LASER INDUCED GAS BREAKDOWN

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AESTRACT

The lecture is divided in three sections. In the first section the ionization mechanism is discussed. Multiphoton ionization and electron cascade ionization theories are exposed and compared with the experimental results. A brief discussion on the origin of the first electrons is also included. In the second section the plasma expansion mechanism and the plasma properties are examined. In the last section a discussion of plasma decay and recombination is provided.
The advent of high power lasers has created a great interest in the phenomena occurring when electromagnetic radiation interacts with matter. They can be roughly divided into two categories. In the first one (generally referred to as nonlinear optics), one is interested in phenomena where matter does not go through any change of state; the important feature here is the nonlinear behaviour of polarizability. Examples are harmonic production, parametric interaction, stimulated scattering, etc... The second category includes those phenomena in which there is a change of state of matter, such as evaporation and ionization at the surface of solids and breakdown of transparent gases. In the latter phenomena, matter loses its individuality and a plasma is produced. Obviously, the most important parameter is the laser intensity (or its electric field). The electric field strength we shall be dealing with are only one or two orders of magnitude lower than the atomic field strength:

$$E_a = \frac{m^{1/2} I_0^{3/2}}{e\hbar}$$

where $I_o$ is the energy which keeps the electron in the atom, and $m$ and $e$ are the electron mass and charge. This corresponds to an intensity
\[ J_a = \frac{c}{4\pi} \frac{m I_o^3}{\alpha^2 h^2} \]

which, for a gas with \( I_o = 10 \text{ eV} \), gives \( J_a = 5 \times 10^{14} \text{ W/cm}^2 \), an intensity not out of reach for present lasers. Indeed, powers of the order of \( 10^{13} \text{ W} \) have already been reported for picosecond lasers. At these intensities, matter loses its individuality in a time of the order of \( 1/\omega \), becoming an ionized gas which then interacts with the electromagnetic field. However, it must be noted that in reality the ionization in the electromagnetic field starts to become important for intensities much lower than \( J_a \). This is explained in part because the ionization is obtained not only by direct ionization of the electrons out of the atomic system, but also by formation of an electron cascade in matter, whose threshold depends on intensity.

We are interested here in laser induced gas breakdown. It consists in the fact that at the very high electric field strengths which can be obtained by focusing a Q-switched laser beam, gases that are usually transparent to optical frequencies break down, i.e., they become highly ionized. This is accompanied by a light flash and a sharp sound, just as in ordinary gas discharges, from which the name "Spark" has been borrowed.

The phenomena was discovered very soon after the technique of Q-switching became available, as a byproduct of some experiments on harmonic production (Maker et al., 1964). Thereafter,
it became a subject on its own right, to which a considerable amount of research has been devoted.

Breakdown occurs only for incident electric fields higher than a rather well defined threshold, below which no macroscopic effects are seen. For most gases at atmospheric pressure and with laser pulses of nanosecond duration, the threshold intensity $I_{\text{th}}$ for breakdown is of the order of $10^{11} \text{W/cm}^2$, which explains why the phenomenon could only be observed after Q-switched, high power lasers became available.

2. IONIZATION MECHANISM

There are two ways in which a gas can be ionized in the field of an intense electromagnetic wave, such as that obtained when focusing laser beam: multiphoton ionization and formation of an electron cascade. While the first process is self-sufficient, the second requires the presence of at leasts one electron in the focal region at the time at which the laser pulse arrives. However, as pointed out by TOZER (1965) the equilibrium density of ions in air is typically less than $10^3 \text{cm}^{-3}$, and the rate of production by natural causes (such as, for example, cosmic radiation) is approximately 10 per second. Hence, the probability that a free electron occurs naturally in the focal region at the time of the laser pulse is negligible. Indeed, the presence of a free electron in a volume of $10^{-5}$-$10^{-8} \text{ cm}^3$ in a time of the order of $10^{-8} \text{ sec}$
requires the gas to be in the state of a tenuous gas discharge.

2.1. Multiphoton ionization

2.1.1. Theory

Due to the small energy of the laser photons, the usual photoelectric effect (i.e., one atom absorbing one photon and releasing one electron) is not possible. Indeed, for ruby laser radiation $h\nu = 1.78$ eV, whereas typical values for the ionization potential $I_o$ of rare gases are of the order of 10-20 eV. However, a multiphoton ionization process is possible in which an atomic electron is released as the result of the simultaneous absorption of $n$ photon, where $n = \langle I_o/h\nu + 1 \rangle$, the brackets indicating the integer part of the quantity. The excess energy, $nh\nu - I_o$, goes as kinetic energy to the released electron.

The multiphoton effect has a nonlinear nature, i.e., the probability of a transition involving many photons is not just the product of the probabilities of absorption of each photon individually. Indeed, this probability is so small that the effect was not detected before the advent of high power lasers. The probability of a multiphoton process involving the simultaneous absorption of $n$ photons in the presence of a field corresponding to a photon flux density $F$ is

$$w_n \sim F^n \sim E^{2n}$$
where $E$ is the electric field of the electromagnetic wave (or, to be more precise, its not mean square value).

Several authors have performed calculations of the probability of multiphoton ionization using time dependent perturbation theory. Consider a system consisting of an atom with associated orbital electron under the influence of an electromagnetic field. The Schrödinger equation for the system is:

$$i\hbar \frac{\partial \psi}{\partial t} = H \psi$$  \hspace{1cm} (2.2)

where $\psi$ is the wave function and $H$ the Hamiltonian of the system.

$$H = H_0 + V(t)$$  \hspace{1cm} (2.3)

$H_0$ is the time independent Hamiltonian of the unperturbed system, which is the sum of the Hamiltonian of the electron and the Hamiltonian of the electromagnetic field (this last one can be quantized). $V(t)$ is the Hamiltonian describing the interaction between the bound electron and the field, and it is different from zero only during the time $\Delta t$ of the perturbation. Generally one uses the electric dipole approximation for $V(t)$.

The unperturbed problem has a set of eigenfunctions

$$u_n(\vec{r},t) = \phi_n(\vec{r}) e^{-i \frac{E_n}{\hbar} t}$$  \hspace{1cm} (2.4)
corresponding to a set of eigenvalues $E_N$.

To solve equation 2.2., one begins by expanding $\psi$ in terms of the set $u_N$, eigenfunctions of $H_0$,

$$\psi = \sum_N a_N(t) \phi_N(t) e^{-i \frac{E_N}{\hbar} t}$$  \hspace{1cm} (2.5)

Before the application of the perturbation the system is in a stationary state $E_M$. After the interaction ($t > \Delta t$), the coefficients $a_N(t)$ take the constant values $a_{NM}(\Delta t)$, functions of the initial state.

Assume that the interaction energy is small compared to the energy of the unperturbed system (the usual assumption in order to be able to apply perturbation theory). Then one can expand each coefficient $a_{NM}$ in a series expansion of successive approximations

$$a_{NM} = a_{NM}^{(0)} + a_{NM}^{(1)} + \ldots$$  \hspace{1cm} (2.5)

The transition probability (per unit time) $W_{NM}$ for the system to move from an initial state $M$ to a final state $N$, under the influence of the perturbation is:

$$W_{NM} = \left| a_{NM}(\Delta t) \right|^2$$  \hspace{1cm} (2.6)
where \( \rho \) is the density of final states and \( |\tau_{NM}^{(n)}| \) a transition matrix element. The perturbation acts to the \( n \)-th order if 
\[
a_{NM}^{(0)} a_{NM}^{(1)} \ldots a_{NM}^{(n-1)} = 0, \quad a_{NM}^{(n)} \neq 0. 
\]
The transition from the initial state \( M \) to the final state \( N \) occurs via a series of transitions through \( (n-1) \) intermediate states called virtual states.

In order to determine \( W_n \) for multiphoton ionization involving the simultaneous absorption of \( n \) photons it is necessary to obtain all the matrix elements and to sum over all possible intermediate states belonging both to the discrete and the continuous spectra. The main difficulty in the calculations lies in this sum. The difference in the results calculated by the various authors mainly comes from the different approximations made in order to perform the sum.

GOLD and BEBB (1965) pointed out that the main contribution to the sum over the intermediate states is made generally by only one or two terms, namely those describing transitions to excited levels whose energy is very close to a multiple of the laser photon energy (quasi-resonant transitions). Their results are summarized in Table I, where the quasi-resonant transitions considered are included together with their order \( f \); the
strength is defined as the difference between the energy of
the level considered and the nearest multiple of the photon
energy (for ruby laser radiation).

CONTIER and TRAHIN (1958) have generalized to n-photon
ionization the perturbation theory for two photon ionization of
a hydrogen atom in the ground state of ZERNIK (1964). They de­
erived a hierarchy of inhomogeneous equations; the solution of the
first (which gives the two-photon results) is used to construct
the inhomogeneous part of the second (which gives the three-
photon results), and so on. Their calculations give rise to a
great importance of levels well removed from the quasi-resonant
levels. They presented numerical results for the ground state
of hydrogen and perturbation orders 2 through 8. Only for certain
frequencies their results agree with those obtained by GOLD and
BEBB (1965). Fig. 1 shows the variation of the quantity \( \frac{\sigma_n}{J^{n-1}} \)
with wavelength of the laser beam, where \( \sigma_n \) is the multiphoton
ionization cross section in cm\(^2\), and \( J \) the intensity in W/cm\(^2\).

However, HENNEBERGER (1968) has pointed out that in
all multiphoton ionization calculations the perturbation theory
is applied to intensities far beyond the limit of validity of
usual theory, and that the effect of the intense electromagnetic
wave on the initial state of the atom has been totally neglected.
His method consists of a transformation to an accelerated frame
of reference. It is shown that, in the nonrelativistic dipole
approximation, an effective intensity dependent ionization potential that binds the electrons can be found. His formulation is equivalent to the usual one at low intensities. However, his results show that, with the values of the electric field normally used, the ionization potential could drop by as much as half of its zero intensity value.

KELDYSH (1965) has developed a semi-classical theory in which the physical basis of the phenomenon is brought out very clearly. He has shown that the multiphoton effect and the tunnel effect (in which one electron is extracted from the atom by a static electric field) have a common nature, and are limiting cases of a single process in which one electron goes from a bound state in the atom into a free state under the influence of an alternating electromagnetic field. The formula for the probability of the tunnel effect is obtained in the limit of low frequencies, and the formula describing the multiphoton effect is obtained in the limit of high frequencies. The optical electric field can be considered static if it varies slightly during the time that the electron takes to cross the potential barrier. The width of this potential barrier is approximately $I_0 / E$; the electron velocity can be written $(I_0 / m)^{1/2}$. This defines a critical frequency $\omega_c = \omega / (2mI_0)^{4/2}$, separating the region of the tunnel effect from that of the multiphoton effect. The multiphoton effect will be dominant if $\gamma = \omega / \omega_c \gg 1$, since then the electron does not have the time to pass through
the potential barrier during one cycle of the alternating electric field. Note that, at 1.06 \mu (the wavelength of the neodymium: glass laser), \( \omega = \omega_c \) for \( E = 2 \times 10^8 \) V/cm. Since, as we shall see, experiments are performed with lower electric fields, the multiphoton effect will be dominant. The formula that describes the probability of a transition from a bound state to the virtual levels of the continuous spectrum can be written, in the multiphoton limit,

\[
W_n = B n^{3/2} \left( \frac{e^2 E^2}{8 m v^2 I_0} \right)^n
\]

where \( B \) is a constant, and \( n' = \langle \tilde{I}_0 / \hbar \nu + 1 \rangle \), with the effective ionization potential \( \tilde{I}_0 \) exceeding \( I_0 \) by a quantity equal to the average oscillation energy of the free electron in the field of the electromagnetic wave, \( \tilde{I}_0 = I_0 + e^2 E^2 / 16 m v^2 \).

