AN EXPERIMENTAL INVESTIGATION OF THE TOTAL EMISSION OF X-RAYS FROM CERTAIN METALS.

By C. S. Brainin.

THE experiments described in this paper were undertaken with the purpose of studying the integral intensity of the emission of x-rays by metals from two standpoints: (1) The variation of the intensity of the radiation from a given metal with the voltage and (2) the dependence of the intensity upon the atomic weight (or number) of the metallic radiator at different voltages. It seemed that the use of the Coolidge cathode and the gas-free x-ray tube gave opportunity for the maintenance of unchanging conditions such as it was hitherto impossible to obtain with the gas-filled tube. Moreover, beside this important consideration, the data upon which rest our ideas of the dependence of the total energy of emitted rays upon the voltage and the atomic weight do not seem to be founded upon experiment sufficiently extensive to preclude the desirability of further investigation upon this important topic.

The experiments upon which has been based a relation between the total intensity of x-rays emitted by a metal and the voltage across the electrodes of the tube are mainly those of Whiddington and Beatty. Their results are in agreement with the conclusion reached by Sir J. J. Thomson.[1] His theoretical investigation, founded upon certain atomic assumptions and upon the Stokes ether pulse or stopped electron hypothesis, brought him to the relation that the intensity of the x-rays produced by the collisions of electrons and atoms should be proportional to the fourth power of the velocity of the moving electrons. It should be proportional, therefore, also to the square of the difference of the potential impressed upon the electrodes of the tube. Furthermore, the results of the above mentioned experimenters in conjunction with those previously obtained by Kaye seemed to indicate that under like conditions of current and voltage the emission of two metals is very nearly directly proportional to their atomic weights. Both these results are summed up in the equation

$$E = KAP^2, \tag{1}$$

where K is a constant, A the atomic weight of the radiating target and P

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[1] Phil. Mag., June, 1907.

the difference between the electrodes of the x-ray tube. Recently Bergen Davis[1] has deduced this formula, basing his development upon the quantum hypothesis of radiation and assuming the x-rays to emanate from the atom itself and not from the impacting electron. Equation (1) is to be taken as applying only to the general or independent (non-characteristic) x-radiation. It is then in place to examine the experimental evidence so far brought forward in favor of this very broad formula.

Whiddington[2] makes the statement in an article on the " Production of Characteristic Roentgen Radiations" that " it comes out that E (per unit cathode ray current) is nearly proportional to the fourth power of the velocity." No experimental data connected directly with this question could be found in any of his papers. At best he was getting primary x-rays from one metal (silver) only; his range of voltages was from 7 to 21 kilovolts. His studies on the other metals were made with secondary rays produced by the primary rays from the silver anticathode. Beatty[3] used a method which was quite direct; he measured the total ionization produced in a chamber which absorbed completely the x-rays that entered it at the voltages used by him. His range was from 8.5 to 21 kilovolts and he investigated four metals, rhodium, silver, copper and aluminum. For the voltages used he found that rhodium, silver and aluminum held well to the voltage square law; his results for copper show a large increase at a voltage approximately corresponding to the critical voltage for the characteristic K-radiation from copper but hardly justify the acceptance of a straight line relation up to this critical voltage. In the case of aluminum the entire voltage range is of course above the critical voltage for its K-radiation. Rutherford[4] took up this question after the appearance of the Coolidge cathode and used a regular (commercial) tungsten target x-ray tube. He compared the emissions at three voltages, 48, 64, and 96 kilovolts and found that though the radiated energy was very nearly proportional to the square of the voltages, at the highest voltage the ratio was perceptibly greater.

As to the other factor contained in the equation (1), the atomic weight A, the direct evidence in its favor is again mainly the above-mentioned paper of Beatty and the work of Kaye.[5] Beatty found that the emissivity of rhodium and silver as well as that of aluminum were in the correct ratio in spite of the fact that the critical voltage for the K-radiation of the latter had been exceeded. Kaye studied the emission of a great many

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[1] PHYS. REV., Jan., 1917.

[2] Roy. Soc. Proc., 85 A, 1911.

[3] Proc. Roy. Soc., 89 A, 1913–14.

[4] Phil. Mag., Sept., 1915.

