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DIRECT AND INDIRECT PRODUCTION OF CHARACTERISTIC X-RAYS

BY DAVID L. WEBSTER

STANFORD UNIVERSITY, CALIFORNIA

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I. *The Problem.*—When electrons are ejected from the *K* orbits of atoms in the target of an X-ray tube, the question arises: Are most of them ejected by direct action of the cathode rays through their repulsive forces; or are they ejected by an indirect process, the photoelectric effect of continuous-spectrum X-rays excited by the cathode rays; or perhaps, do both processes occur often? From the experimental standpoint, this question takes the form: Are the characteristic rays from the target of a tube mostly “direct primary rays,” or are they mostly “indirect primary rays” (really a restricted class of secondary rays), or are they a mixture of comparable amounts of both classes?

Beatty,¹ in 1912, said they were mostly direct and presented such clear experimental evidence that his answer was accepted for many years as conclusive. In 1926, however Balderston,² by calculations from data of wholly different types, came to exactly the reverse conclusion. Evidently, the question calls for further investigation.

II. *Emergence-Angle Experiments.*—Two methods were used in the present work, both based on the fact that such indirect rays as may exist are produced at a variety of depths averaging somewhat greater than the mean depth of production of rays in the continuous spectrum. This is obvious qualitatively from the fact that, in any fairly heavy element, X-rays of the types needed for fluorescence of its *K* series are more penetrating than the cathode rays producing them.

If, therefore, any means can be found for revealing differences in the depths of production of X-rays, of these two classes, the question at hand can be answered. The first of the two methods was an adaptation of Ham's³ experiment on total, or unresolved, X-rays, applied here to the

rays as resolved by the spectrometer. The underlying principle is that if the rays from any atom are emitted with equal intensities in all directions, then the deeper the atom is within the target, the more effect the absorption by the target material will have in causing differences of intensity between rays emerging from its surface nearly perpendicularly or at small angles.

In the present case, the target was a layer of silver, 25 microns thick, electroplated upon copper. When it was bombarded by cathode rays driven by a steady voltage of 35 kv., d. c., the continuous-spectrum rays were produced, as will be shown below, at mean depths of less than a micron, while the indirect characteristic rays came from depths averaging several microns. The difference between these depths is such as to cause considerable changes in the ratio of indirect characteristic rays to continuous-spectrum rays of the same wave-length, when the emergence angle ϵ , between the target surface and the rays to the spectrometer, is reduced below 20° .

For quantitative calculation, this ratio must be defined more specifically in terms of spectrum graphs in which electrometer readings are plotted against glancing angles. As the tube is turned to reduce the angle of emergence, the change in apparent size of the focal spot, as seen from the spectrometer crystal, causes a change in the resolving power of the spectrometer. A simple ratio of the maximum ordinate of the $K\alpha$ peak to that of the continuous spectrum under it would, therefore, be misleading, and the only true basis for comparison is the ratio of the *area*, A , of the $K\alpha$ peak, to the *ordinate*, C , of the continuous spectrum in its neighborhood. This ratio, which is of the dimensions of an interval of glancing angles, gives the width of a block of continuous spectrum carrying the same total energy as the α lines.

The next problem is to calculate this ratio, assuming all the line radiation to be indirect. For this purpose the following assumptions, definitions and data were adopted, and will prove useful in the theory of the second method:

(1) The continuous spectrum, as emitted from any atom, is of the same form and intensity in all directions.

(2) Continuous-spectrum rays of the α -line frequency emerging from the target are reduced by absorption to a fraction, $\exp \{-\mu_\alpha \bar{x}_\alpha \csc \epsilon\}$ where μ_α is their absorption coefficient and \bar{x}_α their mean depth of production.

(3) The mean depth of production for rays of any frequency may be found by the method of Webster and Hennings,⁴ depending on the extent to which the K-absorption limit of the target material affects the observed emission spectrum.

(4) Mean depths as thus found for the present case are small enough

so that for calculation of the indirect radiation one may treat the continuous-spectrum rays as if they all originated at the surface.

(5) The absorption coefficient μ at any frequency above the K limit is $\mu_\alpha R_L^K \eta^{-3}$, where $\eta = \frac{\nu}{\nu_\alpha}$ and R_L^K is the absorption limit ratio, given by Richtmyer⁵ for silver as 7.8; in calculations on the second (more accurate) method, this assumption is replaced by Richtmyer's more accurate formula for μ .

