OPTICAL BREAKDOWN OF COMPRESSED GASES BY CO_2 LASER RADIATION

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The breakdown of compressed gases by CO_2 laser radiation was investigated theoretically and experimentally. The high-pressure breakdown ($p \gtrsim 30$ atm) differed considerably from the breakdown in low-pressure gases: the threshold flux density and the energy of the radiation, which were decreasing functions of p at low pressures, increased with p at high pressures. The pressure dependences of the threshold density and energy of the radiation were different for molecular and atomic gases.

INTRODUCTION

MUCH work has been done on the breakdown of gases by laser radiation. The breakdown of atomic (see, for example, reviews[1–4]) and molecular gases[5,6] has been considered. The case of relatively slow acquisition of energy by electrons from the optical field, followed by instantaneous ionization of an atom by an electron which has reached an energy of the order of the ionization potential I, has been discussed in[1], whereas the case of strong fields when electrons "run away" far outside the range of the energies of effective excitation and ionization is considered in[7]. However, in these investigations the optical breakdown has been studied at relatively low gas pressures p: in all cases, the frequency of laser light ω has been much higher than the frequency $\nu_{eff}(p)$ of the elastic collisions between electrons and gas atoms or molecules. At high pressures $\nu_{eff}(p) \gtrsim \omega$ applies and the nature of the optical breakdown in gases is quite different: the threshold flux density $q^*(p)$ and the threshold energy $W^*(p)$ of the laser radiation, which are decreasing functions of the pressure in the $\nu_{eff}(p) < \omega$ case, are found to rise with p when $\nu_{eff}(p) > \omega$. The dependences $q^*(p)$ and $W^*(p)$ are quite different for atomic and molecular gases.

Investigations of the "optical strength" of compressed gases are of physical and practical interest: knowledge of this strength can be used directly in the determination of the limiting parameters of the lasers emitting infrared radiation and operating at pressures of hundreds of atmospheres.[6,8]

THEORY

At pressures $p \gtrsim 1$ atm the breakdown of gases by laser radiation is the result of avalanche ionization and the rate of development of an avalanche is governed by the electron energy distribution function $F(t, \epsilon)$ which satisfies the transport equation

$$\frac{\partial F}{\partial t} = -\frac{\partial}{\partial \varepsilon} I_q + \left(\frac{\partial F}{\partial t}\right)_{ex}. \tag{1}$$

Here, $(\partial F/\partial t)_{ex}$ is the change in the distribution function due to collisions of electrons with other particles, and

$$I_q = {}^1/_3 \alpha (F - 2\varepsilon \partial F / \partial \varepsilon) \tag{2}$$

is the flux of electrons in the energy space, governed by the radiation field.[1] Equation (2) includes the quantity

$$\alpha = \frac{e^2 E_0^2 \nu_{eff}}{2m(\omega^2 + \nu_{eff}^2)} \tag{3}$$

(where e, m, E_0 are, respectively, the charge and mass of an electron and the amplitude of the electric field) which determines the rate of acquisition of energy by an electron from the laser field in accordance with the relationship

$$\frac{\partial}{\partial t}\left(\int \varepsilon F d\varepsilon\right) = \int_0^\infty \varepsilon \frac{\partial I_q}{\partial \varepsilon} d\varepsilon = \alpha. \tag{4}$$

It follows from Eq. (4) that, in the case of breakdown of atomic gases, the growth constant γ of an avalanche is

$$\gamma = k / \tau, \tag{5}$$

where $\tau = I/\alpha$ is the characteristic time for the acquisition of an electron of an energy ϵ close to the ionization potential I; k is the probability that an electron passes through the excitation zone.

Equation (5) for the avalanche growth constant applies not only to atomic gases but also to molecular gases at relatively high laser flux densities. For example, in the case of N_2, $p \approx 1$ atm, and $\lambda = 10\ \mu$, the radiation flux density must exceed 10^{11} W/cm^2. In the opposite case of lower flux densities (this case is of greater practical importance), the dependence of the avalanche growth constant on the flux density and molecular characteristics of a gas is quite different.

Physically, this is related to the efficiency of the deceleration of electrons in the excitation of the vibrational levels of a molecule. These levels form a potential barrier so that the motion of an electron in the energy space resembles tunneling and an exponentially attenuating factor appears in the dependence of γ on the field.

