

## IONIZATION LOSSES OF $\lambda = 248$ nm UV-RADIATION IN AIR

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*A formula for the losses of KrF-laser radiation ( $\lambda = 248$  nm) due to multiquantum ionization of atmospheric air as a function of the radiation intensity and propagation distance is derived based on experimental data.*

Intense laser radiation with wavelength 200 ... 400 nm, propagating through gaseous media, undergoes nonlinear attenuation and ionization losses. As shown in Ref. 1, the probability of ionization is a power-law function of the radiation power density.

$$W \sim I^k \quad (1)$$

Here  $k$  is a fractional number, equal to the ratio of the ionization potential  $I$  to the photon energy  $h\nu$  ( $I$  is the ionization potential of a molecule (atom) in pure molecular (atomic) gases). This process is substantially different from multiphoton ionization, where  $\kappa$  is equal to the number of photons required for the process to be energetically possible. Multiphoton ionization is observed in rarefied gases and implies that  $\kappa$  photons interact simultaneously with the molecule.<sup>2</sup> In Ref. 1 the ionization of air by laser radiation with wavelength 266 nm (fourth harmonic of YAG: Nd<sup>3+</sup> laser radiation) in gases at pressures close to atmospheric pressure was studied. In this case collisions between molecules are important. The process is rather like a cascade, but it is different from what is customarily called cascade ionization, since a molecule can lose energy in a collision. In what follows we shall call this process multiquantum ionization (MQI). As shown in Ref. 1 this process is efficient and it is important to study it, since it affects the propagation of UV radiation in the atmosphere. Ionization losses can effect the range at which the atmosphere is studied with the help of excimer lasers, such as ArF, KrF, and XeCl lasers. In addition, the ionization process will affect the characteristics of the luminescence spectrum of the gases stimulated by optical radiation in the range 200–300 nm. The use of MQI in spectroscopy obviously opens up new, still unexplored, possibilities.

In this paper we study the ionization of air at atmospheric pressure by a KrF-laser beam with  $\lambda = 248$  nm. We present formulas for the ionization losses  $dj/dx$ , the distance over which the power density is halved, and the dependence  $j(x)$ .

**Experimental apparatus.** The radiation source was a model 1710 KrF laser 1 (Fig. 1) built by the Special Design Office at the Institute of Optical Physics of the Academy of Sciences of the USSR. The

beam was directed through a spatial filter 2 and diaphragms 3 and 4 into the ionization chamber 5 and then onto an FÉU-142 photocathode 6, which was insensitive to visible light. The flat, 35 mm in diameter electrodes<sup>7</sup> of the chamber were separated by a distance of 5.5 mm. One electrode was connected to a high-voltage source; the other (collecting) electrode was connected to the input of a charge-sensitive preamplifier 9 with a noise level of 0.2  $\mu$ V in a  $10^4$  Hz band and with a gain of  $2 \cdot 10^3$ . The signal from the preamplifier was fed into a S1-55 dual-beam oscillograph 10. The pulse from the FÉU was fed into the second input of the oscillograph. The amplitudes of both pulses were recorded by two digital voltmeters 11 and 12 operating synchronously with the sweep of the oscillograph. The linearity of the channels was checked in a 70 dB range with the help of a square-pulse generator. The linearity of the FÉU was checked with the help of a collection of Identical UFS-1 filters. The quantitative dependence of the magnitude of the voltage pulse in the ionization-measuring channel on the number of electrons in the space between the electrodes was determined with the help of  $\alpha$ -particles from a collimated Pu<sup>238</sup> source. The magnitude of the signal from the  $\alpha$ -particles fluctuated around 100 mV ( $98 \pm 2$ ). The rms amplitude of the noise was equal to  $\pm 0.4$  mV. The power of the UV-radiation pulse was measured with the help of an IMO-2M calorimeter. In the process, the pulse repetition frequency was equal to 12.5 Hz.

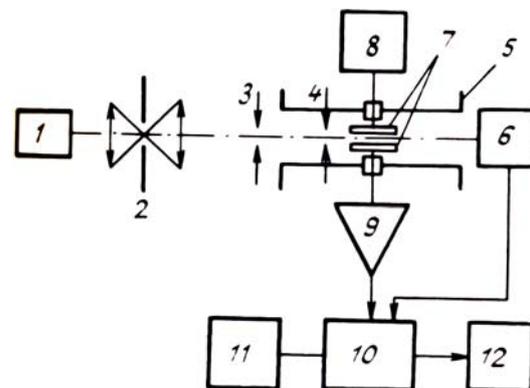


FIG. 1. Diagram of the apparatus.

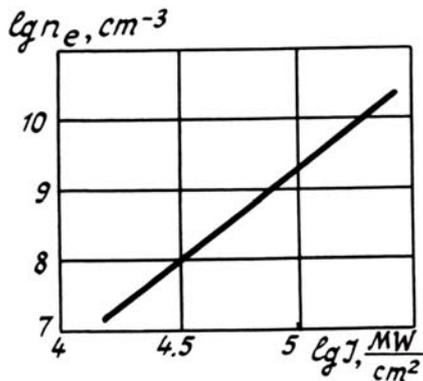


FIG. 2. The dependence  $n_e(j)$  on a logarithmic scale for air under normal conditions. The errors are of the order of the thickness of the line.

