Optical breakdown of gases induced by the thermal explosion of suspended macroscopic particles

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A theoretical analysis is given of a mechanism proposed for the initiation of optical breakdown in gases by "thermal explosion" of macroscopic particles suspended in them. It is shown that in the far infrared (and, in particular, at $\lambda = 10.6\ \mu$) this mechanism is probably largely responsible for the breakdown. The results obtained are used to provide a qualitative and quantitative explanation of many experimental data on the breakdown in gases induced by CO_2 laser radiation, which so far have been difficult to explain. Conditions are discussed under which this breakdown-initiating mechanism may play an important role in the visible and far infrared. Observations of this effect at $\lambda = 1.06\ \mu$ are reported.

1. A macroparticle suspended in a gas exposed to the field of an electromagnetic wave of intensity I absorbs energy from the wave at a rate $Q = \sigma_a I$, where σ_a is the absorption cross section of the particle. If this cross section is large enough, then for certain definite values of I lower than the threshold for optical breakdown in the ambient gas, the particle may undergo a "thermal explosion." This phrase describes the situation when the total energy a/u_s absorbed by the particle, and stored in it while it is held inertially in the vapor state, exceeds the evaporation energy $\rho q v = N_p q_1 v_1$, where a is the linear size of the particle, v is its volume, ρ and N_p are, respectively, the mass density and atomic density of the particles, q and q_1 are the heats of evaporation of the particle material per unit mass and per particle, respectively, and u_s is the velocity of sound in the vapor at temperatures close to the critical temperature T_{crit} of the particle material. It will be assumed throughout that the length of the radiation pulse is $\tau \gg a/u_s$.

During the explosion we can neglect thermal losses by the particle due to thermal conduction and, correspondingly, the threshold intensity I_{exp} for the explosion is given by the condition[1)]

$$\sigma_a I_{exp} a/u_s \approx \rho q v.$$

It will be assumed that the particles are spherical in shape with radius a. We now introduce the absorption efficiency $K_a = \sigma_a/\pi a^2$, so that the threshold intensity for the explosion becomes

$$I_{exp} \approx 4\rho q u_s/3K_a. \quad (1)$$

The particle is initially in the condensed state. If it enters the radiation field of intensity $I \geq I_{exp}$ then afer a time $t < a/u_s$ it is converted into a gas (vapor) region at temperature $T \sim T_{crit}$, density $N \sim N_p \sim 5 \times 10^{22}$ cm^{-2}, and size ~a. This dense vapor region spreads hydrodynamically with velocity u_s and may initiate optical breakdown even when I is much lower than the threshold intensity for the cold ambient gas. In fact, during this expansion process (where the vapor density decreases from $N \sim N_p$) there are always optimum conditions for the development of electrical avalanches when the radiation frequency is $\omega \approx \nu_{eff} = N\langle u_e \sigma_{tr}\rangle$, i.e., it is equal to the effective collision frequency between electrons and the atoms of the vapor. In this situation, the rate of increase in the energy of the electrons is a maximum and is given by

$$(d\varepsilon/dt)_{max} = 2\pi e^2 I/mc\omega$$

(e and m are the charge and mass of the electron and c the velocity of light), and the electron mean free path is $l_e = (N\sigma_{tr})^{-1} \sim u_e/\omega$ ($u_e \sim 10^8$ cm/sec is the mean velocity of free electrons).

Assuming that $a \gg l_e$ (for the CO_2 laser radiation $l_e \sim 10^{-6}$ cm), we find that the breakdown threshold in the above vapor region is approximately determined by the condition

$$(\nu_i - \nu_d) a/u_s \approx k,$$

where $k = \ln(N_e/N_{e0}) \approx 50$ is the number of electron generations necessary for breakdown, $\nu_d = \pi^2 D/a^2$ is the rate of free electron diffusion from the region, and $\nu_i = \Delta^{-1}(d\xi/dt)_{max}$ is the rate at which atoms in the vapor are ionized by the electrons (Δ is the ionization potential of the atoms). Since the diffusion coefficient is $D \approx u_e^2/3\nu_{eff} \approx u_e^2/3\omega$, where $u_e = (\Delta/m)^{1/2}$, we obtain the following expression for the threshold intensity for breakdown in the vapor region:

$$I_0(a) = \frac{c\Delta^2}{2e^2a^2}\left(1 + \frac{k}{3}\frac{a}{u_e/\omega}\frac{u_s}{u_e}\right). \quad (2)$$

