DEPENDENCE OF OPTICAL BREAKDOWN THRESHOLD ON RADIUS OF AEROSOL PARTICLES

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Approximate functions describing all known experimental data on optical breakdown threshold (OBT) for different aerosol particle radii are determined. It is shown that the OBT is determined by the OBT for non- ionized air near the particle focal spot. The OBT for large corundum particles ceased to depend on particle radius, and equals the intensity at which the focal plasma is developed and maintained.

The optical breakdown threshold of may advantageously be characterized by the breakdown intensity of radiation I_b for short ($\leq 10^{-11}$ sec) laser pulses, when an optical breakdown plasma is produced by multiphoton (and tunneling) ionization of air, as well as for very long (≥ 10 μsec) pulses, when the breakdown energy is determined by energy losses of plasma due, for example, to diffusion of electrons and radiation out of the plasma. At the same time, in the intermediate range of pulse durations the use of breakdown radiation energy density q_b may be more suitable. If the energy losses occurring during breakdown are negligible, then q_b will not depend on the duration τ or shape $f(t)$ of the laser pulse. Therefore, if q_b varies with $\tau$and $f(t)$, one can surmise that there is some influence due to prebreakdown stages and/or significant energy losses that occur during the breakdown process. Such behavior of q_b, if observed, would enable one to determine the mechanism of plasma formation more accurately. For this reason, from here on, the breakdown threshold will be given in J/cm^2.

The behavior of q_b as a function of particle radius determines the dynamics[1] and the probability of occurrence of plasmoids due to optical breakdown in aerosols[2]. In this paper, we derive equations that enable one to describe all known experimental of $q_b(a)$. We have also resolved contradictions between experimental data from Ref. 3 and 4 for $q_b(a)$ at a laser wavelength $\lambda = 10.6$ μm for the coarse aerosol component. In the case of submicron aerosol particles, for which there are no direct measurements of $q_b(a)$ available, a technique is proposed, which allows for its restoration using the experimentally measured dependence of laser radiation intensity I_{fw} at the focus of a lens on the lens' focal length, given the probability of plasmoid appearance due to optical breakdown. This technique has been used to reconstruct $q_b(a)$ for the fine-scale aerosol component from the data presented in Ref. 5, 6 at laser wavelengths $\lambda = 10.6$ and $\lambda = 1.06$ μm.

OPTICAL BREAKDOWN THRESHOLDS FOR WATER DROPLETS, IN VISIBLE AND INFRARED WAVELENGTH

The experimental data on the threshold of optical breakdown in water droplets at ruby[7] and neodymium[8] laser wavelengths are presented in Fig. 1.

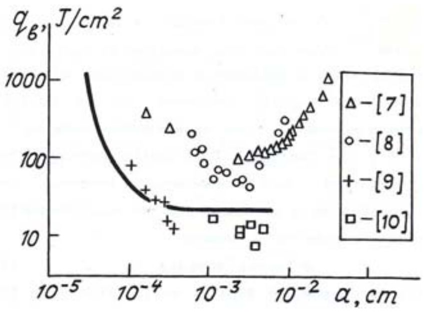

Fig. 1. Optical breakdown threshold as a function of corundum particle radius (l = 1,06 mm, t = 80 nsec [9] and 10 nsec [10]) and water droplets radius (l = 0.69 mm. t = 50 nsec [7] and λ = 1.06. t = 40 nsec [8]). The solid curve represents calculations made using Eq. (2).

A computer program has been developed to approximate these data, in which the optimal approximation coefficients are so chosen as to minimize the symmetrized rms deviation between the experimental $q_b(a_i)$and approximating $q_b(a_i)$ sequences.

$$\delta_1 = (N-1)^{-1}\sum_{i=1}^{N}\left\{\left[q_a(a_i)-q_b(a_i)\right]^2 q_b^{-2}(a_i)+\left[q_a(a_i)-q_b(a_i)\right]^2 q_a^{-2}(a_i)\right\},$$

as well as to the maximum symmetrized deviation between $q_a(a_i)$ and $q_b(a_i)$,

$$\delta_c = \frac{1}{2}\sup_{i=1,N}\left|\left[q_b^2(a_i)-q_a^2(a_i)\right]\left[q_a(a_i)q_b(a_i)\right]^{-1}\right|.$$

