Optical discharges

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Gas breakdown, steady-state maintenance and the continuous generation of a low-temperature plasma, and propagation of the plasma fronts, all are induced by laser radiation. By nature, and in conformity with the fundamental laws, these effects are not different from similar processes that occur in constant and alternating fields and which are a traditional subject of the study of the physics of gas discharge. In fact, a new chapter has been added to the gas-discharge sciences: discharge at optical frequencies. It is a rapidly developing new field which encourages both new experiments and applications. It appears useful at this time to characterize the position occupied by the new field within the general framework of discharge sciences, and to analyze and appraise the latest results.

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1. POSITION OF THE OPTICAL DISCHARGES AMONG OTHER DISCHARGE PHENOMENA

(a) Frequency ranges

During the "pre-laser" era and, more precisely, up to the mid-60's, the physics and technology of gas discharge were committed to fields in three basic frequency ranges: (1) constant electric fields with which, depending on the nature of interaction, relatively short-lived pulsed fields and low-frequency oscillating fields are partially conformable, (2) high frequencies (called "radio frequencies" in the foreign literature), a broad range with a mean around 1 MHz, and (3) superhigh frequencies, designated SHF (called "microwaves" in the foreign literature) and to be found in the gigahertz region that corresponds to the centimeter and millimeter waves. Beyond these lies the optical region: infrared, visible and ultraviolet radiation. However, during the pre-laser era—characterized by weak conventional, non-laser, light sources and the fields they produced—the possibility of occurrence of gas-discharge effects in the light fields was beyond everyone's comprehension.

Historically, gas-discharge phenomena were explored in general in the order of ascending frequency ranges. Thus, constant or short-lived fields generated by condenser discharges were investigated first (hence, incidentally, the term "discharge" which applies to processes occurring in the gas portion of a circuit). Toward the end of the last century and the early part of this century, attention had shifted to rf fields. The early 1940's and the development of rocket technology had advanced the range to microwaves. And, finally, the mid-1960's have moved the field into the optical range.

The development of relatively powerful pulsed and cw lasers had enhanced the discovery and investigation of the many new phenomena induced in a gas by laser radiation, and the interaction of the latter with ionized gases and plasmas. Upon closer examination, it becomes evident that among these effects there are specific processes which naturally and fully belong to gas-discharge physics. The laser technology has essentially bequeathed to the discharge physics a fourth, optical range, thus intrinsically endowing this science with a fundamentally new, exceptionally interesting and highly applicable chapter that deals with discharges in optical fields. Conceivably, the new term—optical discharge—sounds alien to many at this time, but, in fact, it conveys as much sense as the time-honored terms "radio frequency" or "microwave" discharges. The new chapter occupies a proper place among the gas-discharge sciences, and it entails the same fundamentals as the chapters on radio-frequency and microwave discharges.

(b) Classification of "discharge processes"

For simplicity, and in order to clarify the position that effects, arising from the interaction of laser radiation with the ionized gases, occupy among the conventional gas-discharge phenomena, it is expedient to classify all gas-discharge phenomena in some meaningful way. Bearing in mind that the interaction of laser radiation with a gas is unaffected, as a rule, by the presence of solid surfaces, the effects must be classified according to criteria which are dissociated from the effects of electrode-, near-electrode- and boundary-intensive processes. We shall distinguish three basic types of spatial gas-discharge processes:

(1) Gas breakdown, development of a turbulent avalanche ionization in it due to an applied external field, and conversion of initially non-ionized gas into a plasma.

(2) Maintenance of an unstable plasma by a field, in which the temperature of electrons responsible for the ionization is sufficiently high, and the gas containing atoms, molecules and ions remains cold. Normally, this corresponds to a weakly-ionized plasma at fairly high pressures, below 100 torr. The degree of ionization is, moreover, much lower than a value for a stable plasma, which corresponds to electron temperature.

(3) Maintenance of a stable plasma by a field, in which the electron and heavy-particle temperatures are close, and the degree of ionization is close to that of a thermodynamically stable plasma. This is a so-

TABLE I.