Keldysh's calculations give approximately the same values as the Gold and Bebb formula; however, the frequency dependence is somewhat different.

Finally, it is necessary to consider some effects that might slightly change the above picture. The first one is the smearing out of the upper energy levels of an atom in a strong electromagnetic field (Voronov et al, 1966), which leads to their overlapping and merging into a quasi-continuous spectrum. An estimate shows that this effect can produce a decrease of the effective ionization potential by an amount of approximately 1 eV,
thus possibly lowering by one the number $n$ of photons required for multiphoton ionization.

A second effect, which again can lead to a decrease in the required number of photons, $n$, is due to the action of the field on the intermediate states of the atom (Voronov, 1967). This takes the form of a shift of the level energy as a result of Stark effect and a broadening due to the large probability of the transition from this level to the continuum. This effect can be quite important when an atom is ionized via the quasi-resonant intermediate excited states.

In addition, Pereolomov et al (1968) have pointed out that calculations on multiphoton ionization generally neglect the Coulomb interaction of the emitted electron with the atomic rest, and showed that this Coulomb correction is not small and results in a decreased value of $n$.

It must also be remembered that when a laser is operating in many modes the local fields in very small regions can greatly exceed the average value over the focusing volume; multiphoton ionization preferentially occurs in the places with larger fields (Bebb and Gold, 1966). Indeed, for a laser operating on a single mode we expect the flux density to be a well defined "number", and

$$\langle \Phi \rangle = \langle \Phi \rangle^\infty$$
where the brackets denote in this case the expectation value. However, if we consider a large number of modes, so that the fluctuations become similar to a thermal source (Gaussian light), we have

$$\langle F \rangle^n = n! \langle F \rangle^n$$

The radiation from a typical laser falls somewhere in between these two extremes. This means that when comparing theory and experiment, the value of the multiphoton probability has an uncertainty which can be as large as $n!$. However, the values of the exponent $n$ will be changed only if there are changes in the spatio-temporal structure of the laser beam with laser power (LAMBOPOULOS, 1972).

2.1.2. Experiment

At sufficiently low pressures, when the electron mean free path is much larger than the dimension of the focal volume, inverse bremsstrahlung is inhibited; in this condition ionization can only be due to multiphoton absorption. A large number of experiments has been performed in order to find the experimental value $n_{\text{exp}}$ of the number of photons necessary to ionize the atom. The experiments always consist in measuring the number of ions created $N_1$ as a function of the number of photons $F$ arriving at the focal region. Since the number of ions $N_1$ created by multiphoton ionization is proportional to the multiphoton ionization probability $\mathcal{W}_n$, one can write
where \( n_a \) is the atomic density, \( V \) the focal volume, and \( \Delta t \) the laser pulse duration. The physical meaning of \( V_n \) and \( \Delta t_n \) is tied to the fact that the contribution of the volume element \( dV \) to the number of ions created in a time \( dt \) is proportional to the relevant value of intensity raised to the power \( n \). Equation 2.8 shows that the slope of \( \log N_i \) versus \( \log F \) gives the number of photon \( n_{\text{exp}} \) necessary to ionize the atom. Fig. 2 shows a typical experimental plot for He atoms. Care must be taken at high intensity values, when all the atoms in the interaction volume are ionized; in this case saturation effects appear (i.e., the slope of the curve decreases), corresponding to the relation \( w_n \Delta t \sim 1 \). Equation 2.8 also allows one to calculate the multiphoton ionization probability if the interaction volume \( V \) and the pulse duration \( \Delta t \) are known. However, because of the nonuniform intensity distribution of the laser beams generally used, it is experimentally difficult to determine the exact value of \( V \). This results in values of \( W_n \) the precision of which is not very good.

Table II is a summary of the results obtained by various workers. It will be noted that \( \Delta n = 0 \) (i.e., \( n_{\text{exp}} = n \)) only when the field intensity is low, so that the perturbation effects discussed above are negligible.
The influence of a strong electric field on the upper energy levels of the atom has been experimentally shown by DELONE and DELONE (1968). They used the second harmonic of a neodymium:glass laser ($h\nu = 2.36$ eV) on Xenon and Krypton ($n=6.7$ for both atoms). In this experiment the photon energy is comparable with the distance from the first excited level to the boundary of the continuous spectrum. For Xenon the penultimate photon falls into the region close to the boundary of the continuum ($I_o - 5h\nu = 0.3$ eV), whereas for krypton it falls far from the boundary ($I_o - 5h\nu = 2.2$ eV), i.e., the effect of the overlapping of the upper energy levels should be decisive only for Xenon. Indeed, it is found experimentally that $\Delta n$(Xenon) = $1.6 \pm 0.2$, and $\Delta n$(krypton) = $0.5 \pm 0.5$.

DELONE and DELONE (1969) performed an experiment with the aim of demonstrating the importance of quasi-resonant levels of the atom. They used a neodymium : glass laser lasing in a 4 Å bandwidth and tunable from 10.590 Å to 10.680 Å, and potassium vapors ($n=4$). The energy of the 4f level of the potassium atom is close to the energy of three photons, and exactly coincides with it at $\lambda = 10.665$ Å. Fig. 3 shows their experimental results, and it is clearly seen that the dependence of the four photon ionization probability on radiation intensity changes considerably when the energy difference between 3h\nu and the 4f level of potassium decreases. The exponent $n$ at exact resonance is $n_{\text{exp}} = 1.5 \pm 0.1$, whereas far from resonance, at a detuning larger
than 20 Å, one finds \( n_{\text{exp}} = 4.0 \pm 0.1 \), i.e., \( \Delta n = 0 \). The importance of resonances has also been shown in an experiment by HELD et al. (1971). They used cesium and potassium vapors and a neodymium: glass laser. For both atoms \( n = 4 \), and low electric fields of the order of a few times \( 10^5 \) V/cm are sufficient. In the case of potassium it was found that \( \Delta n = 0.1 \pm 0.2 \). As already noted, the only possible resonance is between three photons and the potassium 4f level. However, in this case this difference is 211 cm\(^{-1}\) and remains essentially unchanged by the relatively weak electric field. On the other hand, for cesium one finds \( \Delta n = 0.8 \pm 0.2 \). In this case, the difference between the energy of three photons and the energy of the 6f level is only 9 cm\(^{-1}\), smaller than the bandwidth of the laser radiation (10 cm\(^{-1}\)), and cesium can be iodized via the resonant 6f level.

It is worth noting here that the shift of atomic levels under the influence of non-resonant laser radiation has already been experimentally observed for the mercury atom (PLATZ, 1971). It was found that the \( ^3S_1 \) level is shifted proportionally to the laser electric field at least up to \( 4 \times 10^8 \) W/cm\(^2\), the constant of proportionality being \( 0.28 \times 10^{-6} \) eV/MW cm\(^{-2}\).

In table II, the last two entries refer to experimental results obtained with molecules. It must be noted that in this case one has to deal with two different processes, ionization and dissociation. Ionization may then take place in two ways

a) multiphoton ionization of the molecule followed by dissociation
of the ionized molecule; b) multiphoton dissociation of the molecule followed by multiphoton ionization of the atom. For molecular hydrogen, the two main possibilities may be written

\[
\begin{align*}
&\text{a) } \text{H}_2 + 15.4 \text{ eV} \quad \rightarrow \quad \text{H}_2^+ + \text{e} \\
&\quad \text{H}_2 + 13 \text{ eV} \quad \rightarrow \quad \text{H}^+ + \text{H} \\
&\text{b) } \text{H}_2 + 4.5 \text{ eV} \quad \rightarrow \quad \text{H} + \text{H} \\
&\quad \text{H} + 13.6 \text{ eV} \quad \rightarrow \quad \text{H}^+ + \text{e}
\end{align*}
\]

The first experiments on molecular hydrogen were performed with both ruby and neodymium: glass lasers (Voronov et al., 1965; Berejetskaya et al., 1970). In both cases it was found that the ratio of the number of H\(^+\) ions to the number of H\(_2\)^+ ions is approximately \(10^{-3}\). Thus, it was concluded that H\(_2\) is ionized without dissociation, even though the dissociation energy is about 3 times smaller than the ionization energy. This conclusion is, however, in contrast with the results of Luvan et al. (1972), who studied the multiphoton ionization of molecular hydrogen at 1.06\(\mu\). Indeed, they found that the number of H\(^+\) and H\(_2\)^+ ions is comparable. The experimental results gave \(n_{\text{exp}}(\text{H}^+) = 10.8 \pm 1.0\), and \(n_{\text{exp}}(\text{H}_2^+) = 11.7 \pm 1.0\). From a careful examination of the energy levels, they concluded that the H\(^+\) ions result from an eleven photon transition from the ground state of the molecule to the \(v=12\) vibrational level of the \(B^1\Sigma_u^+\) state, followed by
dissociation of the molecule through an additional two-photon absorption process, and then a three photon ionization of the atomic hydrogen in the \( n=2 \) state. Since these last two absorption processes are saturated, the experimental slope is determined only by the first transition. A similar mechanism is responsible for the production of \( \text{H}_2^+ \).

To summarize, although in some cases experiment and theory agree (especially when the order of nonlinearity \( n \) is low), there are still large discrepancies. The theory for hydrogen atoms, for which the wave functions are well known, should be extended to include the effect of the shift and broadening of the energy levels under the influence of the electromagnetic field, and the Coulomb correction. This should provide very precise values of the multiphoton ionization probability. However, further improvement in the experiments is essential. The use of single longitudinal and transverse mode lasers is necessary to avoid coherence effects, and to allow a precise determination of \( V \) and \( \Delta t \). Atomic hydrogen will have to be used, in order to compare with the only theory which can be made, in principle, without approximations. It is also probable that measurements on line shift and broadening will be essential for the understanding of the influence of quasi-resonant transitions.

Finally, it must be noted that it is also possible to study multiphoton ionization by working at higher pressures (atmospheric) by using picosecond pulses. It is sufficient for
this that the duration of the pulse is shorter than the time needed for the development of an electron cascade (see later).

2.2. Origin of the first electron(s)

Besides performing calculations of multiphoton ionization probabilities, GOLD and BEBB (1965) also calculated the threshold for breakdown by multiphoton absorption. Breakdown is here defined as the photon flux density required to produce $10^{13}$ electrons in a focal volume of $10^{-8} \text{cm}^3$ during the duration of a 10 nsec ruby laser pulse, at an initial atomic density of $10^{20} \text{cm}^{-3}$. The results for the noble gases are summarized in Table III. Since the resulting threshold fluxes are at least two orders of magnitude higher than the values measured experimentally, BEBB and GOLD (1966) also calculated the photon flux necessary to produce one electron, starting with the same conditions. They found $F \approx 10^{29} \text{cm}^{-2} \text{sec}^{-1}$ for Ar, Xe, Kr, and $F \approx 5 \times 10^{30} \text{cm}^{-2} \text{sec}^{-1}$ for Ne and He. These values are in order of magnitude agreement with those observed experimentally for the breakdown thresholds. BEBB and GOLD concluded that although multiphoton ionization may provide the initial electron(s) necessary for an electron cascade to develop, it does not account entirely for the breakdown phenomena, except possibly at very low pressures where the formation of an electron cascade is not possible, due to the fact that the electron mean free path
exceeds in these conditions by some orders of magnitude the dimensions of the breakdown region. They also pointed out that multiphoton ionization predicts a very weak dependence of the threshold flux density on gas pressure, $F_{th} \propto p^{-1/n}$, which does not agree with the experimental results. However, this weak pressure dependence will greatly enhance the role of any low ionization trace impurity present in the gas in providing the initial electron(s). For example, a "hydrogenic model" impurity with an ionization potential of 7 eV (a reasonable value for many organic molecules), for which $n = 4$, will give an initiation flux of $F \gtrsim 10^{29} \text{cm}^{-2}\text{sec}^{-1}$ for a concentration of $10^{14}$ atoms cm$^{-3}$ (an impurity level of a few parts per million).