[5] Phil. Trans. Roy. Soc., 209 *A*, 1908–9.

metals and found that upon partially screening out the soft (characteristic?) rays the intensity of the remaining rays was approximately proportional to the atomic weights. This work was done at a voltage of about 25,000. No attempt was made to obtain complete absorption of the rays in the ionization chamber. However, the experiments certainly show the preponderance of the atomic weight as a factor in determining the emissivity of the substances at a given voltage. It is also interesting to note here that for voltages from 1,500 up to 3,500 volts Whiddington[1] found that no connection existed at all between emission and atomic weight.

A third point of interest which necessarily comes out in study of the variation of the x-ray emission with voltage is the behavior of the metals when the critical voltage for the appearance of the characteristic rays is reached. With the exception of aluminum as noted above in Beatty's paper, all the metals thus far studied, whose critical *K*-voltages lay within the experimental range, show a sharp increase in emissivity when these rays are excited.

The Experiments.

A great difficulty attended the choice of substances for these experiments, as it was badly limited by the necessity of having the element in metallic form and of having the temperature of fusion high enough to withstand the great heating to which they would be subjected. Finally the following six were chosen, so as to obtain the greatest range of atomic weights, and used throughout the work: Platinum, tungsten, silver, molybdenum, copper and cobalt. In order that the intensity of ionization might truly represent the intensity of emission a total absorption of the x-rays which, at any voltage, entered the chamber, was desired and provision for it made as described below. To avoid appreciable absorption of the x-rays in the wall of the bulb, it was planned to have the rays pass only through a very thin window of mica, before entering the ionization chamber so that the effect of this absorption could be neglected even at low voltages.

Description of Apparatus.—Fig. 1 gives a diagrammatic view of the very simple apparatus used. The Coolidge cathode, *C*, was mounted in a horizontal and the anti-cathode, *A*, in a vertical position. The target itself consisted of a brass block 2.5 cm. high and of hexagonal cross-section, to each facet of which a different metal was attached by means of tiny screws at top and bottom. As the little rectangular metallic targets were of different thicknesses, the brass block was trimmed down so that

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[1] Roy. Soc. Proc., 1911 *A*, 85, p. 99.

all the surfaces were at the same distance from the center. Into the base of the brass block was set one end of a narrow brass tube to which a bar of soft iron was fastened, making a right angle with the tube. This tube slipped over an aluminum rod, set up vertically in the x-ray bulb until an iron pin, which was affixed to the upper end of the aluminum rod, touched the inside of the brass block and thus provided a pivot for the latter's rotation. The soft iron bar was cut in two places and hinged there so that it could be folded up along the brass tube and the whole passed into the x-ray bulb through a wide glass tube fused in at the top. When this rotating target had been properly slipped over the aluminum rod, the wings of the soft iron bar were unfolded and the wide glass tube entrance sealed off. The adjustment had previously been made so that the centers of the surfaces of the target came opposite the cathode itself. The anti-cathode could now be rotated from without the tube by means of an electro-magnet. A mirror and a small sighting tube with cross-hairs were used to insure the turning of the surfaces so that the x-rays measured would come off each surface under like conditions. The figure shows the position of the target with respect to the cathode and the path into the ionization chamber.

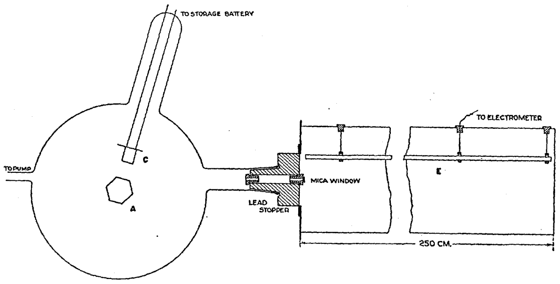

Fig. 1.

The x-rays passed only through a mica window of thickness .001 cm. before entering the ionization chamber. This thin sliver of mica was fastened with De Khotinsky cement to a small lead plug which itself was cemented to the large lead stopper. This in turn fitted into a glass tube fused to the x-ray bulb, the end of which was slightly flared. The lead and the glass were carefully fitted and cemented together. The whole had been previously adjusted so that the channel for the x-rays through the lead pointed toward the area from which the rays were to come. The holes through the small lead plugs were .18 cm. in diameter and had been so calculated that the radiation would traverse the length of the ionization chamber without coming in contact with the side walls. The axis of the ionization chamber had to coincide with the axes of the holes through the lead in order to effect this and the proper adjustments were made before the apparatus was assembled.

