Quantum aspects of laser-induced breakdown in argon

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This paper presents a theoretical study of laser-induced breakdown in argon. It is shown that the Ramsauer minimum in the momentum-transfer cross section of argon causes the mean electron energy in the discharge to be rather small. Thus, in the case of large laser quanta, the process of breakdown must be treated quantum mechanically. In order to calculate breakdown parameters for this case, a computer simulation based upon elementary quantum processes occurring in the laser radiation field is presented. Using this method, the electron multiplication coefficient in the discharge is calculated for several values of laser quanta energies. In the case of large laser quanta, the multiplication coefficient in argon is found to be higher than is predicted by the classical theory. It is shown that helium, not having a Ramsauer minimum, does not reveal a similar feature. Comparing our theoretical results with those of experiments from the literature, information is obtained on the role photoionization from upper excited levels of argon plays in the breakdown process.

INTRODUCTION

The process of laser-induced breakdown in gases was first discovered and investigated in the visible region.[1] Recently, with the development of high-power gas lasers, it has been possible to extend research on breakdown into the infrared region.[2-4]

The opposite has been the case with the development of a theory for the subject. The main problem was to find the ionization frequency of a gas in the laser radiation field. For this purpose, we need to know the electron energy distribution function in the breakdown region. Zeldovich and Raizer[5] (ZR) treated this problem quantum mechanically. In that fundamental paper, the distribution function was found by means of a quantum-kinetic equation, which was solved in the classical limit when the mean electron energy is much greater than the energy of a laser quantum $\hbar\omega$. This condition exists specifically in the infrared region where $\hbar\omega \simeq 0.1$ eV. The first numerical solution of the quantum-kinetic equation in the visible region was given by Phelps[6] for a ruby-laser radiation flux of 7.8×10^{10} W/cm^2. Only recently, attempts have been made to extend the investigation by solving the quantum-kinetic equation numerically[7] and analytically.[8] In those papers it is shown that in nitrogen, when $\hbar\omega$ reaches 1 eV, the classical ZR theory already gives too large a value for the time required to break down the gas. The reason for this is that the ZR theory does not take into account quantum effects which occur when the laser quantum is large enough so that an electron may undergo several collisions with molecules and absorb few quanta from the radiation field before undergoing inelastic collisions. The electron may have gained excess energy so as not to be effective in exciting molecular vibrational levels. Consequently, the rate of growth of electron energy in the laser field is larger than in the case of small $\hbar\omega$, and ionization of the gas is thus made easier.

The purpose of this paper is to show that for large values of $\hbar\omega$ it is necessary to take into account the quantum nature of the energy-exchange processes in laser-induced breakdown even for certain inert gases. The reason for this, as will be shown, is that the Ramsauer minimum of the momentum-transfer cross section causes the mean electron energy $\bar{\epsilon}$ to be rather small. In Sec. I we will see that for breakdown induced by nanosecond pulses from a ruby laser in argon, at pressures above 1 atm, the condition $\hbar\omega/\bar{\epsilon}\ll 1$ is not fulfilled because of the low value of $\bar{\epsilon}$, and it is therefore impossible to apply directly the ZR theory to that case.

As a solution to the problem for the case of large photon energies, we shall suggest in Sec. II the use of a computer simulation of the electron multiplication process. According to ZR, the electron energy distribution in a laser radiation field is only dependent, for a given gas, on $G/\hbar\omega$, where G is the photon flux. We shall see in the same section that for large values of $\hbar\omega$ the distribution function in argon is dependent on $\hbar\omega$ itself. As an additional check for our simulation method, the calculations will also be performed for helium.

I. LASER-INDUCED BREAKDOWN IN THE CLASSICAL CASE: $\hbar\omega/\bar{\epsilon}\ll 1$

The breakdown mechanism in a laser radiation field for gases at high pressures is explained by avalanche multiplication. Let $N(t)$ be the electron density in the breakdown region at time t after the beginning of breakdown. In the early stage of the breakdown, the processes of recombination and diffusion are not important and are neglected. Then

$$N(t)=N_0 e^{\bar{\beta} t}\,,$$

where N_0 is the initial electron density and $\bar{\beta}$ is the coefficient of multiplication. The electrons which are created during the time interval dt are produced as a result of ionization in the gas and begin their "life" with near-zero energies. Therefore, a time τ elapses until their energy distribution function reaches equilibrium. Let us assume that