Multiphoton ionization of residual hydrocarbons in a high vacuum system was observed by CHIN (1970) and EVANS and THONEMANN (1972). The results showed that, even in a vacuum of $10^{-8}$ Torr, a few tens of electrons are created in the focal region for photon fluxes corresponding to the breakdown threshold ones, as a consequence of multiphoton ionization of the residual hydrocarbons. It is calculated that it is sufficient to have a concentration of 1 part in $10^{12}$ of heavy hydrocarbon vapors present in normal gases to provide the first electron(s) necessary for the cascade process.

As a consequence of what we have said above, there is evidence, both theoretical and experimental, that the first few
electrons might be provided by multiphoton ionization.

However, experiments on the breakdown of liquid helium and on breakdown by a CO$_2$ laser beam have pointed out another possible origin. In liquid helium (note that the breakdown threshold should not depend on temperature) it has been found that the breakdown threshold in He II (i.e., helium at a temperature inferior to the $\lambda$ point, $T = 2.2^\circ$K) is at least one order of magnitude higher than in He I (WINTERLING et al, 1969; ABRISKOSOVA and SKRYPNIK, 1971). This seems to be due to the fact that impurity particles sediment out much faster in He II than in He I, and would indicate that the first electron(s) is due to thermoionic or photoelectric emission from microcrystallites of materials which are suspended in the liquid.

Similar conclusions have been arrived at in the study of gas breakdown by a CO$_2$ laser beam. In this case $\lambda = 10.6\mu\mu$, and it must be assumed that multiphoton ionization is not possible ($n\neq 100$) for the intensities at which breakdown is observed. It has been reported (SMITH, 1971) that when dry nitrogen is flown through the focal region breakdown cannot be observed even at the maximum available intensity ($\sim 10^{10}$ W/cm$^2$; in laboratory air the threshold was of a few times $10^9$ W/cm$^2$). This result seems to indicate that the first electron(s) is generated by thermoionic or photoelectric emission from some contaminants present in the air (dust particles, possibly). This
has been clearly shown by HULL et al (1972). They focused a CO₂ laser beam in atmospheric air, and dropped particles into the focal region. Particles with dimensions ranging from 40 to 100 µ were observed to trigger break-down at power densities as low as 10⁸ W/cm². This is a factor of 20 lower than the threshold they found is normal laboratory air (no particles being dropped). However, when the breakdown chamber was evacuated and then filled with air filtered through a 250 Å Millipore filter, no breakdown was observed at intensities up to 10¹⁰ W/cm² for the first few laser shots. Successive laser shots would then produce breakdown, apparently due to particles knocked from the walls of the chamber. This hypothesis was confirmed by sampling the air within the chamber by means of a particle distribution analyser (using forward light scattering to detect particles down to a size of about 1 µm). The results show a clear correlation between breakdown threshold and particle density.

It must be pointed out that the presence of electrons in the gas at power levels well below the breakdown threshold value has been experimentally demonstrated. For example, CHALMETON (1969), using proportional counter techniques, has shown that the first electrons appear at a power of approximately 10 kW, almost two orders of magnitude less than the breakdown threshold.
It can be concluded that the experiments on liquid He and on breakdown with a CO$_2$ laser clearly show that in these conditions the first electron(s) comes from thermoionic or photoelectric emission from contaminants (e.g., dust particles) present in the gases studied. Although this is probably always the case, it cannot be excluded that multiphoton ionization of gas impurities (hydrocarbons) plays a role in gas breakdown at optical frequencies.

2.3. Cascade ionization

2.3.1. Theory

Calculations on the cascade ionization process have been carried out by many authors. It is assumed that a few electrons appear in the focal region at the beginning of the laser pulse, by one of the mechanisms discussed above. The free electrons then absorb light photons by collisions with neutral atoms, in a process that is generally called inverse bremsstrahlung. After acquiring an energy somewhat higher than the ionization potential, an electron may ionize an atom by impact, thus resulting in two electrons being available to start the process again. At sufficiently strong fields it is only necessary for the electron to excite the atom, which is then rapidly ionized by the absorption of a few photons. However, if the field is not strong enough to provide rapid ionization of the excited atoms, the energy lost by the electrons to excitation hinders the development of the cascade.
Apart from this, the main loss processes in the cascade ionization development are the energy loss to elastic collisions and the diffusion of electrons out of the focal volume. The energy loss to elastic collisions is higher the higher the gas and plays accordingly a relatively increasing role the slower the electrons acquire energy from the field, i.e., the weaker the field. The diffusion of electrons out of the focal volume is slower the higher the gas density and the larger the dimensions of the focal region. Note that at low pressures the electrons diffuse rapidly from the very small regions in which large local fields may exist due to the interaction between laser modes, and they become distributed over the entire focal volume so that in general they are subjected to the action of the fields averaged over the volume. On the other hand, at high pressures, diffusion is slow and the cascade develops predominantly in the places where the local fields exceed the average field.

In what follows we will limit ourselves to the case where the electron losses by diffusion outside the focal volume are negligible. These losses can be neglected if the following two conditions are satisfied: a) $l \ll a$, where $l$ is the mean free path of an electron between two collisions with the atoms, and $a$ is the linear dimension of the focal volume; b) $\Delta t \ll a^2/D$, where $D = \bar{V}_e / 3$ is the diffusion coefficient for free electrons and $\bar{V}_e$ is their average velocity. Experimentally one has $a \approx 3 \times 10^{-2}$ cm; thus, for gases at atmospheric pressure and ambient temperature
it is always $l \ll a$, since $l \leq 10^{-4}$ cm. The second condition, taking $\nu_e = 10^8$ cm/sec, is satisfied if $\Delta t \ll 10^{-7}$ sec.

Neglecting diffusion from the focal volume, the threshold field is determined by the condition that a sufficient number of electrons $N_{cr}$ appear during the time of the laser pulse $\Delta t$. At the beginning of the process the number of electrons in the cascade increases exponentially

$$N = N_o e^{t/\tau} = N_o \cdot 2^k$$

where $\tau$ is the cascade time constant. The number of generation of electrons $K$, produced by the end of the pulse, is

$$K_{cr} = \frac{\Delta t / \tau}{\ln 2} = 1.45 \ln \frac{N_{cr}}{N_o}$$

and depends weakly on $N_{cr}$ and the number of initial electrons $N_o$. If one assumes that the cascade starts with one electron ($N_o = 1$) and that the breakdown condition corresponds to $N_{cr} = 10^{13}$, then it is required that $K_{cr} = 43$, and, for a given pulse duration (for example, $\Delta t = 30$ nsec), also the cascade time constant $\tau$ is fixed ($\tau = 1$ nsec). The final number of electrons $N_{cr}$ is very sensitive to the value of the time constant $\tau$, thus explaining the existence of a sharp breakdown threshold.
We are now going to extend the classical microwave breakdown theory to optical frequencies. Classically, a free electron oscillates in the alternating field of the wave, and its mean energy, $e^2E^2/2m\omega^2$, remains constant. The electron acquires energy from the wave only when it collides with an atom and the abrupt change in velocity causes the oscillation energy to go into energy of random translational motion. The rate of growth of the electron energy $E$ is given by

$$\frac{dE}{dt} = \frac{e^2E^2}{m\omega^2} \nu_{\text{eff}} \frac{\omega^2}{\omega^2 + \nu_{\text{eff}}^2}$$

where $\nu_{\text{eff}} = n_a \overline{\nu} \sigma_{\text{tr}}$ is the effective collision frequency, $n_a$ the atomic density, $\overline{\nu}$ the average electron velocity in the energy interval $(0,1)$, and $\sigma_{\text{tr}}$ the transport cross section.

At optical frequencies, for not too high pressures, $\omega \gg \nu_{\text{eff}}$; thus

$$\frac{dE}{dt} = \frac{e^2E^2}{m\omega^2} \nu_{\text{eff}} \hbar \omega \frac{\hbar \omega}{e} \frac{J}{J_o}$$

where the characteristic intensity $J_o$ is given by

$$J_o = \frac{c\hbar^2\omega^4}{8\pi e^2 \overline{\nu}^2}$$

This treatment considers only a single photon bremsstrahlung process, thus neglecting multiphoton bremsstrahlung; this is justified only if one is dealing with radiation intensities such that $J \ll J_o$ (Bunkin and Fedorov, 1966). For ruby ($h\nu = 1.78$ eV)
and $\overline{v}_e = 10^8$ cm/sec, one obtains $J_o = 5 \times 10^{12}$ W/cm$^2$, which is considerably higher than the intensities generally used when dealing with laser pulses of nanosecond duration.

It must be noted that in the optical case one has $h\nu = 1-2\text{eV}$, whereas the oscillation energy is of the order of $10^{-2}\text{eV}$. Therefore, in most of the collisions the electrons do not acquire energy from the electromagnetic field, and only once in a large number they instantaneously acquire the energy $h\nu$ from the radiation. This effect has a quantum character, and therefore one would think that the classical theory is not applicable here. However, ZEL'DOVICH and RAIZER (1965) have performed calculations on the basis of quantum mechanical notions (assuming that there are no losses) and have shown that the quantum mechanical and the classical treatments lead to the same conclusions if the photon energy $h\nu$ is small compared to the total electron energy (not the energy of the oscillation in the field). Since the total electron energy is approximately equal to the ionization potential, this condition can be written $h\nu/I_o \ll 1$, and it is often verified in experiments.

If the electron energy losses to elastic collisions are taken into account, equation 2.12 becomes

$$\frac{de}{dt} = \left( \frac{e^2E^2}{m\omega^2} - \frac{2m}{M} \varepsilon \right) \nu_{eff}$$

2.14
where $M$ is the mass of the atom. This formula clearly shows the failure of the classical description. In the case of large losses $\frac{d\varepsilon}{dt} < 0$, and one would think that the electron energy can never reach the ionization potential. However, due to the quantum character of energy absorption, there is always a finite probability that an electron can jump through the energy loss band and produce ionization. The reason is that the electron motion along the energy axis is made up of random jumps of finite magnitude and has a stochastic character, which is described by the quantum mechanical model.

From equation 2.12 one can write the average threshold for breakdown (classically). The threshold condition is reached if during the laser pulse duration $\Delta t$ the energy gained by the electrons is equal to the ionization potential $I_0$ times the critical number of generations, i.e.,

$$k_c I_o = \int_0^{\Delta t} \frac{d\varepsilon}{dt} \, dt = \frac{d\varepsilon}{dt} \Delta t$$

from which

$$I_{th} = \frac{mc^2k_c\omega^3 I_o}{4\pi\varepsilon_0^2\sqrt{e} n_e \Delta t}$$

Equation 2.15 gives the dependence of the breakdown threshold intensity on the various parameters $\omega$, $I_o$, $n_e$, $\Delta t$. Although it has to be remembered that all energy loss processes
have been ignored in this treatment, it gives a good approximation for the order of magnitude of $J_{th}$.