Field frequency range \ Type of discharge plasma	Breakdown	Maintenance of an unstable plasma	Maintenance of a stable plasma
Constant electric field	In interelectrode gaps	Glow discharge	D.C. arc
Radio frequencies	Radio frequency, electrode or electrodeless	Radio frequency moderate-pressure capacitive discharge	Inductive discharge at atmospheric pressure
Microwave frequencies	In waveguides and resonators	Pulsed discharges in waveguides and resonators	Microwave plasmotrons
Optical frequencies	In gases, induced by a focused laser pulse	Late stages of an optical breakdown	Cw optical discharge, maintained by a CO_2 gas laser radiation

called low-temperature plasma with temperatures of the order of 10,000 K, and at pressures normally of the order of atmospheric.

Each of these processes may occur in any of the foregoing frequency ranges. In fact, nearly all the possible alternatives have been investigated experimentally, and many of these have been found to have occasionally important research and engineering applications. Table I above illustrates the adopted classification, and indicates typical conditions under which one or another process is observed.

(c) Purpose of the paper

Below, we shall consider processes that are induced by laser radiation and belong to the category shown in the bottom line in the table. Having analyzed the salient points, we shall show the gas-discharge nature of these processes and verify that, in principle, they hardly differ from other processes in the same category. We shall also review the current status of investigations and results in this area. The first problem calls for a brief digression into the realm of well-known concepts. The second pertains basically to results obtained after 1972–1973, which were excluded from the author's monograph published in 1974 (Ref. 1).

2. OPTICAL GAS BREAKDOWN

(a) Discovery

The instant of birth of the new chapter of gas discharge physics is etched in the memory of many physicists of the present generation. It is associated with the discovery of a remarkable effect: optical gas breakdown. The first to report this effect were Maker, Terhune and Savage in February 1963.[2]

The discovery of the effect was made possible by the invention of the Q-switched laser, which is capable of producing an especially powerful, so-called giant pulse. When the beam of such a (ruby) laser was focused by a lens, a spark occurred in the focal region, producing a plasma there, as in the case of breakdown in the discharge gap between electrodes (Fig. 1). Very high radiation parameters are required to break down the free air by optical radiation. Air breaks down at the peak power of 30 MW and when a beam is focused to a spot

FIG. 1. Photograph of a laser spark.

10^{-2} cm in diameter (the typical duration of a giant pulse is 30 ns $= 3\times10^{-8}$ s; the energy in such a pulse is 1J). The flux density at the focus for these parameters is 10^5 MW/cm^2, and the electric field intensity in the electromagnetic wave is approximately 6×10^6V/cm.[1)] The breakdown threshold is well defined, and a small decrease in the flux density below a given value will preclude breakdown.

The new effect had evoked such broad interest among physicists that they literally rushed to investigate it. During the next several years, optical breakdown was being studied experimentally and theoretically with such intensity of detail that today our knowledge about it is as extensive as is our understanding of its closest analog, the microwave breakdown, and is certainly superior to our understanding of a more complex process, breakdown of a relatively long gap between electrodes. The bulk of materials dealing with the optical breakdown was generated during the 1960's, as was also the theory of the phenomenon.[1] In recent years, little was added to these data in the way of fundamental knowledge, although some additional experimental numbers have been calculated, refinements of the theory carried out, and allowances for certain subtle and understood details made.

Figure 2[3] shows the threshold fields in an optical wave E_t, which are required to break down several gases by focused radiation from a ruby laser. The threshold values were measured over a broad range of pressures p. By way of comparison, Fig. 3 shows similar data pertaining to breakdown due to microwaves.[4] The overall similarity of the $E_t(p)$ curves should be underscored. As we shall see further, this property has a profound physical meaning.

(b) Avalanche ionization in a field

An electron avalanche develops in a gas under the effect of an electric field associated with an optical wave, as it also does during breakdown in any other field. In the case of breakdown induced by ultrashort pulses from ruby and neodymium-glass lasers, the first, priming electrons appear as a result of a multiphoton emission from atoms, molecules and, possibly, dust which is present in the gas. In this respect,

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1) In a constant field, free air breaks down at the field intensity of 3×10^4 V/cm.