$$\tau \ll 1/\bar{\beta}\,, \tag{1.1}$$

that is, we consider the case where the electron distribution function reaches equilibrium much faster than the time necessary for a new generation of electrons to be formed. In Sec. II we will examine the general case and show that assumption (1.1) holds in the infrared as long as the photon flux is not too large. When (1.1) is fulfilled, the distribution function of all electrons in the discharge is not influenced by the newly born ones, and is equal to the distribution function $F_0(\epsilon)$ of the original N_0 electrons in equilibrium with the laser radiation field. The equation defining $F_0(\epsilon)$ and satisfying the condition $\hbar\omega/\epsilon \ll 1$ can be written[5]

$$-\frac{d}{d\epsilon}J(\epsilon)+\left(\frac{\partial F_0}{\partial t}\right)_c=0\,, \tag{1.2}$$

where

$$J(\epsilon)=\frac{\eta(\epsilon)}{3}\left(F_0-2\frac{dF_0}{d\epsilon}\right). \tag{1.3}$$

Here, $J(\epsilon)$ is the electron flux in energy space, and $\eta(\epsilon)$ is defined by

$$\eta(\epsilon)=\frac{e^2h^2}{\pi m c}\frac{G}{\hbar\omega}\nu_{\text{eff}}(\epsilon)\,, \tag{1.4}$$

where G is the photon flux in units of $\text{cm}^{-2}\,\text{sec}^{-1}$, e and m are the electron charge and mass, respectively, and $\nu_{\text{eff}}(\epsilon)$ is the elastic collision frequency given by $\nu_{\text{eff}}(\epsilon)=N_a(2\epsilon/m)^{1/2}\sigma_m(\epsilon)$, where N_a is the number density of the gas. The collision term $(\partial F_0/\partial t)_c$ is related to excitation and ionization as follows:

$$\left(\frac{\partial F_0}{\partial t}\right)_c=N_a\sum_k\left[\left(\frac{2(\epsilon+\xi_k)}{m}\right)^{1/2}F_0(\epsilon+\xi_k)\sigma_k(\epsilon+\xi_k)-\left(\frac{2\epsilon}{m}\right)^{1/2}F_0(\epsilon)\sigma_k(\epsilon)\right], \tag{1.5}$$

where $\sigma_k(\epsilon)$ is the cross section for excitation (or ionization) of a particular energy level of the atom, and ξ_k is the corresponding excitation (ionization) energy of that level. With the substitution $F_0(\epsilon)=\sqrt{\epsilon}f(\epsilon)$ and integration of (1.2), we obtain an equation defining $f(\epsilon)$ and this is what we shall use henceforth:

$$\frac{df}{d\epsilon}=-\frac{3\pi mc}{2e^2h^2}\frac{\hbar\omega}{G}\frac{1}{\epsilon^2\sigma_m(\epsilon)}\sum_k\int_\epsilon^{\epsilon+\xi_k}\epsilon' f(\epsilon')\sigma_k(\epsilon')\,d\epsilon'\,. \tag{1.6}$$

Equation (1.6) shows that the electron energy distribution function $f(\epsilon)$ depends strongly on the shape of the momentum-transfer cross section $\sigma_m(\epsilon)$. Let us now examine the case of argon, a gas in which $\sigma_m(\epsilon)$ is characterized by a deep Ramsauer minimum at small electron energies. The various relevant cross sections in argon are shown in Fig. 1. As in Ref. 5, we assume that the electrons undergo only two types of inelastic collisions: excitation, with an effective cross section $\sigma_r(\epsilon)$ and energy loss $\xi_r=11.5$ eV, and ionization of the gas. For $\sigma_r(\epsilon)$ we use values from Ref. 9, and for $\sigma_m(\epsilon)$ we use the values obtained by Engelhardt and Phelps[10] from analysis of transport coefficients in argon. The ionization cross section $\sigma_i(\epsilon)$ is taken from Ref. 11.

Using these cross sections, Eq. (1.6) was solved numerically for various values of $G/\hbar\omega$. Calculated distribution functions are shown in Fig. 2. Figure 3 shows the average electron energy $\bar{\epsilon}$ and the average excitation frequency $\bar{\nu}_r=N_a\langle\sigma_r\cdot v\rangle_{\text{av}}$ as a function of $G/\hbar\omega$.