In deriving equation 2.15 we have assumed that $\omega >> \nu_{ef}$; however, this is not true at very high pressures, where one has to use equation 2.11. Then $\frac{dc}{dt}$ has a maximum for $\omega = \nu_{ef}$, and the threshold intensity correspondingly has a minimum (which is also what happens at microwave frequencies, where the condition $\omega >> \nu_{ef}$ is not satisfied). Rough calculations show that for argon the minimum occurs at a pressure of approximately 200 atm (GILL and DOUGAL, 1965).

Let us now look at what happens when the pulse duration $\Delta t$ is decreased (BUNKIN and PROKHOROV, 1969). Obviously, at higher rates of the cascade process the influence of diffusion and radiative losses and recombination diminishes, and the idealized picture drawn above should work better. On the other hand, according to the threshold condition, equation 2.15, the threshold intensity for breakdown, $J_{th}$, should be increased so that the pulse energy remains constant. However, if the laser intensity is increased considerably, the arguments used in the derivation of equation 2.15 are not any more valid. Indeed, as already pointed out, the multiphoton bremmstrahlung absorption cross section can be neglected only if $J_{th} << J_0$, and hence the approximation made for the rate of energy transfer, equation
2.11, is valid only under this condition. However, by decreasing the pulse duration $\Delta t$ this condition can be violated, since according to equation 2.15

$$J_{th} = \frac{mcK_{cr} \omega^2 I_0}{4\pi e^2 v_{cr} n_d} \frac{1}{\Delta t} = J_0 \frac{4K_{cr} \bar{E} I_0}{(\bar{E} \omega)^2} \frac{1}{\Delta t} = J_0 \frac{\Delta t_0}{\Delta t} \tag{2.16}$$

and therefore $J_{th}$ exceeds the characteristic intensity $J_0$ if

$$\Delta t_0 = \frac{4K_{cr} \bar{E} I_0}{(\bar{E} \omega)^2} \geq \Delta t \tag{2.17}$$

For nitrogen at atmospheric pressure and a ruby laser, one finds $\Delta t_0 \approx 3 \times 10^{-10} \text{sec}$. Therefore, for laser pulse durations such that $\Delta t \leq \Delta t_0$, the role of multiphoton bremsstrahlung absorption cannot be neglected. Then the dependence of $J_{th}$ on $\Delta t$ becomes less strong than shown by equation 2.15, because the strong non-linear dependence of the rate of energy transfer $d\bar{E}/dt$ on the intensity $J$ must be taken into account. For $J > J_0$, the rate of energy transfer increases slower than that given by equation 2.11, and reaches a maximum (equal, in order of magnitude, to $\bar{v}_{eff} \bar{E}$) for $J/J_0 \approx (\bar{E}/\hbar \nu)^2$. After this point it decreases as $(J/J_0)^{-1/2}$. The existence of a maximum for $d\bar{E}/dt$ corresponds to the case in which the scattered electron has the maximum probability to emit (or absorb) simultaneously $n = \bar{E}/h\nu$ photons, i.e., almost all of its energy.
The existence of a maximum rate of energy transfer by electrons means that the rate of ionization by a cascade process has also a maximum, and the time interval which is necessary for the arrival of one generation of electrons has a minimum. Since for an avalanche breakdown it is necessary to obtain a well defined number of generations $K_{cr}$ (see equation 2.10), there exists a limit for the pulse duration, $\Delta t_{cr}$, under which it is not possible to have an avalanche breakdown. In order of magnitude, according to equation 2.16,

$$\Delta t_{cr} \approx \frac{4K_{cr}J_0}{\bar{E}v_{eff}}$$

2.18

and the corresponding threshold intensity is

$$J_{th}^{cr} = J_0 \left( \frac{\bar{E}}{K\bar{\nu}} \right)^2$$

2.19

For $\Delta t = \Delta t_{cr}$ the theoretical curve for $J_{th}(\Delta t)$, which takes into account multiphoton bremsstrahlung absorption, crosses the single photon theoretical curve, equation 2.15, and goes up with infinite derivative. For $\Delta t < \Delta t_{cr}$ breakdown can occur only as a result of multiphoton ionization, and in this region the threshold intensity is almost independent of pulse duration, since for multiphoton ionization one has $J_{th} \sim \Delta t^{-1/n}$. The theoretical dependence of $J_{th}$ on $\Delta t$ is shown in Fig. 4. Finally, equation 2.19 shows that the value of $\Delta t_{cr}$ is independent of frequency; for nitrogen at atmospheric pressure it is equal to $10^{-11}$ sec.
AFANAS'EV et al (1969) have calculated the cascade time constant $\tau$ in the case of high power and short pulses; they noted that while at low intensities (when multiphoton bremsstrahlung absorption can be neglected) $1/\tau$ is proportional to the radiation intensity $J$, at high intensity, provided that multiphoton ionization processes are still negligible it becomes a decreasing function of $J$, and the concentration of electrons at a fixed value of time decreases with increasing intensity.

This is physically connected with the fact that now the electron acquires, as a result of each elastic collision, on energy $\tilde{\omega} > I_0$, and falls rapidly into the region of energies where the probability of ionization and excitation by electron impact decreases with increasing energy.

In everything we have said until now loss mechanisms have been neglected. The approach generally used to include the effect of losses (PHELPS, 1966; YOUNG and HERCHER, 1967; MORGAN et al, 1971) is to write down the equation of continuity which governs the growth of electron density before the onset of a visible discharge

$$\frac{\partial n_e}{\partial t} = \nu n_e + D \nabla^2 n_e - R n_e^2$$

where $n_e$ is the electron density, $\nu$ the total ionization collision frequency, $D$ the diffusion coefficient, and $R$ the recombination coefficient. In this equation we have considered only losses due
to diffusion of electrons out of the focal volume and to recombination, thus neglecting other possible loss mechanisms (e.g., attachment); moreover, it is assumed that the first electron(s) appears at relatively low intensity.

By assuming a triangular laser pulse of duration $\Delta t$, replacing $\nabla^2$ by $-1/\Lambda^2$, and by defining the threshold as the intensity for which a degree of ionization $\delta$ is obtained, the solution of equation 2.20 gives the following expression for the breakdown threshold intensity.

$$J_{th} = \frac{1}{k_n a} \left[ \frac{1}{\Delta t} \log \delta n_a V + \frac{2D}{\Lambda^2} + \delta n_a R \left( \frac{n}{2\nu_0 \Delta t} \right)^{1/2} \right]$$

2.21

Here $n_a$ is the atomic density, $k$ the constant that relates $\psi$ and $J$, $\nu_0$ the breakdown ionization rate calculated neglecting recombination, $V$ the focal volume, and $\Lambda$ the characteristic diffusion length of the breakdown region, defined as

$$\frac{1}{\Lambda^2} = \left( \frac{4.9}{d} \right)^2 + \left( \frac{n}{L} \right)^2$$

2.22

$$d = f \theta \quad \text{and} \quad L = (n^2-4)f^2d/a$$

where $f$ is the lens focal length, $\theta$ the laser beam divergence, and $a$ the laser beam diameter.

As equation 2.21 shows, between the diffusion-limited and the recombination-limited cases there is an intermediate
region in which the duration $\Delta t$ of the laser pulse is the dominant factor in determining the breakdown threshold.

Figure 5 is a plot of equation 2.21, showing $\log J_{th}$ versus $\log p$ can be seen that the slope is $-2$ at low pressures (diffusion limited), $-1$ at intermediate pressures (pulse duration limited), and $-1/3$ at high pressures (recombination-limited). The pulse duration-limited region obviously corresponds to the regime we have discussed earlier in terms of classical theory. The difficulty in using this approach is essentially related to the poor knowledge of the constants included in equation 2.21, none of which is known for the particular conditions of laser induced gas breakdown.

2.3.2. Experiment

Experiments aiming at the understanding of the mechanism (or mechanisms) responsible for gas breakdown have been numerous and generally measure the dependence of the threshold electric field (or intensity) on various parameters. In all experiments the threshold value is determined either by observing the appearance of the bright flash in the focal region visually (or by some photographic method) or by observing the abrupt absorption in the laser pulse transmitted through the focal region.
2.3.2.1. External preionization

As it has already been seen, the probability of finding a free electron in the focal volume prior to the arrival of the laser pulse is negligible small. It is then generally thought that the initiating electron(s) is produces by the laser beam itself, through one of the processes discussed.

A few authors have studied the effect of an initial electron density on the development of the cascade. In particular, SMITH (1970) focused a neodymium:glass laser in a dc electrical discharge (atmospheric pressure argon), of electron density $10^{11}\text{cm}^{-3}$, and found that the breakdown threshold was reduced by approximately a factor of two, as compared with the threshold in neutral (atmospheric pressure) argon gas. As can be seen from equation 2.10, the $10^{11}\text{cm}^{-3}$ pre-ionization represents approximately one-half the ionization generations required for full ionization. The reduction of a factor of two in the threshold intensity is therefore in agreement with the theory of the cascade growth of the breakdown process.

2.3.2.2. Pressure dependence

The dependence of the breakdown threshold field on pressure has been studied by several authors. However, it is generally difficult to compare the results of different experiments, since the relevant experimental data (lens focal length,
laser beam divergence, etc.) are not always given. This fact reflects itself on the absolute values of the breakdown threshold but not on the pressure dependence. Typical results for some noble gases are shown in Fig. 6. They were obtained with ruby laser radiation. The data show that the pressure dependence is not compatible with multiphoton ionization (which predicts a very weak $p^{-1/2n}$ dependence for the threshold electric field), whereas it is in qualitative agreement with cascade theory; moreover, calculations show better than order of magnitude agreement for the value of the threshold field. As further evidence, results obtained at very high pressure (Fig. 7) show the minimum in the threshold field predicted by the classical theory.

Of course, comparison with the classical theory, which neglects all loss processes, can only give qualitative agreement. However, if the experimental results are compared with equation 2.21, good agreement is found (MORGAN et al., 1971). The data seem to indicate that for not too high pressures laser induced gas breakdown is pulse duration limited.

Measurements of the breakdown threshold pressure dependence with a CO$_2$ laser (HILL et al., 1972), at 10.6 $\mu$m, have again shown agreement with the cascade theory. Because of the longer wavelength the minimum now occurs at lower pressures.
2.3.2.3. Frequency dependence

Several experiments have been performed to study the frequency dependence of gas breakdown. BUSCHER et al (1965) used the first and second harmonics of ruby and neodymium lasers. Fig. 8 shows their experimental results. The cascade ionization theory predicts a threshold increasing with the square of the frequency; multiphoton ionization predicts a threshold increasing with decreasing frequency (the smaller the frequency, i.e., the photon energy, the greater the number \( n \) of photons necessary for multiphoton ionization). Both predict a monotonic behaviour and cannot therefore explain the experimental dependence. These results have later been confirmed by other authors (BARTHELEMY et al, 1968; ALCOCK et al, 1969). It can be noted that previous experiments (AKHRAMOV et al, 1965; HAUGHT et al, 1966, TOMLINSON et al, 1966), in which only two frequencies were used (the ruby and neodymium : glass laser ones), seemed to indicate qualitative agreement with the prediction of the cascade theory.

