Theory of the Ionization of Gases by Laser Beams

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The ionization of gases by intense pulsed laser beams is discussed. In particular, the production of ions and electrons by direct multiple absorption of photons is considered in simple terms and it is concluded that the experimentally observed gas breakdown is probably initiated by this process, but that the subsequent growth of electron population is governed by some other process, such as inverse bremsstrahlung, or the acceleration of electrons in the oscillatory field. The theory predicts that the variation of threshold photon intensity for breakdown of a gas should exhibit almost pressure-independent low and high limits, that the range of intensities between these limits should be approximately $(10^{13})^{1/N_\nu}$, where N_ν is the number of photons required to raise the atom to its lowest excited state, and that the threshold flux density will vary with change of focal volume as V^{-1/N_ν}.

I. INTRODUCTION

TWO recent papers[1,2] describe experiments in which brightly luminous ionized gas is produced when red light (6943 Å) from a pulsed ruby laser is focused by means of a lens. Meyerand and Haught used a 30 nsec pulse, giving peak powers of 30 MW to produce between 10^{13} and 10^{15} charged particles in a volume of about 10^{-5} cm^3 of argon and helium at pressures between 2 and 150 atm. Minck, operating over a pressure range from 0.3 to 100 atm, produced "sparks" in H_2, N_2, He, and Ar with a 25 nsec pulse having a peak power of 2 MW in a volume of about 10^{-9} cm^3. Minck has sought to explain his results in terms of an extrapolation of the microwave breakdown process[3] and obtained good agreement with theory for nitrogen. Wright[4] has developed an approximate expression for the cross section for the inverse bremsstrahlung process in He and Ar, and has shown that calculations are in good agreement with the results of Meyerand and Haught.[1]

Both the inverse bremsstrahlung and the "microwave" processes can operate only on free electrons and both Wright and Minck find it necessary to assume the presence of an electron in the discharge region at the time of application of the laser pulse. However, the equilibrium density of ions in air is typically less than 10^3 cm^{-3} and the rate of production is approximately 10 per sec.[5,6] The chance that a free electron occurs naturally in the focal region at the time of the pulse must therefore be considered negligible. Indeed, the presence of a random electron in a volume 10^{-9} cm^3 implies a mean charge density in the region of that observed in tenuous glow discharges.

Clearly, then, before the problem of gas ionization by laser beams can be resolved the origin of the first electron must be determined. In this paper the rate of production of ions and free electrons by multiple photon excitation to the lowest level of the atom, and subsequent excitation to the ionization level, is considered in simple terms. Wright[4] has shown that an atom which has been raised to its lowest excited state is very rapidly ionized, and consequently the discussion here centers upon the production of excited atoms. The consideration of statistical fluctuations in photon density, at the atomic scale, strongly affects the theory, and the argument of Meyerand and Haught[1] that an electric field strength ratio of 10^5 between He and Ar breakdown strengths, appears to be in error.

II. THEORY

According to the quantum theory of matter a gas atom can exist only in one of a series of stable states. Therefore, an atom cannot interact with a photon unless that photon can raise the atom to one of its excited states. However, if the atom is subjected to a sufficiently intense bombardment by photons, it is possible for the "simultaneous" absorption of several photons to occur, resulting in the atom reaching one of its excited states.

1. Direct Excitation of a Single Atom

Consider an atom undergoing bombardment by photons with an average flux n_ν cm^{-2} sec^{-1}. The photon energy is $h\nu$, and if the lowest atomic excited state lies E(ergs) above the ground state, then the number of photons which must interact "simultaneously" is $N_\nu = E/h\nu$. Suppose the cross section for "simultaneous" excitation of the atom by N_ν photons is σ cm^2. From the uncertainty principle, the time of interaction between an electron and a photon is uncertain by the amount $t \sim h/h\nu = 1/\nu$. If, during time t, the area σ is crossed by N_ν photons, an excited atom will be formed.

The average number of photons crossing area σ in time t is $n_\nu \sigma t$. However, although the photon distribution over distances comparable with the laser wavelength is governed by the field distribution, it appears to be reasonable to treat the distribution as random over atomic or subatomic dimensions. A Poisson dis-

[1] R. G. Meyerand and A. F. Haught, Phys. Rev. Letters **11**, 401 (1963).
[2] R. W. Minck, J. Appl. Phys. **35**, 252 (1964).
[3] S. C. Brown, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), p. 531.
[4] J. K. Wright, Proc. Phys. Soc. (London) **84**, 41 (1964).
[5] B. F. J. Schonland, *Atmospheric Electricity* (Methuen and Company, Ltd., London, 1953).
[6] J. A. Chalmers, *Atmospheric Electricity* (Pergamon Press Ltd., London, 1957).

tribution then gives for the probability of N_ν photons reaching area σ in time t.

$$p \sim \exp[-n_\nu \sigma t]((n_\nu \sigma t)^{N_\nu}/N_\nu!) \tag{1}$$

provided $N_\nu \gg n_\nu \sigma t < 1$.

