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A. D. MacDonald and Sanborn C. Brown

Abstract

Electrical breakdown of hydrogen at high frequencies has been treated theoretically on the basis of the Boltzmann transport equation. Inelastic collisions are taken into account as a loss term in the Boltzmann equation and measured values of the ionization efficiency are used in the integral determining the ionization rate. The energy distribution function for electrons may be expressed in terms of the confluent hypergeometric function and simple exponentials. The ionization rate and diffusion coefficient are calculated using these distribution functions and kinetic theory and are combined with the diffusion equation to predict breakdown electric fields. These predicted electric fields are compared with experimental values measured at 3000 Mc/sec. They are also compared with older measurements by other workers at frequencies ranging from 3 Mc/sec to 100 Mc/sec. The breakdown equation calculated from kinetic theory and using no gas discharge data other than the collision cross section measurements and involving no adjustable constants predicts breakdown electric fields well within the limits of accuracy determined by these cross sections over a large range of pressure, container size and frequency of applied field.

The theory, based on a solution of the Boltzmann transport equation for electrons, which was used in predicting breakdown in helium (1), has been applied to molecular hydrogen. The method follows closely that used in Reference 1, but some of the simplifications used in treating helium are not permissible in the case of hydrogen. The second-order differential equation derived from the Boltzmann equation is solved for the electron distribution function. The ionization rate and diffusion coefficient are calculated using standard kinetic theory formulas. The breakdown condition is that the number of electrons produced by ionization equal the number diffusing to the walls of the container. This breakdown condition is combined with a solution of the diffusion equation, the ionization rate, and diffusion coefficient to obtain an equation which predicts breakdown electric fields.

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I. The Boltzmann Equation

The phase space continuity equation for electrons is (1)(2)

$$C = \frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f + \vec{a} \cdot \nabla_v f \quad (1)$$

where f is the electron energy distribution function; C is the net rate at which electrons appear in an element in phase space and is calculated in terms of f by determining energy changes due to collision; \vec{v} is the velocity, \vec{a} the acceleration, t the time, and ∇_v the gradient operator in velocity space.

The distribution function may be expanded in spherical harmonics in velocity

$$f = f_0 + \frac{\vec{v} \cdot \vec{f}_1}{v} + \dots \quad (2)$$

The spherically symmetric term f_0 is predominant because collisions tend to disorder any directional motion of the electrons. The series is rapidly convergent and we shall consider only those cases where the first two terms represent a good approximation to the distribution function. The limits of theory discussed in the previous paper indicate those values of the experimental parameters for which this approximation is valid.

The term C arising from collisions may also be expanded in spherical harmonics, since it may be represented in terms of integrals over the distribution function. The rms value of the electric field is given by E , and an energy variable $u = mv^2/2e$ is introduced; m is the mass and e the charge of an electron.

On substitution of these terms and separation of vector and scalar parts, Eq. (1) becomes

$$C_0 = \frac{\partial f_0}{\partial t} - \frac{v}{3u} \frac{\partial}{\partial u} (uE \cdot \vec{f}_1) + \frac{v}{3} \nabla \cdot \vec{f}_1 \quad (3)$$

and

$$\vec{C}_1 = \frac{\partial \vec{f}_1}{\partial t} + v \nabla f_0 - vE \frac{\partial f_0}{\partial u} \quad (4)$$

Elastic collisions are accounted for in the manner of Morse, Allis and Lamar (3) who considered collisions as instantaneous processes and found equivalent energy loss terms by conserving momentum and energy and averaging over space at each collision. It is shown in Reference 3 that these terms are

$$C_{oe\ell} = \frac{2m}{M} \frac{v}{u} \frac{d}{du} \left(\frac{u^2 f_0}{\ell} \right) \quad (5)$$

and

$$\vec{C}_{1e\ell} = -\frac{v}{\ell} \vec{f}_1 \quad (6)$$

where ℓ is the electronic mean free path and M is the mass of the molecule. The

remainder of the C_0 term results from inelastic collisions and may be represented by $-h_x v/l f_0 - h_i v/l f_0$, where h_x and h_i are efficiencies of excitation and ionization, respectively. Inelastic collisions have no angular dependence and therefore do not enter the higher order terms in the distribution function.