It has been shown experimentally[12] that the breakdown threshold for argon at pressures above 1 atm with a ruby-laser 40-nsec pulse is less than $G/\hbar\omega=6\times10^{40}\ \text{erg}^{-1}\,\text{cm}^{-2}\,\text{sec}^{-1}$. From Fig. 2, however, for $G/\hbar\omega=5.3\times10^{40}\ \text{erg}^{-1}\,\text{cm}^{-2}\,\text{sec}^{-1}$, over 70% of the electrons in the discharge have energies less than 3 eV. Therefore, the condition $\hbar\omega/\epsilon\ll1$ does not hold for most of the electrons, and the classical theory fails for this case.

It is known[1] that Eq. (1.2) can be solved analytically with the following assumptions: (i) Electrons with energies $\xi_1\simeq\xi_r$ lose all their energy, exciting gas atoms. (ii) The elastic collision frequency $\nu_{\text{eff}}(\epsilon)$ is energy-independent. With these assump-

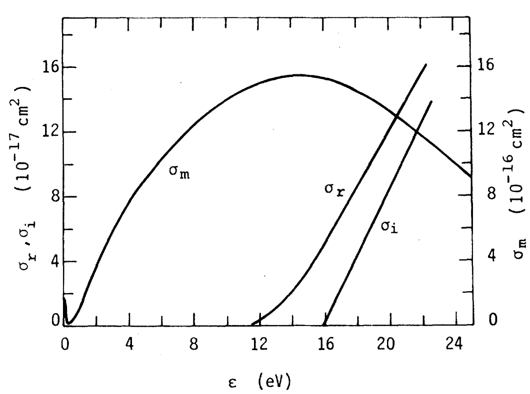

FIG. 1. Argon cross sections used in the calculations: σ_m, momentum transfer; σ_r, excitation; σ_i, ionization.

tions, the average excitation frequency is given by

$$\bar{\nu}_r = \frac{e^2 h^2}{\pi m c} \frac{\nu_{\text{eff}}}{\xi_r} \frac{G}{\hbar\omega} . \tag{1.7}$$

Both assumptions (i) and (ii) seem to be incorrect for argon. However, the linear dependence

$$\frac{\bar{\nu}_r}{N_a} \simeq 5 \times 10^{-52} \frac{G}{\hbar\omega} , \tag{1.8}$$

which is obtained by solving Eq. (1.6) (see Fig. 2), justifies the use of Eq. (1.7) with $\nu_{\text{eff}} \simeq 7.8 \times 10^{-8} N_a$. It should be emphasized that a numerical value for ν_{eff} has been obtained, whereas in other treatments[5,12] ν_{eff} was chosen quite arbitrarily.

II. COMPUTER SIMULATION OF LASER-INDUCED GAS BREAKDOWN

A. Simulation method

It has been shown in the previous section that the ZR theory fails to explain gas breakdown at high pressure of argon by visible radiation. However, to obtain an analytic solution of the quantum kinetic equation is very problematical, since one must take into account the exact energy dependence of $\sigma_r(\epsilon)$, $\sigma_i(\epsilon)$, and $\sigma_m(\epsilon)$. Nevertheless, for large $\hbar\omega$ in particular, the fact that the electrons gain and lose energy from the radiation field in quanta enables one to obtain the electron energy distribution function by a computer simulation of the breakdown process.

The simulation method which is applied here is based on elementary processes which contribute to breakdown. We assume a homogeneous photon flux in the entire breakdown region and consider a single electron of energy ϵ_0 at a time t_0. In the time interval dt after t_0, this electron can gain or lose a quantum of radiation $\hbar\omega$, or it can excite or ionize an atom. Let us denote the probabilities for these various events to occur by $GN_a a(\epsilon_0)dt$, $GN_a b(\epsilon_0)dt$, $\nu_r(\epsilon_0)dt$, and $\nu_i(\epsilon_0)dt$, respectively. The values $\nu_\mu(\epsilon) = N_a(2\epsilon/m)^{1/2}\sigma_\mu(\epsilon)$ ($\mu = r, i$) are the collision frequencies for excitation or ionization by an electron of energy ϵ, and $a(\epsilon)$ and $b(\epsilon)$ are the stimulated absorption and emission coefficients. The function $a(\epsilon)$ is given by[13]

