The Fluorescence Yield from the L_{III} Level of Uranium

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The fluorescence yield, defined as the ratio of the number of fluorescence quanta emitted from the L_{III} level to the total number of incident quanta absorbed in the L_{III} level has been measured for uranium; and the value of 0.67 found. The relative intensities of the α to the β-lines in the K fluorescence spectra of molybdenum have been measured and found to agree with the value from the characteristic spectrum. The relative intensities of the α to the β-lines originating from the L_{III} level have been measured in the fluorescence spectrum of uranium, and found to agree with the value from the characteristic spectrum. The atomic absorption coefficients for uranium for wave-lengths including the L_{III} discontinuity have been measured, and a value for $\delta = 2.27$ found, where δ is the ratio of the absorption coefficients on the short and long wave-length sides of the L_{III} discontinuity.

ACCORDING to the theory of the production of fluorescence x-rays, when an electron in the K energy level absorbs a quantum of x-rays, it is ejected as a photoelectron. An electron from the L or outer levels will then fall into the vacant K level with the emission of K fluorescence radiation. Thus for each quantum of energy absorbed in the K level there should be emitted one quantum of fluorescence K radiation. However, it has been apparent for some years that this simple picture does not give a complete account of the phenomena associated with the reorganization of the atom. In some early experiments on the efficiency of production of fluorescence x-rays, excited by K characteristic radiation, for atoms belonging to the iron group Barkla and Sadler[1] found that only some 30 percent of these atoms ionized in the K level emitted K fluorescence radiation. Barkla[2] and Beatty[3] also found that the excitation of the K series was accompanied by a sudden increase in the emission of the photoelectrons. Auger,[4] using a Wilson cloud-chamber with different inert gases, was able to photograph the tracks of the photoelectrons and measure their energy. He found that in addition to the photoelectron whose energy is given by Einstein's equation $E_{kin} = h\nu - h\nu_K$ for absorption by a K electron; there might be

emitted from the same atom an electron of energy approximately $E = h\nu_K - 2h\nu_L$ and others of a similar type. This simultaneous ejection of several β-particles from the same atom has been called the "compound photoelectric effect." By knowing the energy and number of the β-particles, Auger was able to count the number of quanta absorbed in the K level, and the number of fluorescence K quanta emitted. He defined the fluorescence yield for the K level as the ratio of the number of fluorescence K quanta emitted to the total number of incident quanta absorbed in the K level. A similar definition may be applied to the L levels. Using the symbols w_K and w_L to represent the fluorescence yield from the K and L levels, respectively, Auger found the following results:

Auger's Results on Fluorescence Yield in K and L Series.

Atomic No.	Gas	w_K	w_L
19	argon	0.07	
36	krypton	0.51	0.1
54	xenon	0.71	0.25

Kossel[5] has suggested that the low efficiency of the K series emission may be explained on the assumption that the majority of such atoms undergo radiationless reorganizations of the Klein-Rosseland type resulting in the appearance of high-speed photoelectrons. Bothe,[6] on the other hand, has suggested that the results of Barkla

[1] Barkla, Bakerian Lecture, Phil. Trans. **217**, 317 (1917).
[2] Barkla and Philpot, Phil. Mag. **25**, 489 (1923).
[3] Beatty, Proc. Roy. Soc. **A85**, 329 (1911).
[4] Auger, C. R. **177**, 169 (1923); C. R. **180**, 65 (1925); Ann. de Physique **6**, 183 (1926).

[5] Kossel, Zeits. f. Physik **19**, 333 (1923).
[6] Bothe, Phys. Zeits. **26**, 410 (1925).

