THEORY OF BREAKDOWN OF NOBLE GASES AT OPTICAL FREQUENCIES

D. D. RYUTOV

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A kinetic theory of the breakdown of noble gases by an intense light beam is presented. It is shown that the optical breakdown mechanism is in general similar to the familiar high-frequency breakdown mechanism. The investigation is carried out on the basis of a classical-mechanics electron kinetic equation. Quantum effects are negligible if $\hbar\omega \ll E_i$, where ω is the optical radiation frequency and E_i is the ionization potential of the gas atom. The presence in the equation of small parameters that are characteristic only of breakdown at optical frequencies permits one to obtain an exact solution of the kinetic equation. The dependence of the threshold light-wave electric field $\mathcal{E}$ on the neutral gas atom density N is derived. This dependence can be expressed in the form $\exp(a/\mathcal{E}) = bN^2\mathcal{E}^{17/6}$ where a and b are constants that depend on the nature of the gas. The theory agrees with the experimental results for the breakdown in argon and helium.

1. INTRODUCTION

THE breakdown of noble gases at optical frequencies was investigated experimentally by Minck[1] and by Mayerand and Haught[2]. In these experiments, an intense light beam (wavelength $\lambda = 6934$ Å) was focused on the center of a chamber filled with the investigated gas. The electric field of the light wave caused the gas to break down at the focus. The breakdown was manifest in a bright flash of light and in a cascade-like growth of the electron density. The breakdown took place when the electric field intensity of the focus exceeded a certain threshold value. The dependence of the threshold electric field on the pressure of the argon and helium was investigated in the range from 0.1 to 1000 atm. The purpose of this paper is to explain these relationships.

We note first that the gas breakdown cannot be caused by photoionization: the energy of the light quantum corresponding to 6934 Å is merely 1.8 eV, whereas the ionization energy of argon is 15.76 eV, while that of helium is 24.58 eV. The multiple absorption of photons likewise plays no role[2]. Apparently, the mechanism of breakdown at optical frequencies is close to the mechanism of ordinary high-frequency breakdowns[3]. Qualitatively the breakdown process can be described in the following fashion. A certain residual electron density always appears in the gas at the instant when the light pulse is turned on. The electric field of the light wave increases the energy of the electrons to the ionization value E_i. The neutral atoms are ionized, and the electron intensity increases. The electrons are again accelerated to the ionization energy, the atoms are ionized, etc. The gas breaks down when the increase in the electron density due to the ionization exceeds the decrease in the electron density due to diffusion from the region of the strong electric field at the focus. Electron losses due to adhesion are negligible in noble gases.

A distinguishing feature of breakdown at optical frequencies is that this phenomenon is determined to a considerable degree by the excitation processes. The electrons attaining the energy E_{ex} of the first excited state of the gas atoms can lose their energy to excitation without causing ionization. It turns out that in the field of a light wave the energy of the electrons increases "slowly": the time required for the energy to rise from E_{ex} to E_i is much longer than the time during which the electron loses energy to excitation[1)] (the low efficiency of electron acceleration is connected with the fact that the frequency of the light wave is very high). Therefore most electrons give up energy to excitation without attaining E_i. In other words, the electron energy distribution function $F(E)$ experiences an abrupt decrease in the region $E > E_{ex}$. This is precisely why we can solve exactly the problem of the breakdown limit by using the kinetic equation for the electrons.

To determine the breakdown limit, we can assume that the electron density n_e is small and linearize all equations with respect to n_e. Fol-

[1)] In experiments on gas breakdown by a radio-frequency field, the situation is reversed (see Sec. 3).

lowing this procedure, we neglect recombination and collisions between charged particles. We do not take into account plasma effects such as ambipolar diffusion, assuming that the Debye radius is large compared with the characteristic dimension r_0 of the region in which the electric field differs from zero.[2)] We also disregard collisions between electrons and excited atoms (this effect is essentially quadratic in n_e, so that the rate of formation of the excited atoms, together with their density, is proportional to n_e).

2. KINETIC EQUATION FOR ELECTRONS

Let us investigate the influence of a homogeneous periodic electric field $\mathscr{E} = \mathscr{E}_0 \sin \omega t$ on the electron distribution function (the electric field of the light wave can be regarded as homogeneous if $v \ll c$, where v—electron velocity and c—speed of light). The electron velocity distribution function f(v) satisfies the equation

$$\frac{\partial f}{\partial t} - \frac{e\mathscr{E}_0}{m} \sin \omega t \frac{\partial f}{\partial \mathbf{v}} = I(f), \tag{1}$$

where I(f) is the electron-atom collisions integral, and the remaining notation is standard. Since the frequency of the elastic collisions usually exceeds the frequency of the inelastic collisions, we disregard the latter for the time being.

We introduce in velocity space a polar system of coordinates (v, θ, φ) with polar axis directed along the vector $\mathscr{E}_0'$. In this coordinate system Eq. (1) takes the form

$$\frac{\partial f}{\partial t} - \frac{e\mathscr{E}_0}{m} \sin \omega t \left(\frac{\partial f}{\partial v} \cos \theta - \frac{\sin \theta}{v} \frac{\partial f}{\partial \theta} \right) = I(f). \tag{2}$$

The collision integral I(f) vanishes when we substitute in it any spherically symmetrical distribution function. Therefore, when $\mathscr{E}_0 = 0$ the stationary solution of (2) is any spherically symmetrical function $f_0(v)$.

