ON IONIZATION BY SUCCESSIVE IMPACTS, AND ITS ACTION IN LOW VOLTAGE ARCS.

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SYNOPSIS.

Ionization of gases by successive impacts by electrons; theory.—Stark has shown that the energy acquired by an atom when struck by an electron whose speed exceeds the critical value usually expressed in terms of the minimum radiating potential may be retained for a short interval of time before being lost by radiation, or otherwise. It is assumed that ionization may occur if a second impact occurs within this time interval, provided the sum of the energies of the two electrons exceeds that corresponding to the minimum ionizing potential. Expressions are derived for the *total ionization, intensity of radiation* and *increase in total current when an arc strikes* due to successive impacts.

Low Voltage Arcs; Theory.—Arcs at abnormally low voltage may be accounted for by successive impacts. *Comparison with experiments* by McLennan on the *mercury arc* is made.

It is shown that the phenomena cannot be adequately explained by single impacts of electrons which, for one cause or another, have acquired abnormally high speeds. A combined action of radiant energy and energy of impact may be important.

INTRODUCTION.

THE conditions under which metallic vapors may be ionized by electrons passing through the vapor seem to be quite definitely established by the recent discoveries of a number of investigators.[1] In the case of mercury vapor, which is typical of all metallic vapors, it is found that electrons whose velocity is less than that acquired in falling through a potential difference of 4.9 volts, lose no energy at impacts with mercury molecules and leave the molecules apparently unchanged. As the velocity of the electrons is increased above 4.9 volts, the impacts may become inelastic and the energy of the impinging electron is transferred to the molecule, causing it to emit radiation of wave-length 2536 Å. U. The probability that this transformation of energy may occur increases approximately linearly with the excess of the impact velocity (in equivalent volts) above 4.9 volts. When the velocity is increased above 6.7 volts, a second type of radiation of wave-length 1849 Å. U. appears. At velocities equivalent to 10.4 volts or more,

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[1] Franck and Hertz, Verh. d. Deutsch. Phys. Ges., 10, p. 457, 1913 and 11, p. 512, 1914. McLennan and Henderson, Proc. Roy. Soc., A, 91, p. 485, 1915; Davis and Goucher, PHYS. REV., 10, p. 101, 1917; Tate and Foote, Phil. Mag., 3, p. 64, 1918. For summary see McLennan, Proc. Phys. Soc. Lon., 31, p. 1, 1918.

ionization of the mercury molecules may occur. At this voltage the many lined spectrum of mercury appears, although fairly strong ionization is necessary in order to enable any but the first members of the spectral series lines to be detected. If the ionization is fairly intense an arc strikes, and the normal minimum arcing potential appears to be 10.4 volts.

These minimum radiating potentials V of 4.9 volts and 6.7 volts are related to the frequencies ν of the corresponding radiations by the quantum relation $h\nu = eV$, and the two radiations are, respectively, the first members of the series $(1.5, S) - (m, p_2)$ and $(1.5, S) - (m, P)$, whose common convergence frequency $(1.5, S)$ corresponds, by the quantum relation, to the ionizing potential 10.4 volts.

If the vapor pressure is high and the bombarding electron stream very dense, however, it has been found that an arc will strike and the many lined spectrum appear at applied voltages much less than 10.4 volts.[1] The arc has struck at voltages as low as 4.8 volts and was maintained at voltages as low as 2.8 volts.

This appearance of the many-lined spectrum and the arc at abnormally low voltages has been variously accounted for by ascribing it to electrons emitted from the cathode or from the surrounding vapor by photoelectric action of the radiation excited by 4.9 volt impacts, to impacts of electrons against some of the molecules in such rapid succession that their energy is additive in its effect, to high initial velocities of the electrons emitted by the cathode at the high temperatures at which the phenomenon is observed or to joint action of radiant energy and energy of impact. Increased electron emission from the cathode due to its bombardment by positive ions may aid in sustaining the arc, but obviously cannot be a factor in producing the initial ionization of the gas. Of the above possibilities, photoelectric action on the vapor may probably be eliminated, since the photoelectric long wave-length limit of mercury vapor is almost certainly below 2536 Å. U., although this radiation does act photoelectrically on liquid mercury.

