THE BLACKENING OF PHOTOGRAPHIC EMULSIONS BY LOW SPEED ELECTRONS*

By Kenneth Cole

Abstract

New type of electron spectrograph.—Electrons from an oxide coated filament are accelerated to a cylindrical anode, coaxial with the filament, having a narrow slit for one of its elements. The whole system is placed in a uniform magnetic field parallel to the filament. Those electrons which pass through the slit focus in the plane normal to that containing the filament and the slit. 22 volt electrons have been focused sharply. An extremely accurate direct determination of e/m should be possible with this arrangement.

The results indicate that the *blackening of photographic emulsions by low speed electrons* is due to radiation produced by electron impact on the surface of the emulsion and is a discontinuous function of the electron speed. For electrons with speeds below 100 volts, the blackening apparently depends upon the grain size and gelatine thickness of the emulsion. A thin film of fluorescent lubricating oil on the emulsion greatly increases its sensitivity. Schumann plates have an enormous sensitivity.

I. Introduction

PHOTOGRAPHIC plates have long been used to record the point of impact of electrons and ions.[1] In recent years, they have been used to a certain extent to measure the number of charged particles,[2] since for high velocities it has been found that the blackening of the plate is proportional to the number of electrons striking it.[3] Little, however, seems to be known about the sensitivity of a photographic emulsion as a function of either the energy or the velocity of the charged particles.[4] So far as has been found, no work has been done on the sensitivity to electrons with velocities corresponding to one hundred volts or less.

In the present work it was planned to investigate the mechanism of the blackening of a photographic emulsion by electrons. There seemed previously to be little evidence† to show whether the sensitive crystal

* Presented at the Washington meeting of the American Physical Society, April, 1926.

[1] See for example, J. J. Thomson, Rays of Positive Electricity; F. W. Aston, Isotopes.

[2] See for example, C. D. Ellis, Proc. Roy. Soc. (A) **99,** 261 (1921); H. Robinson, Proc. Roy. Soc. (A) **104,** 455 (1925).

[3] W. Bothe, Zeits. f. Physik **8,** 243 (1922).

[4] O. H. Smith, Phys. Rev. **7,** 625 (1916); O. Klemperer, Zeits. f. Physik **34,** 532 (1925).

† After the present work was begun, Rollefson and Poth (Science **62,** 497, 1925) published a preliminary report on work of a similar nature in which critical potentials of the elements contained in the emulsion were investigated to as low as 173 volts. This indicated that the process was essentially one which involved radiation as an intermediate step.

was rendered developable by the direct impact of the electrons on it, or whether the process was essentially photographic: i.e., the electrons producing upon impact a radiation which in turn affected the plate. In either case, it would seem that there should be a limiting value of electron velocity below which there should be no effect. Accordingly, the present work is concerned with the effect of low speed electrons, i.e., below one hundred volts.

Preliminary experiments, made in this department in the spring of 1925 by H. A. Smith, indicated that there is a lower limit to the velocity of electrons which would produce perceptible blackening upon impact. It was thought that the difference in the average depth of the sensitive material beneath the surface of the gelatine might account for the fact that this threshold was different for different emulsions, since the penetration of low velocity electrons into gelatine is very small.[5]

II. Apparatus

Electrons from an oxide coated filament F, Fig. 1, were accelerated to the anode A, in which a narrow slit was cut. Those electrons which passed through the slit then moved in circular paths under the action of a uniform magnetic field normal to the plane of the figure. When the had traversed a semi-circumference, they fell on the photographic plate.

The magnetic field was produced by a pair of Helmholtz coils. Each coil had an average radius of 31.4 cm and consisted of 142 turns of two No. 18 D.C.C. copper wires wound parallel. These could be connected in series or in multiple. When they were in series, the magnetic field produced at the center of the pair was 8.14 gauss per ampere. At a distance of 8 cm from the axis, the computed value of the field differed from that on the axis by less than .2 percent.[6] The axis of the pair was placed in the direction of the earth's field (approximately 75° dip) as determined by dip needle and flop coil. The magnetic field at the center of the pair was then entirely along the axis, and consisted of two parts: (a) that due to the current in the coils, and (b) that due to the earth's field. These were in the same direction for this work.