and Auger can be correlated on the assumption that they are manifestations of the phenomena of "internal absorption." According to this picture K radiation is always emitted in the reorganization of a K ionized atom, but the probability that this radiation shall be absorbed in the L, M, N, etc., levels of the same atom is very much greater than that it shall be absorbed in the same level of some other atom. In other words, the absorption coefficient for the K series radiation in the L, M, N, etc., levels of the "parent" atom is very much greater than for radiation of the same wave-length emitted from some other atom. There appears, however, to be no method of deciding between the two theories from an experimental standpoint. The excitation of the K series radiations in a solid or gas will be accompanied by a sudden increase in the photoelectric emission as a result of the "internal absorption" of the fluorescence radiations in the outer levels of the atoms. This was observed by Barkla[2] and Beatty[3] in the ionization produced relative to air, in methyl bromide, methyl iodide, and selenium hydride, when the wave-length of the radiation was slightly shorter than the K limit of the gas in question. Martin[7] has calculated the value for w_K from these data for bromine, iodine and selenium, and found it to give reasonable results. The importance of this effect in the measurement of relative intensities of x-ray lines with an ionization chamber has been well shown by Allison and Andrew.[8]

Measurements on the fluorescence yield in the K series have been made by Compton[9] and Martin,[7] where it was shown that w_K increases with the atomic number of the element, but does not vary with the wave-length of the incident x-rays. The latter conclusion involves the assumption, in the calculation of the energy of the x-rays, that the energy required to produce a pair of ions in the gas of the ionization chamber is constant and independent of the wave-length of the x-rays. This assumption appears to be fully justified by quite independent experiments.[10]

Since there are three L levels it was decided to make a determination of the fluorescence yield

for a single one of these; and accordingly the L_{III} level of uranium was chosen. The method used in this experiment is essentially the same as that adopted by Compton.[9]
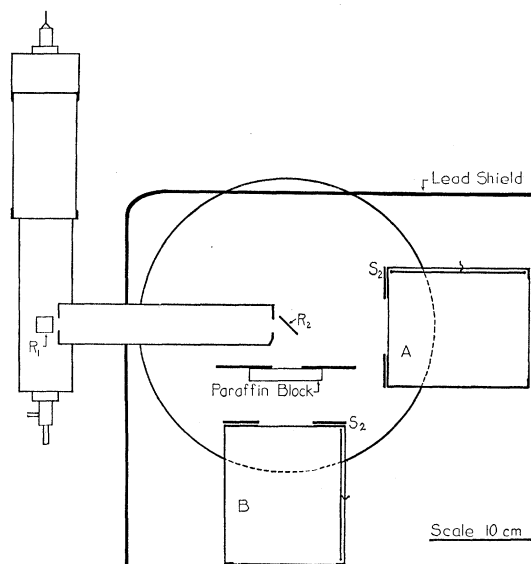

FIG. 1. Diagram of apparatus used for measuring the fluorescence yield from the L_{III} level of uranium.

The wave-lengths of the three L levels of uranium are given by Siegbahn[11] as: $L_I = 0.5680A$; $L_{II} = 0.5913A$; $L_{III} = 0.7208A$; and the wave-length of the molybdenum $K\alpha_1 = 0.7078A$; Mo $K\beta_1 = 0.6309A$; so that if this radiation from Mo is incident on a block of U only the lines arising from the L_{III} level will be excited. Primary x-rays from a rhodium target fell on a block of molybdenum at R_1 producing nearly homogeneous K radiation. A rhodium target was used in the x-ray tube because the wave-lengths of the Rh $K\alpha_1$ and Rh$K\beta_1$ are just a little shorter than the K absorption limit of Mo, and consequently the fluorescence lines from Mo will be excited quite strongly. Fluorescence radiation was used in preference to characteristic, because the former is entirely line radiation.

The fluorescence Mo radiation after passing through collimating slits fell on a piece of U at R_2, and the resulting radiation measured by an ionization chamber placed in position B. The intensity of the direct beam was measured by the ionization chamber in the position A.

[7] Martin, Proc. Roy. Soc. A115, 420 (1927).

[8] Allison and Andrew, Phys. Rev. 38, 441 (1931).

[9] A. H. Compton, Phil. Mag. 8, 961 (1929).

[10] Crowther and Bond, Phil. Mag. 8, 961 (1929).

[11] Siegbahn, Spektroskopie der Röntgenstrahlen, Second Edition, p. 274.