We can always neglect the energy lost by electrons due to inelastic collisions, since the spontaneous emission by the excited atoms (molecules) is "trapped." In fact, the absorption coefficient for radiation on wavelength λ is $\mu_1 = (\lambda/2)^2 NA/\Delta\omega$, where $\Delta\omega$ is the spectral width of the absorption line and A is the probability of spontaneous emission. One can meaningfully consider the energy losses only for $A > u_s/A$. We then have

$$a\mu_1 > \left(\frac{\lambda}{2}\right)^2 N\frac{u_s}{\Delta\omega} = \pi^2\frac{c^2}{\sigma_{tr}}\frac{u_s}{u_e}\frac{\omega}{\Delta\omega}\frac{\omega_L}{\omega^3} > \pi^2\hbar^3\frac{c^2}{\sigma_{tr}}\frac{u_s}{u_e}\frac{\omega}{\Delta\omega}\frac{\omega_L}{\Delta^3},$$

where $\omega_L = Nu_e\sigma_{tr}$ is the laser frequency and $\omega = 2\pi c/\lambda$ is the frequency of the spontaneous emission. Hence, it is clear that when $u_s/u_e \sim 10^{-3}$, $\sigma_{tr} \sim 3 \times 10^{-16}$ cm^2, and $\sigma/\Delta\omega \gtrsim 10$, the optical thickness for the CO_2 laser radiation ($\omega_L = 1.8 \times 10^{14}$ sec^{-1}) is $a\mu_1 > 1$ even for helium which has the maximum ionization energy ($\Delta = 24.5$ eV).

Losses of electrons due to attachment can always be neglected for vapor temperatures $T \sim T_{crit}$. Energy losses due to elastic collisions with the atoms are unimportant when $I > (m/M)mc\omega^2\Delta/\pi e^2$, where M is the mass of the atom. For the CO_2 laser radiation this means means that $I[\text{W/cm}^2] > 10^8\Delta[\text{eV}]/A_1$, where A_1 is the atomic weight.

The development of breakdown in the dense vapor region, which appears as a result of the thermal explosion of the suspended macroparticle, leads to the appearance of a strong spherical shock wave. The front of this wave produces heating and ionization of the "pure" gas which, in turn, leads to the expansion of the absorption zone, i.e., to the breakdown of the ambient gas.[1] The threshold intensity $I_{th}(a)$ for this mechanism of gas breakdown is clearly the larger of the above two intensities, $I_{exp}(a)$ and $I_0(a)$, i.e.,

$$I_{th}(a)=\max\{I_{exp}, I_0\},$$

where for a macroparticle with given optical and thermophysical properties, and given diameter a, the mechanism is significant only provided the threshold breakdown intensity for the "pure" gas (in the presence of initiating electrons) is $I_{th}^{gas} > I_{th}(a)$.

It will be shown below that, in most cases, $I_{exp} > I_0$ and, therefore, $I_{th}(a) = I_{exp}(a)$. In the final analysis, this is connected with the fact that the breakdown-initiating mechanism which we are considering is effective (significant) only when the particle is large enough. Under real conditions, when the radius a of the particle is governed by a certain probability distribution, it is only the large-diameter tail of this distribution which plays the dominant role and is defined by radii $a > a_m$, where a_m is the median radius. The inequality $I_{exp} > I_0$ for sufficiently large a is a direct consequence of the fact that, as a increases, the intensity $I_0(a)$ given by Eq. (2) decreases monotonically while $I_{exp}(a)$ decreases only initially and then tends to a constant value. In fact, the absorption efficiency K_a in Eq. (1) depends on the optical thickness $2a\mu$ of the particle, where μ is the absorption coefficient of the particle material (see[2]). For "weakly absorbing particles" when $2a\mu \ll 1$, we have $K_a \approx 4/3a\mu$; for "highly absorbing particles," when $2a\mu \gg 1$, we have $K_a \approx 1$. Consequently, according to Eq. (1), the intensity I_{exp} is

$$I_{exp}=\begin{cases}\rho qu_s/a\mu, & 2a\mu\ll 1\\ \rho qu_s, & 2a\mu\gg 1\end{cases}. \quad (3)$$

For metal particles the intensity $I_{exp} = I_{exp}^M$ does not, in general, depend on the particle diameter, since for such particles (see[3])

$$K_a=K_a^M\approx 3(\omega/2\pi\sigma)^{1/2}\ll 1$$

where σ is the conductivity and, therefore,

$$I_{exp}^M\approx {}^1/_3\rho qu_s(2\pi\sigma/\omega)^{1/2}\gg\rho qu_s$$

(the metal particles are always weakly absorbing).