It was obviously necessary to eliminate the field due to the heating current of the filament. This was accomplished by means of a motor driven commutator which alternately applied the heating current and the accelerating potential. The heating current was only applied about one tenth of the total time. This arrangement also eliminated the poten-

[5] A. B. Wood, Jour. Inst. E. E. (English) **63**, 1046 (1925).
[6] A. Gray, Absolute Measurements in Electricity and Magnetism.

tial drop along the filament and made it an equipotential source of electrons.

The current for the Helmholtz pair was furnished by four sets of fifteen storage cells each, connected in multiple, and was measured by means of a Leeds and Northrup potentiometer connected across a standard one ohm resistance in series with the coils. The current could easily be measured and kept constant to less than .001 ampere.

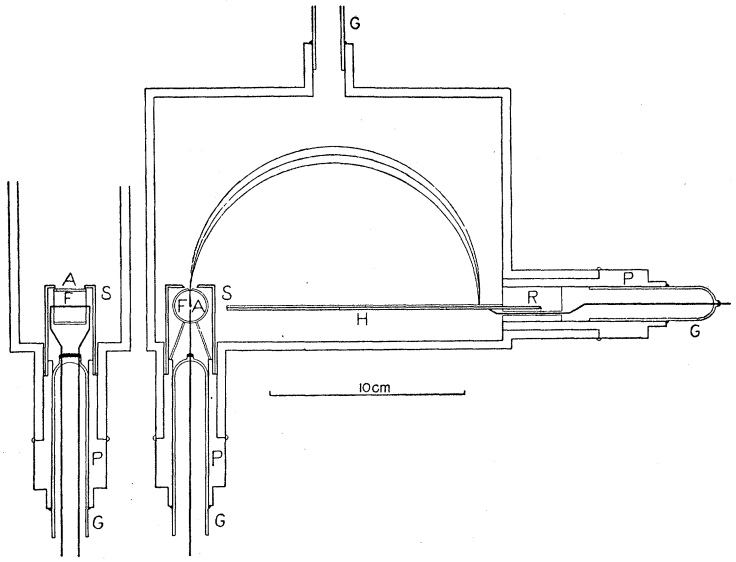


Fig. 1. Diagram of electron spectrograph.

The electron camera proper, as shown in Fig. 1, was constructed from 3/16 in. brass with all joints carefully soldered. The interior of the camera was covered with lampblack in order to reduce the scattering and reflection of stray electrons and light. The two brass plugs *PP* which carried the plate holder *H* and the filament *F* with the slit system were sealed to the outer ends of the tubes in the camera with hard grease so that any vapor had to diffuse through a long narrow path in order to reach the interior of the camera. The leads to the filament *F* and the plate holder *H* were sealed into glass tubes *GG* which in turn were sealed to the plugs *PP* with DeKhotinsky cement. The plate holder *H* was insulated from the plug *P* by the hard rubber plug *R*. The electrons which hit the plate were conducted through the emulsion to *H*, and then through a galvanometer having a sensitivity of 12,000 megohms which thus measured the number of electrons actually striking the emulsion.

Oxide coated filaments were used exclusively. The most satisfactory filaments were made by dipping the thoroughly cleaned platinum wire, about .25 mm in diameter, in a solution of 1 gram of barium nitrate and 1.5 gram of strontium nitrate in 100 cc of water, and then heating to a dull red in an atmosphere of carbon dioxide. When this process was repeated thirty or forty times, the filament had a very uniform and quite adherent white coating. Filaments of this type seem to be inherently unsteady, but the added heat capacity of the large diameter filament aided materially in reducing both the magnitude and the abruptness of the variations.

The vacuum system consisted of a rotary oil pump, a specially designed mercury vapor pump, McLeod gauge, and liquid air trap. The connection to the camera at G was made with DeKhotinsky cement. On the basis of kinetic theory, there was no need to keep the pressure below .1 micron, but it was usually kept below .01 micron. It was found, however, that when the pressure rose to such a value that about 40 percent of the electrons accelerated by 80 volts suffered impact before reaching the plate, there was a general fog over the entire plate. This may have been due to the production of radiation by the collision of the electrons with the gas molecules.