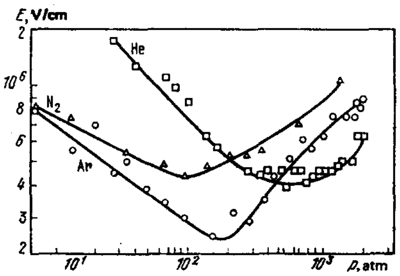

FIG. 2. Thresholds for a ruby-laser-induced breakdown in Ar, He, and N_2. Pulse duration 50 ns, focal spot diameter 10^{-2} cm (Ref. 3).

breakdown in the optical field differs from breakdown in fields of lower frequency, in which the first electrons appear at random (from cosmic rays). Inside the wave field, an electron gradually acquires energy due to collisions with atoms, and it becomes sufficiently energetic to ionize an atom and to produce a new electron. This is the mechanism of electron multiplication.

Avalanche development is determined by an interplay of two opposing processes: energy accumulation by electrons due to the field and energy loss by electrons due to collisions (elastic and inelastic). It is also determined by a loss of electrons due to diffusion or sticking in electronegative gases. Loss of both energy and electrons is relatively independent of the nature of a field, and it occurs in a manner that is more or less the same for all fields. Energy acquisition is the only frequency-dependent process whereby singular features of the optical breakdown, which are associated with the quantum nature of interaction between light and electrons, may be revealed.

In an alternating field, electrons pursue both oscillatory and random motion. According to classical theory, each collision of an electron with a molecule or atom results in a transfer of the mean energy of an oscillating electron $\Delta\varepsilon = e^2E^2/m\omega^2$ into the energy of random motion ε (E is the mean-square electric field and ω is angular frequency). This occurs provided the collisions are relatively infrequent. However, if an electron fails to undergo many oscillations during a period between collisions, i.e., each time oscillations fail to swing "fully," energy transfer from the field to electrons is slowed down. In the general case, the field imparts the following energy per second to an electron

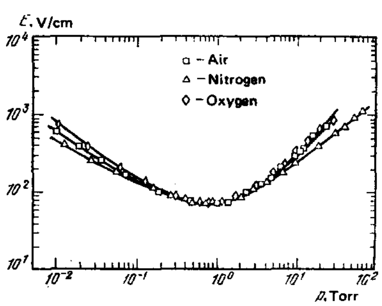

FIG. 3. Breakdown thresholds for N_2, O_2, and air in a microwave field. Frequency 0.994 GHz, diffusion length of discharge volume 1.51 cm (Ref. 4).

$$\left(\frac{d\varepsilon}{dt}\right)_E = \frac{e^2E^2}{m(\omega^2+\nu_m^2)}\nu_m, \qquad (1)$$

where ν_m is the effective frequency of electron collisions with molecules.

Inasmuch as the collision rate is proportional to gas density or pressure, the rate of energy build-up due to the field for each frequency ω at relatively low pressures is proportional to pressure p, and is determined by the ratio E/ω. At relatively high pressures, it is inversely proportional to pressure and independent of ω:

$$\text{at} \quad \nu_m^2 \ll \omega^2 \quad \left(\frac{d\varepsilon}{dt}\right)_E \sim \left(\frac{E}{\omega}\right)^2 p,$$
$$\text{at} \quad \nu_m^2 \gg \omega^2 \quad \left(\frac{d\varepsilon}{dt}\right)_E \sim \frac{E^2}{p}. \qquad (2)$$

(c) Threshold field

In order that an avalanche may develop and breakdown take place, energy losses by electrons and a loss of electrons must be surmounted. In the case of very short field pulses, another requirement is that an appreciable level of ionization must be attained within the pulse width, such that a sufficient number of electron generations is produced. Clearly, an appropriately high rate of energy conversion is required to accomplish this, which is sufficient to provide the required gas ionization frequency ν_i. The latter is the reciprocal of time an electron needs in which to attain energy greater than the ionization potential and to produce ionization. Thus, the breakdown criterion places a specific condition on the parameters $(d\varepsilon/dt)_E$ and $E = E_t$.