CALCULATION OF THE FLUORESCENCE YIELD

Let μ' be the total absorption coefficient of the primary Mo x-rays in the U radiator, then

$$\mu' = \tau' + \sigma'$$

where τ' is the photoelectric absorption, and σ' the absorption due to scattering. In this case the greater part of the photoelectric absorption will be due to the electrons in the L_{III} level, i.e., τ_L'. For the sake of convenience the subscript III will be dropped, and it is to be understood that τ_L and w_L refer only to the L_{III} level of U and not to all of the levels. The Mo fluorescence beam was taken as composed of four lines; α_1, α_2, β_1, β_2, and their relative intensities taken from Meyer's[12] data, namely: $\alpha_1 : \alpha_2 : \beta_1 : \beta_2 = 100 : 50.6 : 23.25 : 3.48$. Each of these lines will make its own contribution to the fluorescence beam from the uranium. Consider first the contribution made by the α_1-line; its intensity is $100/177.3$ of the total intensity from molybdenum. The fluorescence beam coming from the uranium is composed of seven lines: α_1, α_2, β_2, β_5, β_6, β_7, l, and each one of these will make its separate contribution to the ionization current derived from the uranium. Allison's values for the relative intensities in the L_{III} characteristic spectrum from U give $\alpha_1 : \alpha_2 : \beta_2 : \beta_5 : \beta_6 : \beta_7 : l = 100 : 11 : 28 : 6.4 : 1.6 : 0.4 : 2.4$. Let us therefore first obtain an expression for the power entering the ionization chamber due only to the U $L\alpha_1$ and excited by the Mo $K\alpha_1$. The wave-lengths and relative intensities of the Mo and U lines were taken from the characteristic spectrum, but as is shown later this appears to be quite justified.

Suppose a beam of x-rays from the Mo of intensity I' and cross section A' traverses a thick-

ness dx of the U radiator. The number of quanta absorbed by the L level and hence the number of photoelectrons ejected from the L level will be

$$dn = (I'A'dx/h\nu')(100/177.3)\tau_L', \qquad (1)$$

where ν' is the frequency of the Mo $K\alpha_1$. Hence the number of fluorescence L quanta escaping from the U atoms, due to the Mo $K\alpha_1$, will be $dn_1 = w_L dn$. The power in the fluorescence beam due to the U $L\alpha_1$ will be $h\nu''dn_1$, where ν'' is the frequency of the U $L\alpha_1$. Barkla and Sadler have shown that the intensity of the fluorescence beam is the same in all directions. Thus the power in the U fluorescence beam, due to the Mo $K\alpha_1$ and U $L\alpha_1$, uncorrected for absorption; which enters the ionization chamber is

$$dP'' = h\nu''dn_1 \frac{A''}{4\pi r^2} \tau_L'$$

$$= h\nu''w_L P'\tau_L'dx \frac{1}{h\nu'} \frac{A''}{4\pi r^2} \frac{100}{177.3}. \qquad (2)$$

A'' is the area of the diaphragm S_2 and $P' = I'A'$ is the power in the primary Mo beam, and r the distance from the center of the U block to the center of the diaphragm S_2. $A''/4\pi r^2$ is the solid angle which the ionization chamber subtends at the uranium radiator.

If the uranium is set at $45°$ to the primary molybdenum beam and thus at $45°$ to the ionization chamber in the position B, then the paths of the primary and the U fluorescence beam in the U radiator will be the same. Let μ' and μ'' be the absorption coefficients of the Mo $K\alpha_1$ and U $L\alpha_1$, respectively, in the uranium. Then the power in the fluorescence beam entering the ionization chamber will be

$$P'' = w_L \frac{A''}{4\pi r^2} P'\tau_L' \frac{\nu''}{\nu'} \frac{100}{149.8} \frac{100}{177.3} \int_0^\infty e^{-(\mu'+\mu'')x}dx = w_L \frac{A''}{4\pi r^2} \frac{\tau_L'}{\mu'+\mu''} \frac{100}{149.8} \frac{100}{177.3} \frac{\lambda'}{\lambda''} P' \cdots, \qquad (3)$$

where $100/149.8$ is the fractional intensity of the U $L\alpha_1$ to the total U beam λ' and λ'' are the wave-lengths of the Mo $K\alpha_1$ and U $L\alpha_1$ re-

spectively. Similar expressions will give the contribution from the U $L\alpha_2$, $L\beta_2$, $L\beta_5$, $L\beta_6$, $L\beta_7$, l; these added together give the total contribution due to the Mo $K\alpha_1$. To this must be added the total contribution due to the Mo $K\alpha_2$, $K\beta_1$, $K\beta_2$, and then after making the appropriate corrections for the absorption of each of these lines in