(6) The number of α -line quanta emitted indirectly is a constant fraction, u_α , of the number of absorbed quanta of frequency greater than the K-limit frequency ν_K , and u_α is as given by Balderston² for silver, $\frac{5}{6} \times 0.75$, or 0.625.

(7) The total intensity of the α -line rays, or energy leaving the target per unit time and per unit solid angle in the direction ϵ , is E_α , and is made up of two parts, direct and indirect, called E'_α and E''_α , respectively.

(8) The observed α peak has an area $A = A' + A''$, likewise determined by the equation $A = SE_\alpha$, where S is the over-all sensitivity of the spectrometer and its accessories.

(9) The continuous spectrum has its true intensity per unit frequency interval $I(V, \nu)$, and they are related through the formula

$$S = \frac{C d \theta}{I(V, \nu) d\nu} = \frac{C \tan \theta}{\nu I(V, \nu)}$$

where θ is the glancing angle for the frequency ν .

(10) For a single element,

$$I(V, \nu) = K \{ (\nu_0 - \nu) + B\nu_\alpha \}$$

where K is a constant and $h\nu_0 = eV$; the term $B\nu_\alpha$ is expressed as a multiple of ν_α merely for convenience in integration, and is treated as if it were constant with $B = 0.10$, this value being a rough estimate from Webster and Hennings'⁶ data on Mo.

With these assumptions and definitions, it is a simple matter to set up the integral,

$$E''_\alpha = \frac{1}{2} \int_{\nu_K}^{\nu_0} \int_0^{\pi/2} \int_0^\infty u_\alpha \frac{\nu_\alpha}{\nu} \mu I(V, \nu) \exp \{ -\mu r - \mu_\alpha r \cos \psi \sin \epsilon \} \sin \psi dr d\psi d\nu$$

where ψ is the colatitude in a spherical coordinate system.

This integrates readily as far as

$$E''_\alpha = \frac{1}{2} u_\alpha \int_{\nu_K}^{\nu_0} \frac{\nu_\alpha}{\nu} \frac{\mu}{\mu_\alpha \csc \epsilon} I(V, \nu) \log \left(1 + \frac{\mu_\alpha \csc \epsilon}{\mu} \right) d\nu,$$

from which,

$$\frac{A''}{C} = \frac{\frac{1}{2}u_{\alpha}R_L^K \tan \theta \sin \epsilon}{(\eta_0 - 1 + B) \exp \{ -\mu_{\alpha}x_{\alpha} \csc \epsilon \}} \int_{\eta_K}^{\eta_0} \frac{\eta_0 - \eta + B}{\eta^4} \log \left(1 + \eta^3 \frac{\csc \epsilon}{R_L^K} \right) d\eta,$$

where $\eta_0 = \frac{v_0}{v_{\alpha}}$ and $\eta_K = \frac{v_K}{v_{\alpha}}$.

Evaluating this graphically for silver at 35 kv., the results are as follows:

$$\begin{aligned} \epsilon &= 20^{\circ}, 5^{\circ}, 1^{\circ}; \\ \frac{A''}{C} &= 11.9', 7.4', 4.0'. \end{aligned}$$

The observed values of the area-to-ordinate ratios for these angles are obtained from figure 1, as

$$\begin{aligned} \epsilon &= 20^{\circ}, 5^{\circ}, <1^{\circ}; \\ \frac{A}{C} &= 32.4', 30.7', 36'. \end{aligned}$$

In the absolute magnitude of the values calculated for A''/C , there are great opportunities for error, due to errors in the numerous assumptions listed above. Slight changes in them result in changes in A''/C much greater than one might expect; thus a moderate displacement of the center of gravity of the spectrum toward shorter wave-lengths, combined with a moderate asymmetry in space, can give calculated values of A''/C actually greater than the observed A/C . In Balderston's paper, quoted above,² the only means at hand for an estimate of A''/A was a comparison of quantities essentially equivalent to the present A''/C and A/C , for a single value of ϵ . It seems probable, therefore, that some difference in assumptions about the continuous spectrum explains the difference between his conclusions about A''/A and those to be drawn from the values given above.