The collision term in the transport equation, which describes the deceleration of electrons in the excitation of the molecular vibrational levels, can be represented in the form

$$\left(\frac{\partial F}{\partial t}\right)_{ex} = N_0 \sum_m \Big\{ \sigma_{0m}(\varepsilon + \hbar\omega_{0m}) F(\varepsilon + \hbar\omega_{0m}) v(\varepsilon + \hbar\omega_{0m}) - \sigma_{0m}(\varepsilon) F(\varepsilon) v(\varepsilon) \Big\} \approx \frac{\partial}{\partial \varepsilon}[\alpha^*(\varepsilon) F(\varepsilon)], \tag{6}$$

where N_0 is the concentration of the molecules, σ_{0m} is the excitation cross section of the m-th vibrational level with a quantum energy $\hbar\omega_{0m}$, $v(\epsilon)$ is the velocity of the electrons,

$$\alpha^*(\varepsilon)=N_0 v(\varepsilon)\sum_m \sigma_{0m}(\varepsilon)\hbar\omega_{0m}$$

is the rate of loss of the electron energy in the excitation of the vibrational levels. If we approximate $\alpha^*(\epsilon)$ by the expression $\alpha^*(i)\Delta\delta(\epsilon - i)$, where i is the energy corresponding to the maximum of $\alpha^*(\epsilon)$ and Δ is the characteristic "width" of the function $\alpha^*(\epsilon)$ (for example, for N_2 we have $\Delta \approx 2$ eV and $i \approx 1.5$ eV[9]), we find from Eq. (1) the change in the function $F(\epsilon, t)$ on transition through the point $\epsilon = i$:

$$F(t,i+0)=F(t,i-0)\exp\left\{-\frac{3}{2}\frac{\alpha^*(i)\Delta}{\alpha i}\right\}. \tag{7}$$

Physically, the ratio $F(t, i + 0)/F(t, i - 0)$ represents the probability of penetration of an electron across the barrier formed by the vibrational levels of a molecule. If we introduce the frequency $\Omega = i/\alpha$ for the revolution of electrons in the energy band $[0, i]$, we obtain the following expression for the avalanche growth constant:

$$\gamma=k\Omega\exp\left\{-\frac{3}{2}\frac{\alpha^*(i)\Delta}{\alpha i}\right\}, \tag{8}$$

where the coefficient k takes account, as in Eq. (5), of the excitation of the electronic terms of the molecule. The breakdown flux density q^* and the energy W^* of a laser pulse are found from the relationship $\gamma\tau$ = const, where τ is the pulse duration.

EXPERIMENTAL RESULTS

Breakdown in compressed gases was induced by the radiation provided by a high-power ionization-type CO_2 laser, focused by a spherical mirror ($f \approx 10$ mm). The breakdown was deduced from the appearance of a bright flash in the focus of the mirror. The laser, which was filled with a $CO_2:N_2$ mixture (1 : 2) at a pressure p = 2 atm, produced radiation pulses of total energy up to 0.5 J. The shape of the pulses was recorded oscillographically (Fig. 1). A focusing mirror was placed inside a metal chamber of $V \approx 1000$ cm^3 volume. A ZnSe plate, 3 mm thick, was used as the entry window of the chamber. The diameter of the focal spot measured under these conditions was ~50 μ. We investigated pure N_2 and He gases (impurity content < 0.003%), dried air, and CO_2 of technical purity. The gas pressure in the chamber was 1–60 atm.

The energy and shape of the laser pulses were determined by deflecting some of the radiation with a light splitter to a calorimeter and a photoresistor (Ge : Zn cooled with liquid nitrogen, time resolution at least 50 nsec).

The relative values of the optical breakdown threshold were determined to within 10% and the error in the absolute measurements was ~50%.

A 30–40% change in the pulse duration, achieved by varying the pressure of the working mixture in the laser chamber, indicated that the optical breakdown of the molecular gases was governed by the power and not the energy of the radiation pulse. The brightness and the size of the spark produced on breakdown de-

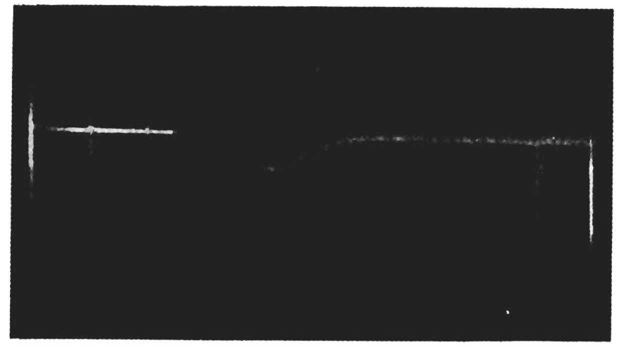

FIG. 1. Shape of laser radiation pulses (one horizontal division corresponds to 1 μsec).

pended on the nature of the gas. Light flashes at the focus of the mirror obtained for an optical flux $q/q^* \approx 1.05$ in CO_2 at p = 1 atm. were photographed (Fig. 2). The structure of the flash was the same for all the gases and pressures: a narrow cone converging at the focus was accompanied by a luminous plasma region expanding toward the mirror—opposite to the radiation flux.