Such a modification of the approximation criteria enables one to significantly increase the efficiency of the minimization procedure compared to that of standard methods. In standard approximation criteria, the range of arguments in which the approximated function reaches its maximum values exercises a decisive influence. In addition, positive and negative deviations of the approximating function from that to be approximated have different effects on the approximation quality. The symmetrized deviations mentioned above eliminate these shortcomings. The original optical breakdown threshold approximation table in energy density units was obtained by multiplying the data on $I_b(a)$ from Ref. 7,8 by the corresponding laser pulse durations.! Of course, this procedure introduced some additional error's in the data. The instant of breakdown was recorded [7,8] from the time behavior of laser light intensity when a noticeable attenuation of light occurred due to the emergence of an optical breakdown plasmoid. Final experimental results [7,8] show only the functions $I_b(a)$,while corresponding durations of the breakdown process are omitted).The data obtained can be satisfactorily described by the relationship

$$q_b(a) = ca^{-s} \exp(ab) \tag{1}$$

over a wide range of droplet radii (from 6 to 120 μm at $\lambda = 1.06$ μm and from 1,5 to 250 μm at $\lambda = 0.69$ μm).Corresponding approximation coefficients for (1) are as follows $c = 5.98 \times 10^{-2}$ J/cm, $s = 1$, $b = 423$ cm^{-1} for $\lambda = 1.06$ μm and $c = 3.9$ J/cm$^{3/2}$, $s = 0.5$, $b = 145$ cm^{-1} for $\lambda = 0,69$ μm.

OPTICAL BREAKDOWN IN CORUNDUM PARTICLES AT λ = 1,06 μm

Experimental data on $q(a)$ obtained[9,10] by initiating optical breakdown in corundum particles with radiation at $\lambda = 1,06$ μm are presented in Fig. 2.

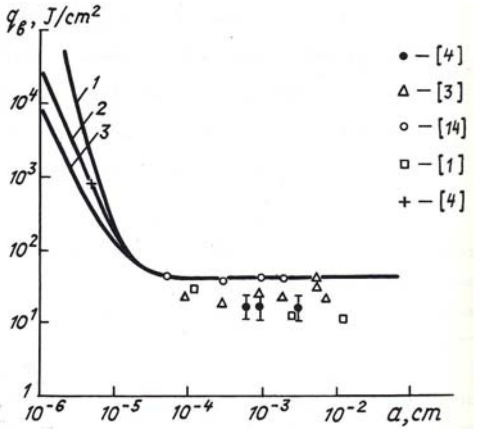

Fig. 2. Optical breakdown threshold initiated by TEA CO_2 laser on particles of different substances (corundum, germanium, magnesium oxide, glass, aluminum, ferric oxide, soot, salt) as a function of their radii. Curves 1 to 3 are calculated using (4).

Of all the data obtained from Ref. 10, only those derived from the lowest mean energy pulse trains are shown in Fig. 2, in order to minimize errors due to poorly determined optical breakdown times obtained by converting from I_b to q_b by multiplying by pulse half-width. It is interesting that the data from Ref. 10 seem to continue or supplement the $q_b(a)$ curve obtained in Ref. 10 for large particle radii. At the same time, both sets of experimental data can be well approximated by the expression

$$q_b(a) = 20 + (3 \times 10^{-4}\ a^{-1}\ [\text{cm}])^3 \tag{2}$$

Calculations using this formula are given in Fig. 2 by a solid curve. The coefficients in expression (2) were obtained from experimental measurements[11] of I_{fw} made in laboratory air under breakdown conditions; see sect. 4. The sharp decrease in $q_b(a)$ in at small radii can be explained by strengthening of optical field at the main maximum, which is located at the pole of the shadow hemisphere[12]. The intensity of the optical field at the main maximum of a weakly absorbing particle with $a\lambda^{-1} > 1$ exceeds the intensity of radiation undisturbed by the particle by a factor of B,