Photoelectric action of the 4.9 volt radiation on the cathode certainly takes place, but a little consideration of the conditions under which it occurs shows that it is entirely insufficient to account for the observed phenomena. Not all the bombarding electrons set up radiation by the mercury molecules. From the geometrical arrangement of the apparatus in which these phenomena have been observed, it is evident that much less than one per cent. of the radiation could fall on the cathode. The

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[1] Hebb, PHYS. REV., 9, p. 372, 1917; 11, p. 170, 1918; 12, p. 482, 1918; McLennan, Proc. Phys. Soc. Lon., 31, p. 30, 1918.

intensity of this radiation would be furthermore enormously decreased by the strong absorption of 2536 radiation by mercury vapor,[1] so that probably not more than 10^{-3000} of this one per cent. would reach the cathode.[2] Of this small amount of radiation reaching the cathode, only a small fraction actually yields equivalent photoelectric emission. A most favorable estimate indicates that the total photoelectric emission from the cathode is an entirely negligible fraction of the primary electron current. Furthermore, an application of Einstein's photoelectric equation shows that the maximum velocity of emission of these electrons from the cathode under the action of 2536 radiation is less than two volts, unless the photoelectric characteristics of the cathode are unexpectedly altered by its high temperature, and, in any case, the initial velocity is too small to account for ionization at voltages as low as those under consideration.

If the phenomenon is to be explained by high kinetic energy of emission of electrons from the hot cathode as a result of its high temperature, it is necessary to suppose that an appreciable number of these electrons are emitted with velocities corresponding to about six volts, or more, in order that their velocities after falling through the applied 4.8 volts may exceed the minimum ionizing velocity. Now it is well established that the emitted electrons have velocities distributed according to Maxwell's law, and with the same mean kinetic energy as that of gas molecules at the temperature of the emitting source.[3] A calculation on this basis, assuming the emitting source to be at 3000° K., shows that less than 10^{-9} of the emitted electrons have sufficient initial velocity to contribute toward ionization by simple impact under the conditions discussed. I have experimentally checked this calculated result by measuring the distribution of velocities of electrons emitted from tungsten near its melting temperature by the ordinary method of accelerating and retarding fields in a three electrode tube. Evidently the observed phenomena cannot be accounted for by high initial velocities alone.

These considerations appear to eliminate those explanations based on ionization by electrons which have, from secondary causes, acquired the 10.4 volt velocity. The remaining suggestions involve the combined

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[1] R. W. Wood, Physical Optics, 2 ed., Chap. 15, and papers.

[2] This assumes true absorption, rather than scattering, of energy, as in the theories of Drude and Lorentz. If the observed reduction of intensity were due to scattering, the reduction in energy would be much less than the amount stated. Experiments on the absorption of 2536 radiation by mercury vapor indicate that some scattering occurs, but that the amount of scattered energy is very small in comparison with the amount otherwise abstracted from the primary beam.

[3] Richardson and Brown, Phil. Mag., 16, p. 353, 1908; Richardson, Phil. Mag., 16, p. 890, 1908; 18, p. 695, 1909.

action of successive impacts, either directly or indirectly. The indirect action may be thought of as arising from the impact of an electron against an atom which has absorbed, but not yet reëmitted, a quantum of 2536 radiation which originated from a 4.9 volt impact against some neighboring atom. Owing to the very strong absorption of this radiation by the vapor, it seems very possible that the radiant energy may be passed on from atom to atom in such a manner as to maintain an appreciable proportion of the atoms in the unstable state in which they may be ionized by the impact of electrons whose velocities exceed about 5.5 volts.

Hebb,[1] who has also come to these conclusions with regard to the impossibility of accounting for the phenomena by simple impact, carried out a test of the hypothesis of a combination of impact and radiant energies by investigating the effect on the low voltage arc produced by illuminating the vapor in the discharge tube by 2536 radiation from a neighboring source, but with negative results. However, the great absorption of this radiation by the outer layers of vapor must have prevented its reaching the region of the vapor through which the electron stream was passing, so that the experiment is probably not a valid test of the hypothesis.

A more promising method of testing the hypothesis is to test the effect on the minimum ionizing potential produced by strongly illuminating the vapor at such low pressures that the light may penetrate strongly to the region where impacts occur. This is being done by Mr. Smyth in this laboratory, and the preliminary experiments have indicated a definite effect of the absorbed radiation in reducing the minimum ionizing potential.