**Results and discussion.** The typical dependence of the electron density in the chamber as a function of the power density of the laser radiation is presented in Fig. 2 on a double logarithmic scale. The straight line was obtained by computer processing of a large volume of experimental data. The slope of the straight line  $\kappa$  is equal to  $2.41 \pm 0.06$ . The error is the rms value with a 5% significance probability. The normalized dependence of the electron density  $n_e$  ( $\text{cm}^{-3}$ ) on the power density  $j$  is given by

$$n_e = C j^{\kappa} = (5_{-2}^{+4}) \cdot 10^{11} j^{2.41} \quad (2)$$

where  $C$  is a function of  $k$ ,  $j = J/J_p$  is the normalized flux density, and  $J_p = 10^6 \text{ W/cm}^2$ . We emphasize that  $k$  is equal to the ratio of the ionization potential of oxygen  $I = 12.07$  to the photon energy  $\hbar\omega = 5.0 \text{ eV}$ . The oxygen comprises approximately 1/5 of the air, but it has the lowest ionization potential among the macroscopic components of the air. Thus for a mixture of gases  $\kappa$  corresponds to the ratio of the ionization potential of the molecular component with the lowest ionization potential to the photon energy. This result is not new.<sup>1</sup> This also occurred in the study of the process of multiquantum ionization of air by 266 nm UV-radiation as well as a 1:1:1 mixture He + CO<sub>2</sub> + N<sub>2</sub>. By measuring  $k$  it is possible to determine an unknown gaseous component — an admixture to a known gas, having a high ionization potential, for example He. Thus it is possible to determine or determine more accurately the ionization potential of the substance of interest in the gas phase. The minimum amount of impurity can be determined approximately from the relation

$$N > \frac{1}{\sigma_t \langle v \rangle \tau} \quad (3)$$

where  $\sigma_t$  is the effective gas-kinetic collision cross section of the molecules,  $\langle v \rangle$  is the average velocity of the molecules, and  $\tau$  is the width of the optical pulse. In equality (3) means that a molecule undergoes several collisions in the field of the wave.

We shall study the ionization losses of radiation with photon energy  $\hbar\omega$  owing to the nonlinear MQI process. The change in the radiation density per unit length is

$$\frac{dj}{dx} = -C' j^{\kappa} \beta \frac{I}{j_n} \quad (4)$$

where  $C' j^{\kappa}$  is the ionization intensity,  $\text{in}^{-1} \cdot \text{cm}^{-3}$ , and the quantity  $\beta$  is the factor by which the energy must exceed  $I$  in order for an average of one pair of ions to be formed.

Integrating Eq. (4) we express the power density at a distance  $x$  in terms of  $x_{1/2}$  — the distance over which one half of the initial power  $j_0$  is lost. In air

$$j(x) = \frac{j_0}{(1 + 1.6x/x_{1/2})^{0.71}} \quad (5)$$

The half-power length is given by the expression

$$x_{1/2} = \frac{j_n / \hbar\omega}{2C' \beta j_0^{1.41}} \quad (6)$$

The quantity  $C'$  appearing in Eqs. (4)–(6) has not yet been determined. To derive a theoretical expression for  $C'$  it is necessary to solve a system of kinetic equations using the matrix elements of the transition of the molecules to higher energy levels in the interaction with the UV-radiation field and to lower energy levels in collisions of the second kind.

We shall determine  $C'$  from the experimental data. The charge collected by the anode in the chamber depends on both the yield of photoelectrons and the rate of the dissipative processes — attachment and recombination of electrons. The balance equation has the form

$$\frac{dn_e}{dt} = C' j^{\kappa} - \alpha n_e^2 - \delta n_e \quad (7)$$

where  $\alpha$  is the recombination coefficient and  $\delta$  is the attachment rate.

Under the conditions of the experiment the second and third terms on the right side of Eq. (7) are much smaller than the first term, since the recombination and attachment times in the electric field of the chamber with a voltage of 5 kV/cm are much longer than the UV-pulse width ( $\tau = 30 \text{ ns}$ ). Neglecting the second and third terms and integrating Eq. (7) we obtain the solution of the simplified equation

$$n_e = C' \tau j^{\kappa} \quad (8)$$

And, comparing with Eq. (2), we obtain

$$C' = \frac{C}{\tau} = 1.5 \cdot 10^{19} \text{ s}^{-1} \cdot \text{cm}^{-3} \quad (9)$$

Given the value of  $C'$  we can find a simple expression for the half-absorption length in air for radiation with  $\lambda = 248 \text{ nm}$ :

$$x_{1/2} = \frac{0.4}{j_0^{1.41}} \quad (10)$$

where  $j_0$  is expressed in  $\text{MW}/\text{cm}^2$  and  $x_{1/2}$  is expressed in km.

When the formulas (5)–(10) are used for practical calculations the beam divergence and the specific conditions of propagation — the distance, linear absorption, etc. — must be taken into account.

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