We note that the intensity $\rho qu_s = N_p q_i u_s$ is independent of the optical properties of the particles and, for most possible macroimpurities, it lies in the range $10^3-3 \times 10^8$ W/cm².

We must first verify that for particles for which the absorption coefficient of the particle material is $\mu \ll 10^5$ cm^{-1}, i.e., for practically all the macroimpurities which are encountered in practice with the exception of metals and media such as soot, which we shall consider separately, we have $I_{exp}(a) > I_0(a)$ in the visible and infrared, at least for $a \geq 10^{-6}$ cm. This follows from the fact that, according to Eqs. (2) and (3), this inequality is satisfied for

$$a>a^*\equiv\frac{\Delta}{2q_1}\mu\lambda_p\frac{u_e^2}{\pi u_s c}\lambda_p,$$

where the plasma wavelength is $\lambda_p \equiv c(\pi m/N_{part}e^2)^{1/2} \approx 10^{-5}$ cm and the quantities $\Delta/2q_1$ and $u_e^2/\pi u_s c$ are of the order of unity so that $a^* \lesssim 10^{-6}$ cm. It will be clear from the ensuing analysis that, under real conditions, radii $a \lesssim 10^{-6}$ cm are unimportant and, therefore, for the broad class of particles which we are considering ($\mu\lambda_p \ll 1$)

$$I_{th}(a)\equiv I_{exp}(a).$$

Let us now consider particles for which the absorption coefficient of the material is $\mu \gtrsim 10^5$ cm^{-1}, i.e., $\mu\lambda_p \gtrsim 1$. These are metal particles and soot particles. For long-wave radiation, when $\sqrt{\lambda_p/\lambda} \ll 1$, i.e., for example, in the case of CO_2 laser radiation (λ = 10.6 μ) and for the type of particles which we have been considering ($\mu\lambda_p \gtrsim 1$), we may suppose, as before, that $I_{th}(a) = I_{exp}(a)$. In fact, in this case, we can use Eqs. (2) and (3) to show quite readily that when

$$a>a^*\equiv\lambda_p[(\Delta/2q_1)(u_e^2/\pi u_s c)]^{1/2}\approx\lambda_p$$

[for metals $a > \sqrt{3}\lambda_p(\omega/2\pi\sigma)^{1/4}$], we have $I_{exp} \approx \rho qu_s$ (for metals $I_{exp} \approx I_{exp}^M$) and this value is greater than $I_0(a)$. Since $\lambda_p \approx 10^{-5}$ cm, particles with radii $a < a^*$ are practically ineffective, just as before. It is only in the visible and near-infrared, where $\sqrt{\lambda_p/\lambda} \sim 1$, that the situation changes and, in a definite real interval of values of a, the threshold intensity I_{th} can be represented by the function $I_0(a)$. In actual fact, in this case, Eqs. (2) and (3) show that $I_{exp} \approx \rho qu_s$, and this exceeds $I_0(a)$ only when

$$a>a^*\equiv\frac{2k}{3}\frac{\Delta}{2q_1}\frac{\lambda_p}{\lambda}\lambda_p.$$

Since $2k/3 \gg 1$, we have $a^* \gg \lambda_p \approx 10^{-5}$ cm and, therefore, the interval of effective values of the radius a is not exhausted by the condition $a > a^*$.

Therefore, in the visible and near-infrared we have from Eq. (2) for soot particles ($\mu\lambda_p \gtrsim 1$)

$$I_{th}(a)\approx\begin{cases}\dfrac{k}{6}\dfrac{mc\omega u_s\Delta}{e^2 a}, & a<\dfrac{2k}{3}\dfrac{\Delta}{2q_1}\dfrac{\lambda_p}{\lambda}\lambda_p\\ \rho qu_s, & a>\dfrac{2k}{3}\dfrac{\Delta}{2q_1}\dfrac{\lambda_p}{\lambda}\lambda_p\end{cases}. \quad (4)$$

For metal particles we must replace ρqu_s in this formula by I_{exp}^M, and the right-hand side of the inequalities must be replaced[2)] by $K_a^M = 3(\omega/2\pi\sigma)^{1/2}$.

2. The above breakdown initiation by thermal explosion of macroparticles can be successfully used to explain both qualitatively and quantitatively many experimental data on the optical breakdown of gases, which could not be explained before.