If the region being irradiated by the laser beam, volume V, contains N atoms cm^{-3}, and N_0 electrons are to be produced in time T (the laser pulse time), then

$$(VNT/t)\exp(-n_\nu \sigma t)((n_\nu \sigma t)^{N_\nu}/N_\nu!) = N_0$$

and

$$n_\nu \sim (1/\sigma t)[(N_0 t N_\nu!/VNT)\exp(n_\nu \sigma t)]^{1/N_\nu} \tag{2}$$

2. Collision of Atoms Partially Excited

The case where two atoms interact "simultaneously" with a total of N_ν photons, and with each other can be dealt with similarly. If there is a cross section σ_1 for the interaction of two atoms, which are within the influence of CN_ν and DN_ν photons, respectively (where CN_ν, DN_ν are the two integers nearest to $\frac{1}{2}N_\nu$ such that $C+D=1$), and if σ_2 is the cross section defining this influence, then N_0 photons will be produced if

$$n_\nu = \frac{1}{\sigma_2 t}\left[\frac{N_0 (CN_\nu)!(DN_\nu)!}{3\sigma_1 V_T V N^2 T \exp(-2n_\nu \sigma_2 t)}\right]^{1/N_\nu}. \tag{3}$$

It is easily shown that the 3-atom case is of negligible importance. V_T is the thermal atomic velocity.

3. Condition for the Production of N_0 Excited Atoms During the Laser Pulse

Combining Eqs. (2) and (3) the condition becomes

$$n_\nu = \frac{1}{t}\frac{N_0^{1/N_\nu}}{(VNT)^{1/N_\nu}}\left[\frac{3\sigma_2^{N}\nu \sigma_1 V_T N \exp(-2n_\nu \sigma_2 t)}{(CN_\nu)!(DN_\nu)!} + \frac{\sigma^{N}\nu}{t}\frac{\exp(-n_\nu \sigma t)}{N_\nu!}\right]^{-1/N_\nu}. \tag{4}$$

III. DISCUSSION

1. General

From Eq. (4) certain predictions can be made independent of the values employed for the various cross sections:

(a) The threshold density n_ν for the production of N_0 electrons will vary with the volume of the focus, and with the time of the laser pulse as V^{-1/N_ν} and as T^{-1/N_ν}. A change of threshold intensity with change in focus volume is revealed by the difference between the results of Minck and of Meyerand and Haught.

(b) The threshold for production of N_0 electrons will vary as N^{-1/N_ν} at low pressures and N^{-2/N_ν} at high pressures. It is to be expected that no breakdown will occur with values n_ν less than the threshold for production of one electron, and that no thresholds will be observed greater than those required to produce $\sim 10^{13}$ electrons from this process alone. All threshold breakdown strengths are therefore expected to lie within a narrow range, and breakdown at pressures of a few Torr should be possible in He, Ar, N_2, and H_2 with photon densities no more than 2 or 3 times those reported in work at atmospheric pressure.

2. Estimated Values of n_ν. The Rare Gases

In order to compare observed photon densities with those predicted by Eq. (4) it is necessary to estimate values of σ, σ_1, σ_2. Minck's results for helium show a sharp break at about 30 atm, below which the threshold density falls more slowly. It seems not unlikely that at pressures about 30 atm the photon threshold is set by the need to produce a single electron before other processes, such as inverse bremsstrahlung, can produce the necessary multiplication. The variation of the threshold with pressure is that to be expected from Eq. (2) with σ set at 10^{-16} cm^2. Figure 1 shows theoretical curves for helium for $N_0 = 1$ (lower) and $N_0 = 10^{13}$ (upper). Experimental results are also shown and are seen to be compatible with the theory. The change in slope of the curves at densities above 10^{22} cm^{-3} results from setting σ_1, and σ_2 both equal to 10^{-16} cm^2 in Eq. (4).

The value of 10^{-16} cm^2 for σ may be considered reasonable on the ground that this is the approximate condition that the N_ν photons are within the atomic dimensions. Substitution of this value in Eq. (2) for argon also results in upper and lower limits which span the range of densities observed experimentally. (Figure 2.)

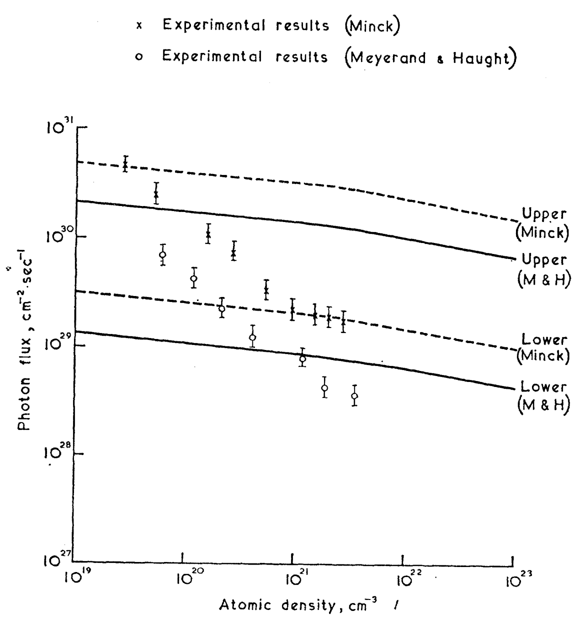

FIG. 1. Ionization of helium by light from a ruby laser.