Each term in the distribution function may be expanded in a Fourier series in time

$$f_n = f_n^0 + f_n^1 e^{j\omega t} + \dots \quad (7)$$

where ω is the radian frequency of the applied electric field. The electric field is represented by $\sqrt{2} E e^{j\omega t}$ and in the expansion of $E f_n$, we must replace the exponential notation by its real part before taking the product, so that

$$\sqrt{2} E f_n = 2 E f_n^0 e^{j\omega t} + E f_n^1 (1 + 2 e^{2j\omega t}) + \dots \quad (8)$$

Combining the results of Eqs. (5), (6), and (8) with Eqs. (3) and (4) and equating terms in like exponents of t , we have

$$-(h_x + h_i) \nu_c f_0^0 = \frac{v}{3} \nabla \cdot \vec{f}_1^0 - \frac{vE}{3u} \cdot \frac{\partial}{\partial u} (u \vec{f}_1^0) - 2 \frac{mv}{Mu} \frac{\partial}{\partial u} \left(\frac{u^2 f_0^0}{l} \right) \quad (9)$$

$$\nu_c \vec{f}_1^0 = -v \nabla f_0^0 \quad (10)$$

$$(\nu_c + j\omega) \vec{f}_1^1 = vE \frac{\partial}{\partial u} f_0^0 \quad (11)$$

In these equations, v/l has been replaced by ν_c and the terms in f_0^1 have been dropped. The f_0^1 term represents the first harmonic of the spherically symmetric part of the distribution function and cannot be generated physically unless there is either a d-c field or the amplitude of oscillation of the electric field is sufficient to sweep out electrons from the container each half cycle.

Equations (10) and (11) may be substituted in Eq. (9) to derive an equation for f_0^0 . From this point on, we shall drop the subscripts and superscripts on f and understand by f the zero-order term f_0^0 . We use the diffusion equation to replace $\nabla^2 f_0^0$ by $-1/\Lambda^2 f_0^0$, where Λ is the characteristic diffusion length, depending only on the geometry of the discharge container (4). Thus

$$-\nu_c (h_x + h_i) f + \frac{2m}{M} \frac{v}{u} \frac{\partial}{\partial u} \left(\frac{u^2 f}{l} \right) = \frac{l v}{3\Lambda^2} f - \frac{v}{3u} \frac{d}{du} \left\{ l u \frac{df}{du} \frac{E^2}{(1 + \omega^2/\nu_c^2)} \right\} \quad (12)$$

where we take only the real part in the last of these terms, as f is real.

II. Collision Cross Section

In Eq. (12), we must now specify h_x and h_i . As was the case in helium, the collision frequency for elastic collisions in hydrogen is constant in energy (5)(1) to a very good approximation. Using Brode's data, we obtain the value of $\nu_c = 5.93 \cdot 10^9 \text{ p}(\text{sec}^{-1})$ (p in mm of Hg).

The excitation and ionization efficiencies for hydrogen have been measured by Ramien (6). A linear approximation to his data gives

$$h_x + h_i = h_{ix}(u - u_x) = 9.0 \times 10^{-3}(u - 8.9), \quad u \geq 8.9 \text{ volts}.$$

The fact that some energy goes into excitation below the dissociation level is explained by Ramien on the basis of wave mechanics.

The ionization efficiency h_i is approximated by

$$h_i = h_{ii}(u - u_i) = 9.4 \times 10^{-3}(u - 16.2), \quad u \geq 16.2 \text{ volts}.$$

These efficiencies are to be substituted in Eq. (12) from which the distribution function is determined. Below the lowest excitation level, the term in $h_x + h_i$ is zero and the distribution function is similar to that found for helium (1). Above this level, the differential equation for the distribution function is different and the solution must be matched in magnitude and slope to the previously determined one.

