THE DYNAMICS OF LASER INDUCED BREAKDOWN
IN GASES

by

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ABSTRACT

The breakdown of gases under the influence of a laser beam is analyzed. Classical diffusion theory is taken as a model, with electrons accelerated by inverse bremsstrahlung until they have sufficient energy to ionize neutral gas atoms. Both diffusion and recombination are included as losses.

For the rare gases in the range below atmospheric pressure it is found that the relation between the laser pulse duration and the plasma growth time is the primary factor which determines the breakdown threshold. The losses play roles of varying importance depending on gas, pressure and geometry. The initial electrons are generally assumed to have come from multiphoton ionization of impurities, although multiphoton ionization of the gas itself becomes a factor at sufficiently low pressures (high photon fluxes).

A number of experimental results are presented. These are used to justify some of the theoretical assumptions and to corroborate the results of the theory. The agreement is good.

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CHAPTER I

INTRODUCTION

At sufficiently high power levels, the focused output of a giant-pulse ruby laser can cause an electrical breakdown, or "spark", in air and other gases at ordinary pressures. The first quantitative measurements of this phenomenon were reported by Meyerand and Haught¹ and by Minck.² These observers measured breakdown field strength or irradiance as a function of pressure for several gases. Meyerand and Haught³ found that in the focal region, the gases were 100% ionized, and they saw abrupt attenuation of the transmitted laser beam after the onset of the discharge. They attributed this attenuation to absorption by the plasma. Later work⁴ indicated that most of the loss of transmitted light was the result of scattering and/or reflection of the incident light by the plasma.

Waynant and Ramsey,⁵ in addition to observing the pressure-dependence of breakdown threshold, measured the time required for the discharge to develop by observing the decrease in transmitted laser light. Others^{6,7} measured the frequency dependence of threshold. Gill and Dougal⁸ noted breakdown minima at very high pressures. They interpreted their results in terms of electron-impact ionization.

Early attempts to explain the laser-induced breakdown of gases were based on the effects of the radiation field on the

individual atom. Gold and Bebb⁹ developed a theory of multiphoton ionization, but detailed calculation¹⁰ indicated that this process could not provide a sufficient number of electrons at the power levels involved.

Meyerand and Haught² suggested inverse bremsstrahlung (the absorption of a photon by an electron in the neighborhood of an atom) as a possible mechanism. Phelps¹¹ was able to fit the time-to-breakdown data of Waynant and Ramsey⁵ by calculating a rate of ionization due to inverse bremsstrahlung followed by electron-impact ionization and by applying classical microwave breakdown theory. Wright¹² and Browne¹³ also considered inverse bremsstrahlung as a mechanism for breakdown, while Tozer¹⁴ concluded from a probabilistic model that multiphoton ionization could account for the initiating few electrons.

In this study we shall use the approach suggested by Phelps¹¹ to calculate the growth of electron density during the laser pulse. A theoretical curve of breakdown power as a function of pressure for the rare gases will be obtained and compared with experiment. The growth of the cascade will be examined at powers below the visible breakdown threshold and will be shown to agree with experiment. Finally, some of the properties of the laser-induced plasma in air will be reported.

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CHAPTER II

THEORY

II-1. Introduction

The electrical breakdown of gases by c.w. microwave fields has been described by diffusion theory.¹ Free electrons are assumed to be created by accelerating a few initial electrons until they suffer ionizing collisions with gas atoms. At a certain electric field strength the electron density begins to grow exponentially. This threshold field strength is determined by equating the (field-dependent) ionization rate to the net loss rate. Loss is generally assumed to be diffusive. Recombination is ordinarily neglected since it is proportional to the square of electron density, and exponential growth begins at low densities. Attachment of electrons to certain molecular species can also occur, but this loss can easily be included by subtracting a rate ν_a from the ionization rate.

For ordinary microwaves the angular frequency of the electric field is often less than the (inelastic) collision frequency of the electron with neutral atoms. For this case, the ionization rate can be related to the first Townsend coefficient α .^{1,2} When the collision frequency somewhat exceeds the frequency of the field, an effective electric field can be introduced.³

Brown and MacDonald⁴ have examined the diffusion theory as it

is described above and concluded that it is not applicable at very high frequencies, when the wavelength of the microwave field becomes comparable to the characteristic diffusion length of the breakdown region.

However, Phelps⁵ has shown that the diffusion theory can be extended even into the optical region by appealing to quantum electrodynamics to obtain an ionization rate. He assumed that the electrons absorb energy from the field by inverse bremsstrahlung, an effect that cannot be described by classical electrodynamics.

When a pulse of power (either optical or microwave) is applied to the gas, the situation is complicated by the fact that the applied field can exceed the c.w. breakdown level, yet not cause breakdown if the pulse width is short compared with the time required for the electron density to grow.⁶ For short, extremely intense pulses, diffusion may cease to play a role, and attachment may be the only significant loss mechanism. In addition, because of the high electron density that can be expected for pulses just below breakdown, recombination (which is proportional to the squared electron density) may well be a significant factor.

In the following sections, ordinary diffusion theory will be extended to the optical region using the method suggested by Phelps.⁵ Recombination, as well as attachment and diffusion, will be included as a loss mechanism. The theory will be developed and shown to be in good agreement with experiment.

II-2. Rate Equation and Definition of Breakdown

The equation of continuity governing the growth of the cascade until the onset of a visible discharge can be written

$$\frac{\partial n}{\partial t} = \sum_N a_N F^N + (\nu - \nu_a) n + D \nabla^2 n - \alpha n^2 + \beta n^2, \quad (2-1)$$

where $\sum_N a_N F^N$ = multiphoton ionization rates, per second, of various molecular species,

$n=n(r,t)$ = number density of electrons and ions, cm^{-3} ,

ν = ionization rate, (electrons created/ $\text{cm}^3\text{-sec}$)/initial electron,

ν_a = attachment rate, (attachments/ $\text{cm}^3\text{-sec}$)/electron,

D = diffusion coefficient, cm^2/sec ,

α = recombination coefficient, cm^3/sec ,

β = additional ionization coefficient discussed below.

In the preceding list, ν is the rate of ionization per electron per second per unit volume, resulting from impact ionization by electrons that have been accelerated by the photon flux. This acceleration occurs when an electron absorbs a photon in the neighborhood of an atom, which must be present to conserve energy and momentum. The last named coefficient, β , describes a rate of ionization ultimately resulting from the acceleration of electrons in the field of an ion (as opposed to a neutral atom). The rate ν can be taken proportional to the photon flux and the density of atoms (assumed constant during initial stages of growth) while the rate βn^2 is proportional to the photon flux and the density of ions. Since the

cross section for an electron-ion interaction is greater than the atomic collision cross section, βn can become comparable to ν even when n is significantly less than the density of atoms, n_a .

Looking somewhat ahead, we will show that the final term, βn^2 , does not appreciably affect the breakdown threshold. In addition to its flux dependence, the ionization rate βn is proportional to n . Near breakdown (when n is small compared with n_a), n must grow exponentially in time. Thus, βn grows exponentially compared with ν . This suggests that we define breakdown as that point where βn becomes a significant fraction of ν , say, a tenth or a hundredth. Specifically, if we assume that during growth the electron temperature is around $1/3$ the atomic ionization potential,⁴ then the "cross section" for the electron-ion-photon interaction⁷ is a few times 10^{-38} cm⁵. The electron-atom-photon cross section⁵ is around 10^{-39} cm⁵.

Accordingly, we somewhat arbitrarily ignore the βn^2 term in equation (2-1), except to define breakdown to occur when the gas is 0.1% ionized. This is in agreement with Phelps' definition⁵ and turns out to be well justified (see Chapter IV). In any case the calculated threshold fluxes will depend only slightly on the precise definition of breakdown.

II-3. Initiating Electrons

A cascade theory such as we are proposing must begin with at least a few electrons in the breakdown region. Bebb and Gold have suggested that these "initiating electrons" result from direct

multiphoton photoionization of the actual gas atoms. If N photons are required to ionize a particular species, then this process is proportional to the N -th power of the photon flux, F photons/cm²-sec. The first term in equation (2-1)

$$\left(\frac{\partial n}{\partial t}\right)_{m.p.i.} = \sum_N a_N F^N \quad (2-2)$$

describes this process for several species including any impurities. The coefficient a_N is proportional to the gas density n_a and the N -th order transition rate w given by reference (10) of the preceding chapter. Bebb and Gold were implicitly treating the case where the threshold for multiphoton ionization is higher than that for inverse bremsstrahlung; hence, the gas could be said to break down only after one electron had been supplied. Following this approach, we should then be able to estimate the breakdown threshold as a function of pressure by calculating the flux required to liberate one electron by multiphoton ionization during, say, the rise time of the laser pulse. Using triangular pulses* with $\tau = 20$ nsec half-width and a maximum flux F_m , we get a threshold condition

$$\int_0^\tau a_N \left(F_m \frac{t}{\tau}\right)^N dt = 1 \quad (2-3)$$

for a gas whose ionization potential is just under $Nh\nu$. The transition rates calculated by Bebb and Gold at the ruby laser wavelength

*See Chapter IV for details of the experimental apparatus.

are given in Table 2-1. Figure (2-1) shows initiation fluxes given by equation (2-3) along with some experimental data.