Finally, the breakdown intensity values, obtained with a CO\(_2\) laser at 10.6\(\mu\)m, are in agreement with the predictions of the cascade theory if they are compared with the results obtained at 1.06\(\mu\)m. Indeed, in this case the breakdown threshold intensity is reduced by approximately two orders of magnitude.
2.3.2.4. Energy loss dependence

The influence of diffusion losses on the breakdown threshold has been investigated by varying the focal lens of the focusing lens, i.e., the dimensions of the focal volume. The smaller the focal length, the smaller the dimensions of the focal volume, the greater the rate of diffusion of electrons out of it and the higher the threshold field. Fig. 9 shows the results of HAUGHT et al (1966), where the breakdown threshold electric field is plotted versus the characteristic diffusion length $\Lambda$. However, some doubts have been expressed on the diffusion like nature of the losses responsible for this behaviour. Indeed, as we have already noted, diffusion can be neglected if the mean free path $l$ of an electron between two collisions is smaller than the characteristic diffusion length $\Lambda$. In the case of figure 10, it is already $l \leq \Lambda$. These doubts have been reinforced by recent experiments with CO$_2$ lasers performed at larger focal diameters. Fig. 10 shows the results of BERGER and SMITH (1972), obtained by focusing a 70 nsec CO$_2$ laser pulse in a He : N$_2$ : CO$_2$ mixture at three different pressure. Initially, the threshold decreases with increasing diameter but for diameters larger than 0.2 cm the threshold seems to level off. The threshold behaviour for focal diameters less than 0.2 cm is quite general, and is the same for all gases studied (both atomic and molecular); it also appears to be independent of the laser radiation wavelength and gas pressure. In all cases the beam diameter dependence
indicates that the loss process controlling breakdown is reduced by increasing the dimensions of the focal volume, leading to a reduced threshold. This behaviour is compatible with electron diffusion out of the focal volume (the threshold intensity should then vary inversely with the square of the focal diameter). However, in the case of figure 10, the time required for an electron to diffuse out of the focal volume during the cascade time would require a beam size of less than $10^{-2}$ cm. Moreover, diffusion losses should decrease as the gas pressure increases. BERGER and SMITH (1972) concluded that the experimental evidence is against electron diffusion as the loss process. The loss process must be fast, faster than the electron diffusion, and must exhibit the same diameter dependence as diffusion. They proposed atomic excitation resonant-radiation-trapping processes as a possible explanation. Physically, this mechanism works as follows: an electron excites an atom and the excited atom loses the excitation energy by a radiative transition. In small beam diameters this excitation energy is lost to the breakdown. By increasing the diameter, a higher probability exists for resonant radiation trapping and therefore less energy loss, which in turn would lead to a lower breakdown threshold. This mechanism has also been used to explain the fact that the addition of a small quantity of neon to argon reduces the breakdown threshold of the mixture below that of pure argon (SMITH and HAUGHT, 1966).
Diffusion losses are, however, important at small focal diameters and low pressures. This has been shown by studying the influence of a magnetic field on the breakdown threshold. COHN et al. (1972) used a CO₂ laser to irradiate argon and helium gases in steady magnetic fields up to 87 kO. The direction of the magnetic field was parallel to that of the laser beam. Since its effect should be stronger at low pressures, they worked at a fixed laser intensity (\( \sim 10^{10} \) W/cm²) and measured the minimum pressure \( p_{th} \) at which breakdown could be obtained. Their results are shown in Fig. 11, and were explained in terms of a simple model of diffusion-controlled cascade breakdown. It is assumed that, to have breakdown, the small number of electrons initially produced must remain in the focal region long enough to acquire an amount of energy from the laser field equal to the ionization potential of the gas. For laser pulses of relatively long duration and pressures low enough for recombination to be negligible, this time is governed by diffusion. Application of the magnetic field will reduce the diffusion rate normal to the field, while axial diffusion will be unaffected. Since the focal region extends farther along the beam than normal to it, the breakdown threshold will be lowered until axial diffusion dominates. At this point the breakdown threshold becomes largely independent of magnetic field, as seen in the figure.

VARDZIGULA et al. (1967) observed a lowering of the breakdown threshold in atmospheric air when the spark was produced
in a 200 Ko e magnetic field parallel to the laser axis. However, when the magnetic field is perpendicular to the laser axis (CHAN et al., 1968) no effect can be detected. These results are consistent with the simple picture described above.

2.3.2.5. Effect of space and time variations of the laser beam

Many authors have pointed out that the average field in the breakdown region could be lower than the actual field in very small regions due to coherence effects when a multimode laser is used. Quantitative comparisons can be made only if lasers with a well defined spatial intensity distribution are used. It has been shown (ALCOCK et al., 1970) that the threshold values obtained with a single mode ruby laser are about 50% lower than those obtained when the same laser is made to lase over many modes.

It is also known that solid state multimode lasers have a complex time structure. In the extreme case in which the laser is mode-locked it has been shown that breakdown is due to a single picosecond pulse and not to a process integrating over the entire pulse train. (ALCOCK et al., 1968).
2.3.2.6. Picosecond pulse breakdown

The advent of mode-locked lasers, with their short duration pulses (of the order of $10^{-11}-10^{-12}$ sec.) has opened a completely new time domain to the study of laser induced gas breakdown. Isolation and amplification of a single pulse have permitted to study the region in which the pulse duration is too short for an electron cascade to develop. Since it is difficult to vary the pulse duration, the transition between cascade and multiphoton ionization has been obtained by working with a fixed pulse duration and varying the pressure. Fig. 12 shows that there is a critical pressure under which the pressure dependence is very weak, in agreement with the prediction of multiphoton ionization theory (for which $J_{th} \propto p^{-1/n}$). At pressures higher than this critical pressure, the pressure dependence agrees with the prediction of inverse proportionality given by the cascade ionization theory. Furthermore, if the breakdown threshold values are compared with those obtained when using nanosecond pulses, the $1/\Delta t$ dependence of equation 2.15 is found, although there is insufficient evidence to conclude that the breakdown process is governed by single photon bremsstrahlung absorption (note that now $J_{th} \gg J_0$) (ALCOCK and RICHARDSON, 1968; KRASYUK et al, 1970).

To conclude it is clear from the experiments that the appearance of the first electron(s) does not determine the
characteristics of breakdown, and that the process by which breakdown is reached is a cascade process. Unfortunately, different experiments are difficult to compare because of the generally poorly known laser characteristics, and this only permits qualitative comparison with the theory. Although the gross characteristics of breakdown agree with the cascade theory, a number of facts remains to be explained (see, for example, the frequency dependence).
3. EXPANSION MECHANISM

3.1. Theory

Let us assume that breakdown has taken place in the focal region. As soon as the ionization becomes appreciable, at temperatures of the order of several thousand degrees, the light is absorbed as a result of free-free transitions of the electrons in the field of the ions (inverse bremsstrahlung). The absorption coefficient, taking into account stimulated emission, is (SPITZER, 1962).

\[
k_{\text{L}} = \frac{4}{3} \left( \frac{2 \pi}{3} \right)^{1/2} \frac{n_e n_i Z^2 e^6}{(m \kappa T)^{3/2} c \nu^2}
\]

where the fact that \( h\nu < kT \) has been taken into account; \( n_e \) and \( n_i \) are the number of electrons and ions per cm\(^3\) and \( T \) is the temperature. The value of \( K_L \) for air at atmospheric pressure, for \( h\nu = 1.78 \text{ eV} \) (ruby laser), in the range of temperature \( 10^5 \text{--} 10^6 \text{ °K} \), is \( 10^2 \text{--} 10^3 \text{ cm}^{-1} \).

It is evident from equation 3.1 that the inverse bremsstrahlung absorption coefficient for light of a given frequency is a function only of the plasma temperature and density, being independent of the radiation intensity. However, when the kinetic energy imparted to a plasma electron by the oscillating electric field of the radiation is greater than the
mean thermal velocity the electron velocity distribution is far from Maxwellian. This will happen when

\[ J \gg \frac{3c\pi m kT v^2}{2a^2} \quad 3.2 \]

In these conditions equation 3.1 cannot be any more used. For strong fields and high temperatures (i.e., condition 3.2 plus the condition \( h\omega > kT \)) the absorption coefficient will be (RAND, 1964; HUGUES and NICHOLSON-FLORENCE, 1968; PERT, 1972)

\[
k_h = \frac{32}{2\pi} \frac{\pi^2}{\frac{2}{3}} \frac{n_e n_i Z^2 e^6}{c E^3} \ln \left[ \frac{16 e^2 E^2}{\nu^2 m kT v^2} \right] \quad 3.3.
\]

In a parallel beam of intensity \( J \) (W/cm\(^2\)) the power absorbed per unit volume in a thin sheet of plasma perpendicular to the direction of propagation of the beam will be \( KJ \) (W/cm\(^3\)).

When equation 3.1 can be used, i.e., for small intensities and high temperatures, the power absorbed per unit volume is

\[
k_L J = 4 \left( \frac{2\pi}{3} \right)^{\frac{1}{2}} \frac{n_e n_i Z^2 e^6}{c (mk)^{3/2}} J \frac{J}{\nu^2 - \frac{3}{2}} \quad 3.4
\]

and it is a linear function of the laser intensity. However, when condition 3.2 is satisfied, i.e., for large intensities and high temperatures, the power absorbed per unit volume will be

\[
k_H J = 4 \left( \frac{\pi}{2} \right)^{\frac{1}{2}} \frac{4}{\sqrt{3}} \frac{n_e n_i Z^2 e^6}{c (mk)^{3/2}} \ln \left[ \frac{64 e^2 J}{c^2 \nu m kT v^2} \right] \frac{\nu}{J^{3/2}} \quad 3.5
\]
and it is inversely proportional to the square root of the laser intensity. Figure 13 shows the power absorbed per unit volume of plasma (normalized to unit ion and electron densities, for ion charge Z=1) as a function of J at several temperatures for the frequency of the neodymium : glass laser. It is evident from the figure that the power absorbed by a plasma at a given temperature will have a maximum value, reached when the directed component of the electron velocity becomes comparable with the random component (this will occur at lower electron velocities and temperatures for lower frequencies). There will be then an optimum laser intensity for plasma heating, given by

\[ J_{\text{opt}} \propto \frac{\pi^2 c m k T \nu^2}{4 e^2} \]

For neodymium : glass laser and \( T=10^6 \text{K} \) one has \( J_{\text{opt}} \approx 3 \times 10^{14} \text{W/cm}^2 \).

Although equation 3.3 is the result of a single photon bremmstrahlung calculation, NICHOLSON-FLORENCE (1971) has shown that multiphoton bremmstrahlung changes only slightly this result. It will be also appreciated that everything we have said is valid in the case \( \omega \ll \omega_p \) (where \( \omega_p=(4\pi e^2 n_e/m)^{1/2} \) is the plasma frequency).

Under the conditions discussed the light is absorbed in a layer of the order of \( l = 1/K \) and heats the gas. It is well known that the plasma expands towards the focusing lens, opposite
to the light flux. Three different and independent mechanisms have been proposed to account for this expansion.

3.1.1. Radiation supported shock wave

Immediately after breakdown the absorption of energy in the gas at the focus of the laser beam causes intense heating and consequent rapid hydrodynamic expansion in the form of a spherical shock wave. The laser light is then preferentially absorbed in that part of the shock wave that is moving toward the laser. This hydrodynamic mechanism, first proposed by Ramsden and Savic (1964), is similar to detonations in reacting gases, and it is generally called the "radiation supported shock wave" or "detonation wave" mechanism.