From a comparison of the first and second terms in the left side of (2) it follows that the second term is small compared with the first, if the electric field $\mathscr{E}_0$ is sufficiently small, namely if

$$e\mathscr{E}_0 / m\omega \ll \bar{v},$$

where $\bar{v}$ is the mean square electron velocity,

2)The Debye radius r_D is proportional to $n_e^{-1/2}$. It is therefore clear that at sufficiently low electron density, the inequality $r_D > r_0$ is satisfied. The inequality $r_D > r_0$ is satisfied when $n_e < 10^{14}$ cm^{-3} under Minck's experimental conditions[1] and when $n_e < 10^{12}$ cm^{-3} under the conditions of the Mayerand and Haught experiments[2].

that is, the deviation of the distribution function from $f_0(v)$ can be represented by a series in powers of $\mathscr{E}_0$:

$$f(v) = f_0(v) + f_1(v, \theta) + f_2(v, \theta) + \ldots,$$

where

$$f_1 = O(\mathscr{E}_0), \quad f_2 = O(\mathscr{E}_0^2)$$

etc.

The equation for the first-order correction $f_1(v, \theta)$ is of the form

$$\frac{\partial f_1}{\partial t} - \frac{e\mathscr{E}_0}{m} \sin \omega t \cos \theta \frac{\partial f_0}{\partial v} = I(f_1). \tag{3}$$

We assume that the atoms are infinitely heavy. In this case, as shown by Bayet et al[4], the collision integral has the following properties:

$$\int \sin \theta I[f(v, \theta)] d\theta = 0, \tag{4}$$

where $f(v, \theta)$ is an arbitrary function of v and θ, and

$$I[g_1(v) \cos \theta] = -g_2(v) \cos \theta, \tag{5}$$

where $g_1(v)$ is an arbitrary function of v, while $g_2(v)$ is connected with $g_1(v)$ by the relation

$$g_2(v) = \nu(v) g_1(v), \qquad \nu(v) = N v \sigma_{tr}(v).$$

Here N—density of the neutral atoms and $\sigma_{tr}(v)$—cross section for the transfer of momentum from the electron to the atom (transport cross section).

Taking (5) into account, we seek the solution of (3) in the form

$$f_1(v, \theta) = g(v) \cos \theta.$$

The function g(v) satisfies the equation

$$\frac{\partial g}{\partial t} + \nu(v) g = \frac{e\mathscr{E}_0}{m} \frac{\partial f_0}{\partial v} \sin \omega t.$$

Hence

$$g = -\frac{e\mathscr{E}_0}{m(\omega^2 + \nu^2)} \frac{\partial f_0}{\partial v} (\omega \cos \omega t - \nu \sin \omega t),$$

$$f_1(v, \theta) = \frac{e\mathscr{E}_0}{m(\omega^2 + \nu^2)} \frac{\partial f_0}{\partial v} \cos \theta (\nu \sin \omega t - \omega \cos \omega t). \tag{6}$$

The second-order correction $f_2(v, \theta)$ is determined from the equation

$$\frac{\partial f_2}{\partial t} = \frac{e\mathscr{E}_0}{m} \left(\frac{\partial f_1}{\partial v} \cos \theta - \frac{\sin \theta}{v} \frac{\partial f_1}{\partial v} \right) \sin \omega t + I(f_2).$$

Let us average this equation over the angles and over the period of the electric field

$$\frac{\partial}{\partial t} \int_0^{\pi} \sin \theta \langle f_2 \rangle d\theta = \frac{e\mathscr{E}_0}{m} \int_0^{\pi} \sin \theta \cos \theta \frac{\partial}{\partial v} \langle \sin \omega t f_1 \rangle d\theta$$

$$-\frac{e\mathscr{E}_0}{m}\int_0^{\pi}\frac{\sin^2\theta}{v}\frac{\partial}{\partial\theta}\langle\sin\omega t\, f_1\rangle\, d\theta \qquad (7)$$

(the angle brackets denote averaging over the period of the electric field). We have taken account of the fact that, in accordance with condition (4), the collision integral averaged over the angles is equal to zero.

Simple transformations enable us to represent (7) in the form

$$\frac{\partial}{\partial t}\int_0^{\pi}\sin\theta\langle f_2\rangle\, d\theta = \frac{e\mathscr{E}_0}{mv^2}\int_0^{\pi}\sin\theta\cos\theta\frac{\partial}{\partial v}v^2\langle\sin\omega t\, f_1\rangle\, d\theta. \qquad (8)$$

It follows from (6) and (8) that, accurate to terms of second order in $\mathscr{E}_0$,

$$\frac{\partial}{\partial t}\int_0^{\pi}\sin\theta\langle f(v,\theta)\rangle\, d\theta = \frac{e^2\mathscr{E}_0^2}{6m^2\omega^2v^2}\frac{\partial}{\partial v}\nu v^2\int_0^{\pi}\sin\theta\langle f(v,\theta)\rangle\, d\theta \qquad (9)$$

(we have used the inequality $\omega \gg \nu$).

