Ionization of gases by 266-nm ultraviolet light

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Experiments show that the ionization probability for various gases is a power-law function of the intensity of optical radiation at $\lambda = 266$ nm. The exponent, not an integer, is approximately equal to the ratio of the ionization potential to the photon energy. The ionization probabilities are estimated.

The interactions of atoms and molecules with optical radiation have been studied in the visible and infrared ranges.[1,2] In the present letter we report a study of the ionization of various gases in the ultraviolet part of the optical range, where the simultaneous application of three or four photons provides enough energy for ionization.

Ultraviolet light with a wavelength of 266 nm is produced by a double frequency doubling of the output from an Nd^{3+} laser (1,2,3 in Fig. 1). It is then amplified (6) and converted in lithium niobate crystals (7,8). The ionization is detected with an ionization chamber (14) with electronic collection. The light extracted by means of a prism (9) and a diaphragm (10) is focused at the center of the gap between the electrodes of the chamber (the diameter of the electrodes is 15 mm, and the gap between them is 4.5 mm). The light is then collected by a lens (13) on the cathode of a photomultiplier (16). The collecting electrode of the ionization chamber is connected to the input of a preamplifier (19). The final amplification and detection of the electronic pulse are performed by an oscilloscope (22). The output signal from the photomultiplier is fed through a 50-ns delay line (20) and a load-matching unit (21) to the same input of the oscilloscope. A camera (23) takes photographs of the oscilloscope screen. Measurements are taken in two recording modes, "fast" and "slow." In the first mode we use preamplifiers (18 and 19) with a rise time of 2 ns and a noise level of 20 μV; in the second mode, the preamplifier (19) has a bandwidth ~ 10 kHz, while the noise level is $\lesssim 0.2$ μV, so that the measurement range can be expanded by two orders of magnitude. The fast measurements tell us where and how the ionization occurs and permit quantitative estimates of the loss of electrons due to recombination and the formation of negative ions. In the slow mode, the ionization measurements are controlled by ionization pulses produced by α particles from a Pu^{239} source. The signal-to-noise ratio in this case is 15. The apparatus that measures the intensity of the ultraviolet light is calibrated through measurements of the energy of the laser pulses. The accuracy of this calibration is no worse than 5%. The double nonlinear frequency conversion of the initial light helps suppress the effect of fluctuations of the mode composition of the light, although these fluctuations remain the primary cause of the scatter in the experimental data.

The experimental results are shown in Figs. 2 and 3. Figure 2 corresponds to the fast measurement mode. Measurements were taken in oxygen, nitrogen, carbon diox-

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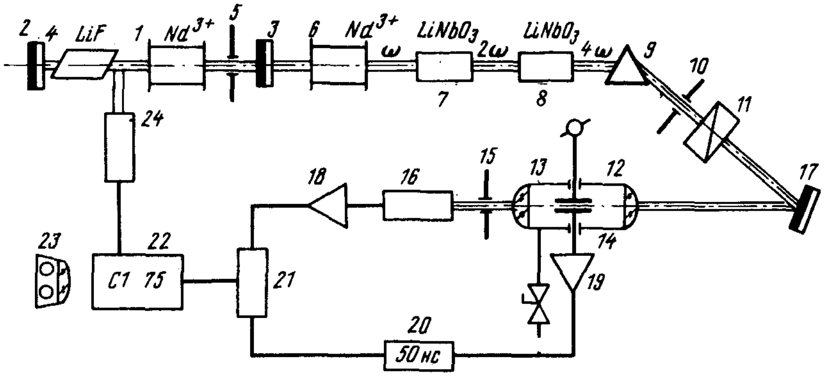

FIG. 1. The experimental arrangement.

ide, and ammonia at a pressure of 750 torr and in water, alcohol, and acetone at pressures of 110, 200, and 250 torr. The intensity of the ultraviolet light, J, is plotted along the logarithmic abscissa, while the relative electron density N is plotted along the logarithmic ordinate. These results show that the number of ion pairs per unit volume is a power-law function of the light intensity. Figure 3 compares results on $\log N(\log J)$ for N_2, O_2, CO_2, and C_2H_2 obtained in the slow measurement mode under standard conditions ($P = 760$ torr, $T = 295$ K). The data on O_2 have been corrected for the loss of electrons due to attachment during drift toward the collecting electrode.