In the first slit system, the electrons were accelerated to a narrow slit and then allowed to pass through a wide slit. Later, the electrons were accelerated between the filament and a wide slit about a centimeter from the filament. The next trial was a portion of a cylinder having a very narrow slit for one of its elements with the filament on its axis. This was very satisfactory for electrons having velocities corresponding to 50 volts or more. This led to the final form in which the filament F is mounted on the axis of the complete cylindrical anode A which contains the slit as is shown in Fig. 1. The shield S was used to cut off both stray light and electrons. All of the plates shown in Fig. 5 were taken with a filament .25 mm in diameter and an anode of .79 cm radius containing a slit .09 mm wide.

Theory of the anode slit. This was suggested by Hull's magnetron[7] and is as follows: Suppose an electron is moving in a uniform magnetic field H which is normal to the plane of Fig. 2, and that the only electric field is radial from the origin of coordinates. Then the angular momentum G of an electron, about the origin, at a distance r is given by

$$dG/dt = Herv_r = Her \cdot dr/dt.$$

[7] A. W. Hull, Phys. Rev. **18**, 31 (1921).

Integrating, $G=\frac{1}{2}Her^2$. But since $G=mrv_\theta$,

$$v_\theta=e/2m \cdot Hr. \tag{1}$$

If we consider the electron to start from the origin with zero velocity then calling the potential zero at the origin and V at r, we have from the conservation of energy,

$$v=\sqrt{2e/m} \cdot \sqrt{V}. \tag{2}$$

The angle ϕ between the tangent to the path and the radius vector is given by

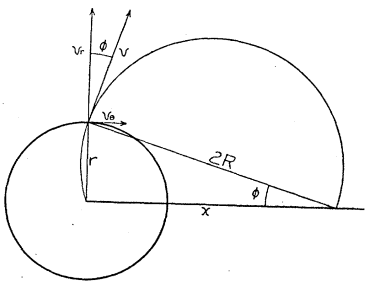


Fig. 2.

$$\sin\phi = v_\theta/v = \sqrt{e/8m} \cdot Hr/\sqrt{V}. \tag{3}$$

(It is interesting to note that when $\phi=\pi/2$, we have Hull's equation for the magnetron, $H=\sqrt{8m/e} \cdot \sqrt{V/r_{max}}$.)

When the electron moves with a velocity v under the action of the uniform magnetic field alone,

$$v=e/m \cdot HR$$

where R is the radius of the circular path. Then by substitution in Eq. (1)

$$\sin\phi=r/2R. \tag{4}$$

The condition for the focusing of the electrons is that they shall traverse a semi-circumference after leaving the narrowest aperture in their path. It is obvious from Fig. 2 that this condition is satisfied on the line through the origin perpendicular to the radius through the slit.*

* There should be no great difficulty in using this arrangement for the direct determination of e/m with a high degree of precision. It has not been possible to make more than a very rough check with the present apparatus due to errors in the alignment of the filament and slit. There is some doubt as to the exact portion of the line that should be measured. The theory of the focusing makes it obvious that the edge of the line away from the slit is the point to be considered, as it probably would be if the electrons were being caught in a Faraday cylinder. However, in the blackening of the plate, the width of the line seems to be due to a lateral spreading of the photographic image for it depends more upon the exposure than upon the error in the focusing or the mutual repulsion of the electrons constituting the beam. With increasing exposure it was found that the line broadened out by about the same amount on each side of the narrowest line that was visible. The narrowest line that could be easily found under a comparator was less than .1 mm wide and it was possible to set on an edge to within .01 mm. The only lower limit to the width of the line seems to be one of visibility. In this work the filament did not remain perfectly straight and it is fairly certain that this is the explanation of most of the bent and tilted lines which appear on some of the plates. It should be noted however that all of the lines, particularly those having small radius of curvature of the electron path, are concave towards the slit due to the "crossfiring" of the electrons.

This means that the electrons, accelerated by a potential difference V, which emerge from the slit will focus on this line according to the relation,

$$HR=\sqrt{2m/e}\cdot\sqrt{V} \tag{5}$$

or

$$HR=3.36\cdot\sqrt{V} \tag{6}$$

when H is in gauss, R in cm, and V in volts.

The nomogram shown in Fig. 3† has been found extremely useful in making approximate computations from Eq. (6). By setting a straight edge on the values of two of the variables, the value of the third may be read on its scale.