However unreliable the present calculated values of A''/C may be, nevertheless the calculated changes of A''/C with ϵ are on a much firmer basis. Even if, by any reasonable changes of assumptions, the calculated A''/C at 20° were made to agree with A/C , the drops in A''/C to about $2/3$ and $1/3$ of the 20° value would still occur nearly as before. The conclusions from these experiments must, therefore, rest primarily on a comparison of A/C and A''/C with regard to their changes.

Returning to the hypothesis that the true A''/C is as calculated, and assuming the rest of the observed peak A to be made up of direct radiation A' , with A'/C the same at all angles, the decrease in A/C in reducing ϵ from 20° to 5° is of the calculated order of magnitude.

The rise in A/C in reducing ϵ still further, from 5° to below 1° , was at the time of the experiments a phenomenon quite unexpected. It may be

nothing but experimental error, since the rays below 1° were very weak, but if it is real an explanation may be found in considering the probable changes in A'/C . Direct line-spectrum rays, as one may show by a thin-target analysis,⁶ should be produced at a mean depth slightly less than that of the continuous spectrum. Both these depths being small compared to

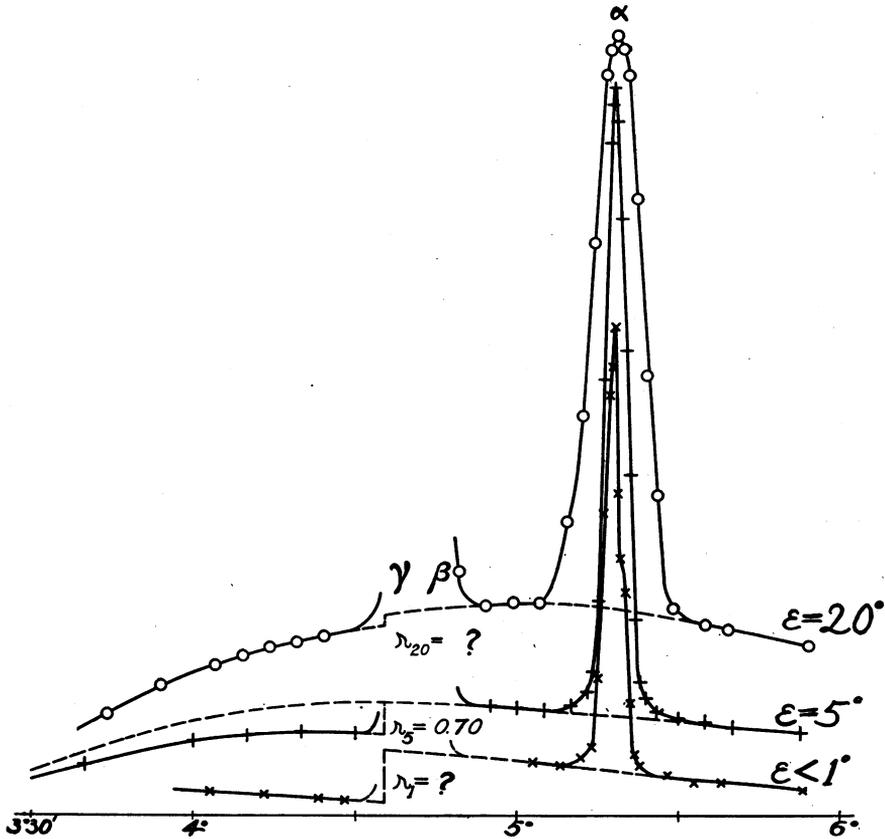


FIGURE 1

Spectra of silver at 35 kv. The scales of these spectra differ, because of differences in slit widths and times of exposure of the ionization chamber to the rays. The depth of production of rays of the continuous spectrum was estimated from the ratio r at $\epsilon = 5^\circ$, as 0.5 micron. The value of r at 20° was too near unity to give a reliable estimate, and its value at 1° too uncertain because of faintness of the rays, but so far as they go, the 20° and 1° ratios are consistent with the above depth.

that of the indirect rays, the effect of the direct rays on A/C should become evident only at emergence angles below a degree or two. There, however, where most of the indirect rays are eliminated by absorption, it should cause an effect of the sort observed.