The pressure dependences of the threshold flux densities of the laser radiation are plotted for various gases in Fig. 3. At relatively low pressures (1–10 atm) the optical breakdown threshold decreased with increasing pressure for all the molecular gases and for the atomic He. At higher pressures the threshold power density for the molecular gases increased rapidly with the pressure, whereas the corresponding density for He did not rise right up to 50 atm.

The optical strength of the $CO_2:N_2$ mixtures was considerably higher than the optical strengths of the pure N_2 and CO_2 gases.

DISCUSSION OF RESULTS

In strong fields ($\alpha \gg \alpha^*$) the avalanche growth constant of atomic and molecular gases is given by Eq. (5). According to Eq. (5), the optical strength of a gas is governed by the energy and not the power of a laser pulse. The dependence of the threshold energy on the gas pressure is given by

$$W^*(p)\propto\frac{\omega^2+\nu_{eff}^2(p)}{\nu_{eff}(p)}. \tag{9}$$

The elastic collision frequency $\nu_{eff}(p)$ is proportional to the pressure p. Thus, at low pressures p we have $W^* \propto 1/p$; conversely, at high pressures p we have $W^* \propto p$.

In the breakdown of helium by the CO_2 laser radiation the minimum value of $W^*(p)$ corresponds to ~30 atm; the experimentally obtained curve (Fig. 3), showing the dependence of the threshold flux density up to ~45 atm, indicates saturation of the dependence $q^* = q^*(p)$ at He pressures of ~40 atm.

FIG. 2. Photograph of a spark in optical breakdown of CO_2 at p = 1 atm.

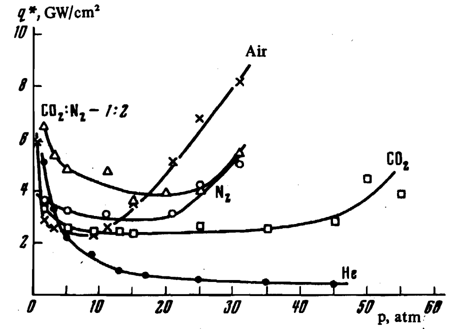

FIG. 3. Dependences of the threshold flux density of CO_2 laser radiation on the pressure of compressed gases.

We shall now estimate the optical strength of a molecular gas. According to Eq. (8), this strength is governed by the lesser flux density (the dependence of q* on the pulse duration is logarithmic). We shall obtain numerical estimates for molecular nitrogen at p = 1 atm for CO_2 laser radiation pulses of $\tau = 10^{-6}$ sec duration and $\lambda = 10\ \mu$ wavelength. According to[9,10],

$$\sum \sigma_{0m}\hbar\omega_{0m} \approx \langle\hbar\omega_{0m}\rangle \sum \sigma_{0m} \approx 0.4\cdot 3\cdot 10^{-16}\,\mathrm{eV\cdot cm^2};$$

$$I \approx \Delta \approx 2\,\mathrm{eV},\quad \nu_{eff} \approx 6\cdot 10^{12}\ \mathrm{sec^{-1}}$$

If we assume that $k \approx 0.1$, we find that Eq. (8) gives $q^* \approx 10^9\ \mathrm{W/cm^2}$ for the breakdown criterion $\gamma\tau = 40$. A similar estimate without allowance for the deceleration of electrons by the vibrational levels gives, in accordance with Eq. (5), the value $q^* \approx 10^7\ \mathrm{W/cm^2}$.

The dependence of the threshold flux density on the gas pressure is as follows. At low pressures we find that $q^* = q^*(p)$ is a slowly decreasing function of p: the pre-exponential factor in Eq. (8) depends on p, but the pressure dependence of $\alpha^*/\alpha \sim 1 + (\nu_{eff}/\omega)^2$ is of little importance at low values of p. At high pressures p, when $\nu_{eff}(p) \gtrsim \omega$, the slow fall of $q^*(p)$ is replaced by a fast rise: $\alpha^*/\alpha \propto \nu_{eff}^3$ and the dependence of q* on p becomes quadratic. According to[10], the collision frequency for N_2 $\nu_{eff} \approx 6\times 10^{12}$ p $\mathrm{sec^{-1}}$ (p in atm) in the range of electron energies of interest to us (1–2 eV). Consequently, the rapid rise of $q^*(p)$ at $\lambda = 10\ \mu$ begins from pressures of 20–30 atm.

The experimental curves plotted in Fig. 3 show clearly the obtained dependences of the threshold flux densities on the gas pressure.

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