$$B_m \approx 20(n-1)^{2}\rho \tag{3}$$

where n is the refractive index of the particle, and $\rho = 2\pi a\lambda^{-1}$ is the Mie parameter. Taking account of Eq. (3), one finds that near the breakdown threshold for particles 1 to 4 μm in size, the of optical field intensity at the pole of the shadow hemisphere is practically unchanged, and is about 3 × 10 W/cm^2. This intensity level provides for efficient outflow of bare electrons from particles due to multiphoton emission (work function for corundum is 3.9 eV) and at the same time this intensity is high enough to break down the non-ionized air in about 10^{-7} sec. In spite of the increase in B_m with increasing a (see Eq. (3)), $q_b(a)$ almost stops decreasing at large a (see Fig. 1). This is probably due to the fact that injection of bare electrons into the focus region does not terminate the breakdown process. At large particle sizes A, the energy density of radiation undisturbed by a particle should be high enough to sustain and develop a bare plasmoid generated in a narrow focus zone behind the particle. The decrease in q_b observed in Ref. 10 with increasing laser pulse slope is obviously due to diffusion loss of electrons from the focus zone bare plasmoid production.

OPTICAL BREAKDOWN THRESHOLD IN THE COARSE COMPONENT OF AEROSOL FOR RADIATION AT λ = 10,6 mm

Paper[4] reported for the first time a $q_b(a)$ independent of particles composition (for aluminum oxide, glass, germanium, soot, salt), for radiation at $\lambda = 10.6$ μm and pulse duration of 10^{-6} to 10^{-7} sec. Investigations carried out in Ref. 4 with individual particles of these materials had demonstrated an optical breakdown threshold not only of independent of composition, but also of particle radius, being about 12 J/cm^2 for a pulse of 0.2 μsec duration. It was also found in Ref. 4 that the threshold of optical breakdown initiated by glass fibers with diameters 6, 10, 30 and 100 μm placed in a focused beam does not depend on the fibers diameter being about 20 J/cm^2. For small particles ($a < 10$ μm), a marked increase in q_b

with increasing a was also observed. In Ref. 4 it was also shown that the optical breakdown threshold is independent of particle composition (at least for the same selection of substances as in Ref. 4 and radius (in the range 0,5 to 50 μm) being about 20 to 40 J/cm^2 for single pulses of 0,2 μsec duration.

We now consider possible explanations for the marked discrepancy between the measurements[3,4] at small a values. In order to measure $q_b(a)$. Smith positioned a particle of known size and composition at into a pre-selected point of a laser beam spot, irradiated it with a laser beam of known energy, and recorded whether a plasmoid was produced or not. By varying the radiation intensity Smith managed to determine the intensity level at the particle at which the probability W of plasmoid production due to optical breakdown was 50 per cent, and this intensity he took to be the value of the breakdown threshold. The reliability of this method is obviously high. The technique used by Lencioni[4] to determine $q_b(a)$ required a complicated procedure for data interpretation. In his work[4], Lencioni focused laser radiation into an aerosol chamber containing a polydisperse aerosol. He then varied the mean energy of laser pulses until W achieved a value of 50 per cent. Lencioni assumed that at $W = 0.5$ the optical breakdown threshold was equal to one half the radiation intensity at the focal point. It was then necessary to relate this threshold to the radius of the particles most likely to initiate optical breakdown. In doing so Lencioni assumed that the threshold falls off with increasing a. This enabled him to relate the focal volume V_c and the number density of particles $n(a)$ whose radii exceed the value a to the breakdown probability $W = 1 - \exp[-V_c(n(a)]$. Lencioni estimated the value of V_c using the homogeneous cylinder model, according to which only the region bounded by the ray caustic must be taken into account in a sharply focused beam. But as shown in Ref. 2, this model results in substantial systematic errors in estimates of the focal volume; as a rule, the error can reach several orders of magnitude. Thus, the discrepancy between the results of Lencioni and Smith are accounted for by this error, as well as by the incorrect assumption of significant variation of $q_b(a)$ in the range of a values, where direct measurements[3] showed no such changes.