Consequently, at low pressures, when $\nu_m^2 \ll \omega^2$, the breakdown threshold field E_t is proportional to frequency and decreases with increasing pressure. Conversely, at high pressures, when $\nu_m^2 \gg \omega^2$, the threshold field, grows with increasing pressure and only weakly depends on the frequency. In alternating fields, the breakdown threshold is minimal at pressures that approximately satisfy the condition $\nu_m = \text{const}\ p \approx \omega$. These considerations explain the behavior of curve $E_t(p)$ in Fig. 3 for a microwave breakdown.[2)]

The behavior of the optical breakdown curve may be explained in the same qualitative way (see Fig. 2). If we proceed from the same equation [Eq. (1)], it becomes evident why breakdown at optical frequencies requires fields considerably stronger than at microwave frequencies ($E_t \sim \omega$, threshold intensity of electromagnetic wave $S_t \sim E_t^2 \sim \omega^2$). It becomes clear why the $E_t(p)$ minimum shifts in the direction of high pressures the order of hundreds of atmospheres (the minimum occurs at $p \sim \omega$). The main issue is to what extent the applicability of Eq. (1) is validated for the quantum case of optical frequencies.

(d) Classics and quanta

The possibility of using a simple and clear formula [Eq. (1)] in the case of optical frequencies was validated

2) Incidentally, the shape of the right-hand side (ascending) branch is, in general, similar to the right-hand side (ascending) branch of the Paschen curve for the breakdown of a gap between electrodes to which a voltage was applied.

in one of the first works dealing with the optical breakdown[5] in which a quantum theory of this effect was formulated. Actually, an electron absorbs energy in quanta, i.e., significant amounts of $\hbar\omega$ equal to 1.78 eV for a ruby laser and 1.17 eV for a neodymium-glass laser. Moreover, the actual energy $\hbar\omega$ acquired by an electron during collision with an atom is much greater than $\Delta\varepsilon = e^2E^2/m\omega^2$, the collisional energy that an electron would have received according to the classical theory. It would seem the latter is totally inapplicable under these conditions.

However, analysis of the quantum kinetic equation for the electron energy distribution function shows that Eq. (1) may be used anyway, even if the actual classical condition $\hbar\omega \ll \Delta\varepsilon$ is not satisfied. This requires a much less stringent condition $\hbar\omega \ll \varepsilon$, where ε is the actual electron energy. In the microwave range, even the trivial requirement $\hbar\omega \ll \Delta\varepsilon$ is satisfied and the question of quantum effects does not generally arise. Conversely, in the optical range, $\Delta\varepsilon \sim 10^{-2}$ eV $\ll \hbar\omega \sim 1$ eV; however, the mean energy of electron spectrum is of the order of the ionization potential, i.e., 10 eV and, therefore, the condition $\hbar\omega \ll \varepsilon$ may be considered satisfied, at least for the frequencies of ruby and neodymium-glass lasers.

Thus, in the case of optical fields, Eq. (1) roughly holds, although it should be treated statistically. Let, for example, $\Delta\varepsilon = 0.01\,\hbar\omega$. An electron, of course, may not receive a hundredth of a photon from the field during collision. This means that, roughly speaking, it gains nothing in the first 99 collisions, and in the hundredth collision it absorbs a full photon all at once. Strict calculations of the electron avalanche and breakdown threshold are normally carried out on the basis of the kinetic equation. Calculations carried out in Ref. 5 and subsequent works (see Ref. 1) are in satisfactory agreement with experimental results.

Alongside the many classical characteristics, certain new details also appear at optical frequencies, which are associated with the quantum nature of interaction between the optical radiation and matter. Thus, for example, ionization of excited atoms is possible by means of two- or three-photon emission of electrons and this sometimes significantly affects the multiplication rate for electrons. However, the avalanche mechanism of the optical breakdown is neither different in principle from a mechanism responsible for microwave breakdown, nor from a spatial breakdown at lower frequencies, including the Townsend (not streamer) gas discharge between electrodes.

