

CASCADE IONIZATION OF A GAS BY A POWERFUL ULTRASHORT PULSE OF LIGHT

Yu. V. AFANAS'EV, E. M. BELENOV, and O. N. KROKHIN

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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We consider the process of cascade ionization of a gas under the influence of a powerful light flux, under conditions corresponding to rapid acquisition of energy by the electrons. We determine the electron energy distribution function as a function of the time, and also the time constant of the cascade development. It is shown that the concentration of electrons at a fixed instant of time decreases with increasing radiation flux, i.e., the time of cascade development increases with increasing field intensity. The results of the paper can be used to interpret experiments on optical breakdown of gases by short radiation pulses operating in the mode-locking regime.

1. Zel'dovich and Raizer^[1] developed a theory for the optical breakdown, observed by Meyerand and Haught^[2] to occur in gases under the influence of laser radiation.¹⁾ The theory proposed in^[1] corresponds to the condition of relatively small rate of acquisition of energy by the electrons in the field of the electromagnetic wave; this is realized in experiments with giant laser pulses of duration on the order of 10⁻⁸ sec. Formally this condition is of the form

$$\eta = \epsilon_0 \sigma_{tr} / I \sigma_i < 1, \tag{1}$$

where $\epsilon_0 = e^2 E_0^2 / 2m\omega^2$ is the effective oscillation energy of an electron with charge e and mass m in the field of a wave with amplitude E_0 and frequency ω ; σ_{tr} and σ_i are respectively the transport cross section for elastic collision between the electron and the atom and the cross section for the ionization of atoms with ionization potential I . According to^[1], the number of electrons in the cascade increases exponentially, $n = n_0 \exp(\gamma_0 t)$, with a growth constant γ_0 proportional to the radiation flux density $q = cE_0^2/8\pi$. When the pulse duration T_0 is decreased, the threshold value of q increases in the Zel'dovich-Raizer theory like $1/T_0$, and condition (1) is violated for short and powerful pulses.

In the case when

$$\eta = \epsilon_0 \sigma_{tr} / I \sigma_i > 1, \tag{2}$$

but the processes of photoionization of the atoms by a strong field are still negligible, the picture of the cascade ionization changes radically. The cascade development constant γ becomes a decreasing function of the flux density q , and the concentration of the electrons at a fixed value of the time decreases with increasing field intensity. This is physically connected with the fact that under the condition (2) the electron acquires, as a result of each elastic collision, an energy $\epsilon_0 > I$ and falls rapidly into the region of energies where the probability of ionization and excitation by electron impact decreases with increasing energy. The indicated mechanism of cascade ionization, as shown by estimates, makes the electron concentration in gases at normal pressure, within a pulse time 10⁻¹²–10⁻¹¹ sec, smaller by several

orders of magnitude than the concentration of the neutral atoms. As a result, the produced plasma is practically transparent to the incident radiation. In this case the only mechanism of the "laser spark" development is the breakdown process^[3]. For this reason, the breakdown region should propagate in the direction of the laser beam at a velocity equal to the velocity of the light pulse, and the length of the spark is determined by the divergence of the beam.

2. In the case under consideration, the number of electrons produced per second in 1 cm³ is given by

$$\frac{dn}{dt} = \sum_n \int F(\epsilon, t) N_0 v \sigma_n(\epsilon) d\epsilon + \int \int F(\epsilon, t) N_0 v \frac{\partial \sigma(\epsilon, \epsilon')}{\partial \epsilon'} d\epsilon' d\epsilon, \tag{3}$$

where $F(\epsilon, t)$ is the distribution function of the electrons interacting with the radiation field, $\sigma_n(\epsilon)$ is the cross section for the excitation of the atom by an electron having a velocity $v = (2\epsilon/m)^{1/2}$, $\sigma(\epsilon, \epsilon')$ is the ionization cross section of the atom, ϵ' is the energy transferred to the atom, and N_0 is the density of the neutral atoms. In relation (3), the summation is carried out over the state of both the continuous and discrete spectra. The latter is connected with the fact that the atom excited by the electron impact is ionized practically instantaneously by the radiation field, and consequently, each act of excitation of the atom is directly connected in this case with the production of an electron. For this reason, in a strong field, the effective ionization cross section should be taken to be the quantity

$$\sigma_i(\epsilon) = \sum_n \sigma_n(\epsilon) + \int \frac{\partial \sigma(\epsilon, \epsilon')}{\partial \epsilon'} d\epsilon'.$$