We can draw some conclusions from these experimental results. 1) At light inten-

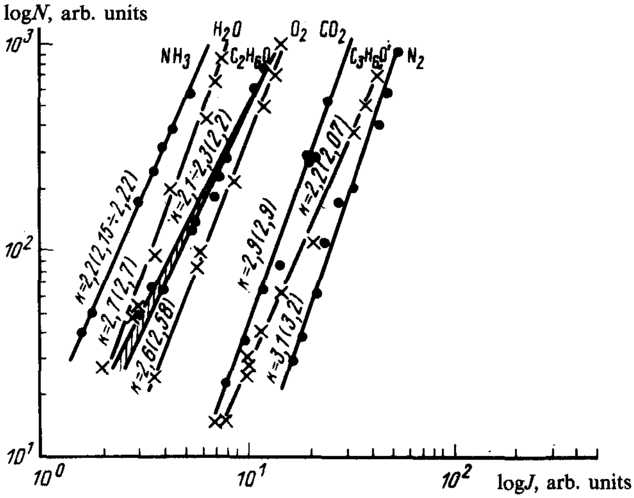

FIG. 2. The electron density N versus the light intensity J, in arbitrary units ($k = \partial \log N/\partial \log J$). The ratio of the ionization potential to the photon energy is shown in parentheses.

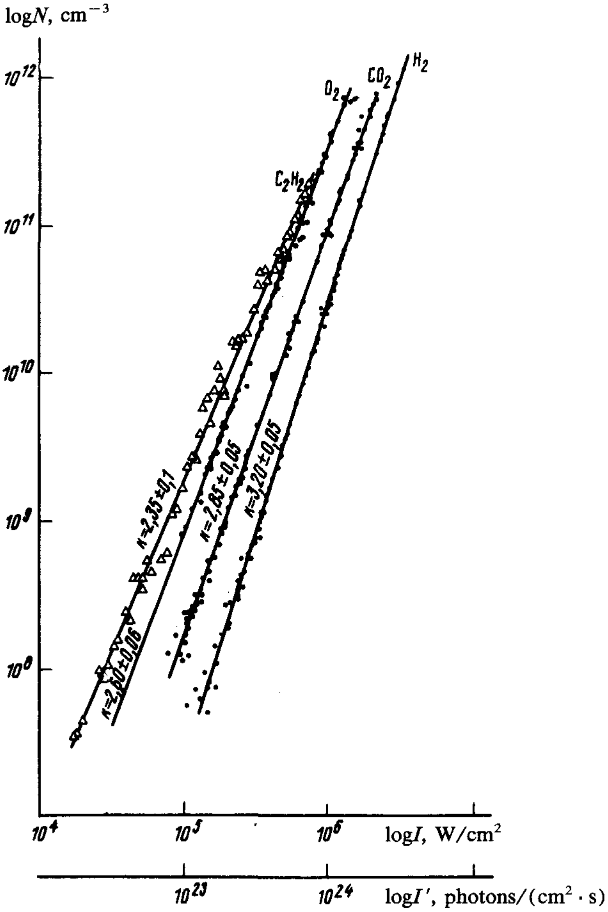

FIG. 3. The electron density N versus the light intensity J ($k = \partial \log N / \partial \log J$).

sities in the range 10^5–10^7 W/cm^2, the ionization probability is described by the power law $w = AJ^k$, where the constant A depends on the particular gas. 2) The exponent k is quite different from an integer. 3) The exponent k is, highly accurately, equal to the ratio of the ionization potential of the gas, I, to the photon energy, $\hbar\omega = 4.66$ eV: $k = I/\hbar\omega \pm (0.05$–$0.1)$. 4) An estimate of the probability of the process yields $2.0^{+2.4}_{-1.2} \times 10^{-78}$ for A in hydrogen, $3.0^{+3.6}_{-1.8} \times 10^{-63}$ in oxygen, $1.0^{+1.2}_{-0.6} \times 10^{-69}$ in carbon dioxide, and $1.0^{+1.2}_{-0.6} \times 10^{-56}$ in acetylene.

Fractional values of k smaller than $k_0 = \left\langle \frac{I}{\hbar\omega} + 1 \right\rangle$ have been encountered in previous studies of multiphoton ionization. In Refs. 3 and 4, for example, in studies of seven-photon ionization in hydrogen and nitrogen, respectively, the values of k were closer to the ratio $I/\hbar\omega = 6.6$ than to the integer 7. The authors of those papers, however, did not attribute much importance to this circumstance because of the uncertainty in the determination of k. In the present experiments, with low fields, 10^3–10^4

V/cm, the shift and broadening of the excited levels cannot be accepted as an argument for reducing k; the same comment applies to resonant effects because of the low multiplicity k and the relatively narrow output line, with the width $\approx 1\ \mathrm{cm}^{-1}$.

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