Negative Current-Voltage Characteristics in Hydrogen at High Pressure Using Plane Parallel Electrodes*

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In conjunction with measurements of current-voltage characteristics in hydrogen, a few characteristics have been obtained which include a region with negative slope. The latter characteristics were obtained with plane parallel electrodes at an electrode separation of 2 cm at a pressure of 400 mm Hg and with three values of externally initiated cathode current. The initial currents ranged from about 10^{-11} to 10^{-9} amp, and the amplified currents reached values as high as 10^{-4} amp. The characteristics corresponding to the larger initial currents become negative at large currents ($\sim 10^{-5}$ amp). The voltage at which a characteristic becomes negative, i.e., the maximum attainable voltage across the electrodes, decreases slightly with increasing initial current. The circuit included a series resistor of 20 megohms.

These characteristics can be explained quantitatively on the basis of the first and second Townsend coefficients (previously measured with the same apparatus) acting in conjunction with space charge, if a not unreasonable discharge area is assumed. These calculations were carried out on an IBM 704 computer.

INTRODUCTION

While measuring static dc breakdown potentials in hydrogen near atmospheric pressure with plane parallel electrodes, Lessin,[1] using apparatus previously described,[2] observed the following effects. With ultraviolet illumination of the cathode, it was noticed that as the generator voltage is gradually raised, the voltage directly across the electrodes initially increases, reaches a maximum value, and then decreases somewhat (by as much as 30 volts), still remaining stable. Further increase in the generator voltage results in the familiar cataclysmic irreversible spark breakdown, with a drastic reduction in the potential across the electrodes. It was also noticed in a rough way that the extent of the above effects depends on the intensity of ultraviolet illumination. The above observations were made with a resistance of 20 megohms in series with the electrodes. This resistance took up the difference between generator and gap voltages. These results imply the existence of a stable negative slope region of the current-voltage characteristics, heretofore unobserved at high pressures.

A number of authors have discussed the existence of a negative characteristic theoretically on the basis of space charge along with a concomitant lowering of the breakdown potential by ultraviolet light. One of the earliest attempts to formulate this problem quantitatively was made by von Engel and Steenbeck.[3] More exact calculations subsequently were made by Crowe, Bragg, and Thomas[4] for nitrogen and by Ward[5] for the rare gases. A lowering of the breakdown potential has been observed experimentally by a number of authors.[6] This lowering cannot be explained by the Townsend theory and must involve the action of space charge. The lowering of the breakdown potential by ultraviolet light may be observed in the absence of an external resistance; the observation of the negative characteristic, however, requires an external resistance. To our knowledge, very few negative characteristics have been observed experimentally, and these have been obtained at very low pressures. We know of no other observations of negative characteristics at high pressure. In conjunction with some static dc breakdown current-voltage measurements,[7] these negative characteristic effects were again noticed and investigated in more detail. The present paper is primarily a study of the negative characteristic feature rather than of the lowering of the breakdown potential, although the two effects are inextricably related.

PROCEDURE AND EXPERIMENTAL DATA

To investigate the negative characteristic at high pressure, a series of current-voltage runs were carried out in hydrogen at three values of the initial current. The measurements were made at a pressure of 400 mm Hg (at 22°C) and at an electrode separation of 2 cm. The measurements were carried out using the equip-

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[1] I. Lessin, Ph.D. thesis, New York University, 1953 (unpublished).

[2] L. H. Fisher and B. Bederson, Phys. Rev. **81**, 109 (1951).

[3] A. von Engel and M. Steenbeck, *Elektrische Gasentladungen* (Julius Springer, Berlin, 1934), Vol. 2, p. 48ff.

[4] R. W. Crowe, J. K. Bragg, and V. G. Thomas, Phys. Rev. **96**, 10 (1954).

[5] A. L. Ward, Phys. Rev. **112**, 1852 (1958).