It is physically clear that the secondary electrons that appear as a result of the excitation of the atom by electron impact with subsequent photoionization have an initial energy much smaller than ϵ_0 , for in this case $\hbar\omega \ll \epsilon_0$. The processes of direct ionization of the atom by electron impact also lead to the appearance of relatively slow electrons. This is connected with the fact that the ionization cross section of the atom $\sigma(\epsilon, \epsilon')$ has a maximum at a produced-electron energy of the order of the atomic energy^[5]. Thus, the overwhelming majority of the produced electrons have an initial energy that is small compared with ϵ_0 . This circumstance allows us to represent the electron energy distribution

¹⁾A view of papers on optical breakdown of gases is contained in the article by Raizer^[3].

function $F(\epsilon, t)$ at the instant of time t in the form

$$F(\epsilon, t) = \int_0^t f(\epsilon, t - \tau) \varphi(\tau) d\tau, \quad (4)$$

where $\varphi(\tau)d\tau$ is the number of electrons produced after a time $d\tau$ at the instant of time τ , $f(\epsilon, t)$ is the distribution function, at the time t , for the electron produced at the time $t = 0$. On the basis of the foregoing, we put $f(\epsilon, 0) = \delta(\epsilon)$. The function $f(\epsilon, t)$ satisfies the kinetic equation

$$\frac{\partial f}{\partial t} = \left(\frac{\partial f}{\partial t}\right)_q + \left(\frac{\partial f}{\partial t}\right)_e + \left(\frac{\partial f}{\partial t}\right)_i, \quad (5)$$

where $(\partial f/\partial t)_q$, $(\partial f/\partial t)_e$, and $(\partial f/\partial t)_i$ are respectively the contributions of the radiation field, and of the elastic and inelastic collisions of the electron to the change of $f(\epsilon, t)$. The electron-electron and electron-ion collisions can obviously be neglected. Then Eq. (5) can be represented in the form

$$\partial f / \partial t = \hat{L}(\epsilon)f, \quad (6)$$

where $\hat{L}(\epsilon)$ is a linear operator.

We shall now show that Eq. (6), together with the integral relation (4), can be reduced to a simpler equation for the function $F(\epsilon, t)$. Indeed, we shall seek the function $F(\epsilon, t)$ in the form $F(\epsilon, t) = F_1(\epsilon)F_2(t)$, which is possible (see below) if $\gamma t > 1$. With the aid of the relation

$$\frac{dn}{dt} = \frac{d}{dt} \int_0^\infty F(\epsilon, t) d\epsilon = N_0 \int_0^\infty F(\epsilon, t) \sigma_i(\epsilon) v d\epsilon \quad (7)$$

we then obtain

$$\frac{1}{F_2} \frac{dF_2}{dt} = N_0 \left(\int_0^\infty F_1(\epsilon) d\epsilon \right)^{-1} \int_0^\infty F_1(\epsilon) \sigma_i(\epsilon) v d\epsilon = \gamma = \text{const.} \quad (8)$$

From (8) it follows that

$$F_2(t) = n_0 e^{\gamma t}, \quad (9)$$

where n_0 is the density of the bare electrons (under the condition $\int_0^\infty F_1(\epsilon) d\epsilon = 1$). Consequently, according to (4), we get

$$n_0 F_1(\epsilon) e^{\gamma t} = \int_0^t \varphi(\tau) f(\epsilon, t - \tau) d\tau. \quad (10)$$

For the function $\varphi(\tau)$ we get as a result of (9)

$$\varphi(\tau) = \gamma n_0 e^{\gamma \tau}. \quad (11)$$

Substituting (11) in (10) and setting the upper limit equal to infinity, which obviously can be done if $\gamma t \gg 1$, we get

$$F_1(\epsilon) = \gamma \int_0^\infty e^{-\gamma t} f(\epsilon, t) dt. \quad (12)$$

Multiplying (6) by $e^{-\gamma t}$ and integrating with respect to t from zero to infinity, we obtain for the function $F_1(\infty)$

$$(\hat{L}(\epsilon) - \gamma)F_1(\epsilon) = -\gamma f(\epsilon, 0). \quad (13)$$

We recall that

$$f(\epsilon, 0) = \delta(\epsilon). \quad (14)$$

The cascade development constant γ can be obtained by solving Eq. (13) with the aid of relation (8).