III. The Distribution Functions

We let

$$\mu = \frac{3m}{M} \frac{m}{e} \Lambda^2 \nu_c^2,$$

$$\beta = \frac{(E\Lambda)^2}{\omega^2/\nu_c^2 + 1},$$

and then Eq. (12) becomes

$$u \frac{d^2 f}{du^2} + \left(\frac{3}{2} + \frac{\mu u}{\beta} \right) \frac{df}{du} + f \left[\frac{3}{2} \frac{\mu}{\beta} - \frac{u}{\beta} - \frac{1}{2} \frac{M}{m} \frac{\mu}{\beta} (h_x + h_i) \right] = 0. \quad (13)$$

For convenience, we introduce a dimensionless independent variable

$$w = \frac{\mu}{\beta} \sqrt{1 + \frac{4\beta}{\mu^2}} u = \frac{\mu}{\beta} bu.$$

Equation (13) becomes

$$\frac{d^2 f}{dw^2} + \left(\frac{1}{b} + \frac{3}{2w} \right) \frac{df}{dw} + f \left[\frac{3}{2bw} - \frac{\beta}{\mu^2 b^2} - \frac{M}{2m} \frac{h_x}{wb} \right] = 0.$$

When $u < 8.9$ volts, we transform the dependent variable

$$f = g \exp \left[-\frac{1}{2} \left(\frac{1}{b} + 1 \right) w \right]$$

and the differential equation becomes

$$w \frac{d^2 g}{dw^2} + \frac{dg}{dw} \left(\frac{3}{2} - w \right) - \alpha g = 0 \quad (15)$$

where

$$\alpha = \frac{3}{4} \left(1 - \frac{1}{b} \right) .$$

Equation (15) is the differential equation for the confluent hypergeometric function; therefore, we may write the two independent solutions

$$g_1 = M \left(\alpha; \frac{3}{2}; w \right) \quad g_2 = w^{-1/2} M \left(\alpha - \frac{1}{2}; \frac{1}{2}; w \right) . \quad (16)$$

Hereafter, for brevity, we shall use the notation

$$M \left(\alpha; \frac{3}{2}; w \right) = M_1(w)$$

$$w^{-1/2} M \left(\alpha - \frac{1}{2}; \frac{1}{2}; w \right) = M_2(w) .$$

Tables of the confluent hypergeometric function are available (7).

For u greater than 8.9 volts, we transform Eq. (14) to the reduced form by letting

$$f = y \left\{ \exp \left[-\frac{1}{2} \int \left(\frac{1}{b} + \frac{3}{2w} \right) dw \right] \right\} . \quad (17)$$

Then

$$\frac{d^2 y}{dw^2} + I(w) y = 0 \quad (18)$$

where

$$I(w) = \frac{3}{16w^2} + \frac{1}{w} \left[\frac{M h_{1x} \beta w_x}{2m\mu b^2} + \frac{3}{4b} \right] - \frac{\beta}{\mu^2 b^2} - \frac{M h_{1x} \beta}{2m\mu b^2} - \frac{1}{4b^2} . \quad (19)$$

When w is that corresponding to a few tenths of a volt more than 8.9, the first term in Eq. (19) becomes negligible and we may write

$$I = -\frac{A^2 w - B}{w} \quad (20)$$

where

$$A^2 = \frac{h_{1x} \beta M}{2m\mu b^2} + \frac{1}{4}$$

$$B = \frac{1}{b} \left(\frac{h_1 x^{M\beta w_x}}{2m\mu b} + \frac{3}{4} \right)$$

and in the case of hydrogen, these are numerically

$$A^2 = 1.03 (E/p)^2 \frac{1}{b^2 \left[\left(\frac{31.8}{p\lambda} \right)^2 + 1 \right]} + \frac{1}{4}$$

$$B = 146/b$$

and λ is the free-space wavelength of the electric field in cm.