TABLE 2-1

<u>Gas</u>	<u>w/F^N</u>	<u>N</u>
He	10 ⁻⁴³⁸	14
Ne	10 ⁻³⁹⁸	13
A	10 ⁻²⁶⁵	9
Kr	10 ⁻²³³	8
Impurity	10 ⁻¹²⁰	4

The fit is not good. First of all, the pressure dependence is wrong. Second, the theoretical curves really define a lower limit for breakdown threshold, since we must include losses and also give the cascade time in which to grow. Furthermore, multiphoton ionization predicts a strong nonlinear dependence of breakdown flux as a function of frequency, but such a dependence is not observed experimentally.⁸ Nor do temporal fluctuations in the laser output, as a result of axial mode beating, appear to affect the breakdown threshold significantly, as they would for any effect nonlinear in intensity.

Finally, an experiment in which free electrons were provided by an external source indicated that the threshold for creating a first electron is not the dominant factor in determining breakdown field strength (see Chapter IV).

We are thus led to believe that the initiating electrons are created early in the laser pulse; that is, at low power levels.

FIGURE 2-1. MINIMUM BREAKDOWN THRESHOLD BY MULTI-PHOTON IONIZATION

Suppose, for example, that the gas has 1 part per million of hydrocarbon impurities whose ionization level is such that $N = 4$. Then $w/F^N \sim 10^{-113}$, and for peak fluxes around the breakdown flux of 10^{30} an electron will be born within the first nanosecond. The dashed line in Figure (2-1) represents the approximate peak irradiance for creating one electron in the first nanosecond of a 20 nsec pulse.

It is for the reasons indicated above that we are inclined to develop a theory of breakdown in which the evolution of the cascade is important. We shall assume that an electron is present in the focal region from the outset of the laser pulse. That is, multiphoton ionization will be neglected, except to provide this initial condition by ionizing impurities. We shall find that the final answer for the breakdown threshold will depend only slightly on the choice of initial electron density.

The foregoing is not meant to imply that the theory of multiphoton ionization is incorrect. To the contrary, it has been verified by a number of Russian workers⁹ at very low pressures, where there is little likelihood of a cascade's building up. In a pressure region above, say, a hundred torr, multiphoton ionization is not the dominant process. However, in Chapter IV, we shall appeal to multiphoton ionization to help explain an experimental result pertinent to argon at low pressures.

II-4. Breakdown Limited by Diffusion and Pulse Duration

In this section, we consider diffusion to be the only loss. Equation (2-1) reduces to

$$\frac{\partial n}{\partial t} = \nu(t) n + D \nabla^2 n, \quad (2-4)$$

where n and ν are functions of both position, $\underline{r}$, and time, t .

Equation (2-4) can be solved most easily if we make the assumption that its solution at any time, t , is the same as the solution to the corresponding time-independent problem (i.e., with the appropriate value of ν). Such an approximation is valid when the collision frequency (of electrons with neutral gas atoms) is considerably greater than the reciprocal of the laser pulse-width. The collision frequency is given by¹⁰

$$\nu_c = p P_c v$$

where p is pressure, P_c , the collision "probability" $\text{cm}^2/(\text{cm}^3\text{-torr})$ and v , the mean electron velocity. Using a pressure of 100 torr and a temperature of a few eV, $\nu_c \approx 10^{11} \text{ sec}^{-1} \gg \frac{1}{\tau}$, where the pulse width $\tau = 2 \times 10^{-8} \text{ sec}$. This is the worst case, so the approximation is justified.

Returning to equation (2-4), we now must solve the problem where ν is independent of time.

$$\frac{\partial n}{\partial t} = \nu n + D \nabla^2 n. \quad (2-4a)$$

This equation is separable. Setting $n(\underline{r}, t) = n(\underline{r}) f(t)$, we immediately obtain

$$D \nabla^2 n(\underline{r}) + \nu(\underline{r}) n(\underline{r}) = \gamma n(\underline{r}) \quad (2-5a)$$

$$f(t) = e^{\gamma(t)t} \quad (2-5b)$$

where γ is a separation constant and we have written $\gamma(t)$ to remind us that this solution is valid only at the instant t . It is easy to show that the solution to the time-dependent equation is

$$n(\underline{r}, t) = n(\underline{r}) e^{\int_0^t \gamma(t') dt'}, \quad (2-6)$$

where $n(\underline{r})$ is the solution to equation (2-5a) at time, t . (Actually, equation (2-5a) is an eigenvalue equation with an infinite number of solutions, so that equation (2-6) really has a sum on the right side (see Appendix). However, we shall have to consider solutions corresponding only to one or two eigenvalues and for simplicity we write down only one term.)

Equation (2-5a) is best solved by assuming a spherical focal volume of radius a , inside which the photon flux is a constant, and outside which it is zero. In this case $\mathcal{V}(\underline{r}) = \mathcal{V}$ inside, and $\mathcal{V}(\underline{r}) = 0$ outside. Assuming the electron density has no angular dependence, $n(\underline{r}) = n(r)$. Equation (2-5a) is now

$$(D\nabla^2 + \mathcal{V})n(r) = \gamma n(r) \quad (r < a), \quad (2-7a)$$

$$D\nabla^2 n(r) = \gamma n(r) \quad (r > a). \quad (2-7b)$$

It is worth pointing out the formal similarity of equations (2-7) to the equations describing a quantum mechanical potential well.¹⁰ The potential well problem leads to an equation similar to equation (2-4a) except for an imaginary coefficient which gives rise to imaginary eigenvalues, or solutions which are oscillatory in time.

The space part of the potential well equation is analogous to equations (2-7) and has the same sort of solutions as (2-7). Some of the properties of these solutions can best be understood in terms of the potential well eigenfunctions.

The solution to equations (2-7) also follows the lines of the potential well problem.¹⁰ Writing the Laplacian operator ∇^2 explicitly in spherical coordinates and suppressing the angular dependence, we obtain Bessel's equation for the spherical coordinates¹¹ from equations (2-7):

$$\frac{d^2 n}{dr^2} + \frac{2}{r} \frac{dn}{dr} + \left(\frac{\nu - \gamma}{D} \right) n = 0 \quad (r < a), \quad (2-8a)$$

$$\frac{d^2 n}{dr^2} + \frac{2}{r} \frac{dn}{dr} - \frac{\gamma}{D} n = 0 \quad (r > a). \quad (2-8b)$$

We are seeking solutions for $n(r, t)$ which grow in time; hence we may take $\gamma > 0$. At $r = a$, we demand continuity of both $n(r)$ and its derivative. Also, we know that $n(\infty) = 0$ and that $n(r)$ must be finite for all r (that is, there can be no singularities in the electron density).

With the restriction $\gamma > 0$, the solutions to (2-8b) are modified spherical Bessel functions of order zero, which can be written

$$\frac{\pi}{2(\gamma/D)^{1/2} r} e^{(\gamma/D)^{1/2} r} \quad \text{and} \quad \frac{\pi}{2(\gamma/D)^{1/2} r} e^{-(\gamma/D)^{1/2} r}. \quad (2-9a)$$

Only the second satisfies the boundary condition at $r = \infty$.

If $\nu < \gamma$, the solutions to equations (2-8a) take the same form as (2-9a) and it is clearly impossible to match derivatives at $r = a$. Thus, there are growing solutions only for $\nu > \gamma$. In this case, solutions to equation (2-8a) are spherical Bessel functions of order zero,

$$\frac{\sin r/r_0}{r/r_0} \quad \text{and} \quad -\frac{\cos r/r_0}{r/r_0}, \quad (2-9b)$$

where $\frac{1}{r_0^2} = \frac{\nu - \gamma}{D}$. Only the first remains finite for all $r < a$.

The solutions to equations (2-8) are therefore

$$n(r) = A \frac{\sin r/r_0}{r/r_0} \quad (r < a) \quad (2-10a)$$

and
$$n(r) = B \frac{\pi}{2(\gamma/D)^{1/2} r} e^{-(\gamma/D)^{1/2} r} \quad (r > a), \quad (2-10b)$$

where A and B are constants.

If we apply boundary conditions at $r = a$ and divide to eliminate A and B, we easily obtain an eigenvalue equation

$$\frac{a}{r_0} \cot \frac{a}{r_0} = -a \left(\frac{\gamma}{D} \right)^{1/2}. \quad (2-11)$$

Defining $x = a/r_0$, we can write (2-11) as

$$\cot x = - \left(\frac{x_0^2}{x^2} - 1 \right)^{1/2}, \quad (2-12)$$

where $x_0^2 = a^2 \nu / D$. This is an equation implicitly linking the eigenvalues, γ , to the ionization rate, ν . It can be solved graphically, as shown in Figure (2-2). The function $f(x) = (x_0^2/x^2 - 1)^{1/2}$ exists only in the range $f(x) < 0$; otherwise it is imaginary. The intersections on the graph of this function with $f(x) = \cot x$ are the solutions to equation (2-12) for various values of x_0 (or of ν). These solutions are indicated by circles.