Data obtained in Ref. 14 also confirmed that the optical breakdown threshold is independent of particle material (quartz, Fe_2O_3, aluminum oxide, soot, magnesium oxide) and particles size (at least in the size range from 0.5 to 20 μm). The results[14] imply that the breakdown threshold in aerosol for a CO_2 laser pulse changes by factor of three at most (the error involved in comparing absolute values of experimentally measured values obtained in different experiments) when the number density of aerosol particles drops by about 6 orders of magnitude(from 10^5 to 0.1 cm^{-3}). Figure 2 presents the results of experimental studies of $q_b(a)$, excluding, those obtained using incorrect estimates of the focal volume. The duration τ of pulses used in measurements of $q_b(a)$ was 0.2 μsec and 0.2 to 4 μsec[4]. Data presented in Fig. 2 were obtained in Ref. 3 using individual glass fibers with diameters from 6 to 100 μm, and aerosol particles ($a > 10$ μm) of Al_2O_3, SiO_2, C and Ge; particles were placed at the focus separately, one by one. The cross in Fig. 2 denotes the optical breakdown threshold in laboratory air cleared of particles of size $2a > 0.1$ μm[4]. In paper[14] optical breakdown threshold were determined by the cutoff of transmission by the optical breakdown plasmoid, from the length of the threshold isophote in the case of aerosols having a narrow spectrum of particle sizes, and from the probability of optical breakdown in aerosols calculated using the correct formula for the focal volume of a Gaussian beam[2]. We see from Fig. 2 that the optical breakdown threshold in aerosol particles with a between 0.5 and 100 μm is independent of a and of particle composition. A comparison shows that the results of different authors are in good agreement.

OPTICAL BREAKDOWN IN PARTICLES OFFINE AEROSOL COMPONENT

Thus far, there are no direct data on $q_b(a)$ for in the literature the finest component of aerosols ($a < 0.5$ μm). At the same time, particles in this size range occur most frequently in air. Measurements of the optical breakdown threshold in the coarse aerosol component[14] have shown that the uncontrolled fine component of laboratory air pollution has a much more higher breakdown threshold than particles with $a > 1$ μm. The simplest formula for $q_b(a)$ allowing significant growth of q_b for small a values and independence of a at large a is

$$q_b(a) = c+(a_*a^{-1})^s, \tag{4}$$

where $c \approx 40$ J/cm^2. The coefficients a_* and s were found from experimental data[6] on the relation between intensity of focused radiation and the conditions of focusing, the probability of optical breakdown W being taken to be 50 per cent. The coefficients a_* and s in Ref. 4 have been determined by minimizing the rms deviation between the calculated average number of particles $N_e = \int v(a)f(a)da$ in the volume V_a and the experimentally measured $N_e = \ln(1 - W)^{-1}$. The calculations used the size distribution function $f(a) = 10^{-15}(2a)^{-5}$ in the size a range $10^{-15} \le a \le 10^{-4}$ cm, and the function $f(a) = 10^{-7}\ (2a)^{-3}$ for $a > 10^{-4}$ cm[5,6]. A narrow enough range of a and s values was identified in this way to enable one to describe the experimental data of [6] satisfactorily. Moreover, within this range ($0.4 \le a_* \le 0.8$ μm, $2.7 \le s \le 3.6$), the optimum values of a_* and s are related by $a_* = (4,88 - s)\ .\ 3.12\ .\ 10^{-5}$. Numerical data on $q_b(a)$ obtained for $s = 3.6$ (curve 1), $s = 2.4$ (curve 2), and $s = 2.0$ (curve 3) (for $c = 40$ J/cm^2 are presented in Fig. 2. It is interesting to note that the experimental value of the breakdown threshold for air filtered through a 0.1 μm mesh[4] is the same as that given by curve 2 of Fig. 2. Thus, one can use the following equation to describe the of optical breakdown threshold in air containing solid particle:

$$q_b(a)[\mathrm{J/cm^2}] = 40 + (7.65 \times 10^{-5} a^{-1}\ [\mathrm{cm}])^{2,4}.$$

In the range of a values where q_b is a strong function of a, the work function typically (due to Coulomb term e^2za^{-1}) increases after the first few dozen (or even fewer) electrons are emitted (for $a < 10^{-6}$ cm). This enables one to relate the growth of $q_b(a)$ at small a values to the fact that the emission of electrons from small particles is inhibited. It is also anticipated that at $a \le 0{,}1$ μm, $q_b(a)$ will become dependent on the particle composition. The functions $q_b(a)$ at $\lambda = 1.06$ μm, as reconstructed from the data[5–11], show that the boundary of weak $q_b(a)$ dependence has been shifted into the large-particle range ($a_* \approx 2.5$ to 4,5 μm) while the variation of s and c are insignificant ($s \approx 2.5$ to 3, $c \approx 15$ to 26 J/cm^2).

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