[6] See for example H. Hertz, Ann. Physik **31**, 983 (1887); J. S. Townsend, *Theory of Ionization of Gases by Collision* (D. Van Nostrand Company, New York, 1910); W. Rogowski and A. Wallraff, Z. Physik **97**, 758 (1935); H. J. White, Phys. Rev. **48**, 113 (1935); W. Fucks, Appl. Sci. Research **5**, 109 (1955).

[7] D. J. DeBitetto and L. H. Fisher, Phys. Rev. **104**, 1213 (1956).

ment, technique, and gas source described in reference 7. Currents were measured for a given run, i.e., a given value of initial current, with increasing voltage up to within about five percent of the maximum stable current for that run. Cataclysmic breakdown occurred on further raising of the generator voltage. At this point, eight determinations of the breakdown potential were made with no ultraviolet illumination of the cathode.[8] The generator voltage was then reapplied at a value corresponding to a current about five percent below that at which instability appeared, and another current-voltage characteristic was then obtained with decreasing voltage. The above procedure was followed for each of the runs. With suitable conditioning of the electrodes, the currents measured in a given run were reproducible to within ten percent of each other for data taken with both increasing and decreasing voltage, despite the large currents and intervening breakdowns. After completion of the the third run, the first run was repeated and the currents were again found to be within ten percent of the original measurements. The conclusions of this paper depend on the observation of rather small effects, and hence the above reproducibility is essential.

The experimental data (average values) are shown in Fig. 1. The data taken with the two larger initial currents each show a point of maximum voltage across the electrodes, and a negative current-voltage characteristic. From values of the first Townsend coefficient previously measured with the same apparatus, the initial currents were evaluated before and after each run by using measured currents at a voltage (14 400 v) low enough so that secondary ionization plays no appreciable role. The vertical line in Fig. 1 represents the average of all breakdown potential measurements made in the absence of ultraviolet illumination. This potential was determined twenty-four times during the observations and was reproducible to within 3 volts.

CALCULATIONS AND INTERPRETATION OF DATA

In the Townsend theory, the steady state current, I, in a uniform field is given by

$$I = I_0 \exp(\alpha d)/\{1-\gamma[\exp(\alpha d)-1]\}, \qquad (1)$$

where I_0 is the initial current released from the cathode, d is the electrode separation, and α and γ are the first and second Townsend coefficients, respectively. The corresponding Townsend breakdown condition is

$$\gamma[\exp(\alpha d)-1]=1. \qquad (2)$$

[8] As previously noted (footnote 15 of reference 7), with the experimental equipment used, the first evidence of a luminous discharge on raising the potential with no ultraviolet illumination of the cathode is not the commonly reported filamentary spark but a visible glow, first covering the anode, and subsequently completely filling the gap. With further increase of the generator voltage of just a few volts the voltage across the chamber falls precipitously with the passage of a filmentary spark. In the present breakdown measurements, no effort was made to distinguish between the potentials for the appearance of the glow and the spark.

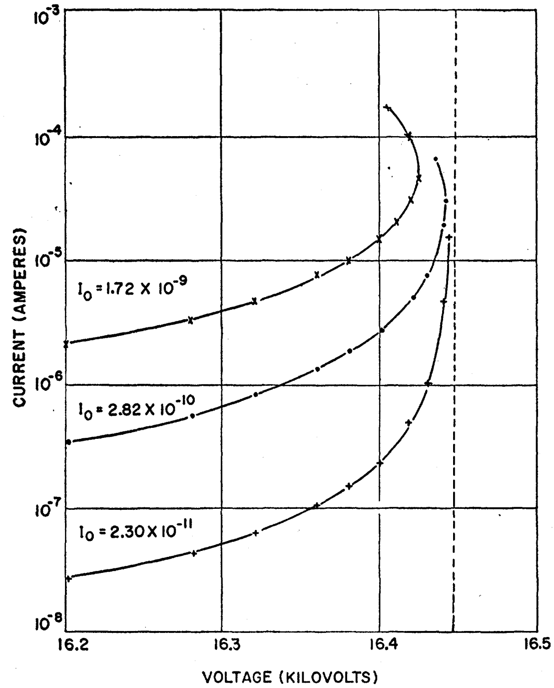

FIG. 1. Experimental current-voltage static characteristics in hydrogen for a pd product of 800 cm-mm Hg. The initial current in amperes is given for each curve. The vertical line indicates the average of 24 independent breakdown measurements made in the absence of ultraviolet illumination.