It can be seen that for $x_0 < \pi/2$, equation (2-12) has no solution. Thus there is no growing mode of diffusion for $x_0 < \pi/2$; losses exceed gains in this region. The point $x_0 = \pi/2$ is the threshold for continuous wave (c.w.) breakdown. For $x_0 = 3\pi/2$, a second mode of diffusion comes into existence. However, we shall show that its effect on the breakdown threshold is small.

Figure (2-3) shows γ as a function of ν , written in terms of the dimensionless parameters $a^2 \nu / \pi^2 D$ and $a^2 \gamma / \pi^2 D$, for the first two modes of diffusion. The use of these dimensionless quantities permits us to divorce our discussion from specific gases or focal spot sizes.

For the first mode, Figure (2-2) indicates that when $x_0 \gtrsim 3\pi$, the solution to equation (2-12) is very nearly $x = \pi$. In this case, the first eigenvalue, γ_1 , becomes

$$\gamma_1 = \nu - \frac{D}{(a/\pi)^2}, \quad (2-13)$$

FIGURE 2-2. GRAPHICAL SOLUTION TO EIGENVALUE EQUATION

FIGURE 2-3. FIRST TWO EIGENVALUES AS FUNCTION OF IONIZATION RATE

where a/π is the characteristic diffusion length Λ of a sphere of radius a . Equation (2-13) or its equivalent would have resulted had we made the approximation $\nabla^2 = -\frac{1}{\Lambda^2}$ in equations (2-4) or (2-5), as is commonly done. Such an approximation is valid only when the ionization rate, ν , is much greater than that involved in c.w. breakdown. We will show in the appendix to this chapter that this is generally the case for our experimental situation. Using the more exact solution shown in Figure (2-3), we will also find that it is necessary to consider only the first or fastest growing diffusion mode.

To summarize, we now have, in the approximation of equation (2-13),

$$n(r,t) = A \frac{\sin r/\Lambda}{r/\Lambda} \exp \int_0^t \left(\nu(t') - \frac{D}{\Lambda^2} \right) dt' \quad (r < a), \quad (2-14)$$

where A is the normalization constant. We take A to be some initial value, n_0 , such that $\frac{4}{3} \pi a^3 n_0 \sim 1$ (see appendix). This corresponds to a single free electron in the focal volume. Taking a triangular pulse of half width τ , and assuming $\nu \propto \text{power}/\text{cm}^2$ (see Chapter II-6), we have

$$n(r, 2\tau) = n_0 \frac{\sin r/\Lambda}{r/\Lambda} \exp \left(\nu_m \tau - \frac{2D\tau}{\Lambda^2} \right). \quad (2-15)$$

The total number of electrons in the focal volume at the end of the pulse is therefore given by

$$n(2\tau) = 4\pi^2 n_0 \Lambda^3 \exp\left(\nu_m \tau - \frac{2D\tau}{\Lambda^2}\right). \quad (2-16)$$

Equation (2-16) is the central result of this section.

II-5. Recombination-limited Breakdown

Here we consider the case when the breakdown threshold is determined only by the pulse duration and by recombination. (The rate of attachment is always ignored for ease of notation. It can be included by replacing ν with $(\nu - \nu_a)$.) The relevant part of equation (2-1) is

$$\frac{\partial n}{\partial t} = \nu(t) n - \alpha n^2. \quad (2-17)$$

The recombination rate is proportional to the ion concentration, which is assumed equal to the electron concentration—hence the term αn^2 . The recombination coefficient α is taken to be constant for temperatures above room temperature.¹²

Equation (2-17) is a non-linear differential equation. However, we can approximate the solution in a region where recombination is not a large effect compared with the limitation due to pulse duration. To this end it is convenient to rewrite (2-17) as an integral equation,

$$n(t) = 4\pi^2 n_0 \Lambda^3 \exp\left[\int_0^t \nu(t') dt' - \alpha \int_0^t n(t') dt'\right], \quad (2-18)$$

where we have written $4\pi^2 n_0 \Lambda^3$ to be in agreement with equation (2-16). (Cf. potential well problem. Even though we are neglecting diffusion—infinite potential well—the eigenfunction inside the well is not constant, but must be zero at the walls. This eigenfunction is still $j_0(r/\Lambda)$, so we can immediately write down (2-18) by comparing with (2-15) and (2-16).)

The assumption that recombination is small permits us approximately to replace $n(t')$ in the second integral with its value in the absence of recombination,

$$n(t') \cong 4\pi^2 n_0 \Lambda^3 \exp \int_0^{t'} \nu(t) dt. \quad (2-19)$$

At the end of the laser pulse, (2-18) becomes

$$n(2\tau) = 4\pi^2 n_0 \Lambda^3 e^{\nu_m \tau} \exp \left[e^{4\pi^2 n_0 \Lambda^3} \int_0^{2\tau} \nu(t) dt \right], \quad (2-20)$$

where $\nu(t)$ is a triangular pulse with half-width τ .

The integral in equation (2-20) can be written

$$I = I_1 + I_2 = \int_0^{\tau} e^{\nu_m t^2 / 2\tau} dt + e^{\nu_m \tau} \int_{\tau}^{2\tau} e^{-\frac{\nu_m}{2\tau} (2\tau - t)^2} dt. \quad (2-21)$$

I_1 is Dawson's integral, while I_2 is the error function, neither of which can be evaluated in closed form, but both of which are tabulated.¹¹ (Physically, we could ignore I_1 anyway, since $n(t)$ is many orders of magnitude smaller during the first half of the laser pulse than it is during the second half.) Evaluating the integrals, we find

$$n(2\tau) = 4\pi^2 n_0 \Lambda^3 e^{\nu_m \tau} \exp \left[4\pi^2 \alpha n_0 \Lambda^3 \left(\frac{\pi \tau}{2\nu_m} \right)^{1/2} e^{\nu_m \tau} \right]. \quad (2-22)$$

It is appropriate at this point to deal with the term in equation (2-1), since we can use the approximation (2-19) to find the solution to an integral equation of the type (2-18), namely

$$n(t) = 4\pi^2 n_0 \Lambda^3 e^{\int_0^t \nu(t') dt'} \exp \left[-4\pi^2 \beta' n_0 \Lambda^3 \int_0^t F(t') n(t') dt' \right], \quad (2-23)$$

where $\beta' F = \beta$. We again use a triangular pulse, so the second integral in (2-23) becomes

$$I_1 + I_2 = \int_0^\tau \frac{F_m t}{\tau} e^{\nu_m t^2/2\tau} dt + e^{\nu_m \tau} \int_\tau^{2\tau} F_m \frac{2\tau - t}{\tau} e^{-\frac{\nu_m}{2\tau}(2\tau - t)^2} dt. \quad (2-24)$$

As before, I_2 is many orders of magnitude greater than I_1 .

Evaluating I_2 , we get from (2-23)

$$n(2\tau) = 4\pi^2 n_0 \Lambda^3 e^{\nu_m \tau} \exp \left[\frac{\beta' F_m \tau}{2} 4\pi^2 n_0 \Lambda^3 e^{\nu_m \tau} \right]. \quad (2-25)$$

This expression will be used shortly to show that the βn^2 -term affects threshold only negligibly.

II-6. Evaluation of Parameters

Equation (2-1) contains a number of parameters that must be evaluated. These are the ratio of ν to the photon flux, ν_a , D , and α .

We can relate ν to photon flux in the following manner. Holstein¹³ has estimated the "cross sections" for absorption of a photon by an electron in the field of helium and argon atoms. They are of the order of 10^{-39} cm⁵. To get the ionization rate per electron we consider a photon flux F photons/cm²-sec and an atom concentration n_a . As before, assuming the electron temperature is $\frac{1}{3}$ the ionization potential I , we can define an average cross section, $\bar{\sigma}$ as the cross section for electrons whose velocity corresponds to $\frac{3}{2} kT = \frac{1}{3} I$.

If free electrons are produced with near zero velocity and must absorb N photons before their energy exceeds I , then the probability per second of gaining that much energy is less than $\bar{\sigma} F/N$.

Assuming that once an electron has sufficient energy it will ionize an atom

$$\nu \lesssim \frac{\bar{\sigma} F n_a}{N} . \quad (2-26)$$

For argon, $\bar{\sigma} = 6 \times 10^{-39}$ cm⁵, $N = 9$; for helium, $\bar{\sigma} = 4 \times 10^{-39}$ cm⁵, $N = 14$. Using irradiance P in watts/cm² at 6943Å (instead of flux) we have $\nu_A \sim 2.5 \times 10^{-21} n_a P$ and $\nu_{He} \sim 1 \times 10^{-21} n_a P$. These results are about a factor of 3 greater than those obtained by Phelps,⁵ who solved the appropriate Boltzmann equation numerically. We will approximate his results by

$$\begin{aligned} \nu_A &= 7 \times 10^{-22} n_a P, \\ \nu_{He} &= 3 \times 10^{-22} n_a P. \end{aligned} \quad (2-27)$$

It is also possible to estimate the ionization rate classically by calculating an effective electric field,^{1,3} $E \nu_c / (\nu_c^2 + \omega^2)^{1/2}$, where E is the rms field strength in the focal volume. Again assuming that an electron whose energy exceeds I will certainly ionize an atom, we obtain an equation analogous to (2-26),

$$\nu \sim \frac{e^2 E^2 \nu_c}{m I \omega^2}, \quad (2-26a)$$

where $\nu_c \ll \omega$. For helium, P_c does not vary much with electron energy, so ν_c may be taken as the value for electrons of energy $\frac{1}{3} I$. Using the fact that irradiance P is proportional to E^2 , we obtain

$$\nu_{He} \sim 5 \times 10^{-23} n_a P, \quad (2-27a)$$

which is less than a factor of 10 smaller than (2-27).