$$a(\epsilon) = \frac{2e^2 h^2}{3\pi m\sqrt{mc}} \frac{\sigma_m(\epsilon')}{(\hbar\omega)^3} \frac{\epsilon'(\epsilon' + \epsilon)}{\epsilon^{1/2}} , \tag{2.1}$$

where $\epsilon' = \epsilon + \hbar\omega$. The stimulated emission coefficient can be written

$$b(\epsilon) = \begin{cases} 0, & \epsilon \leq \hbar\omega \\ a(\epsilon - \hbar\omega)(1 - \hbar\omega/\epsilon)^{1/2}, & \epsilon > \hbar\omega . \end{cases} \tag{2.2}$$

The initial electron energy ϵ_0 remains constant for a time Δt_0 until the electron undergoes an inelastic collision. The time Δt_0 is a random quantity which is derived from computer-generated pseudorandom numbers γ, homogeneously distributed in the interval $[0, 1]$, by the equation

$$\Delta t_0 = -\ln(\gamma)/\nu(\epsilon_0) . \tag{2.3}$$

Here, $\nu(\epsilon_0)$ denotes the total frequency of inelastic collisions $\nu = GN_a(a+b) + \nu_r + \nu_i$. As mentioned, after the time interval Δt_0 the electron undergoes an inelastic collision, and its new energy is $\epsilon_1 = \epsilon_0 - \Delta$, where Δ may have one of the values $-\hbar\omega$, $+\hbar\omega$, ξ_r, or ξ_i, depending on the type of collision. The type of collision can be simulated in the following manner: we generate the next pseudorandom number γ, which lies in one of the four segments

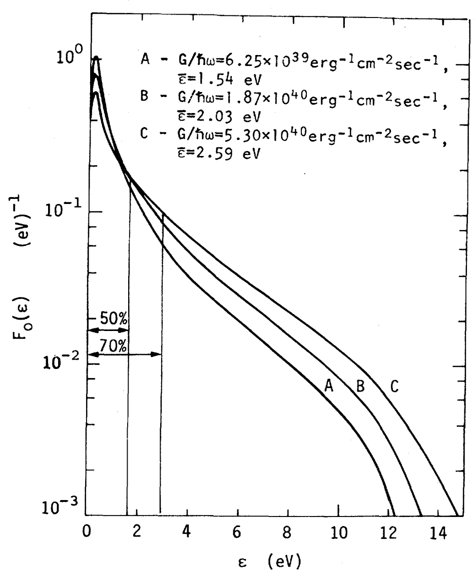

FIG. 2. Electron energy distribution functions in argon for the classical case ($\hbar\omega/\bar{\epsilon} \ll 1$).

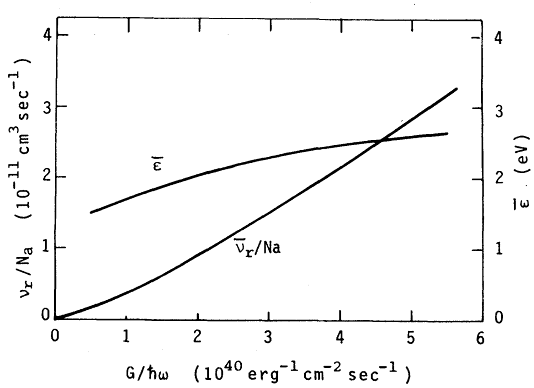

FIG. 3. Mean electron energy $\bar{\epsilon}$ and excitation frequency $\bar{\nu}_r$ in argon vs $G/\hbar\omega$ for the classical case.