The purpose of the present paper is to investigate the rôle played by direct successive impacts of electrons against atoms of the vapor. This possible method of ionization was first clearly pointed out by Van der Bijl.[2] Ionization may occur if an electron strikes an atom which has not yet lost the energy acquired from a preceding impact. Obviously the first impact must be by an electron with at least a 4.9 volt velocity, and the second must be by an electron with at least a 5.5 volt velocity, in order to make up the required 10.4 volts. The extent to which such successive impacts occur under given conditions may be estimated by applying well-known kinetic theory methods, *provided we know the time interval within which the two impacts must occur in order that their effects may be additive.* We have no information regarding this interval in the

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[1] Loc. cit.

[2] Phys. Rev., 10, p. 546, 1917.

case of mercury vapor. Recent work by Stark,[1] however, has shown that energy is not radiated after impacts as quickly as if the radiation began immediately after impact, but that an average interval of the order of 10^{-7} sec. occurs between the instant of impact and the instant at which the intensity of the radiation has fallen to l/e of its original value. Probably the most reasonable explanation is that this average time interval occurs between impact and radiation, since the radiation, once begun, takes place in a relatively short time. The largest time intervals found were $4\ (10)^{-7}$ sec. for the hydrogen H_6 line and $6\ (10)^{-7}$ sec. for the helium D_3 line. It would be of great interest to determine this interval for the mercury 2536 line. Since this has not been done, we shall proceed to develop several general expressions for the magnitude of effects due to successive impacts, and then to make a provisional application to the case of the low voltage mercury arc by assuming the time interval for mercury to be $6\ (10)^{-7}$ sec. This application may be revised when the interval for the 2536 line has been actually determined.

THEORY OF IONIZATION BY SUCCESSIVE IMPACTS.

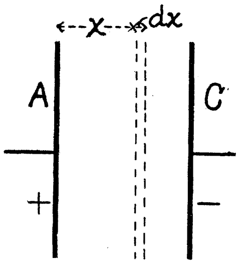

Fig. 1.

Consider the effect produced by electrons crossing a layer dx of the gas, on their way from the cathode C to the anode A under the applied difference of potential V (Fig. 1). If each electron makes, on the average, ν collisions with gas molecules while advancing 1 cm. toward the anode, νdx is the probability that a given electron will collide in the layer dx. Not all collisions are "effective" in the sense that they set up radiation or produce ionization. Let f be the probability that an impact may be "effective." The appropriate form of the function f will be discussed later. Then, if n electrons cross the layer dx per second per square centimeter,

$$\nu nf dx$$

is the number of effective collisions in dx per cm. per sec.

There are $pN'dx$ molecules per cm.^{2} in the layer dx, where p is the gas pressure in millimeters of mercury and N' is the number of molecules per unit volume at 1 mm. pressure. Thus

$$\frac{\nu nf dx}{pN'dx}$$

is the number of effective impacts per molecule per second in the layer dx.

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[1] Ann. d. Phys., 49, p. 731, 1916.

The average time interval between effective impacts against a given molecule is, therefore,

$$T = \frac{pN'}{\nu nf}.$$

By simple kinetic theory considerations it follows that

$$\epsilon^{-\frac{\tau}{T}}, \quad \text{or} \quad \epsilon^{-\frac{\nu nf\tau}{pN'}}$$

is the probability that a molecule will remain for a time equal to or greater than τ without being effectively struck. Therefore

$$P = 1 - \epsilon^{-\frac{\nu nf\tau}{pN'}} \qquad (1)$$

is the probability that a molecule will be effectively struck within a time interval τ. This is, therefore, the fraction of all molecules effectively struck which have been previously effectively struck within a time τ.

If impacts of electrons with gas molecules are inelastic, $\nu = pN$, where N is the number of collisions per electron per centimeter path in any direction—or the reciprocal of the mean free path—at 1 mm. pressure. Thus, for inelastic gases

$$P = 1 - \epsilon^{-\frac{nNf\tau}{N'}}. \qquad (2)$$

Under the most favorable experimental conditions, this expression shows that P is of the order of 10^{-8}, so that successive impacts cannot produce any detectable effects in inelastic gases. It is significant that no ionization at abnormally low voltages has been discovered in such gases.

In the case of gases in which the impacts below a critical speed are elastic, which includes the case of mercury and the other metallic vapors, ν is larger than pN on account of the zig-zag character of the paths of the electrons through the gas. In this case it has been shown that[1]

$$\nu = \frac{4r^2Up^2N^2}{X},$$

where X is the electric intensity at the layer considered, U is the average energy, in equivalent volts, of the electrons crossing this layer and r is a numerical factor whose value depends on the distribution of speeds among the electrons. r equals unity if the electrons all have the same speed, and r equals 0.87 if their speeds, relative to the mean speed of advance, are distributed according to Maxwell's law. Of necessity r lies between these values, and may be taken as unity in view of the

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[1] Benade and Compton, Phys. Rev., 11, p. 194, 1918.

relatively greater uncertainty regarding the values of some of the other quantities involved.