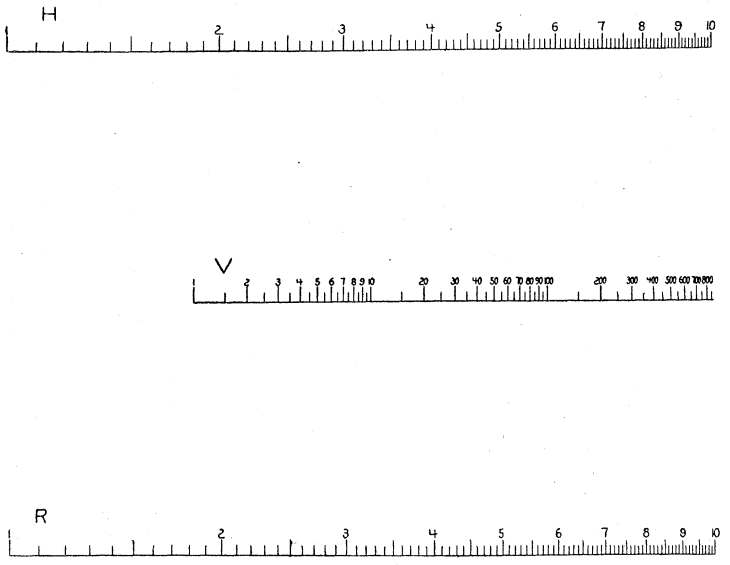


Fig. 3. Nomogram for approximate calculation of H, V, or R.

In the above theory, no account has been taken of space charge or of the finite size of the filament. The effect of space charge can probably be ignored entirely for this work as the electron current to the anode was only a few micro-amperes. In discussing space charge in the magnetron, Hull states that it does not cause an appreciable effect as long as r is less than about $.9r_{max}$. In this work, r was always less than $.1r_{max}$.

† Photographic reproductions (6 by 9 in.) of the carefully ruled original of this figure are available for anyone interested in this field.

As noted above, there is a decided advantage in having a heavy filament from the standpoint of steadiness. If the radius of the filament is r_o, then from Fig. 4

$$\theta \leqq r_o/r.$$

The width of the line δ is approximately

$$\delta = 2R(1-\cos\theta),$$

and so when r_o/r is .28 or less, δ should be less than one percent of the diameter of the path. Since the electrons acquire almost all of their final velocity within a very short distance from the filament, because of the intense electric field in its vicinity, it is all the more important that this field be strictly radial. With the larger filament, the surface intensity is decreased and the effect of such irregularities as are always present on the surface of an oxide coated filament is lessened. From Hull's general equation for the angle between the tangent to the path of the electron and the radius vector from the filament it is seen, however, that when r_o/r is greater than about .1, the error in tan ϕ will be about one percent, so that this is the real limitation on the filament size. In this work the filament was small enough to make r_o/r only about .016.

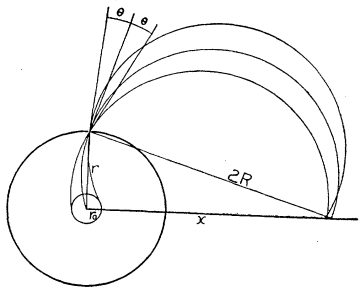


Fig. 4.

The accelerating potential was supplied by forty-eight radio B storage cells connected to a high resistance slide wire rheostat which served as a potentiometer. The potential across 1000 ohms of a 100,000 ohm resistance connected across the accelerating voltage was measured by a Leeds and Northrup potentiometer. It was possible to measure the accelerating potential and keep it constant to about .02 volt. The potentiometer slider was designed to make continuous velocity spectra, as shown in plate N, Fig. 5, for all of the work. When the point of impact of the electrons on the photographic plate moves at a uniform rate and the electron current to the plate is kept constant, the density of the incident charge will be the same over the whole plate. Then any variations in blackening will be due to the variations of the sensitivity with electron velocity. Accordingly, the potentiometer slider was operated by a cam (driven at very low speed) such that the accelerating potential was varied as the square of the time. Then R varied directly