A non-zero rate of attachment, ν_a , can decrease the net ionization rate. In rare gases, the attachment cross section is zero.² However, attachment to impurities may still occur.

Attachment rates are usually given in terms of the number of attachments per collision,^{2,5} h . Taking a large value, $h \sim 10^{-3}$, and $10^{-6} n_a$ impurity density in a gas with density n_a , we find the attachment rate to be on the order of $10^{-16} n_a \text{ sec}^{-1}$. This is equal to the ionization rate when P is less than a kilowatt/cm². Since we are speaking of many megawatts/cm², the impurity attachment rate can

be completely neglected.

It is next necessary to find the diffusion constant $D^{2,5,14}$. We will assume that the electron (and ion) density is always sufficiently high that the electrons cannot diffuse freely. Rather, their diffusion rate is retarded owing to the Coulomb interaction of the electrons with the slower positive ions. Such diffusion of species of both signs together is described by the ambipolar diffusion constant (which is also equal to twice the diffusion constant for the positive ions). It is related to temperature and pressure by²

$$D_P \propto T^{3/2} \quad (2-28)$$

Some values of D_{Op} at NTP are given in Table (2-2). We again take

$\frac{3}{2} kT \sim \frac{1}{3} I$, so that $D \sim 5000 D_O$. This will be justified experimentally in Chapter IV.

TABLE 2-2

gas	$\frac{\nu}{na} \left(\frac{cm^3}{sec} \right)$	$D_{Op} \left(\frac{cm^2-torr}{sec} \right)$	$\alpha \left(\frac{cm^3}{sec} \times 10^7 \right)$
He	$3 \times 10^{-22} P$	540^{14}	0.17^{10}
Ne		115^{14}	2.4^{10}
A	$7 \times 10^{-22} P$		3^{10}
Kr			6^{10}
Air		43^2	17^2
Impurity ²	-10^{-16}		

The recombination coefficients α quoted in references 10 and 2 are also shown in Table 2-2. They are taken to be independent of temperature.^{12,15}

II-7. Breakdown Threshold

We have already defined the breakdown threshold as the peak power for which a density of $10^{-3} n_a$ electrons is created in the focal volume by the end of the laser pulse. If we consider the effects of diffusion and recombination separately or in different pressure domains, we can combine equations (2-16) and (2-22),

$$n(2\tau) = 10^{-3} n_a = 4\pi^2 n_0 \Lambda^3 e^{v_m \tau - D/\Lambda^2} \exp \left[4\pi^2 \alpha n_0 \Lambda^3 \left(\frac{\pi \tau}{2v_0} \right)^{1/2} e^{v_0 \tau} \right]. \quad (2-29)$$

Writing the threshold condition explicitly in terms of peak irradiance,

$$K n_a P_b = \frac{1}{\tau} \log \frac{10^{-3} n_a}{4\pi^2 n_0 \Lambda^3} + \frac{2D}{\Lambda^2} + 4\pi^2 \alpha n_0 \Lambda^3 \left(\frac{\pi}{2v_0 \tau} \right)^{1/2} e^{v_0 \tau}, \quad (2-30)$$

where v_0 in the last term is the value of v_b calculated neglecting recombination. For easier reference, let us refer to the three terms in equation (2-28) by

$$P_b = P_0 + \Delta P_D + \Delta P_R \quad (2-31)$$

where 0 refers to no losses, D to diffusion loss and R to recombination loss. Bear in mind that the losses depend on the pulse width.

Figure (2-4) shows the effects of these three terms on the pressure (or density) dependence in a highly schematic fashion. The dashed line shows P_0 as a function of pressure. At low pressures, D increases to the point where ΔP_D becomes noticeable compared with P_0 , and the breakdown threshold increases faster than P_0 alone.

At higher pressures, where $\Delta P_D \ll P_0$, the term ΔP_R may become significant. To evaluate its role, note that the total number of electrons in the focal volume at breakdown is given by $4\pi^2 n_0 \Lambda^3 e^{V_0 \tau} = 10^{-3} n_a$ so that

$$\Delta P_R = \frac{10^{-3} \alpha}{K} \left(\frac{\pi}{2 V_0 \tau} \right)^{1/2}. \quad (2-32)$$

$V_0 \tau$ is nearly constant over a wide range of pressure. As P_0 decreases (with increasing pressure), ΔP_R becomes increasingly significant, as shown in Figure (2-4). The approximation made in equation (2-19) may break down when $\Delta P_R \gtrsim P_0$.

We now consider the effect of the βn^2 term of equation (2-1). Using equation (2-25) we get an equation analogous to (2-32),

$$\Delta V_b \tau = -\beta' \frac{F_m \tau}{2} \times 10^{-3} n_a. \quad (2-32a)$$

We know $\beta' \lesssim \frac{10^{-38}}{N} \sim 10^{-39}$, $F_m \sim 10^{30}$ and $V_0 \tau \sim 25$ for $n_a = 10^{19}$. Consequently, $\Delta V_b \tau \sim 10^{-1} \ll V_b \tau$. We are therefore justified in ignoring this term in the threshold equation.

The other terms in equation (2-30) will be evaluated in Chapter IV where we will discuss a specific experimental situation.

FIGURE 2-4. EFFECTS OF SEVERAL FACTORS ON BREAKDOWN THRESHOLD

Appendix

In this section, we show some of the details of the solution to equations (2-8). We have already formally solved the problem. Equations (2-10) give $n(r)$ for all r and equations (2-11) and (2-12) represent an eigenvalue equation whose solution is shown in Figures (2-2) and (2-3). We have deduced from Figure (2-2) that when $x_0 \gtrsim 10$, $r_0 = \Lambda$, the characteristic diffusion length of the focal volume. On the basis of this deduction, equation (2-14) was asserted. The calculation in this appendix will show that equation (2-14) is indeed valid.

We begin with equations (2-10). It is necessary to normalize the electron density and determine A and B . These normalization "constants" are in fact time-dependent, since they are implicitly functions both of $\mathcal{V}$ and of the history of the cascade up to the time of interest.

To effect the normalization, we divide the time axis into a large number of equal intervals, Δt . We approximate $\mathcal{V}(t)$ by a discrete function, $\mathcal{V}^{(k)}$, whose value during any interval, t_k to $t_k + \Delta t$, is the mean value of the continuous function. During the k -th interval, we can (at least formally) write the complete solution to the time-independent equation as a sum over all diffusion modes, both growing and decaying,

$$N^{(k)}(r) = \sum_i A_i^{(k)} n_i^{(k)}(r), \quad (2-33)$$

where i is the mode index. We find $A_i^{(k)}$, the coefficient of the fastest-growing mode, for example, by operating on (2-33) with $4\pi \int_0^\infty n_i^{(k)}(r) r^2 dr$. Let us start numbering, $k = 1$, when $\nu^{(k)}$ first exceeds the c.w. breakdown threshold. Then, if we assume initially an even distribution of electrons, $N^{(1)}(r) = n_0$, within the focal volume, we obtain

$$A_i^{(1)} = \frac{n_0 \int_0^a n_i^{(1)}(r) r^2 dr}{\int_0^\infty [n_i^{(1)}(r)]^2 r^2 dr} . \quad (2-34a)$$

After a small interval, Δt , there is still only one growing mode, so we can write

$$N^{(2)}(r) = A_i^{(2)} n_i^{(2)}(r) = A_i^{(1)} n_i^{(1)}(r) e^{\gamma_i^{(1)} \Delta t} . \quad (2-85)$$

Generalizing to time t_k , we immediately find the relation

$$A_i^{(k)} = A_i^{(k-1)} e^{\gamma_i^{(k-1)} \Delta t} \frac{\int_0^\infty n_i^{(k)}(r) n_i^{(k-1)}(r) r^2 dr}{\int_0^\infty [n_i^{(k)}(r)]^2 r^2 dr} . \quad (2-34b)$$

An identical expression is valid for the other growing modes, $2, 3, \dots$.

Equation (2-34b) is a recurrence relation which allows a numerical integration to find $N^{(k)}(r)$. The integrands are all Bessel functions and can be evaluated¹¹ by using the boundary conditions to find B.