Equation (18) may be written

$$\frac{d^2 y}{dw^2} - \frac{A^2 w - B}{w} y = 0$$

whose solution is

$$y = e^{-Aw} w^{B/2A} \left[1 + \frac{a_1}{w} + \frac{a_2}{w^2} + \dots \right]. \quad (21)$$

The series converges rapidly for all values of w which are necessary, and in most cases a_2 is completely negligible. a_1 and a_2 are given in terms of A and B

$$a_1 = \left(1 - \frac{B}{2A} \right) \frac{B}{4A^2}$$

and

$$a_2 = \frac{a_1}{4A} \left[\frac{B}{A} \left(\frac{3}{2} + \frac{B}{4A} \right) - 2 \right].$$

We shall drop the term in a_2 at this point; although the nature of the final result for the ionization coefficient will indicate how this term affects the answer.

Combining Eq. (21) with Eq. (17), we have

$$f = e^{-wS} w^T \left(1 + \frac{a_1}{w} \right) \quad (22)$$

where

$$S = A + \frac{1}{2b}$$

and

$$T = \frac{B}{2A} - \frac{3}{4}.$$

The solution in Eq. (22) is valid for u a few tenths of a volt above the lowest excitation level, so we extrapolate the solution in Eq. (16) to $u = 9.5$ volts and use Eq. (22) for $u > 9.5$ volts.

We are now in a position to write down the distribution function

$$\begin{aligned}
f &= \left[M_1(w) + CM_2(w) \right] \exp \left[-w \left(1 - \frac{2}{3}a \right) \right] & u < 9.5 \text{ volts} \\
&= R w^T \left(1 + \frac{a_1}{w} \right) \exp(-wS) & u > 9.5 \text{ volts}
\end{aligned} \tag{23}$$

where the constants R and C are to be determined by the boundary conditions that the distribution function be continuous in value and slope when $u = 9.5$ volts.

If we let

$$\frac{1}{\Phi} = \frac{T}{w_p} - \frac{a_1}{w_p^2 (1 + a_1/w_p)} - S \tag{24}$$

these conditions give

$$C = \frac{M_1(w_p) - \Phi \left[M_1'(w_p) - \left(1 - \frac{2}{3}a \right) M_1(w_p) \right]}{\Phi \left[M_2'(w_p) - M_2(w_p) \left(1 - \frac{2}{3}a \right) \right] - M_2(w_p)} \tag{25}$$

and

$$R = \frac{\exp \left[-w_p \left(1 - \frac{2}{3}a \right) \right] \left[M_1(w_p) + CM_2(w_p) \right]}{e^{-w_p S} w_p^T \left(1 + \frac{a_1}{w_p} \right)} \tag{26}$$

where w_p is w corresponding to $u = 9.5$ volts, and M' denotes differentiation with respect to w .

IV. The Breakdown Condition

We next compute the ionization rate $n\nu$ by the use of formula 16 of Reference 1

$$\begin{aligned}
n\nu &= -8\pi \frac{e^2}{m^2} \int_{u_i}^{\infty} \frac{u}{v} \nu_c h_i f du \\
&= -16\pi \left(\frac{e}{2m} \right)^{3/2} \left(\frac{\beta}{\mu b} \right)^{5/2} \nu_c R 9.4 \times 10^{-3} \int_{w_i}^{\infty} e^{-wS} \left[w^V + (D - w_i) w^{V-1} - w_i D w^{V-2} \right] dw \quad .
\end{aligned} \tag{27}$$

Evaluation of this integral gives

$$\frac{V!}{S^{V+1}} \left\{ 1 - I(V, Z_i) + \left(\frac{Sa_1 - Z_i}{V} \right) \left[1 - I(V-1, Z_i) \right] - \frac{Sa_1 Z_i}{V(V-1)} \left[1 - I(V-2, Z_i) \right] \right\}$$

where $Z_i = Sw_i$, $V = T + 3/2$, $I(x, p) = \Gamma_x(p+1)/\Gamma(p+1)$, and $\Gamma_x(p+1)$ is the incomplete Γ function. Extensive tables of the I functions are available (8).