$$\left[0, \frac{GN_a a}{\nu}\right];\ \left[\frac{GN_a a}{\nu}, \frac{GN_a(a+b)}{\nu}\right];\ \left[\frac{GN_a(a+b)}{\nu}, \frac{GN_a(a+b)+\nu_r}{\nu}\right];\ \left[\frac{GN_a(a+b)+\nu_r}{\nu}, 1\right],$$

which cover the range from zero to one. Now, the segment to which γ belongs determines whether the electron absorbs a quantum $\hbar\omega$, emits $\hbar\omega$, excites the atom, or ionizes it. We obtain in the same manner the next time Δt_1, for the electron of energy ϵ_1, and determine the type of inelastic collision, and so on. This process allows us to find the electron energy at any given time $t > t_0$. We repeat the procedure for a large number ($\sim 10^4$) of electrons with the same initial energy ϵ_0, and obtain the energy distribution function $F(t, \epsilon)$ for these electrons. At the same time we compute the average number of excitations $N_r(t)$ and ionizations $N_i(t)$ which are caused by an electron during the time $t - t_0$. The average excitation and ionization frequencies are given by $\bar{\nu}_r(t) = dN_r(t)/dt$, and $\bar{\nu}_i(t) = dN_i(t)/dt$. The time dependence of $\bar{\nu}_r/N_a$ is shown in Fig. 4 for the case of zero initial energy of the electrons. The following parameters were used: $\hbar\omega = 1.78$ eV; $G/\hbar\omega = 3.125 \times 10^{40}$ erg^{-1} cm^{-2} sec^{-1}; $N_a = 4 \times 10^{19}$ cm^{-3}. From this graph it can be seen that the excitation frequency rises monotonically as the electron energy distribution tends towards equilibrium with the laser radiation field. In equilibrium, $\bar{\nu}_r(t)$ reaches a constant value $\bar{\nu}_{r_0}$ which is independent of the initial electron energy. The equilibrium is reached after a time τ as a result of inelastic collisions with atoms, by which electrons lose the excitation energy ξ_r and return to the region of small energies. We can therefore approximately define τ as the mean time between inelastic collisions $1/\bar{\nu}_{r_0}$. This also follows from the calculations (see Fig. 4). We will henceforth use the following equation for $\bar{\nu}_r(t)$:

$$\bar{\nu}_r(t) = \bar{\nu}_{r_0}[1 - \exp(-2\bar{\nu}_{r_0} t)] \ . \tag{2.4}$$

This equation describes the results of the calculation quite accurately in all cases which have been treated, e.g., in Fig. 4 it is represented by the dotted line.

The dependence of $\bar{\nu}_{r_0}/N_a$ on $G/\hbar\omega$ is shown in Fig. 5 for several values of $\hbar\omega$. In this figure, the dotted line shows the results of Sec. I for small values of $\hbar\omega$ (see Fig. 3). The ratio $k = \bar{\nu}_{i_0}/\bar{\nu}_{r_0}$, which states the extent to which the electrons have entered the region of excitation $\epsilon > \xi_r$, is shown in Fig. 6. In contradiction to the results of ZR, we find that for a given value of $G/\hbar\omega$, $\bar{\nu}_{r_0}$ increases with $\hbar\omega$. This is so because: (i) For large values of $\hbar\omega$, it is enough for an electron to absorb one or two quanta for it to leave the region of the Ramsauer minimum. The rate of energy growth of the electrons, which is proportional to $\sigma_m(\epsilon)$ [see Eq. (1.2)], therefore increases; (ii) As the energy of the quantum increases, the electrons penetrate the range of electronic excitation more easily and, thus losing the excitation energy ξ_r, begin their motion with an energy larger than the minimum value of $\sigma_m(\epsilon)$, which is about 0.3 eV.

B. Electron multiplication

Until now, we have discussed the behavior of a fixed number of electrons, without taking into account the process of electron multiplication. It is difficult to directly simulate this process because

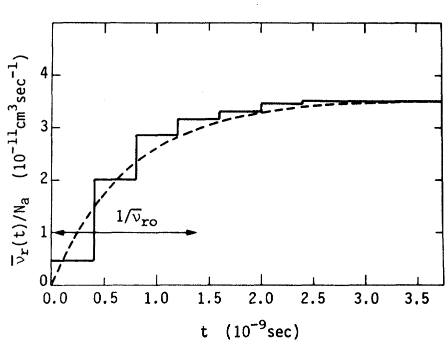

FIG. 4. Time dependence of the average excitation frequency for electrons with zero initial energy in argon. Parameters are: $\hbar\omega = 1.78$ eV, $G/\hbar\omega = 3.125 \times 10^{40}$ erg^{-1} cm^{-2} sec^{-1}, $N_a = 4 \times 10^{19}$ cm^{-3}. The solid curve is the result of the calculations and the dashed line represents the function $\bar{\nu}_r(t) = \bar{\nu}_{r_0}[1 - \exp(-2\bar{\nu}_{r_0} t)]$.