3. Hydrogen and Nitrogen

Calculations of the limiting photon fluxes for H_2 and N_2 have been made on the assumption that the molecule is raised to its first electronically excited state by multiple-photon absorption, that subsequent single-photon absorption results in dissociation into ground-state atoms, and that the atoms are then ionized by the processes discussed above. Setting $\sigma = 10^{-16}$ cm^2, the experimental results are again found to lie within the theoretical range. (Figure 3.)

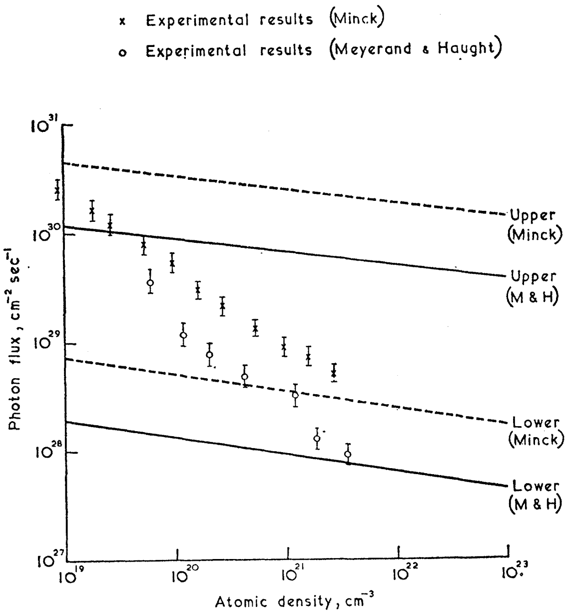

FIG. 2. Ionization of argon by light from a ruby laser.

4. Calculation of Probability of 2-Photon Processes

It is interesting to use Eq. (2) to calculate the probability of 2-photon processes, which have already been determined by exact quantum-mechanical calculations.

Abella[7] has calculated the probability of 2-photon absorption in gaseous caesium using the Bates-Damgaard method, and concluded that in the conditions of his experiment 5×10^{12} transitions should occur. Use of Eq. (2) and $\sigma\sim10^{-16}$ cm^2 yields a value of 3×10^{12}.

Zernik[8] has calculated the probability of 2-photon

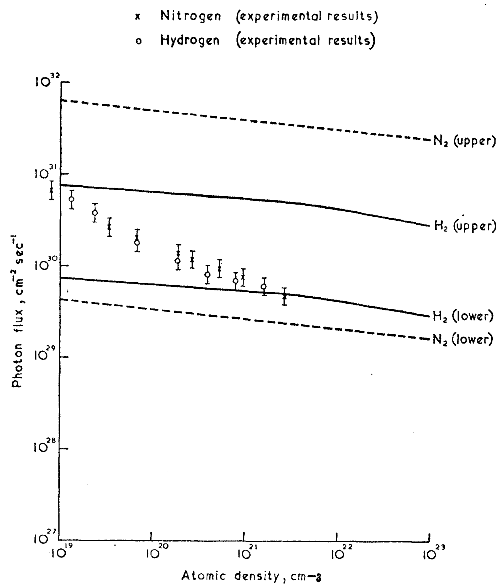

FIG. 3. Ionization of hydrogen and nitrogen by light from a ruby laser.

ionization of excited hydrogen, and obtained a cross section for ruby laser light of 5.3×10^{-29} cm^{4}W^{-1}. Equation (2) and a value of $\sigma\sim10^{-16}$ cm^2 yields a cross section of 3×10^{-29} cm^{4}W^{-1}.

IV. CONCLUSIONS

It has been shown that the free electrons required to explain experimental results on ionization of gases by intense laser beams can originate from multiple photon absorption by gas atoms to the lowest excited state, followed by step-by-step excitation to the ionized level.

According to the theory presented in this paper further experiments on the variation of threshold photon intensity with pressure may be expected to show almost pressure independent low and high limits. The range of intensities covered would be expected to be approximately $(10^{13})^{1/N_p}$. Breakdown of gases at low pressures should be possible with only slight increase in laser-beam intensity. The variation of threshold flux density with change of focal volume would be expected to show a $(V)^{-1/N_p}$ dependence.

ACKNOWLEDGMENT

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[7] I. D. Abella, Phys. Rev. Letters **9**, 453 (1962).
[8] W. Zernik, Phys. Rev. **135**, A51 (1964).