The ionization chamber itself consisted of a thin-walled steel tube of outside diameter 10 cm. and length 250 cm. This was used as a grounded electrode; the other electrode (*E*, Fig. 1) was a lead pipe stiffened by an iron rod within it which ran the entire length of the chamber. It was suspended inside the steel tube from four amber plugs, one of which carried a wire connection out to the electrometer. It was finally decided to use a Braun electrometer with an aluminum needle and measure the rate of leak over a definite range of 500 volts. The needle was always charged so as to indicate over 1,500 volts and the time was taken between 1,500 and 1,000 volts. As no readings were made with the difference of potential between the electrodes less than 1,000 volts, saturation conditions at all times were obtained in the tube. The electrometer needle showed no tendency to stick and gave results which agreed well with one another. The natural leak was practically always a negligible quantity. Proper heavy lead shielding was placed around the x-ray bulb to prevent effects in the electrometer due to secondary or reflected rays.

The x-ray bulb was evacuated with a mercury condensation pump of the Langmuir type made entirely of glass after a design by Prof. G. B. Pegram; it was used in conjunction with an oil fore-vacuum pump and gave extremely rapid service. A freezing-out chamber was fused in between the pump and the x-ray bulb and was kept immersed in a mixture of salt and ice. This served to keep the mercury vapor pressure in the bulb low enough to prevent the formation of a gas discharge. It took several days of alternate pumping and running of the discharge to remove the occluded gases and to make the bulb serviceable for the experiment; but once it had reached a satisfactory state it was found necessary to do but little pumping and several days of observations in succession were sometimes possible without any need of using the pump. In spite of the three De Khotinsky seals, made under difficult conditions, there was at no time a real leak perceptible and most of the runs of observations were made with the pump in readiness but not in actual use. Early in the stage of preparation of the tube it was filled with hydrogen and when this had been nearly all pumped out again a discharge was run for some time with the anti-cathode acting as a cathode. This served to clear perfectly the surfaces of the metal targets, some of which had become slightly oxidized.

The current for heating the tungsten filament of the cathode came from a storage battery placed upon a carefully insulated glass support. Four six-volt $2\frac{1}{2}$ ampere batteries were used, two parallel sets, each composed of two batteries in series, giving twelve volts. The electronic current through the x-ray tube was measured by means of a Weston milliammeter reading directly to tenths of a milliampere. One terminal of this meter was connected to the anticathode and the other grounded. A strip of tinfoil, which was grounded, was fastened around the x-ray tube near the connection of the anticathode and the milliammeter so as to intercept any possible leak over the surface of the glass and thus prevent it from being registered on the ammeter. The main circuit high potential current was obtained from a transformer fed by a 500-cycle, 150-volt alternator. The current from this was rectified by a kenotron system. A very large condenser was placed in parallel in the circuit so that the voltage could be relied upon to be constant and the current unidirectional. The middle of the transformer was grounded. The voltages were measured by means of a spark gap between spheres 12.5 cm. in diameter.

Method.—The data taken in these experiments were obtained in two distinct ways:

1. With a given metal acting as target the voltage was varied by small steps and the rates of the electrometer leak measured; in this way the ionization produced by the energy in the x-ray output of a particular metal could be obtained as a function of the voltage.

2. Keeping the conditions of the voltage and current constant, the different metal targets could one after another be subjected to the bombardment of the electrons and the relative intensities of the emission of x-rays for any particular voltage could be obtained. This was done for a great number of different voltages throughout the range found possible with the apparatus.

The lower limit of usable voltages is naturally determined by the amount of energy necessary to produce readable leaks on the electrometer in a reasonable length of time. It was found impossible to use such high currents through the tube as are possible with the tungsten Coolidge tubes prepared by the General Electric Company. A current over 5 milliamperes soon broke down the vacuum even at very low voltages. The upper limit in an experiment of this nature depends upon how far the voltage can be raised and practically total absorption still be obtained in the ionization chamber. This is really best determined from the results themselves and the shape of the curves obtained by the above method (1) is the best guide as to how high a voltage can be used.

In order to increase the absorption of x-ray energy in the chamber, it was planned to use some dense vapor and the experiments were begun with the chamber containing air saturated with ethyl bromide vapor.