These minor changes in A/C , while consistent enough with the theory, must nevertheless be regarded as unreliable evidence, because they are only of the order of magnitude of the limits of probable error. The more important result is that A/C shows no such large and systematic decrease as would be expected if the line-spectrum rays were nearly all of indirect origin. From that fact it seems reasonable to conclude that for the present case at least (silver at 1.4 times its excitation voltage) a large fraction, probably a majority, of the K-series rays are direct.

III. *Block-and-Film Experiments.*—For a more quantitative estimate of the ratio of direct to indirect rays, in relation to which the above experiments may be regarded primarily as a check against serious errors or misinterpretations, a second method of separating these rays was devised. This method also separates them according to depth of production, and is closely related to Beatty's¹ method, of which it may indeed be considered a modern adaptation. As in his experiments, the cathode rays strike a thin sheet of a metal and the indirect rays come from a thick block of another metal, placed under the sheet. Thanks to the spectrometer, however, in order to distinguish the rays of one metal from those of the other, it is no longer necessary to use metals of very different atomic numbers. In fact, the more nearly alike they are, the better, so long as the lines of one can be resolved from those of the other. In the present experiments, therefore, the composite targets were made of sheets of silver laid on a block of cadmium.

Three such targets were used. One had two sheets of silver foil, made by Baker & Co., each 6 microns thick as judged by its absorption of X-rays, using Richtmyer's⁵ absorption coefficients. Another had a single sheet of 6-micron foil, and the third a half-millimeter plate of graphite electroplated with silver to a thickness of 3.5 microns, also measured by absorption. These targets were placed so as to receive cathode rays at about 45° and send X-rays to the spectrometer with an emergence angle of 77° . Under these conditions they gave the spectra shown in figure 2, all taken with 0.07 ma. at 50 kv. and exposures of the chamber to the X-rays equal to 20 seconds.

To a first approximation, with extremely thin sheets of silver, all rays appearing in the silver lines might be regarded as direct; and if the cathode rays are all stopped before reaching the cadmium, all the cadmium rays must be indirect. With thicknesses such as these, suitable for stopping the cathode rays and for getting reasonably accurate intensity measurements without using cathode rays enough to damage the foils, a large correction must be made for the production of indirect rays in the silver and the consequent loss of opportunity for such action by the cadmium; and another correction must be made for the loss of cadmium rays by absorption in coming out through the silver. A third correction is for the

difference in the frequencies of their absorption limits, preventing the cadmium from making use of some of the rays that would be available for fluorescence in silver. A fourth, occurring in the third film only, is for the absorption by the graphite.

All these corrections are straightforward enough, theoretically at least, though the need for arithmetic integration makes it difficult to express them in formulas. The corrections for absorption of the cadmium rays in coming through silver, and for the loss of primary rays in the silver, are

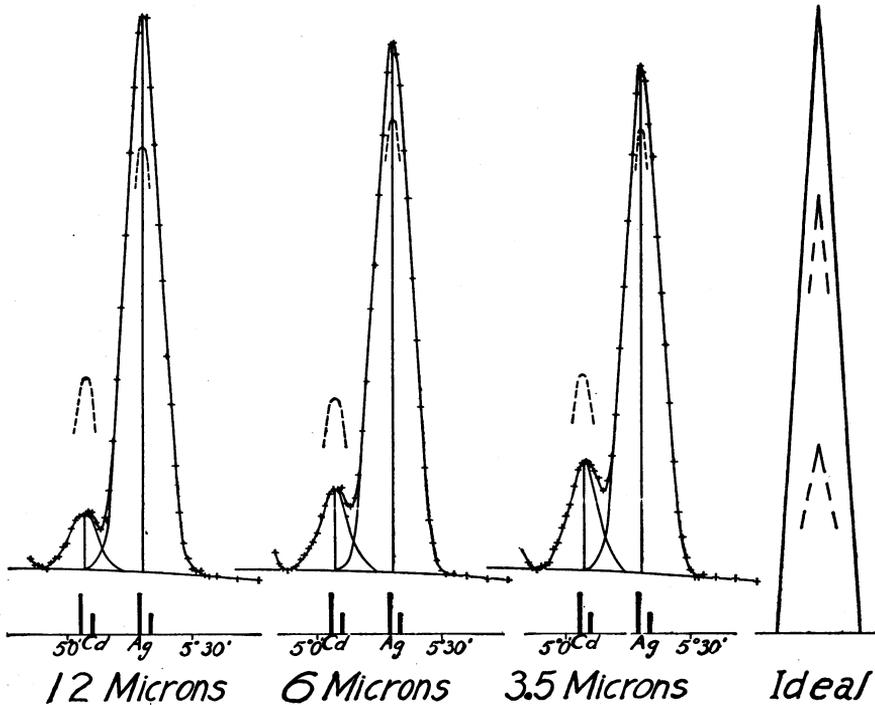


FIGURE 2

$K\alpha$ peaks in spectra of composite targets of Ag on Cd, with thicknesses of Ag as indicated. Full lines, observed data; dotted lines, direct-ray and indirect-ray peaks deduced from these data.