Dropping the mode index, i , we easily obtain

$$A^{(1)} = \frac{2n_0\pi \left(\frac{\sin x_1}{x_1} - \cos x_1 \right)}{\pi \left(1 - \frac{\sin x_1}{x_1} \cos x_1 \right) + \frac{\sin^2 x_1}{(\gamma_1')^{1/2}}} \quad (2-36a)$$

and

$$A^{(k)} = \frac{\pi \left(\frac{\sin(x_k - x_{k-1})}{x_k - x_{k-1}} - \frac{\sin(x_k + x_{k-1})}{x_k + x_{k-1}} \right) + \frac{2 \sin x_k \sin x_{k-1}}{\gamma_k'^{1/2} + \gamma_{k-1}'^{1/2}}}{\pi \left(1 - \frac{\sin x_k}{x_k} \cos x_k \right) + \frac{\sin^2 x_k}{\gamma_k'^{1/2}}} \times \frac{x_k}{x_{k-1}} e^{\gamma_k \Delta t} A^{(k-1)} \quad (2-36b)$$

where $x_k = a/r_0$ and $\gamma_k' = a^2 \gamma^{(k)} / \pi^2 D$. $\gamma^{(k)}$ is the eigenvalue corresponding to $\nu^{(k)}$. Recalling that $1/r_0^2 = (\nu - \gamma)/D \rightarrow 1/\Lambda^2$, we find that for $\nu_k' = \frac{a^2 \nu^{(k)}}{\pi^2 D} \gg 10$, equation (2-36b) becomes

$$A^{(k)} \rightarrow A^{(k-1)} e^{\gamma^{(k-1)} \Delta t} \quad (2-37)$$

so that $n(r, t)$ can be found by integration,

$$A(t) = A_0 \exp \int_{t_k}^t \left[\nu(t') - \frac{D}{\Lambda^2} \right] dt' \quad (2-38)$$

A_0 is the value of $A^{(k)}$ at time t_k where we decide to stop integrating numerically and use (2-38).

We now show that t_k is very near zero and that A_0 is quite close to the initial value, n_0 . The c.w. breakdown threshold is given by

$$\nu' = \frac{a^2 \nu}{\pi^2 D} = \frac{1}{4} . \quad (2-39)$$

Using Table 2-2 with equations (2-28) and (2-30), we can estimate

$$\frac{P_b(\text{c.w.})}{P_b} \lesssim 10^{-2} \quad (2-40)$$

for the worst case, helium. Equation (2-38) is valid from about ten times the c.w. breakdown level on. Thus, t_k necessarily occurs during the first nanosecond of the pulse and can safely be replaced by zero.

The approximation we have made, which corresponds to setting $\gamma = \nu - D/\Lambda^2$, is the worst for helium where this equality is not satisfied for a sizable time interval at both the beginning and the end of the pulse (see Figure (2-3)). In fact the calculated value of γ is larger than $\nu - D/\Lambda^2$ and a growing mode of diffusion exists at ionization rates such that $\nu < D/\Lambda^2$. The effect is greatest at the end of the pulse, when the electron density is growing rapidly. We might therefore expect our approximation for helium (and possibly neon) to predict a breakdown threshold that is somewhat high (see Chapter IV).

Next we consider the effects of the second growing mode of diffusion, $n_2(r)$. It is, of course, orthogonal to $n_1(r)$ so that any growth of electron density which has taken place prior to the onset of n_2 cannot contribute to n_2 . (In cases where the pulsed breakdown threshold approaches the c.w. level, there will not even be a second mode.) Therefore we assume that n_2 is excited by the same even

distribution n_0 that was initially present. With the appropriate value of x_1 , equation (2-36a) gives the number of electrons contributing to n_2 a short time after its inception. Using the same notation as before we find $A_2^{(1)} \sim n_0/3$, while the first mode has grown somewhat from its initial value $A_1^{(1)} \sim n_0$. Thus, the second mode, which starts late, starts smaller, and has twice the diffusion loss, can generally be neglected. The same is true of higher-order modes.

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15. Note added in proof. The recombination coefficients quoted in Table 2-2 pertain to dissociative recombination. Whether this process exists at elevated temperatures, where the formation of molecular species is unlikely, is open to question. However, we use the dissociative recombination coefficients because of the short time involved and because they seem to be of the correct order. See D. R. Bates, ed., Atomic and Molecular Processes, Academic Press, New York (1962).

CHAPTER III

EXPERIMENTAL APPARATUS

III-1. Q-switched Laser and Amplifier

A laser can be described as an amplifying medium with reasonable optical quality placed in a regenerative cavity.^{1,2} In the case of a ruby laser the cavity is generally made of two plane reflectors external to a rod of length ℓ . The ruby laser is most commonly operated in the pulsed mode (a duration of a few milliseconds) or the higher-power Q-switched mode,³ although a continuous wave version has recently been developed.⁴

A Q-switched or giant pulse laser includes in the cavity a device which forestalls the onset of regenerative oscillation until the gain of the amplifying medium is great. Only then is the laser permitted to oscillate, either by decreasing the loss in the cavity or by swinging a mirror into position at the appropriate instant. The cavity is now well above the ordinary threshold for laser action and is therefore unstable. It emits a burst of energy in a short time.

The power output of a laser can conveniently be described by a rate equation.² Consider an amplifying medium containing N_0 "active" atoms or ions/cm³. If we call N the difference between the number of atoms in the excited state and in the ground state, then the amplification coefficient is $\alpha = \alpha_0 N/N_0$, where α_0 is the absorption

coefficient of the unexcited medium. The term $n = N/N_0$ is called the normalized population inversion.

To describe the Q-switching process, suppose a laser consists of a cavity with a total reflector at one end, reflectivity R at the other end. In addition to a laser rod of length ℓ , the cavity contains a slab of material whose transmission is T . We presume that T can be lowered abruptly or that R can effectively be increased by rotating the mirror.

The present work was performed using a "passive" Q-switch consisting of a reversibly bleachable dye solution^{5,6} (cryptocyanine in methyl alcohol). This operates in the following fashion. With a fixed mirror reflectivity R , a pulsed laser will oscillate when the net gain per circuit exceeds the losses. For a dye of transmission T ,

$$e^{2\alpha_0 n \ell} T^2 R \gtrsim 1 \quad (3-1)$$

describes the threshold for laser action. Suppose we adjust T and n so this equation is satisfied and the laser begins to oscillate. At this time the molecular transition causing the dye to be partly opaque may become saturated and the transmission approaches unity. The laser is now well above the new threshold and switching has been effected.

We can describe "fast" Q-switching quite simply.^{2,7} Suppose F is the number of photons in the cavity, L the cavity length, ℓ the rod length, and A the cross section. If γ is the fractional loss per pass and $N\alpha_0\ell$ is not too large, we can write

$$\frac{dF}{dt} = \frac{FN\alpha_0 l}{t_1} - \frac{F\gamma}{t_1}, \quad (3-2)$$

where t_1 is the cavity transit time. Neglecting pumping during the short pulse, the population inversion decreases at twice the rate at which F is amplified,

$$\frac{dN}{dt} = -2 \frac{FN\alpha_0 l}{t_1} \quad (3-3)$$

Making a change of variables,

$$\phi = \frac{F}{A l N_0}, \quad n = \frac{N}{A l N_0} \quad \text{and} \quad T = \frac{t_1}{\gamma}, \quad (3-4)$$

we obtain a set of two coupled differential equations,

$$\frac{d\phi}{dt} = \phi \left(\frac{\alpha_0 n l}{t_1} - \frac{1}{T} \right) \quad (3-5)$$

and
$$\frac{dn}{dt} = -2 \phi \frac{\alpha_0 n l}{t_1}. \quad (3-6)$$

At this point, we assume that Q-switching has been effected instantly at $t = 0$ and that $n(0) = n_i$ is somehow known. Then for all $t > 0$, γ is a constant and we can simplify (3-5) and (3-6) by changing to the time-scale of the laser,

$$\theta = \frac{t}{T}, \quad (3-7)$$

so
$$\frac{d\phi}{d\theta} = \phi \left(\frac{n}{n_p} - 1 \right) \quad (3-8)$$

and
$$\frac{dn}{d\theta} = -2 \phi \frac{n}{n_p}, \quad (3-9)$$

where $\alpha_0 \ell n_p = \gamma$.

n and ϕ are implicit functions of each other. Hence we can combine equations (3-8) and (3-9) by using a chain-rule for differentiating such functions,

$$\frac{d\phi}{dn} = \frac{n_p}{2n} - \frac{1}{2}. \quad (3-10)$$

Integrating,

$$\phi - \phi(0) = \frac{n_p}{2} \log \frac{n}{n_i} - \frac{1}{2} (n - n_i). \quad (3-11)$$

If γ_0 is the fraction of the cavity loss that goes into useful output, we can immediately write down the total energy and peak power outputs,

$$E = \frac{1}{2} (n_i - n_f) N_0 A \ell h \nu \frac{\gamma_0}{\gamma}, \quad (3-12)$$

$$P_m = \phi(n_p) N_0 A \ell h \nu \frac{\gamma_0}{t_i}. \quad (3-13)$$

Dividing E/P_m , we find the approximate pulse width,

$$\tau = \frac{n_i - n_f}{n_p \log n_p/n_i + (n_i - n_p)} T. \quad (3-14)$$

Using $n_p \sim \frac{1}{2} (n_i - n_f)$, $n_i \sim 3 n_p$, we expect pulse widths on the order of 5 T. Under these conditions, using $\gamma_0 = \log 1/R$, $R \sim 50\%$ and $L \sim 30$ cm., peak power will be on the order of hundreds or thousands of megawatts in 10 nsec pulses. Actually, since dye Q-switching is not ideal⁸ we can expect somewhat longer pulses with considerably less power.