The diffusion coefficient D is determined from the equation

$$nD = \frac{2\pi}{3\nu_c} \left(\frac{2e}{m} \right)^{5/2} \int_0^\infty fu^{3/2} du.$$

After substitution of the distribution function from Eq. (23), the expression for nD becomes

$$nD = \frac{2\pi}{3\nu_c} \left(\frac{2e\beta}{m\mu b} \right)^{5/2} \left\{ \int_0^{w_p} w^{3/2} \exp \left[-w \left(1 - \frac{2}{3}a \right) \right] \left[M_1(w) + CM_2(w) \right] dw \right. \\ \left. + R \int_{w_p}^\infty w^V e^{-wS} \left(1 + \frac{a_1}{w} \right) dw \right\}. \quad (28)$$

The confluent hypergeometric functions in the first part of the integral in Eq. (28) have been integrated (7) and those in the second integral result in incomplete Γ functions. The total integral yields

$$\frac{3}{2a} \frac{w_p^{3/2}}{(1 - 2/3a)} \exp \left[-w_p \left(1 - \frac{2}{3}a \right) \right] \left\{ \left[M_1'(w_p) - \frac{2}{3}a M_1(w_p) \right] + C \left[M_2'(w_p) - \frac{2}{3}a M_2(w_p) + \frac{1}{2w_p^{3/2}} \right] \right\} \\ + \frac{RV!}{S^{V+1}} \left\{ 1 - I(V, Z_p) + \frac{Sa_1}{V} \left[1 - I(V-1, Z_p) \right] \right\}.$$

The high-frequency ionization coefficient (4) ξ is then given by

$$\xi = \frac{\nu}{DE^2} = \frac{1}{\Lambda^2 E^2} \left[\frac{282 (p\Lambda)^2 J}{K + FG} \right] \quad (29)$$

where

$$K = 1 - I(V, Z_p) + \frac{Sa_1}{V} \left[1 - I(V-1, Z_p) \right] \\ J = 1 - I(V, Z_i) + \frac{Sa_1}{V} \left[1 - I(V-1, Z_i) \right] - \frac{Z_i}{V} \left\{ 1 - I(V-1, Z_i) + \frac{Sa_1}{V-1} \left[1 - I(V-2, Z_i) \right] \right\} \\ F = \frac{3S^{V+1} \exp \left[- \left(1 - \frac{2}{3}a \right) w_p \right]}{2aR \left(1 - \frac{2}{3}a \right) V!} \\ G = w_p^{3/2} \left\{ M_1'(w_p) - \frac{2}{3}a M_1(w_p) + C \left[M_2'(w_p) - \frac{2}{3}a M_2(w_p) \right] \right\} + \frac{C}{2} \exp \left[w_p \left(1 - \frac{2}{3}a \right) \right]$$

$$w = \frac{16.2 \text{ bu} \left[1 + \left(\frac{31.8}{p\lambda} \right)^2 \right]}{(E/p)^2}$$

$$Z = Sw$$

$$b = \left\{ 1 + \left(\frac{E}{p} \right)^2 \frac{1.52 (10^{-2})}{(p\lambda)^2 \left[1 + \left(\frac{31.8}{p\lambda} \right)^2 \right]} \right\}^{1/2}$$

$$a = 0.75 \frac{b-1}{b}$$

$$S = A + \frac{1}{2b}$$

$$A^2 = 1.03 \left(\frac{E}{p} \right)^2 \frac{1}{b^2 \left[\left(\frac{31.8}{p\lambda} \right)^2 + 1 \right]} + \frac{1}{4}$$

$$V = \frac{1}{2} \left[\frac{146}{Ab} + 3 \right]$$

$$a_1 = \left(1 - \frac{B}{2A} \right) \frac{B}{4A^2}$$

$$B = \frac{146}{b}$$

and ϕ , C and R are defined in Eqs. (24), (25) and (26). The breakdown condition is that $\xi = 1/\Lambda^2 E^2$, so that we obtain the breakdown equation by setting ξ computed from Eq. (29) equal to $1/\Lambda^2 E^2$. This produces a transcendental equation which is very difficult to solve and which is done in practice by successive approximations.