The first phenomenon to explain is why experiments on breakdown in the visible and at λ = 1.06 μ have not shown the presence of any appreciable influence of macroimpurities under "natural" conditions (i.e., without special introduction of highly absorbing particles into the focal region), whilst experiments at λ = 10.6 μ have clearly shown the presence of this effect in a substantial measure.[4-6] Thus, when the CO_2 laser radiation is focused down to a spot of diameter d ≈ 0.1 cm in an ordinary laboratory, air breakdown occurs for $I \approx 2 \times 10^9$ W/cm².[4-6] On the other hand, when dry nitrogen is blown through the focal region, breakdown does not appear even at intensities in excess of 10^{10} W/cm².[4] Finally, when particles with diameter $2a \approx 50$ μ are introduced into the focal region, the threshold falls to about

10^8 W/cm^2, independently of the optical properties of the particles.[6]

The second question is why experiments at $\lambda = 10.6\ \mu$ have shown "diffusion-like electron losses" when the volume of the focal region V is known to exclude the influence of electron diffusion on the breakdown process.[4,5] It is also necessary to explain the result $I_{th} \propto 1/d$ obtained in[4,5] (d is the diameter of the focal spot; more precisely, I_{th} is roughly inversely proportional to the focal length of the lens F for a constant divergence φ of the laser beam or, conversely, to the angle φ for constant F).[3)]

The final question is why the illumination of the focal region of the periodically pulsed CO_2 laser radiation by a sufficiently strong beam of continuous CO_2 laser radiation may lead to a substantial increase in the threshold for the pulse breakdown of gases.[7]

Under real conditions, macroparticles suspended in the gas may differ in their optical and thermophysical properties. Moreover, in general, they have a spectrum of values of a. Therefore, the above gas-breakdown mechanism is essentially statistical, and we may therefore use it to calculate only certain average parameters for the breakdown problem, for example, the mean threshold intensity $\langle I_{th}\rangle$, the relative spread in the threshold intensity $\delta_{I_{th}} = (\langle I_{th}^2\rangle - \langle I_{th}\rangle^2)^{1/2}/\langle I_{th}\rangle$, and so on. We shall consider this problem in the next section.

3. We start by assuming that all the suspended particles have the same optical and thermophysical properties, and differ only in their radii a. Let n be the mean density of macroparticles, independently of their size, and let f(a) be the probability distribution density for the radii a. It will be assumed that the particle distribution inside the focal region remains unaltered during the duration τ of the pulse of radiation.

The probability that breakdown through the above mechanism may occur (independently of the breakdown radiation intensity) is clearly equal to the probability w_1 that at least one macroparticle is present in the focal volume V. It is also assumed that $V \gg a^3$. According to the Poisson distribution, we then have $w_1 = 1 - e^{-Vn}$. On the other hand, the probability that the resulting breakdown occurs for radiation intensity greater than I is

$$P(I)=w_1^{-1}\left\{\left[1-\exp\left(-Vn\int_0^a f\,da'\right)\right]\exp\left(-Vn\int_a^\infty f\,da'\right)\right\}, \tag{5}$$

where a is determined from the equation $I = I_{th}(a)$. The curly brackets give the probability that at least one particle with radius less than a is present in the volume V, but this volume contains no particles with radius greater than a.

According to Eq. (5), the probability density for the breakdown intensity is given by

$$F(I)=-\frac{dP}{dI}=\frac{w_1^{-1}Vnf(a)}{|dI_{th}/da|}\exp\left(-Vn\int_a^\infty f\,da'\right), \tag{6}$$

and the moment of the threshold intensity of order r = 1, 2, 3, ... is given by

$$\begin{aligned}\langle I_{th}^r\rangle &= \int_0^\infty I^r F(I)\,dI=w_1^{-1}Vn\int_0^\infty f(a)I_{th}^r(a)\exp\left(-Vn\int_a^\infty f\,da'\right)da\\ &=w_1^{-1}\int_0^{Vn} e^{-x}I_{th}[a(x)]\,dx,\end{aligned} \tag{7}$$

where a(x) is determined from

$$x=Vn\int_{a(x)}^\infty f(a)\,da. \tag{8}$$

It is clear from Eqs. (7) and (8) that when $Vn > 2$ (there are more than two particles, on the average, in the focal volume), the main contribution of $\langle I_{th}^r\rangle$ is exclusively due to particles whose radii a are greater than the median radius a_m.[4)] As Vn increases, increasingly larger particles become important. On the other hand, when $Vn \ll 1$, all particles in the distribution f(a) are important. According to Eq. (7), we then have

$$\langle I_{th}^r\rangle=\int_0^\infty I_{th}^r(a)f(a)\,da. \tag{9}$$

However, when $Vn \ll 1$, the probability of finding at least one particle in the focal region is $w_1 \approx Vn \ll 1$ and, therefore, breakdown due to the above mechanics is then found to have a sporadic character and can, on the average, appear only once in $1/Vn$ laser pulses. If we ignore such sporadic breakdowns and consider $Vn > 2$, we have $w_1 > 0.86$.