The electron energy distribution function, averaged over the period of the electric field, is connected with $f(v, \theta)$ by the relation

$$F(E)=\frac{2\pi v}{m}\int_0^{\pi}\sin\theta\langle f(v,\theta)\rangle\, d\theta, \qquad E=\frac{mv^2}{2}.$$

Substituting this relation in (9), we get

$$\frac{\partial}{\partial t}F(E)=\frac{\partial}{\partial E}D(E)\sqrt{E}\frac{\partial}{\partial E}\frac{F(E)}{E^{1/2}}, \qquad (10)$$

$$D(E)=\frac{2e^2\mathscr{E}^2}{3m\omega^2}EN\sigma_{tr}(E)\left(\frac{2E}{m}\right)^{1/2}, \qquad (11)$$

where $\mathscr{E} = \mathscr{E}_0/\sqrt{2}$ is the mean-square electric field intensity (we have introduced this quantity exclusively for an easy comparison with the experimental results of Mayerand and Haught[2], who determined just $\mathscr{E}$, and not $\mathscr{E}_0$).

In the derivation of (10) we disregarded the quantum character of the absorption of energy by the electron. The quantum effects are significant only at lower electron energies, when $E \lesssim \hbar\omega$. In the cited experiments[1,2] $\hbar\omega$ = 1.8 eV, that is, Eq. (10) is applicable at practically all energies that are significant in the problem of breakdown of noble gases.

The region of applicability of (10) is bounded on the high-pressure side. As shown in [5], if

$$N^{1/3}v > \omega \qquad (12)$$

the absorption of energy by the electrons apparently begins to be affected by the gas-atom correlation effects characteristic of condensed media. These effects are not taken into account in (10).

Recognizing that the electron velocities of importance in the breakdown of the gas reach 2–3 $\times 10^8$ cm/sec, we obtain from (12) the limit for the applicability of (10) on the high-pressure side, for the conditions of the experiments of [1,2] ($\omega = 2.7 \times 10^{15}$ sec^{-1}):

$$N < 10^{21}\ \text{cm}^{-3}. \qquad (13)$$

If we take into account inelastic collisions as well as electron diffusion, the kinetic equation for the electrons takes the form

$$\frac{\partial}{\partial t}F(E,\mathbf{r})=\frac{\partial}{\partial E}D(E,\mathbf{r})E^{1/2}\frac{\partial}{\partial E}\frac{F(E,\mathbf{r})}{E^{1/2}}+N\int_0^{\infty}[\sigma_{ex}(E,E')+2\sigma_i(E,E')]\left(\frac{2E'}{m}\right)^{1/2}F(E',\mathbf{r})\,dE'-N[\sigma_{ex}(E)+\sigma_i(E)]\times\left(\frac{2E}{m}\right)^{1/2}F(E,\mathbf{r})+d(E)\Delta F(E,\mathbf{r}). \qquad (14)$$

Here

$$d(E)=\frac{1}{3}\frac{(2E/m)^{1/2}}{N\sigma_{tr}(E)} \qquad (15)$$

is the coefficient of diffusion of electrons with energy E, Δ—Laplace operator, and $\sigma_{ex}(E)$ and $\sigma_i(E)$—excitation and ionization cross sections. The functions $\sigma_{ex}(E, E')$ and $\sigma_i(E, E')$ are defined as follows: $\sigma_{ex}(E, E')\,dE$ is the cross section of the process in which an electron of energy E' excites an atom and is left after the excitation with an energy in the interval from E to E + dE; $\sigma_i(E, E')\,dE$ is the cross section of the process in which an electron of energy E' ionizes an atom, and the energy of one of the electrons in the final state lies in the interval from E to E + dE. The factor 2 preceding $\sigma_i(E, E')$ in (14) takes into account the presence of two electrons in the final state (we have neglected ionization with production of other than a singly-charged ion). It is obvious that

$$\int_0^{\infty}\sigma_{ex}(E,E')\,dE=\sigma_{ex}(E'), \quad \int_0^{\infty}\sigma_i(E,E')\,dE=\sigma_i(E'). \qquad (16)$$

From the energy conservation law it follows that

$$\sigma_{ex}(E,E')=0 \quad \text{for} \quad E>E'-E_{ex}, \qquad \sigma_i(E,E')=0 \quad \text{for} \quad E>E'-E_i. \qquad (17)$$

The function $F(E, \mathbf{r})$ should satisfy the conditions

$$\lim_{E\to\infty}EF(E,\mathbf{r})=0, \qquad (18)$$

$$\lim_{E\to 0}E^{1/2}D(E,\mathbf{r})\frac{\partial}{\partial E}\frac{F(E,\mathbf{r})}{E^{1/2}}=0. \qquad (19)$$