V. Experimental Results

Breakdown electric fields have been measured at microwave frequencies using the experimental apparatus shown in Fig. 1. The details of the experiment are similar to those of the helium breakdown measurements (1). Microwave power with a free-space wavelength of approximately 10 cm generated by a c-w magnetron is coupled to a

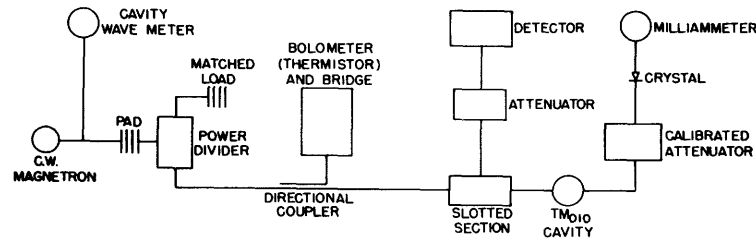


Fig. 1 Block diagram of experimental microwave apparatus.

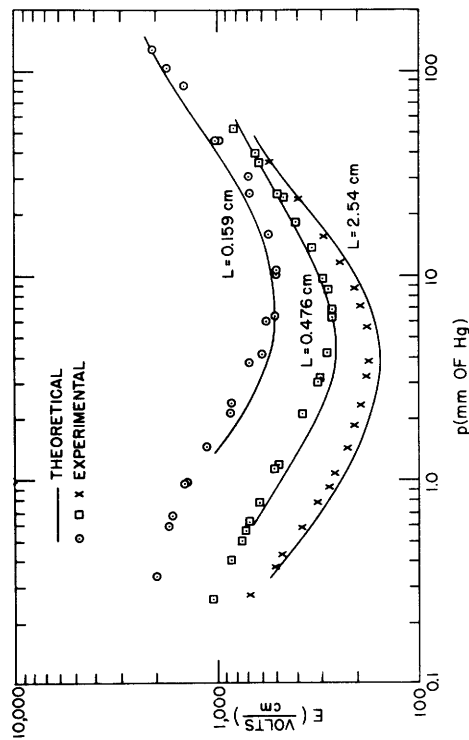


Fig. 2 Comparison of experimental electric fields with those theoretically predicted from Eq. (29) (3000 Mc/sec).

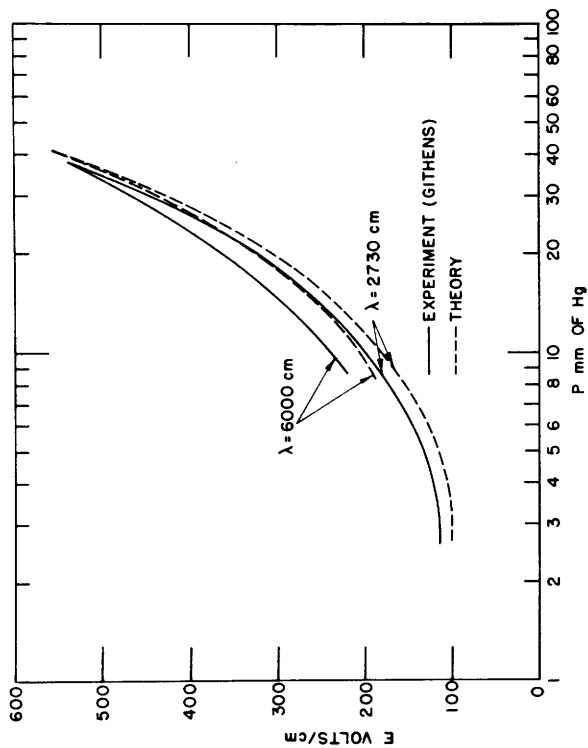


Fig. 3 Comparison of theory of this paper with experiments of Githens (1941) at frequencies of 5 Mc/sec and 11 Mc/sec.