We have, therefore, for metallic vapors and other monatomic gases,

$$P = 1 - \epsilon^{-\frac{4UpN^2nf\tau}{XN'}}. \tag{3}$$

Since P is always a very small quantity, we may use the more convenient expression

$$P = \frac{4UpN^2nf\tau}{XN'} \tag{4}$$

with sufficient accuracy.

The appropriate form of the function f is not known exactly, but can be given accurately enough for the present purpose. It was found by Professor Bergen Davis[1] and by the writer[2] that theoretical expressions for the total ionization current agree very accurately with the actual currents if f is taken to be of the form

$$f = 0 \qquad \text{if} \qquad U \leqq V_0,$$

$$f = \frac{U - V_0}{U} \qquad \text{if} \qquad U \geqq V_0.$$

Experimental determinations of the total ionization by slow moving electrons, made by Johnson[3] show, among other things, that f is given quite accurately by

$$f = \frac{U - V_0}{k} \qquad \text{if} \qquad U \geqq V_0, \tag{5}$$

where k seems to be of the order of magnitude of V_0, though differing somewhat for different gases. Within the range of values of U in which we are interested, these expressions show practically the same variation of f with U.

Substituting for f from equation (5) we find, finally,

$$P = \frac{4pN^2nU(U - V_0)\tau}{XV_0N'}. \tag{6}$$

APPLICATION TO IONIZATION IN MERCURY VAPOR.

In order to test the possibility of accounting for ionization in mercury vapor at abnormally low voltages by the mechanism on which this expression is based, I have substituted in equation (6) experimental values taken from one of the most favorable cases cited by McLennan.[4]

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[1] PHYS. REV., 24, p. 93, 1907.

[2] PHYS. REV., 7, pp. 501, 509, 1916.

[3] PHYS. REV., 10, p. 609, 1917.

[4] Proc. Lon. Phys. Soc., 31, pp. 36, 37, 1918.

Unfortunately few of the quantities involved have been accurately measured, but it is possible to estimate them with certainty of at least the right order of magnitude. These estimates are based on the following considerations:

$p = 20$. This is based on the fact that there appears to have been more than 20 mm. of liquid mercury in the reservoir of the experimental tube, and this mercury was bubbling during the experiment. The electrodes were located immediately above the surface of the mercury.

$U = 5.6$ volts. The applied difference of potential was about 5.0 volts. To this must be added about 0.35 volt for the mean velocity of emission and a small term arising from the contact difference of potential between the tungsten and iron electrodes.

$n = 6\ (10)^{13}$. This assumes a thermionic current density of 10 microamperes. This value is close to that calculated by Langmuir's formula for the current when it is limited by the space charge around the cathode[1] and agrees with actual values found by Richardson and Bazzoni[2] under rather similar conditions.

$X = 2$ volts per cm. The electrodes were about 1.5 cm. apart and the total potential drop was 5 volts. Most of this drop, however, must have been near the cathode because of its much smaller size, whereas the region of the gas in which we are interested is near the anode.

$N = 75$, $N' = 3.6\ (10)^{16}$, $V_0 = 4.9$ volts are definite constants.

$\tau = 6\ (10)^{-7}$ sec. For reasons given above, this is much the most uncertain quantity involved. In view of this, it does not appear to be worth while to attempt closer estimates of the preceding quantities than those which we have made.

Taking these values,

$$P = 0.00018,$$

or about one impact in 5,000 would result in ionization if the applied voltage were about 5.0 volts instead of about 10.4 volts.

This means that the amount of ionization and arc spectrum radiation with the largest electron currents attainable at a 5 volt potential drop would be about the same as that obtained at the real ionizing potential with an electron current about 5,000 times smaller than the maximum. From an experimental study of the various conditions in which ionization may be produced, it seems as if this may accord with the facts, for very strong ionization of the vapor can be obtained with greatly reduced electron currents. Thus, while it is impossible to determine from data now available whether equation (6) *exactly* accounts for the phenomena

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[1] Phys. Rev., 2, p. 450, 1913.

[2] Phil. Mag., 32, p. 426, 1916.

of low voltage arcs, it is at least evident that ionization by successive impacts is a *possible* explanation.