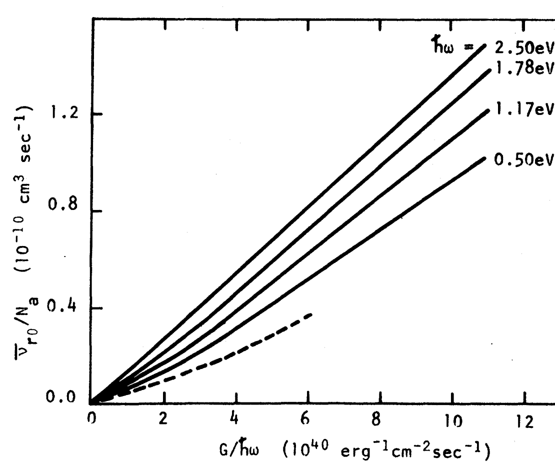

FIG. 5. Calculated average equilibrium excitation frequency in argon vs $G/\hbar\omega$. The dashed curve represents the classical case, $\hbar\omega/\bar{\epsilon} \ll 1$.

of the large number of electrons in a single avalanche. Nevertheless, with a number of reasonable assumptions, one can describe the time development of the avalanche by simulating one generation of electrons.

Let us denote by $n(t)\,dt$ the average number of new electrons which are generated in time dt in breakdown. Let all these electrons start "life" with zero initial energy, and let $n(t)\,dt\beta(t_1)\,dt_1$ be the average number of ions which they produce during dt_1, where t_1 is the time which has elapsed since the electrons were "born." When the laser quantum is small, $\beta(t_1)$ equals the ionization frequency of the gas, $\bar{\nu}_i(t)$. For large values of $\hbar\omega$, however, the process of fast photoionization allows the atoms to be ionized stepwise after being excited,[1] and therefore $\beta(t_1)=\bar{\nu}_i(t_1)+z\bar{\nu}_r(t_1)$, where z $(0<z<1)$ describes the likelihood of this photoionization. For example, as described in Ref. 1, for conditions typical to breakdown by a ruby laser in argon at high pressures, the most likely event is two-photon ionization from upper levels of the gas atom, since ionization of the lower excited levels requires three photons and is therefore less likely.

Let us now construct an equation for the average electron density in the breakdown $N(t)$. Let N_0 be the initial electron density. Then

$$n(t)\,dt=N_0\beta(t)\,dt+dt\int_0^t \beta(t-t_1)n(t_1)\,dt_1\,. \qquad \mathbf{(2.5)}$$

Substitution of $n(t)=dN(t)/dt$ into the last equation gives

$$\frac{dN(t)}{dt}=\int_0^t N(t-t_1)\frac{d\beta(t_1)}{dt_1}\,dt_1\,. \qquad (2.6)$$

Let us further assume that $\beta(t)$ has the same time dependence as $\bar{\nu}_r(t)$ [see (2.4)]:

$$\beta(t)=\beta_0[1-\exp(-2\bar{\nu}_{r_0}t)]\,. \qquad (2.7)$$

After substituting Eq. (2.7) into Eq. (2.6) we obtain the equation

$$\frac{dN(t)}{dt}=2\beta_0\bar{\nu}_{r_0}\int_0^t N(t_1)\exp[-2\bar{\nu}_{r_0}(t-t_1)]\,dt_1\,. \qquad (2.8)$$

We shall again differentiate the last equation to obtain

$$\frac{d^2N(t)}{dt^2}=2\beta_0\bar{\nu}_{r_0}N(t)-2\bar{\nu}_{r_0}\left(2\beta_0\bar{\nu}_{r_0}\int_0^t N(t_1)\exp[-2\bar{\nu}_{r_0}(t-t_1)]\,dt_1\right),$$

or, according to (2.8),

$$\frac{d^2N(t)}{dt^2}+2\bar{\nu}_{r_0}\frac{dN(t)}{dt}-2\beta_0\bar{\nu}_{r_0}N(t)=0\,. \qquad (2.9)$$