The first of these conditions is a consequence of

the finite number of electrons (the integral $\int_0^\infty F(E)\,dE$ should converge). The second condition can be obtained in the following manner: We integrate (14) over the energies, taking the relations (16) into account. The result of the integration takes the form

$$\lim_{E\to 0} D(E,\mathbf{r})E^{1/2}\frac{\partial}{\partial E}\frac{F(E,\mathbf{r})}{E^{1/2}} = -\Big[\frac{\partial}{\partial t}\int_0^\infty F(E,\mathbf{r})\,dE - N\int_0^\infty \sigma_i(E)\left(\sqrt{\frac{2E}{m}}\right)F(E,\mathbf{r})\,dE - \int_0^\infty d(E)\Delta F(E,\mathbf{r})\,dE\Big].$$

It is obvious that the time derivative of the electron density

$$\frac{\partial n_e}{\partial t} = \frac{\partial}{\partial t}\int_0^\infty F(E,\mathbf{r})\,dE$$

is precisely equal to the sum of the last two terms in the square brackets, from which follows condition (19).

3. BREAKDOWN CONDITION

The threshold value of the intensity of the electric field is determined from the conditions for existence of a stationary solution of equation (14): $\partial F/\partial t = 0$. We assume for simplicity that in some region of space the radiation intensity does not depend on the coordinates, and that outside this region the intensity is equal to zero. Such an assumption signifies that the quantity D which enters in (14) and which is proportional to the radiation intensity [see (11)] does not depend on the coordinates in the region where the radiation intensity differs from zero, that is, a stationary solution of (14) can be sought by separating the variables:

$$F(E,\mathbf{r}) = F(E)R(\mathbf{r}).$$

The equation for the function F(E) is

$$\frac{\partial}{\partial E}D\sqrt{E}\frac{\partial}{\partial E}\frac{F(E)}{\sqrt{E}} + N\int_0^\infty [\sigma_{ex}(E,E') + 2\sigma_i(E,E')]\times\sqrt{\frac{2E'}{m}}F(E')\,dE' - N[\sigma_{ex}(E)+\sigma_i(E)]\sqrt{\frac{2E}{m}}F(E) + F(E)\,d(E)\frac{\Delta R(\mathbf{r})}{R(\mathbf{r})} = 0.$$

The last term of this equation can be written

$$F(E)d(E)\frac{\Delta R(\mathbf{r})}{R(\mathbf{r})} = -\frac{\alpha}{r_0^2}d(E)F(E),$$

where α—positive constant of the order of unity, the value of which depends on the boundary conditions.

We note that the breakdown condition can be obtained by solving the stationary equation only in the case when the duration T of the light pulse greatly exceeds the time of establishment of the stationary state, which has in order of magnitude r_0^2/d:

$$T \gg r_0^2/d. \tag{20}$$

Introducing the notation

$$\gamma(E) = \frac{\alpha d(E)}{r_0^2},\quad \Gamma_{ex}(E) = N\Big(\frac{2E}{m}\Big)^{1/2}\sigma_{ex}(E),$$
$$\Gamma_i(E) = N\Big(\frac{2E}{m}\Big)^{1/2}\sigma_i(E),\ \Gamma_{ex}(E,E') = N\Big(\frac{2E'}{m}\Big)^{1/2}\sigma_{ex}(E,E'),$$
$$\Gamma_i(E,E') = N\Big(\frac{2E'}{m}\Big)^{1/2}\sigma_i(E,E'), \tag{21}$$

we can rewrite the equation for the function F(E) in the form

$$\frac{\partial}{\partial E}D(E)E^{1/2}\frac{\partial}{\partial E}\frac{F(E)}{E^{1/2}} + \int_0^\infty [\Gamma_{ex}(E,E') + 2\Gamma_i(E,E')]F(E')\,dE' - [\Gamma_{ex}(E) + \Gamma_i(E)]F(E) - \gamma(E)F(E) = 0. \tag{22}$$

The problem consists of finding the conditions for the existence of a function F(E) that satisfies (22) and the conditions (18) and (19).

We shall show that in the pressure region investigated in the experiments of [1,2] the function F(E) has the following properties: when $0 < E < E_{ex}$ it increases approximately like $E^{1/2}$, and when $E > E_{ex}$ it decreases exponentially

$$F(E) \infty \exp\Big[-\frac{2}{3}\Big(\frac{E-E_{ex}}{E_0}\Big)^{3/2}\Big], \tag{23}$$

with $E_0 \ll E_i - E_{ex}$.

An important role is played in what follows by the integral

$$G(E) = \int_0^\infty [\Gamma_{ex}(E,E') + 2\Gamma_i(E,E')]F(E')\,dE'.$$

From (17), (21), and (23) it follows that this integral differs from zero only when $E \lesssim E_0 \ll E_{ex}$.