To calculate the velocity of the detonation front, Ramsden and Savic (1964) used the Chapman-Jouget theory (Zel'dovich and Kompáneets, 1960), replacing the reaction energy by the energy per unit mass absorbed behind the shock front from the laser beam. If one assumes that the radiant energy absorbed is much larger than the ambient enthalpy of the gas, and uses ideal gas theory (which means assuming that the temperature is sufficiently high to ensure almost complete dissociation and ionization), the velocity of the detonation front is
\[ \nu = \left[ 2 \left( \gamma^2 - 1 \right) \frac{Wk}{\pi r^2 p_0} \right]^{1/3} \] 3.7

where \( \gamma \) is the adiabatic index, \( r \) the radius of the focusing area, and \( p_0 \) the initial gas density. This formula is valid for a flow in a constant area channel. Actually, this is not the real situation, since the flow expands to fill the solid angle subtended by the focusing lens at the breakdown point (see figure 14). We take this into account by writing

\[ r = x \tan \alpha \] 3.8

where \( \alpha \) is the half angle of the focused light beam, \( x \) the distance along the optical axis from the focus to the front, and it has been assumed that \( r_0 \alpha \approx x \tan \alpha \). Then, from equation 3.7 it follows that

\[ x^{2/3} \frac{dx}{dt} = \left[ \frac{2 \left( \gamma^2 - 1 \right) Wk}{\pi p_0 \tan^3 \alpha} \right]^{1/3} \] 3.9

RAMSDEN and SAVIC (1964) integrated this by assuming \( W = \text{const.} \). Then

\[ x = \left( \frac{2}{3} \right)^{3/5} \left[ \frac{2 \left( \gamma^2 - 1 \right) Wk}{\pi p_0 \tan^3 \alpha} \right]^{4/5} t^{3/5} \] 3.10

which predicts that a log-log plot of \( X \) versus \( t \) has a fixed slope of 3/5.
Actually, the assumption of a constant power $W$ is unrealistic. DAIBER and THOMPSON (1967) have integrated equation 3.9 assuming a Gaussian pulse shape. They found that in this case the slope of the log-log plot of $X$ versus $t$ depends on the time to breakdown $t_b$, explaining why the slope should change with gas, pressure, laser energy and lens focal length. For breakdown near the peak power it is found that the variation of $\ln X$ versus $\ln(t-t_b)/\Delta t$ is a straight line with slope 0.60, as predicted by equation 3.10. As breakdown occurs earlier in the pulse, the slope of the straight line, which would be fitted to the calculated points, begins to increase.

Until now the lateral expansion of the gas has been disregarded. The result of this is that equation 3.7 yields an overestimated value for the front velocity (RAIZER, 1965). If the width of the wave $\Delta x$ is comparable with the radius of the light channel $r$, a significant fraction of the released energy is transferred to the gas surrounding the channel during the time of energy release. The velocity of flow of the gas through the lateral surface of the light channel is of the order of the sound velocity $u$. Then, during the time of energy release $\Delta t = \Delta x/v$ the shock wave travels in the radial direction a distance $\Delta r = u\Delta t = u\Delta x/v$, and consequently the released energy goes into the area $\pi (r+\Delta r)^2$ instead of $\pi r^2$. Then, in place of $W$ in equation 3.7 one has to write $W\delta$, where

$$\delta = \frac{\pi r^2}{\pi (r+\Delta r)^2} = \left(1 + \frac{2\Delta x u}{rv}\right)^{-1}$$

3.11
if $\Delta r \ll r$. In the detonation regime $u = v/2$, and therefore $\delta = (1 + \frac{\alpha u}{r})^{-1}$.

Heating in the detonation regime has the maximum possible value, and the specific internal energy that the gas acquires is

$$e = \frac{T}{(\gamma^2 - 1)(\gamma + 1)} v^2$$  \hspace{1cm} 3.12

As a numerical example, consider $J = 2 \times 10^{11} \text{ W/cm}^2$, $\rho_0 = 1.3 \times 10^{-3} \text{ g/cm}^3$, $\gamma = 1.33$ (air at atmospheric pressure). The values uncorrected for the lateral expansion are $v = 1.22 \times 10^7 \text{ cm/sec}$, $\varepsilon = 1.35 \times 10^7 \text{ J/g}$, corresponding at equilibrium to $T = 9.1 \times 10^5 \text{ K}$. The corrected values are, for $\delta = 0.5$, $v = 1.05 \times 10^7 \text{ cm/sec}$, $\varepsilon = 8.5 \times 10^6 \text{ J/g}$ and $T = 7.2 \times 10^5 \text{ K}$.

KEY (1969) noted that the detonation wave theory as developed above neglects the energy used to produce ionization in the gas during the expansion. He modified the conservation equation to introduce explicitly the ionization term. The result of this is a cubic equation relating the instantaneous front velocity to the absorbed laser intensity; this equation reduces to equation 3.7 if ionization is neglected. The net effect of the additional ionization term is to give a lower detonation wave velocity for a given absorbed intensity, and also a lower temperature. Physically, this happens because some of the energy has been used to produce ions and it is not available as internal kinetic energy. The difference is particularly important for the temperature;
in helium, two times ionized, at an initial pressure of 4 atm, this
theeory predicts a temperature three times as low as that deduced
when neglecting ionization.

The rate of energy exchange between electrons and ions
for the conditions discussed is of the order of $3 \times 10^{-10}$ sec. This
gives, for $v=10^7$ cm/sec, an exchange length of $3 \times 10^{-3}$ cm,
comparable with the mean free path. Thus the electron and
ion temperatures cannot be greatly different. Note that the
energy absorbed by the electrons is transferred to the ions not
only by energy exchange during collisions between them, but
also by hydrodynamic means, via the work performed by the electron
pressure forces on the ions, since electrons and ions are
rigidly coupled through the Coulomb interaction.

From the lateral expansion velocity $v_L$ one can
calculate the specific plasma energy, and from this the ion
temperature $T_i$ (RAIZER, 1965). For air at normal density, in
the temperature range $5 \times 10^5$-$10^6$ K, one has

$$ T_i = 1.25 \times 10^2 v_L^{8/4} \quad (°K) \quad 3.13 $$

It has to be stressed out that the high laser intensities
normally used in experiments are only necessary to produce the
initial gas breakdown, but are actually not needed to maintain
the radiation supported shock wave. Indeed, RAIZER (1970) has
shown that the wave can propagate along a parallel light beam in the direction opposite to the beam for laser intensities that are several orders of magnitude lower than the breakdown threshold values. The lower limit for the light intensity that admits the self-maintaining regime of the plasma front propagation is determined by the energy losses due to lateral gas expansion, which become significant at a wave width equal to or greater than the beam radius. Calculations show that the limiting intensity in atmospheric air and for a beam radius of 0.1 cm is approximately $10^8$ W/cm$^2$, corresponding to a limiting wave velocity of about $10^6$ cm/sec.

3.1.2. Breakdown wave

The spark expansion may, under certain conditions, simply indicate a smooth change with time in the point at which the time integrated effect of the laser beam upon the gas has been sufficient to break it down. This is generally called the "breakdown wave" mechanism.

RAIZER(1965) has calculated the expansion velocity for this mechanism in the following way. Under the action of the laser pulse an electron cascade develops in the cold gas. The electron density grows accordingly to

$$\frac{dn_e}{dt} = \frac{n_e}{\tau}$$
and thus

$$n_e = n_{e_0} \exp \left[ \int_0^t \frac{dt}{\tau} \right]$$ \hspace{1cm} (3.14)

where $\tau$ is the time necessary for the electrons to acquire the energy needed for ionization of the atoms, and $n_{e_0}$ is the initial electron density. For large light intensities, but in the range where multiphoton bremsstrahlung absorption can be neglected, $1/\tau = AJ$, where $A$ is a constant. If $n_{e_c}$ is the critical value of the electron density for which breakdown occurs, the moment $t$ of onset of breakdown in the section $X$ of the light channel is determined by

$$\int_0^t \frac{dt}{\tau} = A \int_0^t J(x,t) dt = \ln \frac{n_{e_c}}{n_{e_0}} = B$$ \hspace{1cm} (3.15)

where it can be assumed that $B$ is a constant, since it depends only logarithmically on the ratio $n_{e_c}/n_{e_0}$. For a triangular laser pulse, on the rising portion only, one can write

$$J(x,t) = \frac{W_0}{\pi r^2} \frac{t}{\Delta t}$$ \hspace{1cm} (3.16)

If $t_b$ is the time at which breakdown first occurs at the focus, then

$$A \int_0^{t_b} J(0,t) dt = B$$ \hspace{1cm} (3.17)
From equations 3.15 and 3.17, by using equation 3.16 and putting

\[ r = r_0 + x \tan \alpha \]

(see figure 14), one has

\[
\begin{align*}
\chi &= \frac{r_0}{t \tan \alpha} \frac{t - t_b}{t_b} \\
\nu &= \frac{d\chi}{dt} = \frac{r_0}{t_b \tan \alpha}
\end{align*}
\]

from which it follows that \( \nu \) is a constant during the rising portion of the laser pulse. An estimate of the dependence of the velocity \( \nu \) on peak power \( W_0 \) and pulse duration \( \Delta t \) can be obtained by deriving \( r_b \) from equation 3.17,

\[ t_b \sim \Delta t^{4/2} W_0^{-1/2} r_0 \]

from which, substituting in equation 3.18,

\[ \nu \sim \frac{(W_0/\Delta t)^{4/2}}{t \tan \alpha} \]

This equation shows that the breakdown wave velocity is higher for short powerful pulses and with long focal length lenses.
Actually, this derivation assumes that electrons are present throughout the gas at the time at which the laser pulse arrives, an assumption which has already been shown to be unrealistic. ALCOCK et al. (1968) assumed that electrons are produced ahead of the breakdown wave (for example, by precursor radiation from the spark itself). If electrons appear at point $x$ at time $t_1$, and the wave arrives at time $t_2$, equation 3.15 must be written

$$\int_{t_1}^{t_2} \frac{dt}{t^2} = A \int_{t_1}^{t_2} J(x,t) dt = B$$

3.21

and following the same calculations one obtains in place of equation 3.18

$$r^2 \tan \alpha = \frac{r_0^2}{t_b} (2t_2 - t_b)$$

3.22

where the assumption $t_2 - t_1 = \text{const} = t_b$ has been made.

AFANAS'EV et al (1969) studied theoretically the interaction of powerful ultrashort laser pulses with a gas. They concluded that with picosecond duration pulses, provided that multiphoton ionization can still be neglected, the cascade ionization mechanism is such that the electron concentration in gases at normal pressure is smaller by several orders of magnitude than the concentration of neutral atoms. As the result of this low electron density the produced plasma is practically transparent to the incident radiation, and the only possible mechanism
for the laser spark development is the breakdown wave. Hence, the breakdown region should propagate in the direction of the laser beam at a velocity equal to the velocity of the light pulse, and the length of the spark is determined by the divergence of the beam.

3.1.3. Radiation transport wave

The gas in front of the absorbing layer becomes ionized due to the absorption of thermal radiation from the strongly heated region of the gas and consequently acquires the ability to absorb the laser radiation. This is generally called the "radiation transport wave" mechanism.