The solution of this equation for $N(t)\gg N_0$, and with the boundary conditions $N(0)=N_0$ and $(dN/dt)|_0=0$, is given by

$$N(t)=N_0e^{\bar{\beta}t}\,, \qquad (2.10)$$

where

$$\bar{\beta}=S(\bar{\nu}_{r_0}/\beta_0)\beta_0\,, \qquad (2.11)$$

and

$$S\left(\frac{\bar{\nu}_{r0}}{\beta_0}\right)=-\frac{\bar{\nu}_{r0}}{\beta_0}+\left[\left(\frac{\bar{\nu}_{r0}}{\beta_0}\right)^2+\frac{2\bar{\nu}_{r0}}{\beta_0}\right]^{1/2}\,. \qquad (2.12)$$

Let us examine two extreme cases.

(i). Breakdown induced by infrared radiation. Here $\beta_0\simeq\bar{\nu}_{i0}$ because photoionization is unlikely in this case. Thus, $\bar{\nu}_{r_0}/\beta_0=\bar{\nu}_{r_0}/\bar{\nu}_{i_0}=1/k$, and therefore

$$S\left(\frac{\bar{\nu}_{r0}}{\beta_0}\right)=-\frac{1}{k}+\left(\frac{1}{k^2}+\frac{2}{k}\right)^{1/2}\,. \qquad (2.13)$$

When $G/\hbar\omega$ is not too large, $k=10^{-1}$–10^{-2}, and therefore $0.95<S(\bar{\nu}_{r_0}/\beta_0)<1$, and $\bar{\beta}\simeq\bar{\nu}_{i_0}$.

(ii). Breakdown induced by visible radiation. In this case, $\beta_0\simeq z\bar{\nu}_{r_0}$ and, for $z=1$, $S(\bar{\nu}_{r_0}/\beta_0)=0.73$. In other words, the avalanche multiplication coefficient $\bar{\beta}$ is about 27% smaller than the ionization frequency β_0 in the state of equilibrium.

C. Comparison with experimental data

It was shown that the dependence of $\bar{\nu}_{r_0}/N_a$ on $G/\hbar\omega$ for argon is practically linear for all values

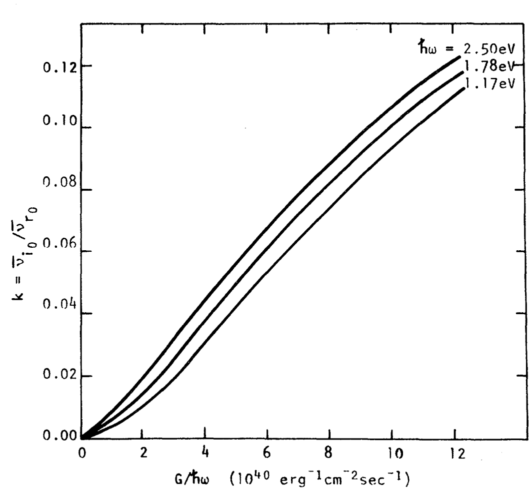

FIG. 6. Ratio $k=\bar{\nu}_{i_0}/\bar{\nu}_{r_0}$ vs $G/\hbar\omega$.

of $\hbar\omega$ examined. Therefore, we can obtain numerical agreement between these results and those given by the simplified formula (1.7), by finding an appropriate collision frequency ν_{eff}. Thus, for example, for $\hbar\omega = 1.78$ eV, we obtained (Fig. 5)

$$\bar{\nu}_{r_0}/N_a = 1.16\times 10^{-51} G/\hbar\omega\ . \tag{2.14}$$

This corresponds to expression (1.7), when $\nu_{\text{eff}} = 1.8\times 10^{-7} N_a$. If we substitute (2.14) into (2.11) and remember that in this case $\beta_0 \simeq z\bar{\nu}_{r_0}$, then

$$\bar{\beta}/N_a = 1.16\times 10^{-51}[-1+(1+2z)^{1/2}](G/\hbar\omega)\ . \tag{2.15}$$

Morgan, Evans, and Morgan[12] have shown, by analyzing the experimental results for breakdown threshold with a 40-nsec ruby-laser pulse, that the best agreement can be obtained for argon at pressures higher than 1 atm when the multiplication coefficient is $\bar{\beta}/N_a = 0.3\times 10^{-51} G/\hbar\omega$. If we compare this result with Eq. (2.15), we find that $z = 0.3$, i.e., only $\frac{1}{3}$ of the atoms are photoionized after being excited. Pressures lower than 1 atm were investigated in argon by Yong and Hercher.[14] They showed that the expression $\bar{\beta}/N_a = 0.56\times 10^{-51} \times G/\hbar\omega$ gives good agreement with experiment. From Eq. (2.15), we then obtain $z = 0.6$. The increase of photoionization at lower pressures can be explained by the larger radiation field necessary for breakdown and the possibility of three-photon ionization of the first excited levels of argon under these conditions.