Equations (1) and (2) are based on the assumption that space charge in the gas plays no role whatsoever in determining the current or breakdown condition. That space charge plays a relatively minor role in determining the current and the breakdown potential, provided that the primary current is kept small enough, has been verified experimentally. This was first demonstrated long ago by Townsend and co-workers in gases at low pressure and more recently in gases near half an atmosphere.[9] However, Eqs. (1) and (2) can only hold approximately for any finite current, the approximation becoming better the smaller the current. Thus, space charge must play some role, albeit sometimes a small one, in determining the current and the breakdown condition.

Ward[5] used the differential equation and boundary condition which yield Eq. (1), together with Poisson's equation and an empirical expression for the dependence of α/p (p=pressure) on E/p (E=field strength) to calculate the effect of space charge on current-voltage static characteristics in rare gases. Similar calculations have now been made for hydrogen on an IBM 704 computer to cover the experimental data of Fig. 1. The expression

$$\alpha/p = A \exp(-Bp/E) \qquad (3)$$

[9] See for example F. Llewellyn Jones and A. B. Parker, Proc. Roy. Soc. (London) **A213**, 185 (1952); J. Dutton, S. C. Haydon, and F. Llewellyn Jones, Proc. Roy. Soc. (London) **A213**, 203 (1952). See also reference 7.

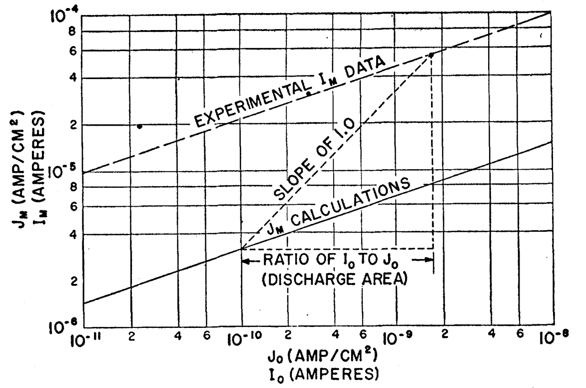

FIG. 2. Experimental variation (three points) of the current for maximum voltage, I_{max}, as a function of initial current, I_0, and calculated variation of current density for maximum voltage, J_{max}, as a function of initial current density, J_0. The line for the experimental points is drawn with the theoretical slope of $\frac{1}{3}$. A simple construction is shown to obtain the ratio of I_0 to J_0, i.e., the discharge area.

has been used in the calculations where A and B are constants. A fit of the experimental data of reference 7 yields $A=9.1$ cm^{-1} (mm Hg)$^{-1}$ and $B=143$ volt (cm mm Hg)$^{-1}$. These values of A and B yield α/p values that agree within a maximum error of two percent (approximately the scatter of the experimental data) for $18<E/p<22$ volt (cm mm Hg)$^{-1}$. The average breakdown potential for the data in Fig. 1 yields a value of γ of 9.68×10^{-4} if one uses Eqs. (2) and (3).