We note that although sporadic breakdowns are rare, fluctuations in their threshold intensity may, under certain definite conditions, turn out to be small, i.e., $\delta_{I_{th}} \ll 1$. This occurs, for example, in the case of a "sharp" distribution f(a), when

$$\delta_a=\langle\Delta a^2\rangle^{1/2}/a_1\ll1,$$

$$a_1=\int_0^\infty af(a)\,da,\quad \langle\Delta a^2\rangle=\int_0^\infty(a^2-a_1^2)f(a)\,da,$$

where a_1 is the mean radius and $\langle\Delta a^2\rangle$ is the spread in radii. According to Eq. (9), we then have

$$\langle I_{th}\rangle\approx I_{th}(a_1),\quad \delta_{I_{th}}\approx a_1|I'/I|_{a=a_1},\ \delta_a\ll1,$$

independently of the mean optical thickness of the particles $a_1\mu$. In the case of a "broad" distribution f(a), when $\delta_a \sim 1$, fluctuations in the threshold intensity for sporadic breakdown are small only for highly absorbing particles, when $a_1\mu \gg 1$; when $a_1\mu \ll 1$ we have $\delta_{I_{th}} \sim 1$, i.e., breakdown is not only rare but occurs with a large spread of threshold intensities.

Since $I_{th}(a)$ decreases monotonically with increasing a, and tends to a constant ρqu (or I_{exp}^M), it follows from Eq. (7) that, as Vn increases, the mean intensity $\langle I_{th}\rangle$ should also show a monotonic fall, tending to ρqu (or I_{exp}^M); the relative spread $\delta_{I_{th}}$ must then obviously tend to zero. In the experiment, this dependence of $\langle I_{th}\rangle$ on Vn (for $Vn > 2$) is probably seen as a diffusion-like electron loss. We shall consider this in greater detail below.

It is known (see[2]) that for aerosol particles with radii $a > a_m$ the distribution f(a) can be approximated by

$$f(a)=\frac{\beta-1}{2a_m}\left(\frac{a_m}{a}\right)^\beta,\quad a\geqslant a_m, \tag{10}$$

where β lies between 2 and 5, depending on the type of aerosol; in most cases, $\beta \approx 4$. The above formula can probably be used to approximate not only the distribution of large-particle aerosol but, in general, the distribution of any large particles "naturally" suspended in other experimental gases.

Substituting Eq. (10) in Eq. (7) with $Vn > 2$, we have

$$\langle I_{th}^r\rangle\approx\int_0^\infty e^{-x}I_{th}^r\left[a_m\left(\frac{Vn}{2x}\right)^{1/(\beta-1)}\right]dx. \tag{11}$$

Let us now use the foregoing analysis (see end of Sec. 1) for a broad class of particles and distributions (10) characterized by the following conditions: a) $\mu\lambda_p \ll 1$, $a_m \gtrsim (\mu\lambda_p)\lambda_p$ or b) $\mu\lambda_p \gtrsim 1$, $\sqrt{\lambda_p/\lambda} \ll 1$, $a_m \gtrsim \lambda_p$ ($\lambda_p \approx 10^{-5}$ cm). The function $I_{th}(a)$ in the integrand of Eq. (11) can now be replaced by $I_{exp}(a)$ defined by Eqs. (1) and (3). The quantities $\langle I_{th}\rangle$ and $\delta_{I_{th}}$ are then described by the following expressions, depending on the "mean effective thickness" of the focusing region:

$$z_0 = {}^1/_2 Vn(a_m\mu)^{\beta-1}. \quad (12)$$

When $z_0 \ll 1$,

$$\langle I_{th}\rangle \approx \Gamma\left(\frac{\beta}{\beta-1}\right)\frac{\rho q u_s}{z_0^{1/(\beta-1)}} = \Gamma\left(\frac{\beta}{\beta-1}\right)\frac{\rho q u_s}{a_m\mu}\left(\frac{2}{Vn}\right)^{1/(\beta-1)}, \quad (13)$$

$$\delta^2_{I_{th}} \approx \Gamma\left(\frac{\beta+1}{\beta-1}\right)\Big/\Gamma\left(\frac{\beta}{\beta-1}\right) - 1 \quad (14)$$

where $\Gamma(x)$ is the gamma function.