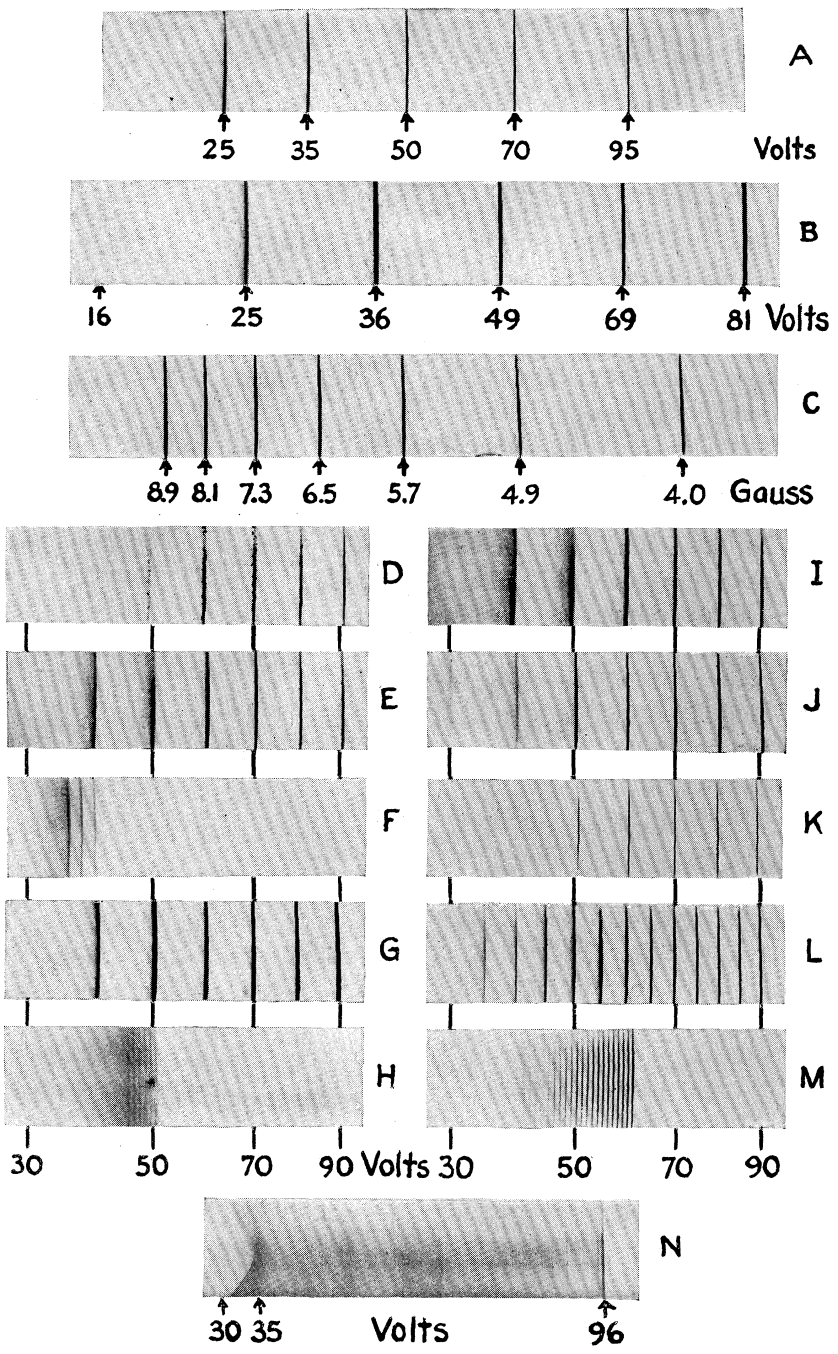


Fig. 5. Electronic spectrograms (full size). See Table I for explanation.

as the time and the point of impact moved across the plate with approximately constant speed. The current to the plate was maintained constant by adjusting the filament current. Only a few plates were taken by this method because of the difficulty of maintaining the plate current constant with the varying potential and also because of the length of exposure required for the unoiled* plates.

III. Explanation of Plates

The plates in Fig. 5 show the sharpness of the focusing of the electrons by the anode slit and illustrate the dispersion which can be obtained with the apparatus. Plates *A* and *B* were taken when the alignment of the filament and slit was fairly good. There is no fog on these plates, the lines are sharp and straight, and they have about the same density throughout their length. They also show to a certain extent the effect of exposure on the width of the lines for it was difficult to measure the charge accurately with the present apparatus before the electron emission of the filament had steadied down. Plate *C* illustrates the effect of the strength of the magnetic field upon the position of the line.