made in one step, by partitioning the secondary rays between the cadmium and silver as if both the film and the block were made of silver. This is mathematically easy, because the ratio of the absorption coefficients of any two frequencies, primary and fluorescent, is practically the same for each of these metals, and it is done by simply splitting the triple integral given in section II for E_g'' into two parts. Thus for a silver film of thickness x_1 , the range of r for the silver is from zero to $x_1 \sec \psi$ for any given value of the colatitude ψ , and the cadmium has r from $x_1 \sec \psi$ to infinity.

The third correction, for the difference in absorption limits, is made by changing the lower limit of the frequency integral for cadmium, leaving that for silver fixed.

The fourth correction, for the effect of the graphite, is made by the use of absorption data taken by Mr. R. M. Yeatman for a thick block of graphite from the same piece from which this plate was cut. This plate stops about 2 per cent of the outgoing cadmium rays and about 1 per cent of the harder primary rays.

Calculating thus the fractions of the theoretical indirect rays that should be received from the cadmium, we find for the three cases, in order as in the figure,

$$E''_{\alpha Cd} = E''_{\alpha} \times 0.29, 0.47 \text{ and } 0.56.$$

Likewise calculating the fractions produced in the silver,

$$E''_{\alpha Ag} = E''_{\alpha} \times 0.67, 0.44 \text{ and } 0.325.$$

With these fractions the observed cadmium peaks can be used to calculate the total indirect radiation from a solid silver target, shown as the dotted peaks above the cadmium peaks. Then these quantities can be used to calculate the indirect rays from the present silver foils, which can be deducted from the observed silver rays, leaving the direct rays alone, as shown in the dotted peaks under the observed peaks for silver.

Finally, the results of these calculations have been averaged for the three cases and are shown in an idealized form at the right-hand side of the figure. This represents what could be expected of a solid silver target, omitting the continuous spectrum and showing only the $K\alpha$ peak, in full lines, with its direct and indirect components in dotted lines within it. In this average, the ratio of the direct rays to the indirect is 2.36, with a mean deviation of 7% for the three individual cases.

With regard to errors in this result, the greatest was probably in the spreading of the primary continuous-spectrum rays on their way from the silver to the cadmium. This occurred in the 12 and 6 micron targets because the silver foils would not lie flat when heated by the cathode rays, but became curved, raising the whole middle part of the foil about a millimeter; and in the third target, the graphite had a finite thickness.

To minimize this error, the source used was limited by the spectrometer slits to a width not over 1.5 millimeters, and the focus of the cathode rays, as measured across the plane of the slits, was made about a centimeter in diameter. Thus, whatever rays were lost to view from the spectrometer crystal by spreading out of the region used, the loss was probably almost balanced by the gain from rays coming into it.

Other errors, probably small, are due largely to differences in properties between cadmium and silver, other than those corrected for. Among such errors may be listed fluorescence produced by cadmium β -line rays

in the silver, contributing to the silver α peak an amount equal to a small fraction of the amount deducted as described above. Another error is due to the very small difference in the ratios between the absorption coefficients of rays of any two wave-lengths. Another is due to differences in the over-all sensitivity of the spectrometer and its accessories for rays of the α -line frequencies of the two elements. Finally there is an error, not of this type, due to lack of uniformity in the thickness of the silver films. How much this last error amounts to, it is hard to say. For the film on graphite, which was slightly rough, it may be considerable and it is difficult to estimate; for the foils, the presence of minute holes throws doubt on the assumption of uniformity, but absorption tests of two pieces showed average thicknesses equal within limits of error, about 2 or 3%.