When $z_0 \gg 1$, we have

$$\langle I_{th}\rangle \approx \rho q u_s, \quad (13a)$$

and $\delta_{I_{th}}$ is a small quantity of order higher than $1/z_0$.

For metal particles, and when $\sqrt{\lambda_p/\lambda} \ll 1$ and $a_m \gtrsim \sqrt{3}\lambda_p(\omega/2\pi\sigma)^{1/4}$, the mean intensity is $\langle I_{th}\rangle \approx I^M_{exp}$ and the relative spread $\delta_{I_{th}}$ is also vanishingly small.

Consider now the case of "soot" particles and metal particles ($\mu\lambda_p \gtrsim 1$) in the visible and near-infrared, when $\sqrt{\lambda_p/\lambda} \sim 1$. The function $I_{th}(a)$ is then given by Eq. (4) and the mean effective thickness of the focal region is

$$x_0 = \frac{Vn}{2}\left[\frac{2q_1/\Delta}{2k/3}\frac{\lambda}{\lambda_p}\frac{a_m}{\lambda_p}\right]^{\beta-1}. \quad (15)$$

When $a_m > 3u_e^2/k\omega u$ (see Footnote 2) we have from Eq. (11)

$$\langle I_{th}\rangle \approx \rho q u_s\left[1 - e^{-x_0} + x_0^{-1/(\beta-1)}\Gamma\left(\frac{\beta}{\beta-1}; x_0\right)\right] \quad (16)$$

where $\Gamma(\alpha; x)$ is the incomplete gamma function (see, for example,[8]).

When $x_0 \ll 1$, we then have [for small x, $\Gamma(\alpha; x) \approx \Gamma(\alpha)$]:

$$\langle I_{th}\rangle \approx \Gamma\left(\frac{\beta}{\beta-1}\right)\frac{\rho q u_s}{x_0^{1/(\beta-1)}} = \frac{k}{6}\frac{mc\omega u_s\Delta}{e^2 a_m}\left(\frac{2}{Vn}\right)^{1/(\beta-1)}. \quad (17)$$

It is readily verified that under these conditions the relative spread $\Delta_{I_{th}}$ is given by Eq. (14).

When $x_0 \gg 1$, we find from Eq. (16) that the mean intensity $\langle I_{th}\rangle$ is given by Eq. (13a), and the relative spread $\delta_{I_{th}}$ is again vanishingly small.

Equations (15)—(16) refer to soot particles. For metal particles, the quantity ρqu in these expressions must be replaced by

$$I^M_{exp} = {}^1/_3\rho q u_s(2\pi\sigma/\omega)^{1/2},$$

and x_0 by

$$x_0^M = x_0[3(\omega/2\pi\sigma)^{1/2}]^{\beta-1}.$$

When $x_0^M \ll 1$, we have Eq. (17); when $x_0^M \gg 1$, we have $\langle I_{th}\rangle = I^M_{exp}$.

Equations (11)—(17) enable us to draw the following general conclusion. For high particle densities n, or large focal regions V, when $Vn > 2$ and the thermal explosion of the particles is of a regular character, the mean threshold intensity for breakdown, $\langle I_{th}\rangle$, and the fluctuations in it, $\delta_{I_{th}}$, are essentially determined by the mean effective thickness of the focal region. For long-wave radiation, when $\sqrt{\lambda_p/\lambda} \ll 1$, this quantity is equal to z_0 (independently of the absorptive properties of the particles themselves). For the visible and near-infrared, when $\sqrt{\lambda_p/\lambda} \sim 1$, it is, as before, equal to z_0 for weakly absorbing particles for which $\mu\lambda_p \ll 1$, whilst for soot-type particles and metal particles for which $\mu\lambda_p \gtrsim 1$, it is equal to x_0. If the mean effective thickness of the focal region is small in comparison with unity, then $\langle I_{th}\rangle \sim (1/Vn)^{1/(\beta-1)}$. In other words, the gas breakdown initiated by the thermal explosion mechanism is, in this case, subject to diffusion-like electron losses. The threshold breakdown intensity is then subject to fluctuations, the relative spread of which is given by Eq. (14); when $\beta \approx 4$ we have $\delta_{I_{th}} \approx 0.37$.