D. Application of simulation method to helium

An additional effect which must be taken into account in the case of helium is the energy loss by electrons due to elastic collisions with atoms. This effect is included in our simulation procedure in the following way. Let ϵ_i be the electron energy after the ith inelastic collision. We then compute the time Δt_i which passes until the next inelastic collision, in the same manner as described in Sec. II A. Electron energy losses due to elastic collisions in this time interval, are given by

$$\Delta\epsilon_i \simeq \epsilon_i (2m/M_a)\nu_{\text{eff}}(\epsilon_i)\Delta t_i\ , \tag{2.16}$$

where M_a is the atomic mass. Thus the type of the $(i+1)$th inelastic collision is simulating considering the electron with energy $\epsilon_i' = \epsilon_i - \Delta\epsilon_i$.

Figure 7 shows the dependence of the average excitation frequency $\bar{\nu}_{r_0}/N_a$ on $G/\hbar\omega$ as calculated with the cross sections given in Ref. 15. The dashed curve in Fig. 7 represents the results of the calculations without taking into account energy losses due to elastic collisions. It can be seen that $\bar{\nu}_{r_0}/N_a$ can be approximated by

$$\bar{\nu}_{r_0}/N_a \simeq 2.67\times 10^{-52} G/\hbar\omega\ . \tag{2.17}$$

Substituting Eq. (2.17) into Eq. (2.11), and considering the case of large photon energy (when $z = 1$), we obtain

$$\bar{\beta}/N_a \simeq 1.96\times 10^{-52} G/\hbar\omega\ . \tag{2.18}$$

This result is in striking agreement with the results of Ref. 12, where it is shown that exactly the same dependence explains with good accuracy experimental values for the breakdown threshold by ruby-laser radiation in helium at high pressures.

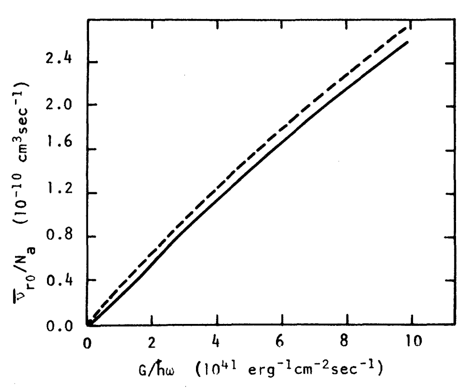

FIG. 7. Average excitation frequency in helium vs $G/\hbar\omega$. The dashed curve shows the results of the calculations without taking into account energy losses by electrons due to elastic collisions.

III. CONCLUSIONS

(i) The classical theory of Zeldovich and Raizer[5] fails in the visible region for gases which exhibit a Ramsauer minimum. This is so because the latter causes a reduction of the average electron energy $\bar{\epsilon}$ in the discharge. In these cases $\bar{\epsilon}$ may be comparable to the laser quantum $\hbar\omega$, and one must consider the quantum nature of the stimulated emission and absorption of the laser quanta by electrons.

(ii) The simulation method offers a more natural solution of the quantum-kinetic equation than those used in Refs. 7 and 8. With relatively few assumptions, one obtains parameters of the discharge, such as the average frequencies of ionization and excitation in the laser radiation field. The simulation method is in particular very effective because it enables one to follow the temporal growth of the avalanche by simulating the temporal development of one generation of electrons only.

(iii) The simulation method was applied for the cases of argon and helium. It was indeed shown that, in contrast to helium, the ionization and excitation frequencies of argon by electrons are strongly dependent on $\hbar\omega$ itself, and not only on $G/\hbar\omega$, as in the classical theory.

ACKNOWLEDGMENTS

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