Table I

Data explanatory of Fig. 5.

	H gauss	*V* volts	Charge e.s.u.	Plate
A	6.5	—	240 approx.	Com. oiled
B	4.46	—	90 approx.	Com. oiled
C	——	50	60	Com. oiled
D	7.3	90 to 30 by 10 volt steps.	2400.	L.S. unoiled
E	7.3	90 to 30 by 10 volt steps.	2400.	Com. unoiled
F	7.3	40 to 28 by 2 volt steps.	1800. approx.	Com. unoiled
G	7.3	90 to 30 by 10 volt steps.	5.	Sch. A unoiled
H	7.3	50 to 25 by 1 volt steps.	1.	Sch. B unoiled
I	7.3	90 to 30 by 10 volt steps.	2400.	Com. unoiled
J	7.3	90 to 30 by 10 volt steps.	22.	Com. oiled
K	7.3	90 to 30 by 10 volt steps.	45.	L.S. oiled
L	7.3	90 to 30 by 5 volt steps.	30.	Com. oiled
M	7.3	60 to 40 by 1 volt steps.	30.	L.S. oiled
N	5.7	96 to 15 continuous	35 per mm	Com. oiled

Abbreviations: L.S. Eastman lantern slide (slow); Com. Eastman commercial plate emulsion No. 1834; Sch. A Hilger Schumann plate; Sch. B Hilger Schumann plate (another plate from same box).

The rest of the plates in Fig. 5, with the exception of *N*, were taken with rather small dispersion in order that all of the lines should fall on all of the plates, some of which were available in short lengths only. Plates *D*, *E*, and *G*, show the relative sensitivities of three different

* See below.

emulsions. From the table, it seems that greater charge is required to give plates of lower light sensitivity the same blackening as those of higher sensitivity. With the exception of the Schumann plates, it was found that those plates having the higher electron and light sensitivities were also blackened by lower velocity electrons. *D* shows a distinct difference in blackening between 60 and 50 volts and again between 50 and 40 volts. On *E*, the difference between 60 and 50 volts is very little, while the 40 volt electrons produced a very decided line and nothing could be found at 30 volts. The sharpness of this discontinuity is shown in *F*. The Schumann plates, *G* and *H*, showed enormous sensitivity to electrons accelerated by more than 40 and 45 volts, respectively. No trace of lines below 40 volts could be found on *G* even though it was very much over-exposed, while *H* which was cut from another plate in the same box showed a sharp discontinuity at 45 volts and also showed very faint lines down to 25 volts which show clearly on the original plate but did not reproduce in the illustration. This difference in the sensitivity is quite often found in different parts of the same plate as can be seen in *D* and *F*.

The effect of applying a thin film of oil to different emulsions is shown in plates *I* and *J* and those which follow. Extremely black finger prints were sometimes found on the early plates, but the reason for them was assumed to be entirely chemical until it was found that oiled plates have been used extensively in the spectroscopy of the ultra-violet. For spectroscopic work, the difference in sensitivity between the oiled and unoiled plates is not great, while a comparison of plates *I* and *J* shows the great increase in sensitivity to electron bombardment due to the oiling. These plates also show that the oiled plate is blackened by lower velocity electrons than the unoiled. Many different types of oils were tried, but the best found were ordinary lubricating oils which showed a green fluorescence in the visible spectrum. Only enough oil was used to give the emulsion a shiny surface. This was accomplished by rubbing the emulsion with a piece of absorbent cotton which had a very small drop of oil on it. Usually the surface was then rubbed with a clean piece of cotton to remove any surplus oil. The oil caused a slight darkening of the emulsion. It was not found necessary to remove the oil from the plate before development. *J* and *K* are two different plates oiled as nearly alike as possible, while the smaller steps of *L* and *M* show the gradual diminution in blackening as the lower voltages are reached. Plate *N* was an attempt at a continuous velocity spectra. The control of the electron emission was rather poor and accounts

for the high velocity fine structure. The oil was thicker at the edges of the plates than at the center.