3. In the classical limit $\hbar\omega/\epsilon \ll 1$, the right side of

(6), which is connected with the diffusion of the electrons in energy space upon interaction with the radiation field only, is of the form^[1]:

$$\left(\frac{\partial f}{\partial t}\right)_q = -\frac{1}{2} \frac{\partial}{\partial \epsilon} \left[\frac{D(\epsilon)}{\epsilon} f \right] + \frac{\partial}{\partial \epsilon} \left[D(\epsilon) \frac{\partial f}{\partial \epsilon} \right], \quad (15)$$

where $D(\epsilon) = SN_0(\hbar\omega)^2 a(\epsilon)$ is the diffusion coefficient in energy space, S is the quantum flux, and $a(\epsilon)$ is the coefficient of (2) absorption of light by electrons in scattering by neutral atoms, calculated per electron and per atom. When $\hbar\omega/\epsilon \ll 1$ we have $a(\epsilon) = (2/3)a_\omega \epsilon / \hbar\omega$ ^[1], where $a_\omega = 4\pi e^2 \sigma_{\text{tr}} / mc \omega^2$ is the classical effective absorption coefficient in a weakly ionized gas. The term connected with the inelastic losses is given by

$$\left(\frac{\partial f}{\partial t}\right)_i = N_0 \int d\sigma_i(\epsilon, \epsilon') \left[f(\epsilon + \epsilon') v(\epsilon + \epsilon') \frac{d\sigma_i(\epsilon + \epsilon', \epsilon')}{d\sigma_i(\epsilon, \epsilon')} - f(\epsilon) v(\epsilon) \right]. \quad (16)$$

Inasmuch, as already noted, in the case of fast electrons the principal role is played by collisions with energy transfer $\epsilon' \ll \epsilon$, the integrand can be expanded in powers of ϵ'/ϵ . Neglecting terms of second order of smallness²⁾ in ϵ'/ϵ and recognizing that $d\sigma_i(\epsilon, \epsilon') \sim \epsilon^{-1}$ ^[5], we obtain

$$\left(\frac{\partial f}{\partial t}\right)_i = -\frac{1}{2} \frac{v(\epsilon) N_0}{\epsilon} \kappa(\epsilon) \left[f(\epsilon) - 2\epsilon \frac{\partial f}{\partial \epsilon} \right], \quad (17)$$

where $\kappa(\epsilon)$ is the effective deceleration^[5].

The kinetic equation (13) with the terms represented in the form (15) and (17) cannot be solved in general form. In a real case, however, when $\sigma_{\text{tr}} \sim \epsilon^{-2}$ and $\kappa(\epsilon) \sim \epsilon^{-1}$ ^[5], it is possible to obtain an approximate analytic solution of the equation under consideration. Expressions (15) and (17) can in this case be written in the form

$$\left(\frac{\partial f}{\partial t}\right)_q = C \frac{\partial^2}{\partial \epsilon^2} \epsilon^{-1/2} f, \quad \left(\frac{\partial f}{\partial t}\right)_i = B \frac{\partial}{\partial \epsilon} \epsilon^{-1/2} f, \quad (18)$$

where C and B are logarithmic functions of ϵ , which we shall assume constant. Thus, Eq. (13) takes the form³⁾

$$\frac{d^2 \Phi(\epsilon)}{d\epsilon^2} + a \frac{d\Phi}{d\epsilon} - b \sqrt{\epsilon} \Phi(\epsilon) = -b \delta(\epsilon), \quad (19)$$

where

$$\Phi(\epsilon) = \epsilon^{-1/2} F(\epsilon), \quad a = \frac{3}{2} \frac{N_0 \kappa(\epsilon)}{\epsilon \sigma_{\text{tr}}(\epsilon) \epsilon_0} \approx \text{const.},$$

$$b = \frac{\gamma b_0}{\epsilon_0} = \frac{\gamma}{2 \gamma_{\text{eff}} \epsilon^{3/2} \epsilon_0} = \text{const.}, \quad \gamma_{\text{eff}} = N_0 \sigma_{\text{tr}}(\epsilon) v \sim \epsilon^{-1/2}. \quad (20)$$