(e) A link between microwaves and light

A particularly convincing experimental result in this respect is the fact that the classical laws $E_t \sim \omega$ or $S_t \sim \omega^2$ are satisfied for the threshold values over a broad range of optical frequencies, up to the microwave range. As far as the latter is concerned, the law $E_t \sim \omega$ is theoretically valid only at low pressures that correspond to the left-hand side of the curve $E_t(p)$. However, even the atmospheric pressure in the optical region is "low" in this sense.

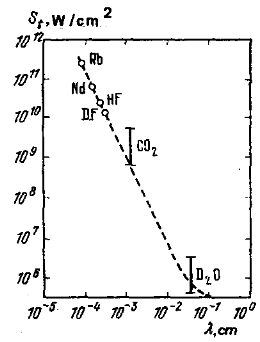

FIG. 4. Thresholds for atmospheric-pressure air breakdown induced by various lasers. Dashed line corresponds to the classical function $S_t \sim (\omega^2 + \nu_m^2)$ which, with the exception of very long-wave region, yields a law $S_t \sim \omega^2$, i.e., a straight line in the logarithmic scale.

To validate the law, we have numerous data for the air breakdown by ruby ($\lambda = 0.694$ μm), neodymium-glass ($\lambda = 1.06$ μm) and CO_2($\lambda = 10.6\,\mu$m) lasers. Quite recently, other results were obtained in the intermediate infrared range by means of HF ($\lambda = 2.7$ μm) and DF ($\lambda = 3.8$ μm) lasers,[6] and a heavy water laser ($\lambda = 385$ μm $= 0.38$ mm) was used to establish a point in the broadest unknown region of the spectrum between the infrared and microwave regions (the submillimeter region).[7]

Threshold intensities are shown on a logarithmic scale in Figure 4; the experimental data points are plotted on the curve which follows the law $S_t \sim \omega^2$. As can be seen, data points bunch closely near the curve, although strict obedience of the law is never expected. The fact is that work with different lasers is performed under different experimental conditions. The pulse width of ruby and neodymium-glass lasers is approximately 30 ns; CO_2 laser, in this case, 80ns; HF, 120ns; DF, 90ns; and D_2O, 75ns. The focal spot diameters are also different (10^{-2}–10^{-3} cm). At the long wavelength, threshold essentially depends on either presence of dust particles in the air or the preionization conditions, since the occurrence of priming electrons in these cases is difficult. Deviation of a point at $\lambda = 0.38$mm from the curve $S_t \sim \omega^2$ is associated with the fact that the frequency ω is already comparable with collisional frequency ν_m and the law must be corrected for the latter $[S_t \sim (\omega^2 + \nu_m^2)]$. Allowance for this makes the theory more compatible with experiment.

The law $S_t \sim \omega^2$ is violated significantly in the short-wave region of the spectrum as a result of breakdown by the second harmonics of neodymium-glass and ruby lasers. Instead of increasing, the threshold intensity decreases sharply with increasing frequency (quantum growth). Here, quantum effects are fully in evidence; the second harmonic of a ruby laser is very large, 3.56 eV.

(f) A long spark

At a moderately high intensity above threshold, laser radiation must be sharply focused to produce breakdown, which occurs only in a small focal region. However, at very high intensities in the case of a beam weakly collected by a long-focus lens, the intensity is sufficient to produce air breakdown over a long distance along the lens axis and beyond. This results in an extended optical breakdown, a highly impressive phenomenon called the "long spark."

A two-meter long spark was observed for the first time in 1967, when a 1-GW 18-ns giant-pulse neodymium-glass laser was focused through an $f = 2.5$-m lens.[8] Two years later, a 25-m spark was produced [by a 90-J 4-GW(peak) neodymium-glass laser, with a beam divergence of 4×10^{-5} rad and focused by a $f = 28$-m lens].[9] A 15-m section of the spark extended in front of the focus and a 10-m section, behind. A record-length spark—longer than 60 m—was obtained in 1976 by means of a two-stage neodymium-glass laser setup, with the combined energy of 160J and average power of 5GW, using an $f = 40$m lens.[10] The spark, produced in a courtyard (at an institute) is well defined against the background of a building (Fig. 5). Long sparks are never continuous, but consist of ionized sections alternating with those unaffected by breakdown. Clearly, this is associated with the statistical origin of the priming electrons which occur at selected points and, probably, the time-dependent variations in the field at various points due to a complex spatial-temporal and angular structure of the intense light beam.