Qualitatively, these considerations are more strikingly brought out by an estimate of the *total ionization* produced in the vapor by successive impacts, and the way in which this varies with the experimental conditions.

$\nu nfdx$ is the number of effective collisions in the layer dx per square centimeter per second. The fraction P of these follow a preceding effective impact within the time τ. Thus $P\nu nf$ is the number of neutral molecules ionized by successive impacts in the layer dx per second per unit volume. Calling this quantity I, and substituting, we have

$$I = \frac{16p^3N^4n^2U^2(U - V_0)^2\tau}{X^2V_0^2N'}. \tag{7}$$

The essential conditions for obtaining low voltage arcs are high vapor pressure, large electron current density and high filament temperature. The reason for the importance of these factors is at once evident in equation (7). In particular, the effect of high filament temperature is both to increase the current density n and to increase the factor $(U - V_0)^2$, which is very sensitive to the small changes in U due to variations in the mean velocity of emission as the temperature of the filament is altered. Obviously, ionizing by successive impacts cannot occur unless U exceeds V_0, which seems to be well established experimentally.

THE ARC SPECTRUM.

Since the rate of ionization is always so small that the number of neutral molecules is greatly in excess of the number of ions, the number of ions present will be proportional to their rate of production, or equal to kI per unit volume. Thus the number of ions struck per second per unit volume will be νnkI.

If we assume, with Stark, and as indicated by recent experiments on the subject, that visible radiation comes from the disturbance of atoms already ionized (while the disturbed neutral atom in general emits ultra-violet light), then the arc spectrum in the low voltage arc must be due to these impacts, and its intensity must be proportional to the number of such impacts. Thus we should have the intensity of radiation R given by

$$R = K\nu nI = \frac{64Kp^5N^6n^3U^3(U - V_0)^2\tau}{X^3V_0^2N'},$$

whence

$$R \propto \frac{U^3(U - V_0)^2p^5n^3}{X^3}. \tag{8}$$

As before, we see the great importance of large vapor pressure and current and high filament temperature.

Increase of Current when the Arc Strikes, and Subsequent Maintainance of the Arc.

It is commonly observed that the total current suddenly increases as the arc strikes. The increase is apparently much too large to be accounted for by a photoelectric effect of the radiation on the cathode, and can obviously not be accounted for directly by the addition of the ionization current. However the ionization current will act indirectly through the effect of the positive ions in partially neutralizing the space charge around the cathode, thus permitting the escape of additional electrons from the cathode. Owing to the large mass and consequent small speed of the positive ions, each will remain in the region of the effective space charge for a much longer time than an escaping electron, with the result that each positive ion will permit the escape of a large number of additional electrons.

If there were no electric field in the vapor, the speeds of the electrons and positive ions would obviously be inversely proportional to the square roots of their respective masses, or in the ratio of 600 to 1. Thus, if we assume that both electrons and ions are in thermal equilibrium with the gas molecules in the region surrounding the filament where the electric intensity is brought approximately to zero by the space charge, we see that, on the average, each positive ion remains in this region about six hundred times as long as an electron, and consequently permits the escape of six hundred additional electrons. This additional current will cause additional ionization, which will still further reduce the space charge, and so on. Calculating on this basis, I find that the total current should increase by approximately 15 per cent. in the example considered.

But there is an additional factor tending to still further increase the effectiveness of the positive ions in breaking down the space charge. The effect of the space charge is due to the accumulation of electrons around the filament to such an extent that the minimum potential of the system may not be at the cathode, but at a surface in space surrounding the cathode. The field thus set up is such as to cause an accumulation of positive ions in this region. In the extreme cases of high filament temperatures and low voltages in which we are interested, this feature of the space charge effects must be of considerable importance, so that it seems quite possible that ionization by successive impacts may adequately account for the phenomena of increased total current when the arc strikes.

CONCLUSIONS.

The considerations advanced in this paper lead to the conclusion that ionization and the arc type of radiation, observed under certain conditions, cannot be adequately accounted for by single electron impacts, but must be due to some sort of cumulative effect of successive impacts. The analysis of the problem shows that the additive effect of successive impacts is probably very important, and may prove to be sufficient to account for the phenomena. The cumulative effect of absorbed radiant energy and direct impacts may also play an important part.

It is desirable to make some experimental measurements of the quantities involved in this analysis, particularly of τ for the mercury 2536 line, in order that a more accurate test of the action of successive impacts may be made.

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