Let us consider first a case corresponding to a relatively small role of the inelastic energy losses, corresponding to large radiation fluxes, i.e., we put $a \approx 0$. Then the solution of (19) is given by

$$\Phi(\epsilon) = \frac{2b}{\pi} k^{-1/2} \sin \frac{2\pi}{5} \Gamma\left(\frac{8}{5}\right) \epsilon^{1/4} K_{7/5}(2k\epsilon^{3/4}), \quad (21)$$

where $k = (2/5)b^{1/2}$ and K_ν is the Macdonald function. The cascade development constant γ can then be calculated with the aid of the distribution function (21) in accordance with formula (8), and is connected with the

²⁾The diffusion term in (17) contains the small parameter $\epsilon'/\epsilon \epsilon_0$ relative to the analogous term in (15).

³⁾The elastic losses can obviously be neglected.

maximum value of the ionization $\sigma_m(\epsilon_m)$ with the energy ϵ_0 of the electron oscillations in the field, by the relation

$$\begin{aligned} \gamma &= N_0 \int_0^{\infty} \epsilon^{1/2} \Phi(\epsilon) \sigma_i(\epsilon) v d\epsilon \\ &= \left[\frac{\Gamma(4/5) \Gamma(2/5)}{\Gamma(3/5)} N_0 \left(\frac{2}{m} \right)^{1/2} \sigma_m b_0^{1/2} \epsilon_m \right]^{5/4} \epsilon_0^{-1/4}. \end{aligned} \quad (22)$$

It follows from (22), that γ decreases with increasing radiation flux density q , like $q^{-1/4}$.

When account is taken of the inelastic energy loss, Eq. (19) can be integrated approximately. By using the substitution $\Phi(\epsilon) = \psi(\epsilon) \exp(-a\epsilon/2)$, we reduce Eq. (19) at $\epsilon > 0$ to the form

$$\frac{d^2\psi}{d\epsilon^2} + \left(-b\bar{\gamma}\epsilon - \frac{1}{4}a^2 \right) \psi = 0. \quad (23)$$

Replacement of (19) by the homogeneous equation (23) is equivalent to introducing an additional condition for the normalization of the distribution function:

$$\int_0^{\infty} \bar{\gamma}\epsilon \Phi d\epsilon = 1.$$

An investigation of (23) is best carried out by regarding it as a Schrödinger equation for the wave function ψ :

$$\begin{aligned} \frac{\hbar^2}{2\bar{m}} \psi'' + (E - U)\psi &= 0, \\ \hbar &= 2/a, \quad \bar{m} = 1/2, \quad E = -1, \quad U = \hbar^2 b \bar{\gamma} \epsilon. \end{aligned} \quad (24)$$

At small ϵ we have $\psi(\epsilon) = A \exp\{-\epsilon/\hbar\}$. At large ϵ , we can find ψ by using the quasiclassical approximation^[4]

$$\psi(\epsilon) = \frac{A i}{\sqrt{p}} \exp\left\{ -\frac{i}{\hbar} \int_0^{\epsilon} p d\epsilon \right\}, \quad (25)$$

where

$$p = [2\bar{m}(E - U)]^{1/2} = i(1 + 4a^{-2}b\bar{\gamma}\epsilon)^{1/2}.$$

Consequently

$$\psi = \frac{A}{(1 + 4a^{-2}b\bar{\gamma}\epsilon)^{1/4}} \exp\left\{ -\frac{a}{2} \int_0^{\epsilon} \left(1 + \frac{4}{a^2} b \bar{\gamma} \epsilon \right)^{1/2} d\epsilon \right\}. \quad (26)$$

It is easy to see that in the limit as $\epsilon \rightarrow 0$ the function (26) coincides with the solution of (24) at small values of ϵ . For this reason, the function (26) can be regarded as a satisfactory approximation for the solution of Eq. (24) in the entire range of variation of ϵ .