However, the study of certain phenomena which were found and are hereinafter described made it desirable to have the air alone present, in order to avoid the great increase in ionization which takes place in the vapor when the x-rays absorbed by it contain wave-lengths which correspond to the characteristic *K*-radiation from one of the component elements of the gas. These would begin to appear, of course, when the voltage across the tube is equal to the critical voltage for the excitation of this radiation. This increased ionization is entirely out of proportion to the increased energy of the x-radiation due to the increased voltage alone, and difficult to correct for. With the long ionization chamber containing only dry air it was found that the highest voltage at which the results could be relied upon was in the neighborhood of 33,000 volts. Above 35,000 volts there appeared on all the curves a sudden great increase due to the impinging of a large amount of the ionization upon the end wall of the chamber. Tests with an electroscope showed that no appreciable amount of x-ray energy remained unabsorbed below that voltage. The lowest it was found possible to use was 4,700 volts, though in general the readings were taken above 7,000 volts.

During the progress of the experiments the first x-ray tube set up became so blackened that its further use was very difficult and at approximately the same time the glass pump cracked at one of its fused joints. The whole apparatus was, therefore, taken down; the pump was repaired and a new tube set up differing only in slight details from the former one. Data were then obtained on the metals which had not been examined and much of the data already obtained was carefully gone over. It was found that while the actual quantitative data differed by a constant factor, the general results remained exactly the same.

Experimental Results.

I.

In the results presented below in the form of curves, the energy, as represented by the rate of leak of the electrometer reduced to unit electron current, through the x-ray tube, is plotted as ordinate and the square of the voltage used is plotted as abscissa. If the resulting curve is a straight line pointing to the origin, the energy of emission is really proportional to the square of the voltage as expressed by the equation (1). A deviation from the straight line passing through the origin and before the critical voltage for the characteristic radiation is reached necessarily means a failure of the equation to represent truly the phenomenon.

Platinum and *tungsten*, the two metals of highest atomic weight in the list and the only two whose critical voltage for the *L*-radiations are within the range of the experiments, show a similarity in that they both deviate from the " square of the voltage " law. It is to be noticed particularly that these curves do not become straight lines through the origin until the voltage of 22,000 volts, approximately, has been reached in the case of platinum, and 19,000 volts, approximately, in the case of tungsten. It is to be noticed also that for a short interval of voltage the radiation of tungsten seems to be more powerful than that of platinum, a fact which is brought out again later in connection with the results from method (2).

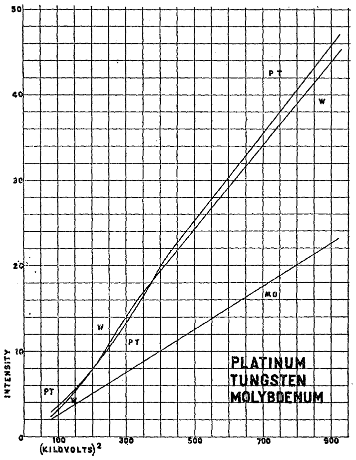

Fig. 2.

Molybdenum, as shown on Fig. 2, seems to hold well to the law for the range of voltages given and, not only that, but also appears not to deviate from this law upon the voltage reaching and passing the critical voltage for its characteristic K-radiations. For molybdenum this is about 20,000 volts. All the curves on Fig. 2 are mean curves based on a number of separate sets of observations on each metal.

Copper and *cobalt* also show a striking similarity in behavior and they are plotted together in Fig. 3. Especially in the case of copper is it possible to say that a straight line really represents the data up to the point where the curvature, due to the characteristic, begins to be noticeable. For cobalt also the curve may be said to be fairly close to a straight line. The critical voltage for cobalt is about 10,000 volts; that for copper is 11,000 volts very nearly. Upon approaching these voltages both curves bend gradually upward and show what we may be allowed, for the sake of brevity, to call a striking increase of " efficiency." This increase is greater for the metal of lower atomic weight, cobalt, at least within the range of the experiment.