If, on the other hand, the mean effective thickness is large in comparison with unity, the threshold breakdown intensity for the thermal explosion mechanism assumes its minimum value which is independent of the volume V of the focal region (and of the density n) and is equal to ρqu [for metals it is equal to $1/3\,\rho q u_s(2\pi\sigma/\omega)^{1/2}$]. Fluctuations in the threshold intensity are then practically absent.

So far we have assumed that there is only one type of particle with uniform optical and thermophysical properties in the focal region. We shall now suppose that there is a variety of particles. Since the contribution of a particular type of particle to the initiation of breakdown in the gas is independent of the contribution of the other kinds of particle, we can, in fact, use the above results in the analysis of this more complicated case. Firstly, to eliminate sporadic breakdowns from our analysis we must also exclude all the minor types of particle for which $Vn \ll 1$. Secondly, under "natural conditions," the most probable situation is possibly that in which several types of weakly absorbed particle are represented (for which $Vn > 2$ and $\mu\lambda_p \ll 1$). In this case, the breakdown initiation threshold is clearly determined by the particular type of particle for which the mean effective thickness is a maximum.

Particles such as soot and metal particles ($\mu\lambda_p \gtrsim 1$) are not well represented under "natural conditions" ($Vn \ll 1$). However, when the gas is artificially contaminated by them, and $Vn > 2$, their contribution to breakdown initiation is most frequently the predominant one. In the case of long-wave radiation ($\sqrt{\lambda_p/\lambda} \ll 1$), this is always so because for soot particles we have $z_0 > 1$ from Eq. (12) (for $a_m > \lambda_p$) and, consequently, the mean intensity $\langle I_{th}\rangle$ is close to its minimum value $\rho q u_s$. For metal particles, in this case, the mean intensity is always a minimum and equal to I^M_{exp}. In the case of the visible and near-infrared, on the other hand ($\sqrt{\lambda_p/\lambda} \sim 1$), the mean effective thickness x_0 (or x_0^M) is not, in general, necessarily greater than unity [if Vn is not too large; see Eq. (15)]. However, if, for example, $a_m \gg \lambda_p$ [or $a_m \gg \lambda_p 3(\omega/2\pi\sigma)^{1/2}$], then $x_0 \gtrsim 1$ (or $x_0^M \gtrsim 1$) and, consequently, the mean intensity $\langle I_{th}\rangle$ is again close to its minimum value.

4. Let us now consider the problems formulated in Sec. 2. It is immediately clear from Eqs. (12)—(13) why the effect of macroimpurities on gas breakdown under "natural conditions" has not been detected in the visible

and at $\lambda = 1.06\ \mu$. The fact is that, in this range, the absorption coefficients of all the possible impurities (apart from soot and metal particles) which, under these conditions, are not strongly represented ($Vn \ll 1$) are relatively low ($\mu \lesssim 10\ cm^{-1}$). The median radius a_m of "natural" particles will probably not exceed 3×10^{-3} (see[2]) so that $a_m\mu \lesssim 3 \times 10^{-4}$ and, therefore, we have from Eq. (13)

$$\langle I_{th} \rangle \gtrsim (3\div10) \cdot 10^{11} (2/Vn)^{1/(\beta-1)}\ [W/cm^2].$$

Under typical conditions ($Vn \sim 3-10$ and $\beta \approx 4$) we have $\langle I_{th} \rangle \gtrsim (3-10) \times 10^{11}\ W/cm^2$, which exceeds the threshold intensity for breakdown in a "pure" gas. This, in turn, means that the breakdown initiation mechanism associated with the thermal explosion of the particles is ineffective. However, the impurity particles can act as sources of electrons for the avalanche in the "pure" gas. At $\lambda = 10.6\ \mu$ the situation is radically altered because most of the impurities exhibit much greater absorption at this wavelength. Typical values lie between 100 and 1000 cm^{-1} (for example, for water $\mu = 800\ cm^{-1}$; see[2]) and, therefore, under "natural conditions" the typical maximum value is $a_m\mu \sim 0.03$. Although the mean effective thickness of the focal region is then still small ($z_0 \ll 1$), the mean breakdown intensity is, according to Eq. (13), still small

$$\langle I_{th} \rangle \sim (3\div10) \cdot 10^{9} (2/Vn)^{1/(\beta-1)}\ [W/cm^2].$$