Let us find the solution of (22) in the region $E < E_{ex}$. In this region $\Gamma_{ex}(E) = \Gamma_i(E) = 0$. We shall first make a rough comparison of the first and last terms in the right side of (22). The order of magnitude of their ratio is

$$\gamma(E)F(E)\Big/\frac{\partial}{\partial E}D(E)E^{1/2}\frac{\partial}{\partial E}\frac{F(E)}{E^{1/2}} \sim \frac{\gamma E^2}{D}.$$

Under the conditions of [1,2], this ratio is much smaller than unity[3]. Consequently, the solution

[3] The quantity $\gamma E^2/D$ has a simple physical meaning: it is the ratio of the time necessary to accelerate the electron to an energy E to the time within which the electron leaves the region of strong electric field as a result of diffusion.

can be represented by a series in powers of γ:

$$F(E) = F_0(E) + F_1(E) + \ldots,$$

where

$$F_0(E) = O(\gamma^0), \qquad F_1(E) = O(\gamma)$$

etc. A solution of (22) in the region $E < E_{ex}$, accurate to terms of first order in γ and satisfying condition (19), is of the form

$$F(E) = AE^{1/2}\left[1 + \int_0^E \frac{dE_1}{D(E_1)E_1^{1/2}} \int_0^{E_1} \gamma(E_2)E_2^{1/2}dE_2\right] - E^{1/2}\left[\int_0^E \frac{dE_1}{D(E_1)E_1^{1/2}} \int_0^{E_1} G(E_2)\,dE_2 + \int_0^E \frac{dE_1}{D(E_1)E_1^{1/2}} \times \int_0^{E_1} \gamma(E_2)E_2^{1/2}dE_2 \int_0^{E_2} \frac{dE_3}{D(E_3)E_3^{1/2}} \int_0^{E_3} G(E_4)\,dE_4\right], \quad (24)$$

where A—arbitrary constant.

Let us investigate now the equation (21) with $E > E_{ex}$. As indicated above, in this region $G(E) = 0$. In addition, it is easy to verify that $\Gamma_{ex}(E) \gg \gamma(E)$ when $E > E_{ex}$ (see [1,2]). Consequently, when $E > E_{ex}$ the function $F(E)$ satisfies the equation

$$\frac{\partial}{\partial E} D(E)E^{1/2} \frac{\partial}{\partial E} \frac{F(E)}{E^{1/2}} - [\Gamma_{ex}(E) + \Gamma_i(E)]F(E) = 0. \quad (25)$$

Let us integrate this equation over the energies from E_{ex} to ∞. As a result we obtain

$$F'(E_{ex}) - \frac{F(E_{ex})}{2E_{ex}} = -\frac{1}{D(E_{ex})} \int_{E_{ex}} [\Gamma_{ex}(E) + \Gamma_i(E)]F(E)\,dE. \quad (26)$$

On the other hand, it follows from (24) that

$$F'(E_{ex}) - \frac{F(E_{ex})}{2E_{ex}} = \frac{A}{D(E_{ex})} \int_0^{E_{ex}} \gamma(E)\sqrt{E}\,dE - \frac{1}{D(E_{ex})} \int_0^{E_{ex}} G(E)\,dE - \frac{1}{D(E_{ex})} \int_0^{E_{ex}} \gamma(E_1)\,dE_1 \int_0^{E_1} \frac{dE_2}{D(E_2)E_2^{1/2}} \times \int_0^{E_2} G(E_3)\,dE_3. \quad (27)$$

Noting that, in accordance with the foregoing,

$$\int_0^{E_{ex}} G(E)\,dE = \int_0^{\infty} G(E)\,dE = \int_0^{\infty} [\Gamma_{ex}(E) + \Gamma_i(E)]F(E)\,dE,$$

and comparing (26) with (27), we determine the constant A:

$$A = \left[\int_0^{E_{ex}} \gamma(E)E^{1/2}dE\right]^{-1} \left[\int_{E_i}^{\infty} \Gamma_i(E)F(E)\,dE + \int_0^{E_{ex}} \gamma(E_1)E_1^{1/2}dE_1 \int_0^{E_1} \frac{dE_2}{D(E_2)E_2^{1/2}} \int_0^{E_2} G(E_3)\,dE_3\right].$$

By finding the constant A we have completely determined the solution in the region $E < E_{ex}$. We shall need in what follows the value of the function $F(E)$ at the point $E = E_{ex}$. Accurate to terms of first order in γ we have

$$F(E_{ex}) = E_{ex}^{1/2}\left[\int_0^{E_{ex}} \gamma(E)E^{1/2}dE\right]^{-1} \left\{\int_{E_i}^{\infty} \Gamma_i(E)F(E)\,dE - \int_0^{E_{ex}} \gamma(E_1)E_1^{1/2}dE_1 \int_{E_1}^{E_{ex}} \frac{dE_2}{D(E_2)E_2^{1/2}} \times \left[\int_0^{\infty} G(E_3)\,dE_3 - \int_{E_2}^{\infty} G(E_3)\,dE_3\right]\right\}.$$

The second integral in the square brackets differs from zero only when $E_2 \lesssim E_0 \ll E_{ex}$. Therefore, if we neglect terms of order E_0/E_{ex}, then this relation takes the form

$$F(E_{ex}) = E_{ex}^{1/2}\left[\int_0^{E_{ex}} \gamma(E)E^{1/2}dE\right]^{-1} \times \left\{\int_0^{\infty} \Gamma_i(E)F(E)\,dE - \int_0^{\infty} [\Gamma_{ex}(E) + 2\Gamma_i(E)]F(E)\,dE \int_0^{E_{ex}} \gamma(E_1)E_1^{1/2}dE_1 \int_{E_1}^{E_{ex}} \frac{dE_2}{D(E_2)E_2^{1/2}}\right\}. \quad (28)$$