Thus, we obtain ultimately for the electron energy distribution function

$$F_1(\epsilon) = \frac{A \bar{\gamma} \epsilon}{(a^2 + 4b \bar{\gamma} \epsilon)^{1/4}} \exp\left\{ -\frac{a}{2} \left[\int_0^{\epsilon} \left(1 + \frac{4}{a^2} b \bar{\gamma} \epsilon \right)^{1/2} d\epsilon + \epsilon \right] \right\}. \quad (27)$$

The parameters of the electron cascade can be calculated by direct integration using the obtained distribution function (27), but the explicit expressions turn out to be rather cumbersome. However, the calculations can be simplified, since the factor $(1 + 4a^{-2}b\bar{\gamma}\epsilon)^{1/2}$ depends little on ϵ , and consequently the distribution function (27) is close to Maxwellian. Replacing in the expression $(1 + 4a^{-2}b\bar{\gamma}\epsilon)^{1/2}$ the energy by its mean value $\sim 3T/2$, we can represent (27) in the form

$$F_1(\epsilon) = \frac{2}{\sqrt{\pi T}} e^{-\epsilon/T} \bar{\gamma} \epsilon, \quad (28)$$

where the electron temperature T is determined from the equation

$$\frac{2}{aT} = 1 + \left(1 + \frac{4}{a^2} b \bar{\gamma}^{3/2} T \right)^{1/2}. \quad (29)$$

At large radiation fluxes q , when $4a^{-2}b\bar{\gamma}T \gg 1$, we have

$$\gamma = \left(\frac{4\sigma_m \epsilon_m N_0}{m} \right)^{3/4} \left(\frac{m}{2\pi} \right)^{3/4} \left(\frac{b_0}{\epsilon_0} \right)^{1/4} \sim q^{-1/4}, \quad (30)$$

$$T = (3/2)^{-1/2} b^{-1/2} \sim q^{1/2}. \quad (31)$$

We note that the quantities γ , calculated with the aid of the distribution functions (21) and (28), differ only in the numerical factor of the order of unity. In the opposite case of small q ($4a^{-2}b\bar{\gamma}T^{1/2} \ll 1$), the expressions for γ and T are

$$\gamma = \left(\frac{4\sigma_m \epsilon_m N_0}{m} \right)^{1/2} \left(\frac{m}{2\pi} \right)^{1/2} a^{1/2} \sim q^{-1/2}, \quad (32)$$

$$T = 1/a \sim q. \quad (33)$$

Thus, γ is a decreasing function of the flux, and the growth of the field intensity hinders the cascade development.

4. It is of interest to consider an approximate model of the cascade-ionization theory developed above, in which it is possible to avoid of integration of (13). We replace the distribution function $f(\epsilon, t)$ by a function describing the trajectory of the "average" electron,

$$f(\epsilon, t) = \frac{\text{const}}{\alpha(\epsilon) - \beta(\epsilon)} \delta\left(t - \int_0^{\epsilon} \frac{d\epsilon}{\alpha(\epsilon) - \beta(\epsilon)} \right), \quad (34)$$

where

$$\alpha(\epsilon) = \epsilon_0 \bar{\gamma} \epsilon_{\text{eff}}, \quad \beta(\epsilon) = N_0 \alpha(\epsilon) v.$$

Formula (34) is a consequence of the expression for the average rate at which the electron acquires energy:

$$\frac{d\bar{\epsilon}}{dt} = \int \epsilon \hat{L}(\epsilon) f(\epsilon, t) d\epsilon = \overline{\epsilon_0 \bar{\gamma} \epsilon_{\text{eff}} (\epsilon) - \beta(\epsilon)}. \quad (35)$$

Substituting (34) in (12), we obtain

$$F_1(\bar{\epsilon}) = \frac{\text{const}}{\alpha(\bar{\epsilon}) - \beta(\bar{\epsilon})} \exp\left\{ -\gamma \int_0^{\bar{\epsilon}} \frac{d\epsilon}{\alpha(\epsilon) - \beta(\epsilon)} \right\}. \quad (36)$$

We can then use for the determination of γ the equation

$$N_0 \sqrt{\frac{2}{m}} \int_0^{\infty} \exp\left\{ -\gamma \int_0^{\epsilon} \frac{d\epsilon}{\alpha(\epsilon) - \beta(\epsilon)} \right\} \frac{\overline{\sigma_i(\epsilon) \bar{\epsilon}^{1/2} d\epsilon}}{\alpha(\epsilon) - \beta(\epsilon)} = 1, \quad (37)$$

which is equivalent to Eq. (8).