Long sparks were also obtained in air by means of high-power electroionization CO_2 lasers[11,12] (of the order of 1 m[11]). The purpose of one work[12] was to establish the maximum power and intensity of the CO_2 laser radiation that can be propagated through air, a problem of considerable importance. The laser output was 160J, of which 30J was produced during 50 ns and the remainder, 130J, during μs; the peak power was 0.56 GW. The longest spark (7.5m) was achieved by expanding the initial beam by means of a telescope to be 40 cm in diameter and, subsequently, focusing it by means of a long-focus mirror ($f = 54$m) directly outdoors; the angle of beam convergence (d/f) was 1/135 and the least cross-section diameter, 0.5 cm. The spark occurred at intensities of $1-2 \times 10^8$ W/cm^2. A considerable portion of radiation (of the order of tens of percent) was absorbed in the process by the plasma. Both the plasma generation threshold and amount of

FIG. 5. Photograph of a long spark obtained by means of a neodymium-glass laser. Spark length 8 m, focal length of lens 10 m (Ref. 10).

energy absorbed in the plasma depend on the dust content of air, the presence in air of sub-micron size aerosol particles, and humidity. In purified air, the threshold increases to 3×10^9 W/cm^2. Schlieren photography shows that each particle serves as a plasma focus from which an optical detonation wave propagates (see below) and leaves an absorbing plasma cloud behind it.

Actually, the plasma generation threshold measured in the experiment (10^8 W/cm^2) is not the same as the breakdown threshold, i.e., occurrence of avalanche ionization in a gas induced by a priming electron; the latter is an order of magnitude higher. Instead, it represents a threshold at which the plasma foci occur as a result of heating of gas particles and the subsequent ionization of ambient air by laser radiation. The question of what is the real mechanism for breaking down dusty air by CO_2 laser radiation remains unclear. Citations concerning this subject may be found in Refs. 12 and 13. The breakdown threshold is further reduced if the radiation is focused near a solid surface.[14,15 (and references therein)]

(g) Discharge initiation by a laser spark

It was observed some time ago that concurrent interaction of the laser radiation and other fields—microwave, constant—with a gas considerably enhances its breakdown by the other field. In this manner, directed breakdown is achieved between electrodes under a constant potential: The spark discharge develops along the optical channel and may be directed either at an angle to the constant field or even be fractured (for references see Refs. 1 and 11). The lowering of the electric breakdown threshold and a very rapid laser interaction effect have contributed to the development of laser-fixed dischargers.[16] The long laser spark has been used effectively to initiate discharge in long interelectrode gaps.[11,17-19] This procedure may replace the conventional method of using thin exploding wires for initiating electrical discharges, which has many disadvantages. The long laser-spark path provides a conduit for the electrode gap discharge. Moreover, breakdown electric field intensity is reduced considerably to 250V/cm.[19] Normally, electric breakdown of long gaps is due to a leader mechanism: A bright channel leader, propagates from the anode, and is preceded by a darker streamer. The dense portion of a long laser spark, which lies relatively close to the focus, is an equivalent of a leader thus formed.[19]

3. MAINTENANCE OF AN UNSTABLE PLASMA

Glow discharge is one of the most common discharge processes in a constant field at pressures below tens of torr. Unstable, weakly-ionized stationary plasmas may be produced at both radio and microwave frequencies at low pressures. At optical frequencies, however, the steady-state process is totally atypical; instead, it calls for much higher radiation intensities. The power of currently available cw lasers is sufficient, as a rule, to maintain a stable plasma only.

Steady-state maintenance of an unstable plasma al-

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