Thus far, nothing has been said of the spectral or spatial distribution of the output. Owing to effective inhomogeneous broadening⁹ of the laser line in a standing wave cavity, one need not expect a Q-switched laser to operate in a single axial mode. However, Hercher¹⁰ has shown that by using a dye Q-switch and a special resonant reflector, it is possible to force the laser into one axial mode at some sacrifice in total energy output. It is also possible to operate a dye Q-switched ruby laser in a single transverse mode,¹¹ but this has been reported only at relatively low powers.

The dye Q-switched oscillator with a resonant reflector has other important properties. Because the dye always switches at the same population inversion, the peak power of single axial-mode pulses is repeatable from shot to shot, independent of small fluctuations in pumping power. In addition to this, the resonant reflector keeps the output wavelength reproducible to $\sim .005 \text{ \AA}$ with only nominal temperature control on the ruby.¹⁰

The major difficulty in using a high power laser system is damage to the components, especially to the ruby rod itself. We have found that an oscillator of the type described has an upper limit, using even the best available rods, of about 200 MW/cm^2 output power. (Inside a standing wave cavity, this corresponds to 2 or more times that power at the peaks of the radiation field.) At this power level, we begin to observe damage both on the surface and in the interior of the rod, particularly in the plane where the rod becomes obscured from the flashlamps by its holder. Occurring as it

does within the resonator, this damage soon increases the cavity losses, changes the laser's threshold and otherwise affects the output. The damage is cumulative, and the laser will not operate indefinitely.

A lower power ($\leq 50 \text{ MW/cm}^2$) oscillator synchronized with an amplifier alleviates some of these problems. First of all, an anti-reflection coated amplifier is a traveling-wave device. Consequently we do not require 400 or 500 MW/cm^2 inside the rod in order to get a useful output of 200 MW/cm^2 . This has the effect of increasing the damage threshold as a function of useful output.

Second, the slight damage that does occur in the amplifier does not radically affect the system's performance, since the losses are not inside the resonator. An amplifier also offers an easy way to make gross adjustments in the total output without substantially affecting other aspects of the system performance. For high power applications, small fluctuations in the amplifier pumping also do not affect the output power since the rod is often saturated either as a result of high output or high gain (see below).

The system we have finally devised is shown in Figure (3-1). The oscillator consists of a 9 cm. rod in a cavity with a Brewster angle roof prism at one end and a resonant reflector at the other. Q-switching is effected by a saturable dye solution which must be replaced every few days. The amplifier power supply is synchronized with the oscillator supply. Both rods are anti-reflected on the ends with a magnesium fluoride coating. They are cooled by the same water bath.

FIGURE 3-1
LASER OSCILLATOR-AMPLIFIER SYSTEM

(SCHEMATIC: WATER COOLING OF
LASER RODS AND FLASHLAMPS
NOT SHOWN)

The characteristics of a particular amplifier rod are shown in Figure (3-2). For low inputs, the amplification is $e^{n\alpha_0 l - \gamma}$, which can be related approximately to flashlamp voltage by fitting to two experimental points. γ is some loss term. At high pump powers, we can begin to see gain saturation, $n \rightarrow 1$, while for high input energies another form of saturation owing to depletion of the excited state population is evident.

All the rods used (especially in the oscillator) are of good optical quality, with typically only a few fringes across a 1 cm. aperture in a Twyman-Green interferometer. The rods are generally lightly doped (0.03 wt. % Cr^{+3}).

The laser produces 20 nsec pulses, repeatable to better than $\pm 5\%$ of peak power and $\pm .01 \text{ \AA}$ with a band width of $\sim 300 \text{ Mc}$. The beam divergence is approximately 1 mrad, which ensures high power densities at the focus of a lens. The system can be operated indefinitely at 150 MW/cm^2 output, although the unfocussed beam often damages glass prisms and filters in a few shots.

Figures (3-3) - (3-5) show some of the characteristics of the output beam. Figure (3-3) is a photograph of the focal spot of a 6 cm lens. Figure (3-4) shows the time development of the pulse monitored by a fast photodiode and oscilloscope with a time constant less than 1 nsec. The upper oscillogram was taken when the laser accidentally operated in two modes. Figure (3-5) is a Fabry-Perot spectrogram used to verify that the laser operated in a single axial mode whenever the oscillogram indicated a smooth pulse shape.

The need for a single axial mode is demonstrated rather

FIGURE 3-2. CHARACTERISTICS OF A PARTICULAR AMPLIFIER

2 milliradians

FIGURE 3-3. PHOTOGRAPHS OF FOCAL SPOT

FIGURE 3-4. LASER PULSE SHAPE. Lower trace shows single axial mode output.

Q-SWITCHED

PULSED

FREE SPECTRAL RANGE 0.12 cm^{-1}

FIGURE 3-5. SPECTROGRAM SHOWING OUTPUT IN SINGLE AXIAL MODE

clearly in Figure (3-4). In the upper trace, the laser output varies strongly as a function of time; it is nearly impossible to determine the power output accurately. In addition, nonlinear processes (or processes that are fast compared with the beat frequency of the two modes) will be strongly affected by the high instantaneous power produced by the mode beating. Unfortunately, when two or more modes are present, the detailed output of the laser is not repeatable from pulse to pulse, so this beating cannot effectively be utilized. It is therefore clear that the smooth pulse shown in the second trace is desirable for studying unknown or nonlinear processes. With this sort of output, a constant peak power can be obtained or at least monitored accurately. On the other hand, processes which are known to depend only on total energy will not require such strict mode control.

For accurate control of the irradiance from the laser, a set of cells containing copper sulfate solution was made. The cells were calibrated in a spectrophotometer in density steps of 0.05 at the laser wavelength. Their density was checked at the highest power level that would not damage the cells. No evidence of saturation was observed. The calibration of the cells seems to be even more precise than the repeatability of the laser system requires.

III-2. A Few Notes on High-vacuum Technology

To study the properties of gas breakdown at low pressures, a vacuum system was required. Commercial reagent grade rare gases have over 1 ppm hydrocarbon and low ionization potential impurities. Since

no attempt to purify them further was envisioned, the vacuum system to be designed did not have to meet very stringent requirements.

Our intention was to seal off the vacuum chamber and fill it with a gas sample whose pressure would always exceed, say, 10 torr. In this case, the partial pressure of impurities would be no less than 10^{-5} torr. It would be necessary, therefore, to build a vacuum chamber which could be evacuated to about 10^{-6} torr, and which would not rise above 10^{-5} to 10^{-4} torr after a few hours of being sealed off.

A vacuum system can be made effectively leak tight; the problem in separating a part of the system from its pumps is that the gasket materials and sealants adsorb gases from the atmosphere and contaminate the vacuum roughly in proportion to their exposed surface area.¹² This outgassing is a function of time, temperature and history and has been extensively reported.¹³⁻¹⁵ After careful consideration of most of the common gasket materials, it was concluded that Viton A would meet the requirements, provided it was not baked to over 150°C. Teflon is an excellent vacuum material¹⁷ and was used for insulation of electrical wires. Small amounts of epoxy did not seem to affect the observed outgassing rate after a bakeout at 150°C.

The system finally built consisted of a chamber (about 1 liter) pumped by a 2 inch (nominal) oil diffusion pump and a fast forepump to handle any accidentally large gas loads. To prevent streaming of pump oil, a chevron baffle was introduced between chamber and pump. A low vapor pressure silicone oil¹² was chosen

for the diffusion pump.

The chamber was also provided with a side-arm containing zeolite sorbent material. This could be baked at 300°C. (Cooling to 77°K resulted in pumping the rare gases at a prohibitive rate.) The sorbent very likely had the effect of purifying the samples somewhat.

The system was leak-tested and found to have a leak rate less than 10^{-9} torr-liter/sec. It had an ultimate pressure $\sim 10^{-6}$ torr when baked properly as indicated. When sealed off, the chamber filled up to a few microns in over 30 hours.

The system also contained two high-purity aluminum electrodes about 1 cm² in area and 1 cm apart. A glow discharge between the electrodes was used to provide a source of free electrons in the focal volume during some gas breakdown experiments.

The same electrodes were also used to determine the amount of charge liberated in the gas at powers below threshold for a visible discharge. In this case, it was necessary to place a negative "grid" between the input and outgo windows (lenses) to prevent electrons knocked out of the glass from making their way to the collecting electrodes. The grid was biased at 30-100V.

Figure (3-6) shows the arrangement inside the vacuum chamber. The lenses serve as windows and have about 6 cm focal length. The focal point in red light is located directly between the electrodes.

When the electrodes are to be used for charge collection the "grid bias" is adjusted so that no electrons can be observed in a hard vacuum. Most of the experiments in which charge collection was important were carried out at 150 torr. With the voltages and

FIGURE 3-6. LAYOUT FOR GAS BREAKDOWN EXPERIMENTS

pressures used, amplification by the gas was not a factor. Thus, any electrons collected must have been liberated directly from the gas by the laser radiation.

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CHAPTER IV

EXPERIMENTAL RESULTS

IV-1. Introduction

In this chapter we describe a number of experimental results that serve to verify the validity of the mechanism described in Chapter II. In the first section an experiment to determine the possible role of multiphoton ionization is presented. This is accomplished by providing an external source of electrons, which are found to have no effect on the breakdown threshold. On the basis of this finding, it is tentatively concluded that the initiating electron does indeed arise from an impurity molecule, as suggested in Chapter II.