If we assume in the presently considered model the same relations for the cross sections as in Sec. 3, then $\alpha(\epsilon) \sim q/\bar{\epsilon}^{3/2}$ and $\beta(\epsilon) \sim \bar{\epsilon}^{-1/2}$. Then we can neglect the contribution of $\beta(\epsilon)$ in the calculation of the integral (37) at large values of q . As a result we get $\gamma \sim q^{-1/4}$. At small fluxes, the main contribution to the integral (37) is made by the integration region $\epsilon \sim \epsilon^*$, where $\alpha(\epsilon^*) - \beta(\epsilon^*) = 0$. In this case the integral under the exponential sign depends logarithmically on $\epsilon^* - \epsilon$ and $\gamma \sim q^{-1/2}$. Thus, the results coincide qualitatively with the more rigorous results of Sec. 3.

By way of one more example, we present in explicit form the expressions for the electron concentration

$$n(t) = \int_0^{\infty} F(\epsilon, t) d\epsilon,$$

for the average electron energy

$$\bar{\varepsilon} = \frac{1}{n(t)} \int_0^{\infty} \varepsilon F(\varepsilon, t) d\varepsilon$$

and for the coefficient of absorption $K(t)$ of the laser emission, calculated with the aid of the distribution (36) in the case when $\gamma_{\text{eff}} = \text{const}$ and $\sigma_i(\varepsilon) \sim \varepsilon^{-1}$, and neglecting the inelastic losses

$$n = n_0 e^{\gamma t}, \quad \bar{\varepsilon}(t) = \frac{\alpha}{\gamma}, \quad K(t) = \frac{\alpha}{q} n_0 e^{\gamma t}. \quad (38)$$

The cascade development constant γ is then given by

$$\gamma = \frac{2\pi}{am} N_0^2 \varepsilon_m^2 \sigma_m^2. \quad (39)$$

In the indicated case, the time of cascade development $1/\gamma$ is proportional to the flux density, the electron concentration at a fixed value of the time decreases with increasing q , and the average energy is independent of the time and increases in proportion to q^2 .

5. As was noted above, the limit of applicability of the developed theory, on the high flux-density side, is determined by the photoionization effect. We note first that condition (2) can be rewritten in the following form, by introducing the parameter $\gamma^0 = \omega \sqrt{2mI}/eE_0$, which determines the processes of photoionization in a strong field^[6]:

$$\eta = \frac{1}{\gamma^{02}} \frac{\sigma_{tr}}{\sigma_i} > 1. \quad (40)$$

The limiting flux density corresponds to the minimum possible values of γ^0 , at which photoionization still does not occur. The quantity γ_{min}^0 depends on the ratio $I/\hbar\omega$, and decreases with increase of the latter. Since the photoionization in a strong field has a threshold character, γ_{min}^0 can be determined from the condition $W(\gamma_{\text{min}}^0, I/\hbar\omega) T_0 \approx 1$, where $W(\gamma^0, I/\hbar\omega)$ is the atom photoionization probability^[6]. As shown by numerical estimates, when $I/\hbar\omega > 10$ and $\gamma_{\text{min}}^0 < 1$, this corresponds to ionization via the tunnel effect. The condition (2) is then satisfied in a relatively wide range of flux densities. When $I/\hbar\omega < 10$ and $\gamma_{\text{min}}^0 > 1$ (corresponding physically to multiphoton ionization), the region of applicability of the considered breakdown mechanism becomes narrower.

In particular, for atoms with an ionization potential $I = 20$ eV and for a neodymium-laser pulse duration 5×10^{-12} sec, the range of current densities in which the ionization picture considered here is realized turns out to be $(\sigma_i/\sigma_{tr}) \times 10^{14} < q < 10^{15}$ W/cm². It must be

emphasized that the region of applicability of the breakdown mechanism considered here is greatly broadened for lower-frequency radiation, in the infrared range of the spectrum, since $\omega_0 \sim \omega^{-2}$.

In conclusion we note that the expression given here for the cascade development constant γ depends very strongly on the cross sections of the elastic and inelastic collisions. For this reason, it is meaningful to perform in each concrete case numerical calculations corresponding to definite experimental conditions, on the basis of an exact integration of (8). Similar remarks should be made also with respect to the determination of the region of applicability of the ionization mechanism considered here, since at the present time there is still no sufficiently accurate verification of the theoretical results^[7].

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