One concludes, therefore, that:

A. For unoiled plates

1. The electron sensitivity is roughly proportional to the light sensitivity.

2. The discontinuities in the velocity sensitivity are quite sharp for any one point in a plate, but may vary from point to point and plate to plate.

3. The velocities at which the discontinuities in the sensitivity occur decrease as the electron sensitivities increase, except for the Schumann plates.

B. For oiled plates

1. The electron sensitivity is 50 to 100 times as great as for the unoiled plates.

2. The electron sensitivity is still roughly proportional to the light sensitivity.

3. The discontinuities in the velocity sensitivity are usually much less abrupt than for the unoiled plates.

4. The velocities at which the discontinuities in the sensitivity occur decrease as the electron sensitivity and the oil thickness increase.

IV. Discussion

That the blackening of the unoiled emulsion is due to the production of radiation at the surface where the charged particles strike seems very probable. Otherwise we might expect that the bombardment of the neutral air molecules would make the emulsion developable, as of course is not observed. The fact that the lines broaden as the exposure is increased may then be explained by assuming that the radiation produced along a narrow line on the surface of the gelatine is strongly absorbed by the gelatine. Then as the exposure is increased, crystals at greater and greater distances from this line are rendered developable. It was found that when electrons were allowed to strike the back glass surface of the plate, a broad and fuzzy band was produced on the emulsion—yet no electrons had struck it. This blackening was of the same order of magnitude as for the direct impact on the emulsion. This shows that it was radiation not too far in the ultra-violet which produced the blackening. It was interesting to notice that the emulsion was blackened on the glass side only, when the electrons fell on the back side of the plate. This seems to indicate that the gelatine absorbed the radiation somewhat more strongly than the glass. On this basis,

we should expect plates having comparatively little gelatine to be very sensitive and this expectation is amply justified in the tremendous sensitivity of the Schumann plate. The variations from plate to plate of the discontinuity in the velocity sensitivity are then probably due to the absorption of the gelatine. This would make it seem inadvisable to attempt to extend the determination of critical potentials by the method of Rollefson and Poth below 100 volts until more is known of the mechanism of the process and the exact role played by the gelatine.

With the oiled emulsions, we are apparently dealing almost entirely with fluorescence excited by electron impact. It was found that there was little difference in the blackening of an oiled and unoiled emulsion, when the electrons hit the clean glass back surface of the plate. But when this surface was oiled and the emulsion unoiled, the speed of the arrangement was nearly as great as for the direct impact of the electrons on the oiled emulsion.

V. Suggestions for Further Work

The field which may be called either the extreme ultra-violet or the long x-ray region should yield many interesting and important results when attacked by this method.

The most important problems in the method itself are those of the effect of the grain size and the gelatine thickness of the emulsion. One of the most promising methods of attack on the latter is suggested by the work of Duclaux and Jeantet[8] who made extremely sensitive plates for the ultra-violet by dissolving off the greater part of the gelatine of an ordinary plate. The radiation and fluorescence of materials, solid, liquid, or colloidal, may be investigated by placing them in, or on the surface of, the emulsion.

This line of investigation was suggested by Professor F. K. Richtmyer, and this step is due to his enthusiastic interest. The author wishes to express his appreciation of the suggestions and assistance of the many people who interested themselves in the problem. The work is being continued.

Rockefeller Hall,
Ithaca, N. Y.
June 1926.