With regard to other voltages than this 50 kv., Beatty's¹ work of 1912 showed for copper that the direct and indirect rays increased in much the same proportion, though in that case the ratio was about 6 or 7, some two or three times the value for this case. In rhodium, likewise, it has been noted before⁷ that the total intensity of any line increased much as one might expect of indirect rays, though for a different reason. Preliminary work at 40 and 55 kv. with the 6 micron foil used here, showed practically the same ratio as in this work at 50, but as the current was less, due to a desire to find out the main facts before running too much risk of damaging the target, the results are not so exact.

The study of this direct-to-indirect ratio is being continued, using other voltages, and will be reported more fully. In the meantime, it is safe to assume that a value of 2.4 at 50 kv., about twice the excitation voltage, is fairly representative of the ratio for silver at all ordinary X-ray voltages.

IV. *Ratio of Indirect Rays to the Continuous Spectrum.*—The assumption that the ratio of direct to indirect rays is approximately constant at 2.4, for all voltages, leads at once to the inference that in the emergence-angle experiments of section II, the values observed for A/C should bear a ratio to the calculated values of A''/C something like 3.4. Actually, at the 20° emergence angle, where A'' is least affected by absorption, the ratio is somewhat less than 3.4, but of the right order of magnitude.

A more reliable check on calculated values for A''/C can be obtained from the 50 kv. data of section III, where the values of A'' are given by the corrected cadmium peaks, without reference to A' . The theoretical value of E''_{α} is $\frac{1}{2} K u_{\alpha} \nu_{\alpha} \nu_K \times 0.338$, where the symbols are as defined in section II and the 0.338 is the result of arithmetic integration for the special case of 50 kv. Therefore,

$$\frac{A''}{C} = \frac{\frac{1}{2} u_{\alpha} \nu_K \times 0.338 \tan \theta}{\nu_0 - \nu_{\alpha} + B \nu_{\alpha}}$$

This gives numerically, 28.4', and the experimental value, for the average of the three cases, is 30.2'.

The ordinate C was itself only 30.5 mm. and the electrometer was liable to erratic variations of a millimeter or two either way, resulting apparently from the fluctuations in numbers of α and β rays producing the natural ionization. Thus errors in C alone might readily account for the difference between these figures, and there are several other sources of error in the calculation, especially assumptions (1) and (10) of section II, capable of producing more error than this. The agreement found here must, therefore, be discounted somewhat, but it shows that the basic ideas are not unreasonable. When the form of the continuous spectrum and its distribution in space are more exactly known, such calculations may prove useful as a check on determinations of the fluorescent efficiency constant, u_α ; or conversely, since Balderston's u_α is probably the better known factor at present, they tend to confirm the above assumptions about the continuous spectrum.

V. *Fluorescent Rays in Thin-Target Spectra.*—An important problem, on which these data throw some light, is that of line spectra from thin targets, and the relations of their intensity to voltage. Bergen Davis⁸ proposed a method of getting such data by using thin films of silver plated on copper; and in a research in progress in this laboratory the method has been found quite practical. If, however, it had appeared here that nearly all the line-spectrum rays from a thick target were indirect, such rays as came from the thin target might well contain an unknown amount of indirect rays, due to continuous-spectrum rays from the copper. According to the present calculations, if the silver is made well under a micron thick, as it must be for cathode-ray penetration without undue loss of speed, the line-spectrum rays from it are nearly all direct; the next step, to eliminate the indirect rays almost completely, is to reduce the continuous-spectrum rays by replacing the copper with beryllium.

VI. *Quantum Probability Ratios.*—When a cathode ray passes through an atom, it may lose a large fraction of its energy, either in ejecting an atomic electron or in emitting a quantum of continuous-spectrum radiation, but there is no other obvious way for it to lose energy in anything like such large quantities. Consequently one may well inquire whether either one of these types of action occurs very much more often than the other and, if so, which?

To make the question more definite, since the ejection of an electron cannot occur without the loss of at least as much energy as $h\nu_K$, we may best compare the probability of the ejection of a K electron with that of emitting any continuous-spectrum quantum of frequency above ν_K . In view of the confirmation of the theoretical relations between A'' and C , noted above, it seems reasonable to apply to this problem the same basic assumptions used for that.