For further calculations we shall need the function $F(E)$ only in the region of energies which do not exceed greatly the ionization energy, namely in the region where $\Gamma_i(E) \ll \Gamma_{ex}(E)$. We therefore neglect the term with $\Gamma_i(E)$ in (25). In the section from E_{ex} to E_i the function $\Gamma_{ex}(E)$ can be approximated, accurate to 20—30%, by the linear function (see Fig. 1)

$$\Gamma_{ex}(E) = \Gamma_{ex}'(E - E_{ex}),$$

where Γ'_{ex} does not depend on the energy.

Thus, when $E > E_{ex}$ Eq. (25) takes the form

$$\frac{\partial}{\partial E} D(E)E^{1/2} \frac{\partial}{\partial E} \frac{F(E)}{E^{1/2}} - \Gamma_{ex}'(E - E_{ex})F(E) = 0. \quad (29)$$

So long as the inequality $E - E_0 \ll E_{ex}$ is satisfied, we can assume near the point $E = E_{ex}$ that $D(E) = D(E_{ex})$ and $E^{1/2} = E_{ex}^{1/2}$, and (29) simplifies to

$$D(E_{ex}) \frac{\partial^2 F(E)}{\partial E^2} - \Gamma_{ex}'(E - E_{ex})F(E) = 0. \quad (30)$$

We introduce the notation

$$[D(E_{ex}) / \Gamma_{ex}']^{1/3} = E_0.$$

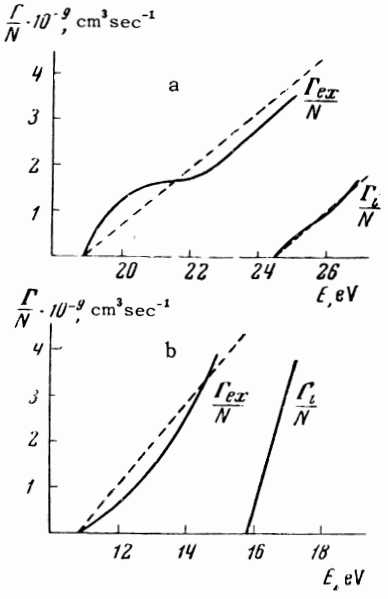

FIG. 1. Approximation of the excitation and polarization cross sections: a – for helium, b – for argon. Solid lines – experimental results of [3]. Dashed lines – lines used for approximation.

E_0 is the characteristic scale over which an appreciable change takes place in the function $F(E)$ when $E > E_{ex}$. Using relation (11), we write

$$E_0 = \left[\frac{N}{\Gamma_{ex}'}\,\frac{2e^2\mathscr{E}^2E_{ex}\sigma_{tr}(E_{ex})\,(2E/m)^{1/2}}{3m\omega^2}\right]^{1/3}.$$

This quantity does not depend on the gas density (since Γ'_{ex} is proportional to N). The theory developed above is based on the inequality

$$E_0 < E_i - E_{ex}.$$

Using the definition of E_0 given above, we can write down this inequality for argon and helium in the form

$$\mathscr{E} \lesssim 4\cdot 10^{-9}\,\omega, \quad \text{V/cm}$$

(ω is measured in sec^{-1}). As applied to the experimental conditions of [1,2], it yields

$$\mathscr{E} < 10^7 \text{ V/cm},$$

which is realized in practically the entire experimentally investigated region of electric field intensities.

We note that in experiments on radio-frequency breakdown of noble gases (see [3]), the situation is reversed:

$$E_0 > E_i - E_{ex}$$

(which is equivalent to $\mathscr{E} > 4\times 10^{-9}\omega$ in V/cm). The theory proposed here can therefore not be applied to experiments on radio frequency breakdown.

A solution of (30), finite when $E - E_{ex} \gg E_0$, is known to be (see [6], p. 584):

$$F = F(E_{ex})\int_0^\infty \cos\left[u\frac{E-E_{ex}}{E_0} + \frac{u^3}{3}\right]du \Big/ \int_0^\infty \cos\frac{u^3}{3}\,du \quad (31)$$

(the constant factor is chosen to make the function $F(E)$ continuous at the point $E = E_{ex}$). An asymptotic expression for the function $F(E)$ with $E - E_{ex} \gg E_0$ is given by formula (23). The solution (31) is valid only in the region $E - E_{ex} \ll E_{ex}$.

An asymptotic solution of (29) in the region $E - E_{ex} \gg E_0$ can be obtained without making any assumption that $D(E) = \text{const}$ or $E^{1/2} = \text{const}$. This solution is ([6], page 196)

$$F(E) = \frac{\sqrt{\pi}}{2}F(E_{ex})\left[\int_0^\infty \cos\frac{u^3}{3}\,du\right]^{-1}\left[\frac{\Gamma_{ex}'}{D(E_{ex})}\right]^{1/6}\left[\frac{D(E)}{\Gamma_{ex}(E)}\right]^{1/4}$$

$$\times \exp\left[-\int_{E_{ex}}^{E}\left[\frac{\Gamma_{ex}(E)}{D(E)}\right]^{1/2}dE\right] \quad (32)$$

(we have neglected terms of order E_0/E_{ex} in the exponential and in the pre-exponential factor). In the region $E_0 \ll E - E_{ex} \ll E_{ex}$ this solution goes over into (31).