The calculations for this model are rather more complicated than those for the two former ones; they have been carried out by RAIZER (1965), and here only the results are reported. At temperatures of the order of $10^5$ K or more, the mean free path of photons with energy $h\nu > kT$, radiated by the heated gas, is $l = 10^{-1}$-10 cm, and so it is much larger than $l = 1/K$ and every characteristic dimension of the heated region. The heated gas is hence transparent to its own radiation and emits from its entire volume. This radiation is absorbed in the colder layers, where the ionization is small; the corresponding mean free path in atmospheric air is of the order of $10^{-2}$-10$^{-1}$ cm (for $h\nu \approx 20-200$ eV).
As soon as the ionization due to absorption of thermal radiation reaches a sufficient value the new ionized layer begins to absorb the laser light. The intensity of the focused laser radiation is much larger than the flux of thermal radiation, so that the new layer becomes rapidly heated and the boundary of the high temperature region moves towards it. To calculate the expansion velocity of this boundary RAIZER (1965) considered the stationary conditions in a coordinate system in which the wave is at rest, as done in the theory of shock wave structure. Approximate calculations show that, both in order of magnitude and in dependence on laser power, the velocity of the radiation transport wave mechanism is close to that due to the detonation mechanism.

3.1.4 Other mechanisms

EVANS and GREY-MORGAN (1969) pointed out another explanation for the development of the plasma and the existence of the discrete breakdown spots experimentally observed. They proposed that this is due to the effect of primary spherical aberrations caused by the simple lenses generally used to focus the laser radiation. The intensity distribution of the diffraction pattern in the neighborhood of the focus is computed numerically from the HUYGENS-FRESNEL-KIRCHHOFF equation. The results show the existence of discrete regions of high intensity maxima lying along the optical axis; separations of approximately 0.3 mm for a 5 cm
focal length lens were calculated. Discrete breakdown plasmas are then created one after the other at intervals of a few nanoseconds and can give the appearance of a plasma moving towards the lens with a velocity of propagation of approximately $10^7$ cm/sec. This apparent velocity of propagation depends very weakly on the lens focal length and laser beam radius.

HORA (1969) considered the possibility of self-focusing of the laser beam occurring in the plasma (after breakdown has already occurred). The ponderomotive forces (due to the gradient of the light intensity) accelerate the plasma in the radial direction of the beam, producing a lower density in the central region and causing self-focusing of the following radiation. The minimum necessary laser power is evaluated from conditions of diffraction and total reflection, using the condition of equilibrium between the ponderomotive and hydrodynamic forces. The time characteristic for this process is of the order of $10^{-8}$ sec. As in the case of self-focusing in liquids and solids, it is found that for the phenomenon to occur the power has to be higher than a lower limit, independent of beam diameter. This minimum necessary laser power is of the order of 1 MW, and the smallest possible beam diameter due to self-focusing is of the order of a few microns. However, it must be noted that this is the power necessary to sustain a self-trapped filament, and that it corresponds to a self-focal length of the order of one meter, far too long to be relevant to laser induced gas breakdown. PALMER (1971) has calculated the threshold conditions for self-focusing
and stimulated Thomson scattering due to ponderomotive forces and thermal deposition using a linearized theory analogous to the one employed for liquids. Figure 15 shows the order of magnitude range of ruby laser intensity and plasma density within the limits of the theory which permit a net positive gain for self-focusing and stimulated scattering in a 10 eV, fully ionized, hydrogen plasma. Note that in the figure are also included the limits on laser power imposed by saturation of the nonlinear processes. The plasma density is limited to a range allowing simultaneous validity of the hydrodynamic theory and of the slowly varying approximation used in the linearised theory. For the conditions of gas breakdown the ponderomotive self-focusing threshold is of the order of $10^{13} \text{ W/cm}^2$.

3.2 Experiment

3.2.1. Spark expansion

Very soon after the discovery of gas breakdown, it was found that, after breakdown, the spark does not expand symmetrically, but moves towards the focusing lens, against the laser beam. This was initially discovered (RAMSDEN and DAVIES, 1964) because the laser light scattered at 90° by the plasma is shifted from the incident laser wavelength towards the blue by up to 3 Å. This displacement was interpreted (and then confirmed by streak photography) as a Doppler due to the movement of the plasma towards
the laser with a velocity of approximately $10^7$ cm/sec.

After this, a fair amount of work has been devoted to the understanding of the expansion mechanism. The experiments consist in taking time resolved photography of the plasma luminosity, or time resolved Schlieren photography, and plotting the plasma displacement versus time. Most of the results obtained in this way agree with the detonation wave theory. In particular, log-log plots of displacement versus time (RAMSDEN and DAVIES, 1964; DABBER and THOMPSON, 1967), during the time in which the laser pulse is acting on the spark, are in agreement with the detonation wave theory, as developed above. Moreover, the Doppler shift magnitude varies with the incident laser intensity as $I^{1/3}$, in agreement with equation 3.7 (MANDEL'SHTAM et al 1966).

A few authors have interpreted their results as evidence for the breakdown wave theory. Noteworthy among these is the work of ALCOCK et al (1968), in which picosecond time resolved Schlieren photography gave excellent agreement with the breakdown wave theory, modified to take into account the effect of preionization ahead of the wave.

As a conclusion, it can be said that most of the experiments are satisfactorily explained by the detonation wave theory; there is, however, also evidence pointing to the other mechanisms.
Here again there is a considerable difficulty in comparing data, due to the different (and not always clear) experimental conditions. Moreover, there is evidence that at least in some cases different mechanisms operate simultaneously.

As already pointed out, the high intensities used in experiments are necessary not so much to maintain the spark itself as to break down the gas. If the ignition is provided by an external plasma source, the propagation of the laser spark in the detonation regime can occur at intensities a few orders of magnitude lower than those necessary for gas breakdown. This has been experimentally demonstrated by MUL'CHENKO et al (1970) by breaking down argon gas with a ruby laser and maintaining the plasma propagation by another ruby laser operating in a quasi-continuous mode for 1.5 ms. The threshold intensity necessary to maintain the plasma was $4 \times 10^7 \text{ W/cm}^2$ at a pressure of 20 atm, whereas the breakdown threshold in these conditions is of the order of $2 \times 10^{10} \text{ W/cm}^2$. After the gas has been broken down, the plasma front propagates along the quasi-continuous laser beam, opposite to it. The front covers a distance an order of magnitude larger than the focal dimension, and stops in the cross-section of the light cone where the intensity is not strong enough. At the end of the laser pulse the front recedes. Recently, by using a cw CO$_2$ laser (with a power of a few hundred watts) to maintain the plasma, continuous plasmas have been obtained in several gases,
3.2.2. Density measurements

The density of sparks is measured by using laser interferometry. In these experiments, classical interferometry (generally Mach-Zender) or holographic interferometry is used to time resolve the plasma density (Alcock et al., 1966; Alcock and Ramsden, 1966). The most accurate measurements have been performed at two wavelengths (for example, the first and second harmonics of a ruby laser) in order to separate the contribution of neutrals to the fringe shift. Unfortunately, these experiments present the difficulty that interferograms obtained during the first tens of nanoseconds after breakdown are difficult to interpret. Fig. 16 shows the experimental results of Alcock and Ramsden (1966). There is general agreement that the initial electron density of sparks in atmospheric air is of the order of $2 \times 10^{19}$ cm$^{-3}$. This value of $n_e$ is consistent with the assumption that complete ionization occurs within the focal volume during the initial phase of spark.

3.2.3. Temperature measurements

The electron temperature is obtained by measuring the intensity of the soft x-ray radiation around $\lambda \approx 10$ Å, by a foil absorber method. The ratio of the intensities at two different
wavelengths yields the maximum value of $T_e$ (ALCOCK et al., 1966; DAIBER and THOMPSON, 1970; NEUSSER et al., 1971; AHMAD and KEY, 1972). Note that this method has no temporal or spatial resolution. Electron temperatures varying between a few tens of eV and 100 eV, depending on the experimental conditions, have been measured. These values are in order of magnitude agreement with those that can be derived from equation 3.12, knowing the spark expansion velocity and assuming equilibrium.

As already pointed out, the ion temperature cannot be very different from the electron temperature, because of the short equipartition time. Although direct measurements of $T_i$ have not been performed, the values obtained from the measured lateral expansion velocity, by means of equation 3.13, agree with the assumption that $T_i = T_e$.

It should be pointed out that in gas breakdown produced by conventional nanosecond laser pulses it is difficult to increase the temperature, since increasing the incident power simply results in a larger amount of heated matter (because of the hydrodynamic motion of the spark). It has been suggested (BOBIN et al., 1968) that a way of increasing the temperature would be to deliver the same energy in a shorter (a few nanoseconds) laser pulse. However, experiments show that this is not possible due to self-focusing (see later) and the resulting low amount.
of energy absorbed by the spark (ALCOCK et al, 1970; BELLAND et al, 1971).

3.2.4. Picosecond pulse breakdown

Some people have investigated breakdown in air produced by mode-locked neodymium : glass and CO\textsubscript{2} lasers (KARTMAZOV et al, 1968; ALCOCK et al, 1968; MAC PHERSON and GRAVEL, 1971). In this case the spark is periodic, breakdown occurring in points, each point corresponding to a different light pulse. After breakdown, and the end of the laser radiation of one pulse, of duration much shorter than the interval between pulses, a spherical shock wave propagates on all sides from the point of breakdown. The gas is ionized behind the shock wave and breakdown from each successive light pulse occurs at the place where the laser pulse meets the shock front formed by the preceding pulses.

3.2.5 Self-focusing effects

Recently, evidence for self-focusing in gas breakdown produced by single mode ruby laser has been reported (KOROBKIN and ALCOCK, 1968, ALCOCK et al, 1969, ALCOCK et al, 1970). This evidence rested on the observation of scattered laser light originating in filamentary regions having transverse dimensions of \( \leq 5\mu \), a value which was over an order of magnitude less than
the diameter of the focal region. The nature of these filaments is different in molecular and noble gases. In molecular gases the tendency is to have one long filament, whereas in noble gases breakdown occurs in points, of longitudinal dimensions comparable to the transverse ones. In addition, a large amount of the incident laser radiation is scattered at angles up to 30° from the forward direction, the spectrum of this light being both broadened and shifted from the laser wavelength. For argon, as much as 85% of the incident laser light is scattered in the forward direction. Some of these effects have also been observed in gas breakdown produced by a multimode laser (Tomlinson, 1969). They have been interpreted as providing evidence for the occurrence of self-focusing in gas breakdown, but it was noted that self-focusing is unlikely to occur in the neutral gas prior to breakdown, probably appearing after the breakdown process has already started. However, the observations reported above yield only indirect evidence for the existence of self-focusing since neither the laser beam within the focal region nor the plasma produced by it is observed directly. In addition, some authors (Tomlinson, 1969, Ahmad et al, 1969) have pointed out that the observations of very small spots in 90° scattering could arise either from plasma blobs (due to Fresnel reflection at the plasma boundary) or from true filaments of plasma. However, this ambiguity has been removed by Schlieren measurements (Key et al, 1970, Belland et al, 1971), where one observes the plasma refractive index gradients, and by inter-
ferometric measurements (RICHARDSON and ALCOCK, 1971), where one observes the plasma refractive index. These experiments have indeed shown the existence of the plasma filaments.

Similar effects have been observed in sparks produced by mode-locked ruby and neodymium-glass laser pulses (ALCOCK et al, 1969), and by single picosecond pulses (BUNKIN et al, 1971).