In the second section, we verify that the spark growth can be diffusion limited and that it is appropriate to use the ambipolar diffusion constant and a temperature given by $1/3$ to $1/2$ the gas ionization potential to describe the diffusion process.

Next, we apply the theory of Chapter II quantitatively to some specific breakdown problems. First, we calculate breakdown threshold as a function of pressure for four rare gases and compare the results with experiment. The agreement is found to be good. Second, the number of charges liberated at irradiance levels below the breakdown threshold are determined experimentally and compared with theory. Here the agreement with theory is good in the cases of

helium and neon, but not argon. To explain the discrepancy of argon from theory, we reintroduce multiphoton ionization and find that it begins to become a significant effect in this gas at lower pressures.

Finally, we shall conclude that the basic mechanism described in Chapter II is generally correct with the initial electrons coming from impurities, although in argon and krypton one might expect the multiphoton effect to begin to provide initiating electrons at pressures of a few hundred torr or lower.

IV-2. Initial Charge Density

To determine the role of multiphoton ionization in the formation of a gas spark, an external source of electrons was provided by means of a glow discharge. The discharge ensured 100 to 1000 free electrons in the focal volume at the start of the pulse. The effect of the discharge on the threshold flux density helps answer an important question about the mechanism of breakdown.

There are two distinct possibilities, as discussed above. First of all, if the threshold is determined by the creation of a single electron, with the cascade forming rapidly, then the presence of an initial charge density should lower the threshold noticeably. On the other hand, if free electrons are liberated from impurities at the beginning of the laser pulse, and breakdown results from pumping sufficient energy into a growing cascade, the effect of a glow discharge should be minimal.

In our experiments the first conjecture turns out to be untenable. Experimentally, no difference in threshold could be seen

in either helium or argon, with an external source of ionization.

We conclude tentatively that the hypothesis advanced in Chapter II on theoretical grounds is essentially correct, and that the dynamics of the growth of the cascade do, in fact, govern the properties of the breakdown threshold.

It should be pointed out here that this conclusion can be reached because the effects of the glow discharge were not striking. Any moderate change in threshold (up or down) could have been attributed to the complexities of the glow discharge—increased ion temperature, lower density, etc. Consequently, the conclusion offered here is only tentative, but encourages us to attempt to verify the hypothesis in more detail.

IV-3. Diffusion Constant

The first experiment we shall discuss concerns verifying that the value of the diffusion constant, D , chosen in Chapter II-6, is reasonable. In that section, we estimated that the product of pressure and diffusion constant, Dp , appropriate to the growing cascade was related to the value, D_0p , at room temperature, T_0 , by

$$Dp = D_0p \left(\frac{kT}{kT_0} \right)^{3/2} \approx D_0p \left(\frac{\frac{1}{3}I}{\frac{3}{2}kT_0} \right)^{3/2}. \quad (4-1)$$

In air, $I \sim 15$ eV, so we can expect Dp to be ~ 5000 times the tabulated value of D_0p .

It is possible to determine the effective diffusion constant by measuring the breakdown threshold as a function of focal size.

Using equations (2-30) and (2-31) we get

$$\frac{P_b}{P_o'} = 1 + \frac{2\pi^2 D \tau}{P_o'} \cdot \frac{1}{a^2}, \quad (4-2)$$

where P_o' is the threshold for no diffusion (large focal volume),

$$P_o' = P_o + \Delta P_R.$$

The experiment can be carried out by determining the breakdown irradiance with lenses of varying focal length. With a beam divergence of 1 mrad, lens aberrations should play no role, so the spot size is proportional to the lens focal length.

Figure (4-1) shows the results of such an experiment, together with theoretical curves for D/D_o equal to 10^4 and 2×10^4 for comparison. These results agree with our estimate (5×10^3) to within a factor of 2 or 3.

IV-4. Threshold vs. Pressure

Using the apparatus described earlier, the breakdown thresholds for four rare gases were measured at pressures ranging from 600 torr to 10 torr, wherever possible. Figure (4-2) shows the results of these measurements. The laser emitted 20 nsec pulses, nearly triangular in shape. Using the 6 cm lens on the vacuum system gives a measured spot radius of 5.5×10^{-3} cm.

The measured points show a trend toward decreasing threshold with increasing pressure. Furthermore, the magnitude of the slope decreases noticeably from helium through neon and argon to krypton. (The scatter in the data is regular and reproducible. Although no

FIGURE 4-1. DEPENDENCE OF BREAKDOWN THRESHOLD ON FOCAL SPOT SIZE

FIGURE 4-2. BREAKDOWN THRESHOLD AS A FUNCTION OF PRESSURE

attempt to explain such scatter is made here, one would guess it is related to the number of inverse bremsstrahlung-free paths across the focal spot.)

Judging by slope alone, most of the data appears to fall in the right-hand region of Figure (2-4). Since recombination coefficients increase towards the heavier rare gases, the dependence of slope on atomic weight strongly suggests that recombination is a dominant loss.

This conjecture is verified by applying equation (2-30). We use the calculated ionization rates for helium and argon. D is given by equation (4-1) and α is found in Table(2-2). We can estimate ionization rates ν for neon and krypton by comparing their elastic collision cross sections at $\frac{1}{3} I$ with those of helium and argon, respectively. This is in agreement with the Holstein approximation for the inverse bremsstrahlung cross section.

The theory is shown as solid curves in Figure (4-2). The agreement is good in the case of the two heavier gases, where recombination and pulse duration are found to be the limiting mechanisms. As we anticipated in Chapter II, the theory predicts too high a threshold for helium and neon. Here diffusion is the primary loss mechanism and the observed threshold is relatively close to what we have called the c.w. threshold.

IV-5. Charge Collection below Threshold

Further to substantiate our theory, we present the results of some charge collection experiments at power levels somewhat below

the threshold for a visible spark. As indicated previously, charges were observed by biasing the two electrodes well below the breakdown potential and observing a voltage pulse across a resistor. The beginning of the pulse was verified simultaneous with the laser output, and the "grid" bias was adjusted so that no electrons could be found in a hard ($\sim 10^{-6}$ torr) vacuum.

Figure (4-3) shows the results of this experiment in the form of a graph. The number of charges collected is plotted as a function of peak power divided by breakdown threshold. All the measurements were made at 150 torr. The results have been checked at other pressures and found to be consistent.

Theoretical curves are shown as solid lines that break abruptly at 0.1% ionization. The theory is seen to fit the data well for helium and neon, and the experimental curves do, indeed, seem to break at 0.1% ionization. To the left of the break ($P/P_b < 1.0$) no spark is visible, while to the right of the break a spark is always seen. Right at threshold, helium does not become fully ionized as do the other gases, but this is easily ascribed to growing diffusion losses as a hot plasma is formed.

In the case of argon, no electrons could be found more than a few per cent. below threshold. Data had to be obtained by setting the laser carefully within a few per cent of threshold so that fluctuations in the output would cause the gas sometimes to break down, sometimes not. In this way it was verified that "pre-breakdown" electrons are present in argon as in helium and neon, although the theory does not properly explain the data. In the case of krypton,

FIGURE 4-3. RESULTS OF CHARGE COLLECTION EXPERIMENTS COMPARED WITH THEORY

no pre-breakdown electrons were found.

Attempts to redefine the breakdown criterion at some value below 0.1% ionization for argon and krypton by considering the effects of electron temperature are fruitless. (Recall that the cross section for absorption in the field of an ion¹ is proportional to $T^{-1/2}$.)

The large discrepancy of argon from theory can be ascribed to the multiphoton effect. In Chapter II, we calculated the flux required to create one electron by the end of the pulse by direct ionization of the gas atoms. The results of this calculation are shown in Figure (2-1). At 150 torr, a power of approximately 3×10^{10} watt/cm² is required. Recalling that $N = 9$ for argon, so that multiphoton ionization is a ninth-order process, we can immediately write

$$n(2\tau) = \left(\frac{P_m}{3 \times 10^{10}} \right)^9 \quad (4-3)$$

for a triangular pulse of half-width τ at 150 torr, neglecting subsequent impact ionization. Breakdown occurs at 3×10^{11} watt/cm at this pressure. Hence we would expect 10^9 electrons from this process alone, whereas for helium and neon multiphoton ionization contributes no electrons at the breakdown threshold.

We can better estimate the effect of multiphoton ionization by assuming all the electrons are born at the peak of the pulse, $t = \tau$. (Recall we are discussing a ninth-order process.) Then, neglecting loss, we have

$$n(2\tau) \sim \left[e^{\nu_m \tau/2} + \left(\frac{P_m}{3 \times 10^{10}} \right)^9 \right] e^{\nu_m \tau/2}, \quad (4-4)$$

when amplification is considered. The multiphoton effect will not become significant until the two terms in brackets are comparable; for argon at 150 torr, this occurs at just about the breakdown threshold, 3×10^{11} watt/cm². At this pressure, the calculated value of P_b will be affected negligibly by the correction in equation (4-4). Nevertheless, the expected number of electrons collected below threshold decreases somewhat faster because of (4-4) than one would expect. The curve for argon in Figure (4-3) takes this correction into account. While not perfect, it helps to explain the discrepancy of argon from the original prediction.