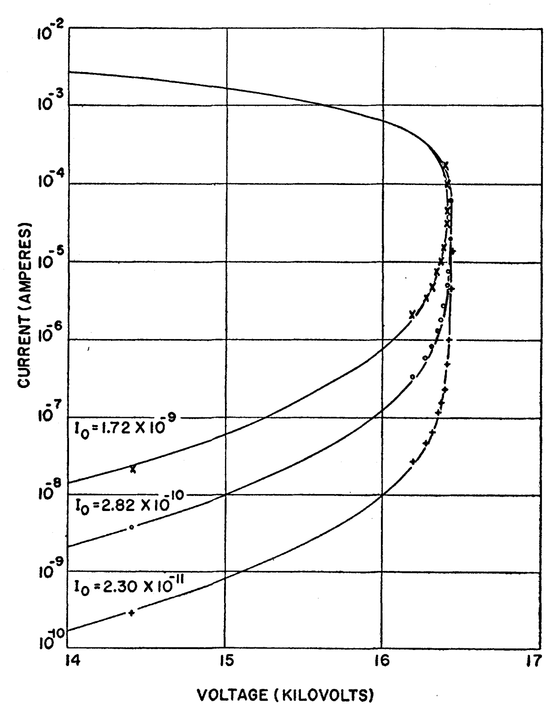

FIG. 3. Comparison of calculated (solid lines) and experimental (points) current-voltage static characteristics in hydrogen. A discharge area of 17 cm² is assumed for the calculations.

For the calculations γ was assumed constant over the voltage range of Fig. 1, and was rounded off to 10^{-3}. In addition, the positive ion and electron mobilities at atmospheric pressure, μ_+ and μ_-, were taken to be 14 and 460 cm^2 (volt sec)$^{-1}$, respectively.[10,11]

Computer calculations yield *current density*, J, vs voltage, V, whereas measurements yield *current* vs voltage. It is therefore necessary to assign a value for the discharge area, and this was done in a somewhat indirect way. The approximate calculations of von Engel and Steenbeck[3] showed that the current density for maximum voltage, J_m, varies as $J_0^{\frac{1}{3}}$, where J_0 is the initial current density, providing $J_0 \ll J_m$. Figure 2 shows both a log-log plot of values of J_m vs J_0 obtained from computer calculations for $p=400$ mm Hg and $d=2$ cm using the above values of A, B, γ, μ_+, and μ_-, as well as a log-log plot of the current for maximum voltage, I_m vs I_0 for the experimental data.[12] Excluding the point

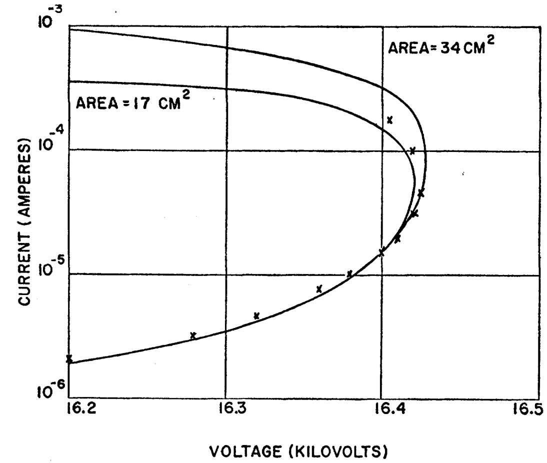

FIG. 4. Effect of assuming different areas for calculation of static characteristics for an initial current of 1.72×10^{-9} amp. Again, the experimental data are indicated by points.

corresponding to $I_0=2.3\times10^{-11}$ amp (which was obtained by questionable extrapolation of the parabola), the experimental data display very nearly the theoretical slope of $\frac{1}{3}$. The area which, when multiplied by both J_m and J_0, will bring the calculated and experimental curves into agreement is 17 cm². Values of J_0 were then calculated from the experimental values of I_0 and the above area. Using these values of J_0, and the same values of A, B, γ, μ_+, μ_-, p, and d as above, machine calculations were then made to yield values of V vs J. These J values were then transformed to V vs I values, again using the above area.