It must be stressed out that, although there is overwhelming evidence for the occurrence of self-focusing in laser induced gas breakdown, the experiments reported do not permit to have any information on the processes leading to it. It is generally thought that self-focusing occurs prior to breakdown during the cascade process through a mechanism involving the existence of a considerable amount of excited atoms (since the polarizability of excited atoms, or molecules, is much greater than the polarizability of unexcited particles). Under these conditions, it can be thought that this would affect the development of the cascade, and that the theory developed earlier (see equation 2.21) cannot truly reflect the experimental situation (BELLAND et al, 1971; BUNKIN et al, 1971).
4. DECAY AND RECOMBINATION

At the end of the laser pulse a strongly heated volume has been produced with a shape elongated along the laser beam axis. The length of this region is obviously of the order of the path travelled by the absorption wave, i.e., $v\Delta t$, which is typically of the order of a few millimeters. The transverse dimensions are determined by the dimensions of the converging light channel and are typically of the order of some tenths of millimeter. The process in this stage is very similar to a strong explosion in a gas. From the point of view of energy release in the gas, a shock wave propagates in all directions and attenuates with time. Its surface is not fully spherical, since the energy was released in a volume of elongated form; however, during the course of time, the form of the shock wave surface becomes nearly spherical. This is not seen in photographs of the spark luminosity because the boundary of the luminous region can coincide with the shock wave front only at the start of the explosion process. Indeed, the luminosity is due to the highly heated gas; therefore, from the moment at which the amplitude of the shock wave decreases to a value insufficient for strong heating the wave becomes invisible and travels forward, leaving behind it a strongly heated luminous region of smaller dimensions. Gradually, as the gas cools down, the luminosity also attenuates.
If one applies Taylor's blast wave theory (TAYLOR, 1949), the expansion obeys the law

\[ x = \left( \frac{e^*}{\rho_0} \right)^{4/5} t^{2/5} \]  

where \( e^* \) is the total energy in the gas. Log-log plots of displacement versus time, obtained from streak photographs after the end of the laser pulse, agree with equation 4.1 (RAMSDEN and DAVIES, 1964). Moreover, interferometric measurements (ALCOCK et al, 1966) have shown that after the end of the laser pulse the hot gas core continues to expand as a blast wave, the density behind the front being in accord with the theory if the perturbed spherical symmetry is taken into account (PANARELLA and SAVIC, 1968).

Contrary to these observations, which seem to confirm the blast wave nature of this stage of the breakdown process, DAIBAND THOMSON (1967) and ALCOCK et al (1968) found that a log-log plot of displacement versus time, after the end of the laser pulse, gives a straight line of slope approximately 0.2, independent of the experimental conditions. AHLBORN (1972) has shown that this behaviour is consistent with a shock driven by a subsonic radiadian, which is powered by black body radiation.

Spectroscopic measurements on the plasma decay have been reported by a number of authors, especially on hydrogen...
or helium sparks (LITVAK and EDWARDS, 1966; BBAERMAN et al., 1969). The results indicate temperature decay because of radiation and expansion cooling and electron loss because of collisional radiative recombination. The space and time resolved structure of a helium plasma, produced by a 1 MW CO$_2$ laser, has been studied by GEORGE et al. (1971) during the first 15 nsec of the afterglow. It was found that the plasma is comprised of a dense hot core which emits primarily ionic lines, and a well defined tenuous outer shell which is primarily the source of neutral emission lines. This plasma structure develops approximately 400 nsec after breakdown, at about the time when the shock front peals off from the luminous fireball, which is then greatly showed down. Fig. 17 shows some of their results for the electron temperature and density. These results were not Abel inverted and thus represent averages along the line of sight. It is seen that for times longer than 400 nsec the ionic line yields a higher average density than does the atomic line. Since the line of sight is the same in the two cases, this is due to the fact that the ionic and atomic emission originate from different regions of the plasma.

Of crucial importance for the determination of density and temperature from spectroscopic measurements is the assumption of local thermodynamic equilibrium. It has been shown by HUGENSCHMIDT (1971), using laser interferometry, Schlieren and shadow techniques, that, because of the high density, LTE is established in the central part of the plasma 10-15 nsec after breakdown.
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TABLE CAPTIONS

TABLE I - Summary of cross-section calculations for multiphoton ionization of rare gases by ruby laser radiation (from Gold and Bebb, 1965).

TABLE II- Summary of experimental results on multiphoton ionization

FIGURE CAPTIONS

Figure 1: Plot of $\sigma_n/J^7$ for eight photon ionization of atomic hydrogen in the ground state. Solid line: GONTIER and TRAHIN (1968); broken line: BEBB and GOLD (1966).

Figure 2: Number of ions produced by multiphoton ionization in helium versus laser power $W$ (after AGOSTINI et al. 1968).

Figure 3: Value of exponent $n_{\text{exp}}$ versus laser wavelength for potassium vapors (after DELONE and DELONE, 1969).

Figure 4: Theoretical dependence of the breakdown threshold intensity $J_{\text{th}}$ versus laser pulswidth $\Delta t$ (after BUNKIN and PROKHOROV, 1969).

Figure 5: Theoretical dependence of the breakdown threshold $J_{\text{th}}$ versus gas pressure $p$ (both in arbitrary units), after equation 2.21 (after YOUNG and HERCHER, 1967).

Figure 6: Breakdown threshold electric field $E_{\text{th}}$ versus gas pressure $p$ for neon, helium, and argon, using ruby laser radiation (after HAUGHT et al., 1966).
Figure 7 : Breakdown threshold electric field $E_{th}$ versus gas pressure $p$ for helium, nitrogen, and argon, using ruby laser radiation (adapted after Gill and Dougall, 1965).

Figure 8 : Breakdown threshold intensity $J_{th}$ versus laser pulse wavelength $\lambda$ for argon and xenon at a pressure of $10^3$ Torr (after Buscher et al, 1965).

Figure 9 : Breakdown threshold fields $E_{th}$ versus characteristic diffusion length $\ell$ for air, helium, and argon at a pressure of 6,800 Torr (after Haught et al, 1966).

Figure 10 : Breakdown threshold intensity $J_{th}$ versus focal diameter dimension for a laser mixture (He: N$_2$:CO$_2$), using CO$_2$ laser (after Berger and Smith, 1972).

Figure 11 : Breakdown threshold pressure $P_{th}$ at a constant laser power, versus magnetic field, $B$, using a CO$_2$ laser (Cohn et al, 1972).

Figure 12 : Breakdown threshold intensity $J_{th}$ versus pressure $p$ for nitrogen, helium, and argon under ruby laser picosecond pulse irradiation (after Krasyuk et al, 1970).

Figure 13 : Power absorbed per unit volume of plasma (normalized to unit ion and electron densities, for ion charge $Z=1$) versus laser intensity $J$ at several temperatures for the frequency of neodymium:glass laser radiation (after Hughes and Nicholson-Florence, 1968).
Figure 14: Geometry of the breakdown region.

Figure 15: Ruby laser threshold intensity for a net positive gain for self focucusing and stimulated scattering versus plasma electron density. In the calculations a 10 eV, fully ionized, hydrogen plasma was used. Also in the figure are the saturation limits. I: ponderomotive stimulated scattering; II: ponderomotive self-focusing; III: thermal stimulated scattering IV: thermal self-focusing (after PALMER, 1971).

Figure 16: Electron density $n_e$ versus time after breakdown for an air spark produced by ruby laser irradiation; the experimental values were deduced from two-wavelength interferometry (after ALCOCK and RAMSDEN, 1966).

Figure 17: Electron temperature and density as a function of time, measured relative to the beginning of the laser pulse, for a helium spark produced by a CO$_2$ laser (after GEORGE et al, 1972).
TABLE I

SUMMARY OF CALCULATIONS FOR MULTIPHOTON IONIZATION OF RARE GASES
BY RUBY LASER RADIATION (FROM GOLD AND BEBB, 1965).

<table>
<thead>
<tr>
<th>Gas</th>
<th>$I_0$ (eV)</th>
<th>n</th>
<th>f</th>
<th>State</th>
<th>strength (eV)</th>
<th>cross section (cm$^2$)</th>
<th>$\sigma^n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>12.127</td>
<td>7</td>
<td>6</td>
<td>$7p^3S_1$</td>
<td>0.188</td>
<td>$8.76 \times 10^{-211}$</td>
<td>$p^6$</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>5</td>
<td>$6s^3P^0$</td>
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<td></td>
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<td>$6s^3P_1$</td>
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<td>$F^7$</td>
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<td>6</td>
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<tr>
<td>Kr</td>
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<td>8</td>
<td>7</td>
<td>$6s^3P_1$</td>
<td>0.114</td>
<td>$3.52 \times 10^{-245}$</td>
<td>$F^7$</td>
</tr>
<tr>
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<td></td>
<td></td>
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<td>9</td>
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<tr>
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**TABLE II**

**SUMMARY OF EXPERIMENTAL RESULTS ON MULTIPHOTON IONIZATION**

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<tr>
<th>Gas</th>
<th>$I_0$ (eV)</th>
<th>$\hbar\omega$ (eV)</th>
<th>$n$</th>
<th>$n_{\text{exp}}$</th>
<th>$\Delta n = n - n_{\text{exp}}$</th>
<th>$E$ (V/cm)</th>
<th>$p$ (Torr)</th>
<th>Ref.</th>
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<td>12.13</td>
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<td>a</td>
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<tr>
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<td>11</td>
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<td>$4\times10^7$</td>
<td>$&lt;10^{-3}$</td>
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<tr>
<td></td>
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<tr>
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<td>$1^{+1}_{-1}$</td>
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<td>$10^{-3}$</td>
<td>K</td>
</tr>
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<td>$10^{-3}$</td>
<td>l</td>
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<td>$12(H₂⁺)</td>
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<td>$4\times10^7$</td>
<td>$10^{-4}$</td>
<td>K</td>
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<tr>
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<td>12</td>
<td>$10.8^{+1}_{-1}$</td>
<td>$1.2^{+1}_{-1}$</td>
<td>$5\times10^7$</td>
<td>$10^{-4}$</td>
<td>l</td>
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<td>$9(H⁺)</td>
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<td>$1.3^{+0.4}_{-0.4}$</td>
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<td>$10^{-4}$</td>
</tr>
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<td>7</td>
<td>$6.4^{+0.5}_{-0.5}$</td>
<td>$0.6^{+0.5}_{-0.5}$</td>
<td>$2\times10^7$</td>
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<tr>
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<td>2.36</td>
<td>$5.7^{+0.5}_{-0.5}$</td>
<td>$0.3^{+0.5}_{-0.5}$</td>
<td>$5\times10^7$</td>
<td>$2\times10^{-4}$</td>
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TABLE III

MULTIPHOTON BREAKDOWN THRESHOLDS OF RARE GASES BY RUBY LASER RADIATION (FROM GOLD AND BEBB, 1965).

<table>
<thead>
<tr>
<th></th>
<th>Xe</th>
<th>Kr</th>
<th>Ar</th>
<th>Ne</th>
<th>He</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_{th}^{30}$ (10$^3$ photons cm$^{-2}$ sec$^{-1}$)</td>
<td>6.62</td>
<td>18.9</td>
<td>69.7</td>
<td>42.0</td>
<td>439</td>
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</tbody>
</table>
\[ n_{\text{exp}} = 48 \pm 0.3 \]

Fig. 2

- Power (W)
- He ions
- \(10^4\) to \(10^9\)
- \(10^5\) to \(10^6\)
$4 - a^2 = 0$

Fig. 3
fig. 4
fig. 5
fig. 6

\[ \Lambda = 2.44 \times 10^{-3} \text{ cm} \]
Fig. 7
fig. 8
fig. 9

- Air
- He
- A

$p = 6800 \text{ Torr}$
fig. 11
fig. 12
\( J \) (W/cm²)

\( n_e \) (cm⁻³)

fig. 15
fig. 16
fig. 17