IV-6. Properties of Air Spark²

When breakdown is achieved in air, an intense blue-white spark is formed in the vicinity of the focal point of the lens. The spark appears to radiate uniformly in all directions. However, it appears quite red when viewed along the lens axis, looking back towards the laser. This suggests a large amount of forward-scattered laser light. The spark is also accompanied by a loud report which increases audibly with increasing power, as does the apparent volume of the spark.

A number of close-up photographs of various sparks were taken on color film. Examination of these photographs reveal several distinct regions as sketched in Figure (4-4). (Unfortunately this detail could not be reproduced in a black and white reproduction or photograph.) In the center of the spark, running along the lens axis, is an intense core orders of magnitude brighter than the rest of the

FIGURE 4-4. SPARK IN AIR

FIGURE 4-5. SIZE OF SPARK AS FUNCTION OF INCREASING
LASER ENERGY

spark. The region surrounding the core is generally divided into lobes which are a reproducible function of geometry and power alone. With increasing input power, the spark grows, mostly in the direction of the focussing lens as shown in Figure (4-5).

Figure (4-6) at the top shows a typical single mode laser pulse. At a well-defined point in the pulse the plasma begins to form and abruptly becomes opaque to the laser radiation. This is seen in the second oscillogram in Figure (4-6), in which the vertical scale is magnified 10 times. The final trace in that figure shows the light emitted by the plasma. This emission continues for 20 or 30 nsec after the end of the laser pulse.

To measure the angular distribution of light emitted by the plasma, it was first necessary to determine the angular distribution of scattered laser light. The laser was polarized in a horizontal plane and scattered intensity was observed in three orthogonal planes. The measurements were made with a silicon photodiode operating in an integrating mode. A $6942\text{\AA}$ interference filter with a $70\text{\AA}$ bandwidth (FWHM) was used to reject emission from the spark. The results of these measurements are shown in Figure (4-7).

In the vertical plane containing the polarization vector, Figure (4-7a), the scattered intensity falls off as $\cos^2\theta$. This is consistent with incoherent classical Thomson scattering from free electrons. In the other two planes, scattering is strongly peaked in the forward direction, with a smaller peak in the backward direction. Figure (4-7b) shows the scattering in the horizontal plane. The result is quite similar in the vertical plane containing the lens

FIGURE 4-6. (a) TYPICAL LASER PULSE; (b) PULSE ATTENUATED BY PLASMA FORMATION, 10x VERTICAL SCALE; (c) RADIANT EMISSION FROM SPARK

FIGURE 4-7. ANGULAR DISTRIBUTION OF LIGHT SCATTERED BY SPARK

axis, except for the addition of the $\cos^2\theta$ factor at 90° , where the scattering is relatively small.

The forward scattering profile is suggestive of relativistic Thomson scattering,³ although the effect could equally well result from reflection from the dense conducting plasma (see below).

The angular distribution of light emitted from the plasma itself was determined by repeating the measurements of scattered light without using the band-pass filter. The two sets of data could be subtracted, after allowing for the transmission of the filter, to obtain the radiance of the spark as a function of angle. As nearly as we can determine (± 10 or 20%) the radiance of the plasma is the same in all directions.

Most of the data described in this section was made with a laser capable of emitting 5 Mw in 30 nsec pulses (0.15 joule). Approximately 1 to 3% of this incident energy was converted into visible and near UV radiation. Using the higher power laser and the silicon photodiode, we have been able to determine the radiant output of the air sparks as a function of energy input. This is shown in Figure (4-8), for 20 nsec pulses focussed by a 3-cm lens.

We have also measured the acoustic energy using the 0.15 joule laser and found it to be on the order of only 5 ergs. This was accomplished by constructing a ballistic pendulum suspended by a quartz fiber and measuring the deflection of the pendulum by the shock wave emanating from the spark.

The (time-integrated) spectrum of the spark was obtained using a 1-meter spectrograph and a fast film. Linewidths on the order of

FIGURE 4-8. ENERGY RADIATED BY SPARK AS FUNCTION OF INPUT LASER ENERGY

$0.3\text{\AA}$ could be resolved with this apparatus. The data obtained is shown in Figure (4-9).

The spectrographic data was converted to absolute radiometric data using an iodine-cycle tungsten lamp that had been calibrated by the Bureau of Standards. A set of $D \log E$ curves for the combination film and spectrometer was obtained at various wavelengths. These were used as calibration curves, assuming reciprocity failure did not occur.⁴ The total radiant energy obtained in this way agreed with an estimate made with the silicon photodiode.

The spectrum consists of a strong line spectrum superimposed on a continuum whose peak is roughly at $4000\text{\AA}$. The lines can be identified primarily as NI and NII spectra, but they are considerably Stark broadened.⁵ Owing to the time averaged nature of the spectra, it would seem fruitless to make any comment about the temperature of the plasma.

One can make the following generalizations about the laser-induced plasma. Of the energy falling on the plasma after its formation has begun, over 90% is scattered or reflected away. A few per cent is absorbed and re-emitted as the broad-band radiation we have been discussing. A minute fraction of the incident energy goes into the initiation, acoustic energy, etc.

The plasma is an extremely intense light source, particularly in the vicinity of the line spectra. It should be useful itself as a source of light, especially in experiments where synchronization with the laser is important. One can estimate the peak power in any spectral interval from figures (4-6), (4-8) and (4-9). The peak of

FIGURE 4-9. SPECTRAL OUTPUT OF AIR SPARK

the emission from the plasma is coincident with the peak of the laser pulse.

A comment on the nature of the light scattered or reflected by the spark is in order here. We conjecture that the phenomenon is primarily due to ordinary reflection from the electron gas making up the core of the plasma. This mechanism accounts qualitatively for the forward scattering and we can show that it is approximately correct quantitatively in the following manner.

The complex refractive index $\tilde{n}$ of a free electron gas whose density is n_e is given by⁶

$$\tilde{n}^2 - 1 = \frac{4\pi n_e e^2}{m(-\omega^2 + i\omega/\gamma)} \quad (4-5)$$

where e , m and ω have their usual meanings and γ is a relaxation time generally taken to be the collision time of the electrons with the ions or atoms. In air at atmospheric pressure, $\omega > \gamma^{-1}$, so the complex index becomes

$$\tilde{n}^2 - 1 = - \frac{4\pi n_e e^2}{m\omega^2} \quad (4-6)$$

or

$$\tilde{n} - 1 = - \frac{1}{2} \frac{4\pi n_e e^2}{m\omega^2} \quad (4-7)$$

since the term on the right of (4-6) is small compared with unity.

Thus, the reflectivity of the plasma at normal incidence is

$$R = \left| \frac{\tilde{n} - 1}{\tilde{n} + 1} \right|^2 = \left(\frac{\pi n_e e^2}{m\omega^2} \right)^2. \quad (4-8)$$

R is on the order of 10^{-4} . We obtain an experimental value for R from Figure (4-7b), using the sensitivity of the photodiode at $6943\text{\AA}$ and including the solid angles subtended by the lens and detector at the location of the spark. The experimental value is also on the order of 10^{-4} , in good agreement with the result of equation (4-8). One is therefore inclined to regard the curious scattering profile of the incident beam as merely the result of reflection by the plasma.

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CHAPTER V

SUMMARY

V-1. Conclusions

In the preceding chapters, we have taken the several conjectures concerning the laser-induced breakdown process, put them in quantitative form, and compared the theory with experimental results. It is concluded that the formation of laser-induced gas sparks is governed by the growth of a cascade resulting from the heating of an initial charge density by the laser beam.

For helium and neon, the lighter gases with high ionization potentials, the growth of this cascade is limited primarily by diffusion out of the focal volume. The initial charge density is liberated early in the pulse by multiphoton ionization of impurity molecules initially present in the focal volume.

Diffusion loss is smaller in argon and krypton than in helium and neon. In these gases, it turns out that the growth of the cascade is limited by recombination, and that the breakdown threshold is governed largely by the pulse duration. At pressures under a few hundred torr, amplified multiphoton ionization begins to make its effect felt and the impurities play little part.

V-2. Suggestions for Further Work

Several thoughts for further work arise. First of all it would be interesting to build an ultrahigh vacuum system and purify the gas samples to the extent that an impurity molecule in the focal volume is unlikely. Then, at, say, 100 torr, we would expect the breakdown threshold for argon and krypton to be relatively unchanged, while that for helium and neon should increase dramatically. Such an experiment could be a striking demonstration of the role of impurities in initiating breakdown.

Second, it might be fruitful to extend the calculation of recombination-limited breakdown into the domain of high pressure gases and, especially, solids. This would be done either by numerically integrating the appropriate differential equation or by successive approximation to the integral equation. In this way, one would hope to gain insight into the nature of the damage mechanism in transparent solids. Certain features of the damage in solids are, in fact, quite similar to the features of gas breakdown—in particular the existence of a sharp threshold and the accompaniment of a visible discharge. Preliminary experimental results indicate little difference between, say, ruby and sapphire. All of these facts suggest that the theory that has been presented here will be applicable with appropriate modification in solids or liquids.