The results of these calculations are shown as solid curves in Fig. 3. The points of Fig. 3 are the experimental values of Fig. 1. The fit of the calculations to

[10] E. J. Lauer, J. Appl. Phys. 23, 300 (1952).
[11] N. E. Bradbury and R. A. Nielsen, Phys. Rev. 49, 388 (1936).
[12] In both cases, J_m and I_m were calculated by a three point parabolic interpolation of log$I$ (or of log$J$) vs V values.

the experimental points seems satisfactory, considering that both the area and γ were assumed to remain constant. This agreement would seem to indicate the reasonableness of the space charge explanation of the negative characteristics. The lowering of the breakdown voltage is also given by the calculations with reasonable accuracy.

At the largest experimentally measured current, the field at the cathode is calculated to be seven percent higher than the average applied field. Thus, for the currents for which experimental data are available, the E/p values are well within the region for which Eq. (3), with the values of the constants A and B as used, is valid, namely $18<E/p<22$ volt (cm mm Hg)$^{-1}$.

Although the assumed discharge area for the calculations is 17 cm^2, the illuminated area (image of the arc) of the cathode was only about 10 cm^2, but this is a lower limit if the possibility of reflections of ultraviolet light from the anode and walls onto the cathode is considered. The previously mentioned glow covering the entire anode suggests possibly a much larger discharge area. The calculations are not too sensitive to the precise area chosen. This may be seen in Fig. 4 where calculations are shown for the largest value of I_0 for 34 as well as for 17 cm^2. Taking all of the above factors into consideration, 17 cm^2 is not an unreasonable area to assume, although the proper value is somewhat uncertain. In fact, it seems reasonable to assume that the effective discharge area varies somewhat with current even below breakdown.

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Cubic Field Splitting of *D* Levels in Metals

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The splitting of the fivefold degeneracy of free atom d electron states by nonspherical components of a crystalline field is calculated. The crystal potential employed is that of a lattice of positive point charges screened by a uniform distribution of electrons. The calculation is done to first order in the cubic field, using hydrogenic electron wave functions. The triply degenerate d state is lowered with respect to the doubly degenerate one in both body-centered and face-centered cubic lattices. Numerical results are given for both lattices. Finally, analytic atomic wave functions are used to estimate the splitting in iron and copper at the observed lattice spacing. The crystal field splitting of these levels is found to be much smaller than the overlap splitting as obtained in previous calculations for both materials.

I. INTRODUCTION

In a recent calculation, perturbation theory was used to study d bands in the body-centered cubic lattice.[1] The crystal potential employed was that produced by a lattice of point charges of atomic number Z and lattice parameter a, screened by a uniform distribution of valence electrons. The energy of an electron state in this potential can be expressed as (atomic units throughout)

$$E=(Z/a)f(Za)$$

where $f(Za)$ is a function proportional to $(Za)^{-1}$ for small Za. The parameter Za is a measure of the tightness of the binding, and the perturbation calculation reported contained the first three terms in the expansion of the function $f(Za)$ for certain interesting states. The calculation could only be expected to be valid for small Za, but the apparent behavior of the series for large Za led to the conjecture that the d bands in this limit would split into two sub-bands. Within the model considered, such a split could arise only from the departure of the crystal potential around a lattice site from spherical symmetry.

The situation is similar in many respects to that considered in crystal field theory, in which the presence of cubic terms in the crystal potential produces a split of the fivefold degeneracy of the d states of a single electron into a triply degenerate state T_{2g} and a doubly degenerate state E_g.[2] We estimate this splitting for a metal. The calculation, which utilizes the ideas of the tight-binding approximation, considers the potential produced by the lattice of point charges previously mentioned; it can probably be generalized to more complicated situations.

To understand the physical situation in more detail,

[1] J. Callaway, Phys. Rev. **115**, 346 (1959). For a review of energy band calculations, see J. Callaway in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1958), Vol. 7.

[2] A recent review of crystal field theory is given by W. Moffitt and C. J. Ballhausen, Ann. Revs. Phys. Chem. **7**, 107 (1956).