The number of continuous-spectrum quanta of the required range of frequencies is of course readily found as

$$N_C = 4\pi \int_{\nu_K}^{\nu_0} \frac{I(V, \nu)}{h\nu} d\nu = \frac{4\pi K}{h} \left\{ (\nu_0 + B\nu_\alpha) \log \frac{\nu_0}{\nu_K} - \nu_0 + \nu_K \right\}.$$

The number of direct α -line quanta is

$$N'_\alpha = \frac{4\pi E'_{\alpha_0}}{h\nu_\alpha}$$

where E'_{α_0} is the intensity of direct characteristic rays as emitted from the atoms. The correction for absorption, to get E'_{α_0} from E'_α , may be estimated from earlier data on molybdenum to be not over 1.02, and is unimportant. It will, therefore, be neglected here. This quantity N'_α is, of course, less than the number of K electrons directly ejected, which we may call N'_K , partly because some of the K-series energy is in the β and γ lines and partly because some K ionizations result in secondary photoelectrons rather than quanta of characteristic rays. Since the subsequent disposal of any ionization energy by an atom is probably independent of the method of ionization, we may assume that the ratio N'_α/N'_K is the same as N''_α/N''_K , which by definition is u_α .

Now let P equal the ratio of the direct radiation to indirect, determined above as 2.4, and Q the ratio to be finally determined, namely that of the number of direct K ionizations to the number of continuous-spectrum quanta of frequency above ν_K . Then

$$Q = \frac{N'_K}{N_C} = \frac{N'_\alpha}{u_\alpha N_C} = \frac{4\pi E'_\alpha}{h\nu_\alpha u_\alpha N_C} = \frac{4\pi P E''_\alpha}{h\nu_\alpha u_\alpha N_C}.$$

Inserting the value of E''_α as calculated from the integral of section III, and that of N_C just given, this becomes, for 50 kv.,

$$Q = \frac{\frac{1}{2} \times 0.338 P}{\frac{\nu_0 + B\nu_\alpha}{\nu_K} \log \frac{\nu_0}{\nu_K} - \frac{\nu_0}{\nu_K} + 1},$$

where the 0.338 is, as in section IV, a result of arithmetic integration for this special case. For this case, with $P = 2.4$ and $\nu_0 = \frac{e}{h} \times 50$ kv., this has the numerical value

$$Q = 0.965.$$

The fact that this particular value of Q is equal within limits of error to unity suggests that Q may possibly be unity always. This idea, however, is probably false. For if it were true, and a comparison were made of different elements at corresponding voltages, that is, equal values of

ν_0/ν_K , the value calculated for Q would be practically proportional to P and, as we have seen, Beatty's data for copper make P two or three times as large as in the present work on silver. A check on this comparison may be obtained from Unnewehr's² data on ratios of the type A/C for various elements including these two, from which one may calculate the ratios of A to the continuous-spectrum energies in corresponding intervals of the spectrum. These ratios must differ qualitatively much like the values of $(1 + P)$, and the data at hand confirm, so far as they go, the above conclusions about the changes in P and Q from one element to another. On this topic, however, more work is needed before definite laws can be formulated. The important points for the present are: first, that Q , the ratio of the numbers of losses of energy from cathode rays, by direct K ionization and by corresponding continuous-spectrum radiation, is probably not constant for different metals; and second, in the absence of information on the point, no definite statement can be made as to the variation of Q with voltage for any one metal. For the case tested, silver at 50 kv., its value is nearly unity.

VII. *Summary.*—1. By experiments of two types (sections II and III) it has been proved that the characteristic rays from silver are largely of direct origin, the experiments of the second type giving a ratio of direct rays to indirect equal to 2.4 at 50 kv., nearly twice the excitation voltage.

2. The ratio of indirect radiation to continuous-spectrum radiation can be calculated reasonably accurately (section III) from known data on the form of the continuous spectrum and the fluorescent-energy transformation constant.

3. Indirect rays will not appear notably in spectra from extremely thin targets, especially with light backing materials for the targets (section IV).

4. The ratio of the probability that a cathode ray will remove a K electron from some atom by direct action to the probability that it will radiate a quantum of energy in the continuous spectrum as great as the K ionization energy, is of the order of unity for silver at 50 kv. It is probably greater for copper under corresponding conditions and may vary somewhat with voltage (section VI).

In conclusion, I wish to express most hearty thanks to Professor L. S. Jacobsen and Messrs. B. G. Stuart and R. H. Varian for assistance in making these measurements.

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