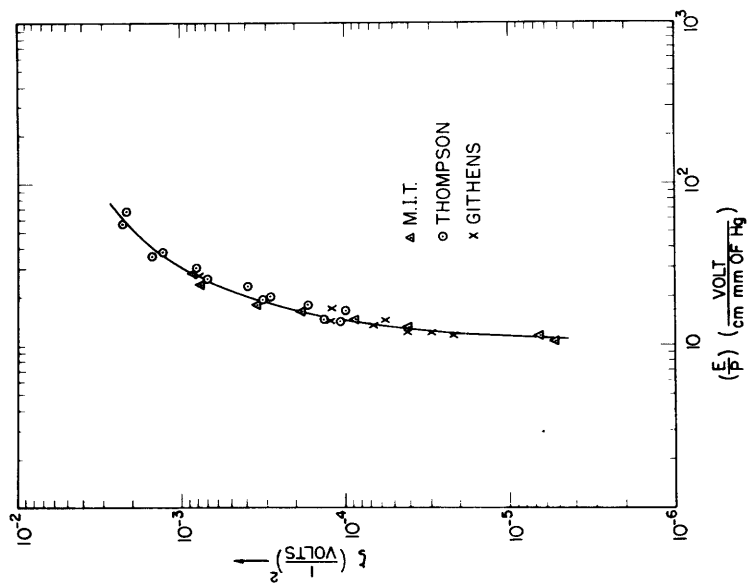


Fig. 4 Experimental ζ curves of Thompson (1937), Githens (1940) and the present work. The data cover a frequency range of 5 to 3000 Mc/sec and a range of diffusion length from $\Lambda = 0.0505$ cm to $\Lambda = 0.50$ cm.

microwave resonant cavity through coaxial transmission lines. A known fraction of the power delivered is measured by a bolometer. The power absorbed by the cavity is combined with the cavity Q and the known field configuration to determine the electric field by standard methods (9)(10). The cavities in which breakdown takes place are made of oxygen-free high-conductivity copper and connected through Kovar to an all-glass vacuum system. The vacuum system holds at a pressure of better than 10^{-7} mm of Hg for a period of about two hours with the pumps turned off. A single series of breakdown measurements takes about this time. The pressure is measured by an ionization gauge. Air Reduction Company spectroscopically pure hydrogen was used. Measurements were made in cylindrical cavities having heights of 0.1586 cm, 0.476 cm and 2.54 cm. The experimental data are presented in Fig. 2, which gives breakdown electric field as a function of pressure, and theoretical curves of E computed from Eq. (29).

The theory of breakdown derived in this paper is not restricted to microwave frequencies but applies to any high frequency discharge in which electrons are produced by field ionization and lost by diffusion to the walls of the container. Theoretical electric fields have been computed for electric field wavelengths of 6000 cm and 2730 cm. In Fig. 3, these are compared with the experimental data obtained by Githens in 1940 (11). Figure 4 presents ionization coefficients experimentally obtained by Githens (11) and Thompson (12), as well as the experimental data of this paper.

VI. Discussion

Breakdown electric fields at high frequencies have been derived theoretically on the basis of kinetic theory, the only experimental data used being collision cross sections. The elastic collision cross section for hydrogen used in this theory is probably correct within 10 percent. Calculations of the theory indicated that this will not introduce more than 2 or 3 percent error in electric fields. The excitation and ionization efficiencies are very difficult to measure and the experimental error in the best measurements in hydrogen may be as high as 20 percent. These introduce an error of approximately 14 percent in the theoretical electric fields. These effects combine to give a possible error of 16 percent in theoretical fields and indicate a need for more precise collision cross section measurements. The maximum error in the experimental electric fields in the 10-cm wavelength region is 5 percent and in pressure is 1 percent. The derivation of the equation for the distribution function implicitly assumed that each electron dropped back to zero energy after an inelastic collision. Since excitation takes place over a certain range of energy, this is not exactly correct, but the error which it introduces is small.

Equation (29), calculated from kinetic theory and using no gas discharge data other than collision cross section measurements and involving no adjustable constants, predicts breakdown electric fields well within the limits of accuracy over a large range of pressure, container size and frequency.

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