[12] Meyer, Wissenschaftliche Veröffentlichungen aus dem Siemens-Konzen VII, 2 (1929).

Also, Siegbahn, *Spektroskopie der Röntgenstrahlen*, Second Edition.

the gas of the ionization chamber, the sum total will give values proportional to the observed ionization currents.

The absorption coefficients of these various wave-lengths in uranium need to be determined, as well as the value of τ_L. It is also necessary to test whether the relative intensities of lines of the Mo characteristic spectrum agree with those from the fluorescence spectrum. This must also be checked for the lines from the L_{III} level of uranium.

ABSORPTION COEFFICIENTS IN URANIUM

Because of the high absorption or uranium it is impractical to use foil, even if it were available; consequently a solution of uranyl nitrate was employed. A parallel sided cell was made 0.320 cm thick, the front and back faces covered with mica and held in position with rubber gaskets. A single crystal ionization spectrometer was used, the cell being placed between the two defining slits. A motor generator set supplied current at 540 cycles to the primary of a transformer; the secondary current being rectified by kenotrons and then passed into condensers to smooth out the ripple. The high voltage was read on an electrostatic voltmeter and kept constant during the experiment. The current through the x-ray tube was read on a milliammeter and also kept constant, so that the power through the tube remained the same during an experiment. This equipment was used in all the experiments described in this paper.

Solutions of 2, 5 and 10 g of uranyl nitrate crystals dissolved in 100 g of water were used in the cell and the three served as checks. Since the absorption coefficient of water is also found this served as an additional check, the results agreeing very well with the published data.[13]

The results shown in Table I were obtained for the atomic absorption coefficient for uranium. The value of the L_{III} discontinuity, or the ratio of μ_a on the short and long wave-length sides of the L_{III} absorption limit is found from the graph to equal 2.27. Kustner[14] gives the value of 2.2 for the L_{III} discontinuity. From these results it would appear that the wave-length of the L_{III}

[13] Dershem, Phys. Rev. 21, 30 (1923); Hewlett, Phys. Rev. 17, 284 (1921).

[14] Kustner, Phys. Zeits. 46, 845 (1932).

TABLE I. *Atomic absorption coefficients in U.*

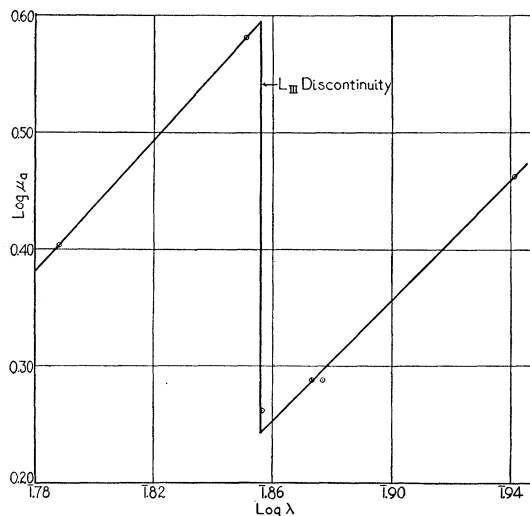| Line	λ in A	μ_a
Sr $K\alpha_1$	0.8734	2.90×10^{-20}
U $L\beta_2$	.7531	1.94
Cb $K\alpha_1$	.7446	1.94
U $L\beta_1$	.7185	1.84
U $L\beta_3$	.7088	3.81
U $L\gamma_1$	.6136	2.53

FIG. 2. Graph showing logarithm of atomic absorption coefficient for uranium plotted against logarithm of wave-length (log μ_a as a function of log λ).

absorption limit is less than 0.7208A, since the line U $L\beta_1 = 0.7185$A lies on the long wave-length side of the limit.