If we substitute the function $F(E)$ given by (31) and (32) in (28) and exclude $F(E_{ex})$ from the latter, we obtain a connection between the threshold electric field intensity and the neutral gas density. Thus, the problem of determining the breakdown limit has been solved in principle.

Let us stop to evaluate the integrals in (28). To calculate the integral

$$\int_{E_i}^\infty \Gamma_i(E)F(E)\,dE,$$

we can make use of the asymptotic representation (32). Since the function $F(E)$ decreases exponentially when $E > E_{ex}$, the contribution to this integral is made by an interval of width $\sim E_0$ (from E_i to $E_i + E_0$). In calculating the integral we can therefore expand the function in the exponential of (32) in powers of $E - E_i$, confining ourselves to the zeroth and first terms of the expansion, and the pre-exponential factor can be assumed constant. The function $\Gamma_i(E)$ is well approximated by the linear function (Fig. 1):

$$\Gamma_i(E) = \Gamma_i'(E - E_i),$$

where Γ_i' does not depend on the energy. As a result of integration we obtain

$$\int_{E_i}^\infty \Gamma_i(E)F(E)\,dE = F(E_{ex})\left[\int_0^\infty \cos\frac{u^3}{3}\,du\right]^{-1}$$

$$\times \frac{\sqrt{\pi}}{2}\Gamma_i'\left[\frac{\Gamma_{ex}'}{D(E_{ex})}\right]^{1/6}\left[\frac{D(E_i)}{\Gamma_{ex}(E_i)}\right]^{1/4}$$

$$\times \exp\left[-\int_{E_{ex}}^{E_i}\left(\frac{\Gamma_{ex}(E)}{D(E)}\right)^{1/2}dE\right]. \quad (33)$$

Accurate to within an exponentially small quantity, we have

$$\int_{E_{ex}}^{\infty}[\Gamma_{ex}(E)+2\Gamma_i(E)]F(E)\,dE=\int_{E_{ex}}^{\infty}\Gamma_{ex}(E)F(E)\,dE.$$

The last integral can be calculated with the aid of (29):

$$\int_{E_{ex}}^{\infty}\Gamma_{ex}(E)F(E)\,dE=-D(E_{ex})F'(E_{ex})+\frac{D(E_{ex})F(E_{ex})}{2E_{ex}}$$

Substituting here (31) we obtain (accurate to terms of order E_0/E_{ex})

$$\int_{E_{ex}}^{\infty}\Gamma_{ex}(E)F(E)\,dE$$

$$=\frac{D(E_{ex})F(E_{ex})}{E_0}\int_0^{\infty}u\sin\frac{u^3}{3}du\Big/\int_0^{\infty}\cos\frac{u^3}{3}du. \quad (34)$$

With the aid of (28), (33), and (34) we obtain the breakdown condition (neglecting terms of order E_0/E_{ex}):

$$\exp\int_{E_{ex}}^{E_i}\left[\frac{\Gamma_{ex}(E)}{D(E)}\right]^{1/2}dE=\frac{\sqrt{\pi}\,3^{5/6}}{\Gamma(5/3)}\left[\frac{D(E_i)}{\Gamma_{ex}(E_i)}\right]^{5/4}\left[\frac{D(E_{ex})}{\Gamma_{ex}'}\right]^{1/6}$$

$$\times\Gamma_i'\left[D(E_{ex})\int_0^{E_{ex}}\gamma(E_1)E_1^{1/2}dE_1\int_{E_1}^{E_{ex}}\frac{dE_2}{D(E_2)E_2^{1/2}}\right]^{-1}. \quad (35)$$

Using formulas (11) and (15), we can rewrite this relation in the form of a connection between the threshold electric field intensity and the neutral gas density:

$$e^{a/\mathscr{E}}=bN^2\mathscr{E}^{17/6}; \quad (36)$$

$$a=\left(\frac{3m\omega^2}{2e^2}\right)^{1/2}\int_{E_{ex}}^{E_i}\left(\frac{\sigma_{ex}(E)}{E\sigma_{tr}(E)}\right)^{1/2}dE, \quad (37)$$

$$b=\frac{\sqrt{\pi}}{\Gamma(5/3)}\,2^{17/12}3^{5/12}\,\frac{r_0^2}{\alpha Q}\left[\frac{e^2}{m\omega^2}\,\frac{E_i\sigma_{tr}(E_i)}{\sigma_{ex}(E_i)}\right]^{5/4}$$

$$\times\left[\frac{e^2}{m\omega^2}\,\frac{NE_{ex}^{3/2}\sqrt{2/m}\,\sigma_{tr}(E_{ex})}{\Gamma_{ex}'}\right]^{1/6}\sqrt{\frac{m}{2}}\,\frac{\Gamma_i'}{N}\,\sigma_{tr}(E_{ex})E_{ex}^{-5/2},$$

$$Q=\frac{\sigma_{tr}^2(E_{ex})}{E_{ex}}\int_0^{E_{ex}}\frac{E_1\,dE_1}{\sigma_{tr}(E_1)}\int_{E_1}^{E_{ex}}\frac{dE_2}{E_2^2\,\sigma_{tr}(E_2)} \quad (38)$$

(we emphasize once more that the quantities Γ_{ex}'/N and Γ_r'/N in (38) do not depend on the density N).