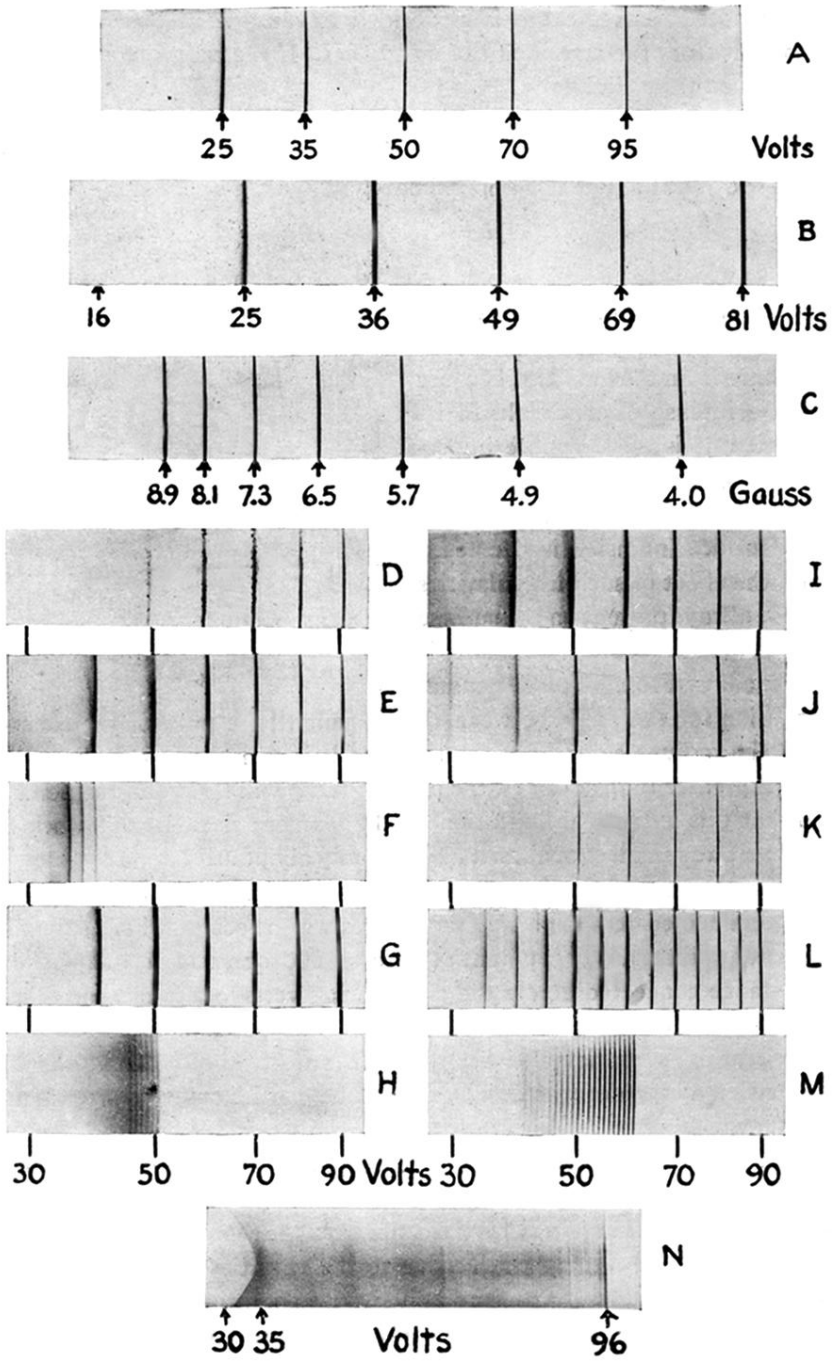
Note added with proof, September 3, 1926.—A "Note on the Photographic Effect of Slow Electrons," by G. F. Brett, Proc. Leeds Phil. Soc., Vol. I, Part I, Page 1, October, 1925 (Sci. Abs. **29**, 1564 (1926))

[8] Duclaux and Jeantet, J. de Physique et Rad. **2**, 156 (1921).

escaped notice until pointed out to the author after this article was submitted for publication. Stop-cock grease in ether was applied to kodak duplitized x-ray film, "the effect of the grease layer being to increase the sensitivity to 100 volt electrons several times. At the lowest voltage so far tried, viz., 65 volts, the sensitivity of the treated film was still maintained, although not to the same degree as with 100 volts." Grease was used because of its low vapor pressure. The "magnetic spectrum" method was used and the vacuum was tested by spark discharge.

As mentioned above, H. A. Smith in his senior thesis (unpublished), submitted in June, 1925, found that Eastman Portrait film was sensitive to electrons above a certain critical velocity (between 30 and 40 volts) but was not affected at lower velocities.

Although the data is not presented in this article, both oiled and unoiled duplitized films have been used. They were found somewhat more sensitive than the Eastman commercial emulsion 1834 and had about the same increase of sensitivity due to oiling. The "cut-off" was about 28 volts for the unoiled and 22 volts for the oiled emulsions. No difficulty was experienced from the vapor pressure of the oil or the curling of the film.



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