The results for silver are shown separately in Fig. 4. The upper curve was obtained with the air in the ionization chamber saturated with ethyl bromide vapor, the other with air alone, without this vapor. Upon plotting the very first results obtained with silver, when the chamber still contained the vapor, and calculating the ratios between the ionization intensities and the squares of the voltages, the writer was surprised to find the slight downward break in the curve as shown in the figure, upon reaching the vicinity of the critical voltage for the silver K-characteristic radiations, 25,000 volts. Up to this point the curve is surely a straight line pointing toward the origin. To make certain of the real existence of this phenomenon, the data for silver were taken several times alternately with data for metals already examined, copper, platinum and molybdenum. In each case the results were a repetition of the data previously obtained. Then the ethyl bromide vapor was removed from the ionization chamber and dry air only used as absorber. The curve of the results so obtained shows the break much more prominently. The lessening of the effect in ethyl bromide vapor was no doubt due to the reaching of a voltage greater than the critical voltage for the *K*-characteristic radiations from the bromine atoms in the vapor. This voltage is about 17,000. In this case the increase in relative ionization power served to mask partly the decrease in the so-called efficiency of the silver radiating mechanism. Nothing could be found in the apparatus to explain away the anomaly, which would not apply equally well to the other metals by which, however, it was not shown. Beatty and Whiddington had not found this phenomenon because their highest voltages were below the voltage at which it appeared.

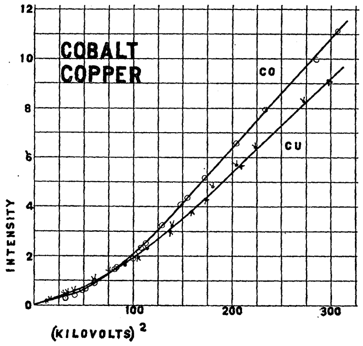

Fig. 3.

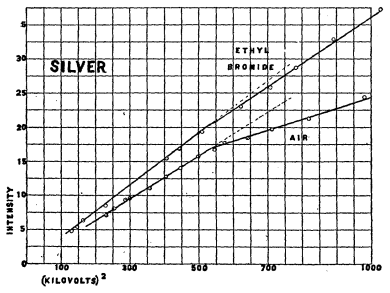

Fig. 4.

II.

The results obtained from the second experimental procedure are given in Fig. 5 below. The actual voltages and not the voltage squared are here used as abscissæ; the ordinates represent the ratio that the emission energy of the particular metal bears to the emission of molybdenum at the given voltage. Molybdenum was chosen as the unit of reference because it gave (in Fig. 2 above) the straight-line relation between voltage squared and emission intensity postulated by the formula $E=KAP^2$ and was therefore best suited to show deviations from this relation. Its own emissivity is naturally then represented by a straight line of unit ordinate. For a better understanding of the data presented by the curves, a table is added giving the atomic numbers and weights and their ratios to those of molybdenum.

TABLE I.

Metal.	At. Wt.	Ratio of At. Wt.	At. No.	Ratio of At. No.
Pt.	195	2.17	78	1.86
W.	184	1.92	74	1.72
Ag.	107	1.10	47	1.12
Mo.	96	1.00	42	1.00
Cu.	63	.66	29	.69
Co.	59	.62	27	.64

Examination of Fig. 5 brings out some interesting information and further confirmation of some of the results discussed above. The deviation of platinum and tungsten from proportionality in their emission intensity to atomic weight (or number) is very striking. If they really obeyed this relation exactly, the curves representing each would be a straight line parallel to the voltage axis and to the line representing molybdenum and at the proper distances from these. However, at the lowest voltage shown, 4,700, the relative emission is very much below this and it rises rapidly with the voltage, becoming actually the required straight lines in the neighborhood of 20,000 volts. At this stage the ratio of the emission of platinum to that of molybdenum, from the curve, is about 2.06 and that of tungsten about 1.93, which is closely in proportion to the atomic weights. Again there is a short interval of voltages where tungsten is a better radiator than platinum. We might also, perhaps, call attention to the fact that tungsten seems to reach the straight line relation before platinum and that the voltages at which this is reached are very roughly twice the critical voltages for their respective characteristic L-radiations.

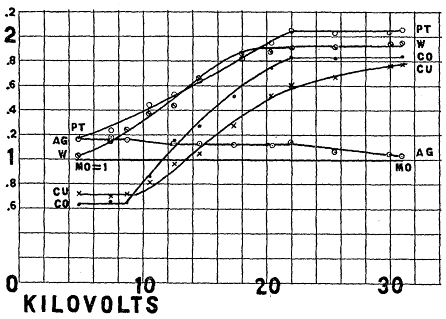

Fig. 5.