It must be noted that the approximation of $\Gamma_{ex}(E)$ by means of a linear function is not used when calculating the constant a, since the asymptotic representation (32) is valid regardless of the concrete type of the function $\Gamma_{ex}(E)$. The constant b also depends weakly on this approximation, $b\propto(\Gamma_{ex}')^{1/6}$.

4. COMPARISON WITH EXPERIMENT

Let us ascertain first the limits of the theory. The region of its applicability is bounded on the side of strong electric fields by the inequality

$$\mathscr{E}\ll 10^7\ \text{V/cm}.$$

On the high-pressure side, the region is bounded by inequalities (13) and (20). Simple calculations show that under the conditions of Minck's experiments[1] ($r_0=6\times10^{-4}$ cm, $T=25\times10^{-9}$ sec), the more stringent of these two inequalities is (13), which yields

$$N\ll 10^{21}\ \text{cm}^{-3}.$$

Under the conditions of the experiments of Mayerand and Haught[2] ($r_0=10^{-2}$ cm, $T=30\times10^{-9}$ sec), the inequality (19) is more stringent. For helium it yields

$$N\ll 3\cdot10^{20}\ \text{cm}^{-3},$$

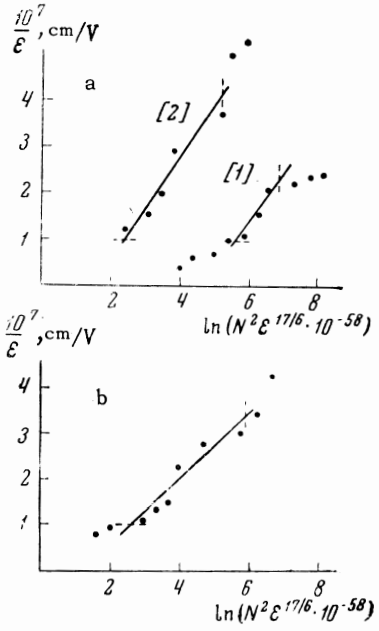

FIG. 2. Comparison of theory with the experiments of [1] and [2]: a – for helium, b – for argon. The straight lines were drawn by the least squares method, and the only points included in the calculation were those lying within the limits of applicability of the theory (the applicability limits are denoted by the dashed lines).

Experiment	Gas	$a_{theor} \cdot 10^{-7}$	$a_{exp} \cdot 10^{-7}$	$b_{theor} \cdot 10^{-59}$	$b_{exp} \cdot 10^{-59}$
[1]	Ar	1.6	1.5	~ 3	3.0
	He	1.0	0.9	$\sim 10^{-2}$	$9 \cdot 10^{-3}$
[2]	He	1.0	0.9	~ 3	2.2

and for argon

$$N \lesssim 3 \cdot 10^{19}\ \text{cm}^{-3}. \qquad (39)$$

We shall not consider below the experimental results obtained for values of the parameters $\mathscr{E}$ and M which do not satisfy the conditions of applicability of the theory. In particular, we exclude from the comparison with theory all the results of [2] on the breakdown of argon, since they were obtained at pressures for which condition (39) is not satisfied.

To verify the correctness of (36), we plotted $1/\mathscr{E}$ against $\ln N^2 \mathscr{E}^{17/6}$ (Fig. 2). In accordance with the predictions of the theory, these relations are close to linear. With the aid of Fig. 2, we can obtain the experimental values of the constants [4] a and b. The theoretical values of these constants are calculated from formulas (36) and (37). Results of comparison of the theory with experiment are listed in the table (a and b are measured respectively in units of V/cm and $\text{cm}^6\,(\text{V/cm}^{-17/6})$.

To calculate the constant a we use the cross sections given in Brown's book [3]. Exact calculation of the constant b by means of formula (37) is difficult, because we do not know the parameter α, which depends on the distribution of the electric field at the focus. We therefore made only a crude estimate of the constant b under the assumption that $\alpha \sim 1$ and $Q \sim 1$.

The good agreement between the experiments and a theory based on classical mechanics indicates that quantum effects do not play a noticeable role in optical breakdown of noble gases (in direct opposition to the opinions expressed in [2]). The quantum effects become significant only in investigations of a gas with a low ionization potential, or when the frequency of the optical radiation is increased.

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[4] The experimental results of [1,2] were given only in graphic form. The use of these results to plot $1/\mathscr{E}$ against $\ln N^2 \mathscr{E}^{17/6}$ and to calculate the constants a and b possibly causes an error of 10–15%.

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