10^7$ erg/(deg·mol)); M is the molecular weight of nitrogen (28); ε is the energy of the ionizing radiation; ε_0 is the ionization potential of N_2 (15.576 eV); $\varepsilon_{\max}$ is the maximum energy of radiation in the spectrum; $F(\varepsilon)$ is the spectral energy distribution function; $g(\varepsilon)$ is the ionization function; E is the energy of activation of the reaction (7.4 kcal/mol); σ is the coefficient of ion recombination, under normal conditions in the air $\sigma \approx 1.6 \cdot 10^{-6}$ cm^3/s; T is the temperature of air, in °C.

Figure 3 shows the dependences of the NO_2 concentration on the distance to the plasma formation for two values of the laser radiation intensity at the time of 10^{-7} and $5 \cdot 10^{-7}$ s. The concentration of NO_2 near the plasma cell was calculated by the equation for the rate of formation of the nitrogen dioxide (7) with the allowance made for the maximum NO_2 concentration observed in the air under irradiation of the air by the ionizing radiation (~ 6%), presented in Ref. 14.

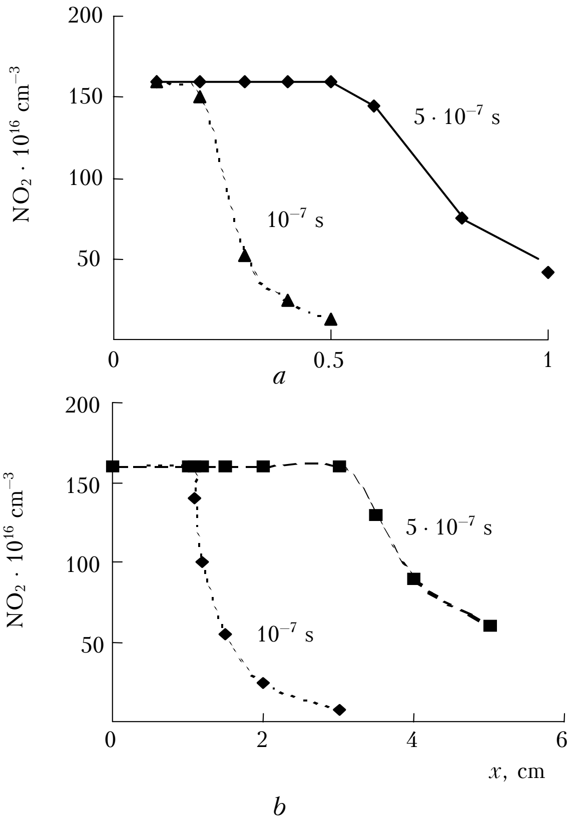

Fig. 3. Concentration of NO_2 in the air as a function of the distance to the plasma formation at the laser radiation intensity of 10^8 (*a*) and 10^9 W/cm^2 (*b*).

Based on the obtained data on the NO_2 concentration in the halo around the plasma formation, we have drawn the dependence of optical thickness D for the radiation with a wavelength of 0.4 μm (the cross section of attenuation by the nitrogen dioxide molecule is $2 \cdot 10^{-19}$ cm^2) on the power density of the laser radiation at the time of 10^{-7} and $5 \cdot 10^{-7}$ s (Fig. 4).

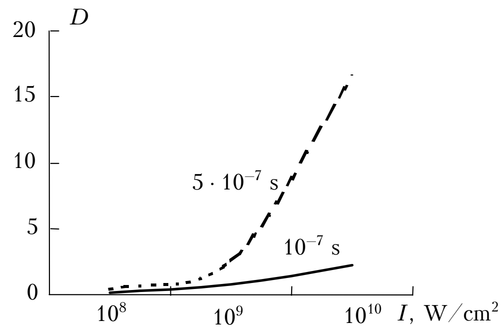

Fig. 4. Optical thickness D of the NO_2 gas mantle for radiation with the wavelength of 0.4 μm as a function of the laser radiation intensity I.

It can be seen from Fig. 4 that, at the laser radiation intensity higher than 10^9 W/cm^2, the optical density of the mantle increases sharply during $5 \cdot 10^{-7}$ s, significantly exceeding unity. As a result, it virtually screens the thermal radiation from the plasma cell.

The shock wave of optical breakdown of the air propagates in the gas with the high concentration of nitrogen dioxide, drastically increasing its temperature. In Ref. 16, the investigations were carried out in a shock tube filled with NO_2 mixed with argon. It has been found that at the gas temperature $T > 2500$ K the decomposition of NO_2 leads to the nonequilibrium distribution of the energy over the vibrational degrees of freedom of NO_2 molecules. The superequilibrium vibrational energy is stored during the isothermal stage $O + NO_2 \rightarrow \rightarrow NO + O^*$, and the fast quasiresonance exchange of vibrational quanta between O_2 and NO_2 leads to the excitation of NO_2 and to the separation of the vibrational temperature from the gas one. As some critical threshold in the temperature and the NO_2 concentration is exceeded, the reaction of decomposition becomes self-accelerating, and its rate exceeds the equilibrium one. Physically, pulsations of the parameters of nonequilibrium decomposition of NO_2 in the shock wave can be explained as follows: there are two energy pools – thermalized degrees of freedom T of a molecule and vibrational degrees of freedom T_v of the ensemble of O_2 and NO_2. The reaction of NO_2 decomposition forms a break between the temperatures of these pools $T_v > T$. If the initial direction of changes in T and T_v turns out opposite, then the reaction proceeds in the mode, which involves the oscillating energy exchange between the pools. The frequency of oscillation of the observed parameters in the shock waves from 50 (2600 K) to 500 kHz (4000 K) [Ref. 17] agrees with the frequencies of power fluctuations of the radiation from the center of optical breakdown of the air by the CO_2 laser radiation in the 0.4 μm absorption band of nitrogen dioxide.

With the increase of the air temperature, the spectra shift to the long-wave region and broaden.[18] A molecular "hot" band is formed, which is associated with the increase of the concentration of excited molecules. The absence of radiation from the plasma formation in the red spectral range at the laser radiation intensity higher than 10^9 W/cm^2 is caused by the attenuation of this radiation by the NO_2 molecular hot band, as well as by the radiation absorption during the photodissociation of ions of oxygen atoms (cross section of photodissociation of O^- is $6.5 \cdot 10^{-16}$ cm^2).

Thus, the presented model of emission properties of plasma formation appears to be capable of explaining the experimentally observed regularities in the luminescence of the optical breakdown cells in the air by the CO_2 laser radiation of microsecond duration with the intensity higher than 10^9 W/cm^2 [Ref. 19].

Conclusions

This paper presents the results of experimental investigations into the spectral and temporal characteristics of optical-breakdown plasma formations arising in the air during the propagation of microsecond CO_2 laser pulses with the power density of $5 \cdot 10^6$–$5 \cdot 10^9$ W/cm^2.

Three types of the temporal regularities have been revealed in the plasma luminescence at different values of the laser radiation intensity.

The mechanisms have been proposed for the processes leading to the obtained regularities of luminescence in different spectral ranges from plasma cells arising in the air at the laser radiation intensity up to 10^8 W/cm^2. The application of these mechanisms to the diagnostics of plasma dynamics has been discussed.[20]

The possibility of formation of the gas mantle with the increased NO_2 concentration around the breakdown cells, as well as its influence on the experimentally confirmed regularities of luminescence of air breakdown cells in the visible spectral region, has been demonstrated.

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