In experiments with CO_2 lasers, typical volumes V are greater by one or two orders of magnitude than the corresponding volumes in the visible region. When $Vn \sim 30$ and $\beta \approx 4$ we have $\langle I_{th} \rangle \sim (1-3) \times 10^9\ W/cm^2$, which is in agreement, for example, with experimental data on breakdown in laboratory air.[4,6] The function $\langle I_{th} \rangle \sim (1/Vn)^{1/(\beta-1)}$ explains the diffusion-like electron losses reported in[4,5]. Since $V \sim F^4\varphi^3$, we have $\langle I_{th} \rangle \sim 1/F^{4/3}\varphi$ for $\beta = 4$, which is in good agreement with experimental results.[4,5]

The artificial introduction of particles with diameter $2a \approx 50\ \mu$ into the focal region of the CO_2 laser, as was done in[6], with density $n \gg 1/V$, clearly corresponds to the realization of the case of large mean effective thickness of the focal region ($z_0 \gg 1$). Under these conditions, we have from Eq. (13a) $\langle I_{th} \rangle \approx \rho q u_s \approx (1-3) \times 10^8\ W/cm^2$, independently of the optical properties of the extraneously introduced particles. This explains the results obtained in[6].

Finally, the results of the experiments reported in[7] can also be explained in a qualitative fashion. When the focal region of the periodically pulsed CO_2 laser is "illuminated" by continuous radiation with $\lambda = 10.6\ \mu$, there is a reduction in the maximum value of $a_m\mu$ (due to the heating of the particles and partial evaporation from the surface between successive pulses), and this leads to an increase in $\langle I_{th} \rangle$.

The fact that the above mechanism for breakdown initiation in the visible and near-infrared by thermal explosion of macroparticles under "natural conditions" is ineffective does not mean that the mechanism will not operate in this wavelength range. A substantial reduction in the breakdown threshold in this range is observed, for example, in experiments with millisecond laser pulses if a target consisting of a highly absorbing material is placed at a certain distance beyond the focal point of the lens.[5)] In this case, the "main" part of the pulse produces ablation from the target surface. As a result, macroparticles of the target material enter the focal region and initiate gas breakdown (usually by individual laser pulse spikes). In the case of nanosecond laser pulses, this effect on target-gas breakdown beyond the focus is usually absent because the particles from the target do not succeed in reaching the focal region during the pulses.

We have, however, performed an experiment on the initiation of breakdown in air at $\lambda = 1.06\ \mu$ with monopulse length $\tau \sim 100$ nsec by artificially introducing highly absorbing particles into the focal region, namely, soot-type particles produced by burning rubber (black smoke). The laser beam had a divergence $\varphi \sim 10^{-3}$ rad and a diameter $D_0 \approx 1.5$ cm. We used a lens with a focal length of about 10 cm. Under these conditions $V \approx 2 \times 10^{-6}\ cm^3$. The smoke particle density apparently satisfied the condition $Vn \gg 1$. In "pure" air the breakdown occurred when the energy per pulse was $W \approx 0.55$ J; in the presence of smoke, this energy fell to less than 0.05 J.

The same giant-pulse laser system was used to observe a substantial reduction in the threshold for breakdown in air with a practically regular repeatability when the radiation was focused on mist produced by condensing a jet of water vapor issuing into the atmosphere from a tube attached to a retort containing boiling water. This reduction in the breakdown threshold could not be associated with the change in the composition of the gas (high partial pressure of water vapor) in the cloud of mist because the ionization potentials of the H_2O and N_2 molecules was quite close to one another.

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1)The concept of the "thermal explosion" of a macroparticle in the radiation field is the same as the generally accepted concept of a thermal explosion due to exothermic combustion reactions in the sense that a stationary thermal state cannot be established.

2)When the first line in Eq. (4) was derived, it was assumed that $a > 3u_e^2/k\omega u_s$, i. e., that the unity in Eq. (2) could be neglected (diffusion losses need not be taken into account). For $\lambda = 1.06\ \mu$ and $k \approx 50$, this gives $a > 3 \times 10^{-6}$ cm.

3)We recall that, in the case of the mechanism involving electron losses by free diffusion, $I_{th} \sim 1/d^2 \sim 1/F^2\varphi^2$.

4)By definition of a_m, we have

$$\int_{a_m}^{\infty} f(a)\,da = 1/2.$$

5)In practice, this target is frequently black photographic paper; under these conditions the threshold for breakdown in air may fall by a factor of 1.5.

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