Silver appears to give an amount of radiation which is very slightly out of proportion to its atomic weight at the lower voltages, but falls pretty well into line above 10,000; at the lowest voltages its emissivity is as high as that of platinum and higher than that of tungsten. This was to be expected if silver maintains the straight-line relation between emission and voltage squared, but the slight apparent rise with respect to molybdenum itself cannot, of course, be thus accounted for. The decrease of the ratio for silver above 22,000 volts is a further confirmation of the curious downward break in the curve for this metal as shown in Fig. 4. Between these two changes the ratio to the emission of molybdenum is 1.13 nearly.

In the case of copper and cobalt the results are fairly well given by straight lines parallel to the molybdenum unity line until the appearance of the characteristic *K*-radiation energy. They both then increase rapidly, crossing the curves for molybdenum and silver, finally appearing to approach a limiting value, and do not increase indefinitely in relative emissivity. In the region below the critical voltages for their *K*-rays, the average of the ratios of their emissions to molybdenum are .71 for copper and .64 for cobalt, which is in good agreement with the values in the table. The constant ratio of 1.83, to which cobalt rises after 20 kv., has no easily interpretable meaning.

III.

Three distinct modes of behavior on the part of the metals, when the voltage is reached which corresponds to their critical voltages for the *K*-rays, have so far been discovered:

1. Most elements increase rapidly in the " efficiency " of their radiated energy output which is evidenced by a break upward, when the plotting of the curves is made, as in this paper; such substances are copper, iron, selenium, nickel, zinc, etc., which have already been studied, and cobalt which this paper adds to the list.

2. Aluminum was found early to be an exception in that it seemed to show no such break at all but gives evidence that it continues on in an unbroken straight line; as mentioned at the beginning of this article, Beatty bases part of his confirmation of the law of proportionality of emission to atomic weight upon the fact that aluminum maintains this relation throughout the range of his experiments. Molybdenum may also now be added to aluminum as an example of such behavior.

3. The behavior of silver, which decreases in " efficiency," with the appearance of its characteristic *K*-radiations.

It seems to the writer that the different modes of behavior recited above are rather antagonistic to the theory that independent and characteristic rays come from different sources of radiation. It has been sometimes held that the former are sent out from the impacting electron and the latter from the electrons of the impacted atom, in a similar way probably as the line radiation of the visible spectrum. One may then attempt to explain away the aluminum and molybdenum behavior by saying that the characteristic radiations in the case of these metals do not contain relatively as much energy as, for example, the characteristics of copper; and it is possible that a very slight increase of energy may go unnoticed in such experiments as these. On the other hand, the characteristic radiation from both aluminum and molybdenum have been found to be not at all weak. However, the one example of silver, unless refuted by later work, makes the above theory unlikely; for it would be rather difficult to understand why the energy emission from the impacting electron should not continue in proportion to the fourth power of the velocity. It seems to make it probable that the entire radiation comes from the atomic radiating structure itself, and, furthermore, that the reaction of this mechanism is not parallel in all atoms when their respective critical voltages are reached. The behavior of the independent radiations in the neighborhood of the characteristic lines of rhodium, as found by D. L. Webster[1] may be perhaps taken as a support in this direction.

SUMMARY.

1. A study has been made of the total intensity of the x-ray emission from the six metals, platinum, tungsten, silver, molybdenum, copper and cobalt over a range of voltages extending from about 5,000 volts to about 33,000 volts. The relations between the energy radiated and the voltage and between the energy and the atomic weight were investigated; this includes a test of the validity of the equation $E = KAP^2$.

2. Below 20,000 volts approximately the energy radiated by platinum and tungsten was found not to be proportional to A and P^2; above this voltage, however, it was in agreement with the above equation.

3. Molybdenum obeyed this relation throughout the range of voltages, and showed no deviation from this relation when the voltage was increased above the critical voltage for the K-radiation.

4. Copper and cobalt seemed to obey this relation below their critical voltages for the K-radiations, but above those voltages their emission increased *more rapidly* than is required by the "voltage squared law."

5. Silver also obeyed this law below the critical voltage for its characteristic K-radiation, but above that voltage the emission energy increased *less rapidly* than is required by the "voltage squared law."

In conclusion I desire to express my sincerest thanks to Prof. Bergen Davis who suggested the problem and whose kindly interest was a continual source of help and encouragement.

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[1] PHYS. REV., June, 1916.