The quantity $1.27/2.27 = 0.56$ represents the fraction of the radiation on the short wave-length side of the L_{III} limit which is absorbed photoelectrically in the L_{III} level. Since the scattering term may be assumed negligible, $\mu' = \tau'$ and hence $\tau_L' = 0.56\mu'$.

RELATIVE INTENSITIES OF THE Mo $K\alpha$ TO THE Mo $K\beta$ IN THE FLUORESCENCE SPECTRUM

A metal x-ray tube with a rhodium target was operated at 40 kv and 10 milliamperes, the molybdenum scatterer being placed immediately in front of the aluminum window of the tube. The distance from the center of the rhodium target to the molybdenum scatterer was approximately 2.5 cm. A single crystal ionization spectrometer was used, the molybdenum fluorescence beam being collimated by two slits 0.8 mm wide, 2.4

cm high, and 25 cm apart. The ionization chamber was made of copper 20.3 cm long with mica windows, and filled with methyl iodide at 20.8 cm pressure. The ratio of the peak intensities of the α_{12} to β_{12} read from the graph is 4.85 to 1.
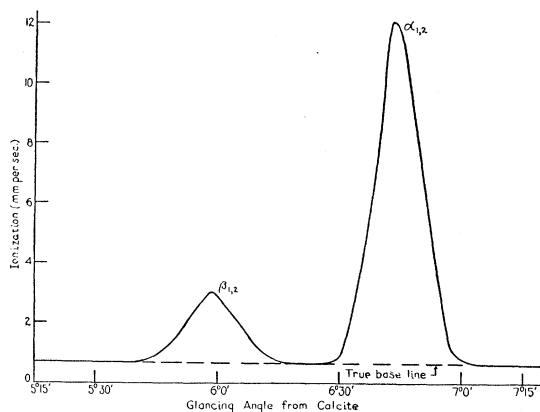

FIG. 3. Graph of intensity against crystal angle for K fluorescence spectrum from molybdenum.

The true base line occurs at 0.048 mm per sec., found by moving the ionization chamber from its correct position for reflection from the crystal. This intensity ratio has to be corrected for: (1) the relative absorption in the air between the scatterer and the window of the ionization chamber, a factor of 1.014; (2) the relative absorption in the mica window of the ionization chamber, a factor of 1.014; (3) the relative absorption in the methyl iodide, a factor of 0.815; (4) the relative absorption in the Mo scatterer, a factor of 1.375; and (5) the reflection coefficients of the two wave-lengths from the calcite crystal, a factor of 0.965. This gives a final result of 5.42 for the relative intensities of the Mo $K\alpha_{12}$, Mo $K\beta_{12}$ in the fluorescence spectrum. In order to check this value with that given by Meyer for these lines in the characteristic spectrum, curves were drawn for the $K\alpha_1$, $K\alpha_2$ with peak intensities of 100 and 50.6 and displaced relative to one another by 2.5 minutes of arc, and similarly for the $K\beta_1$, $K\beta_2$. The value obtained in this manner gave 5.9, which would appear to be within the limits of experimental error. Measurements are at the present time being carried on in this laboratory by Dr. J. H. Williams on relative intensities of lines in the characteristic spectrum, and preliminary results indicate that Meyer's value of β_1

is somewhat low. The calcite crystal which was used here was one previously calibrated by Allison[15] (crystal VA) by means of a double crystal spectrometer.

THE RELATIVE INTENSITIES OF THE $L\alpha_{12}$ TO THE $L\beta_2$ LINES IN THE FLUORESCENCE SPECTRUM FROM THE L_{III} LEVEL OF URANIUM

The apparatus used in this experiment was the same as in the previous one, except the rhodium target was replaced by a molybdenum target, and the molybdenum scatterer by a uranium scatterer. The x-ray tube was operated at 40 kv and 20 milliamperes, and the ionization chamber filled with methyl bromide at atmospheric pressure. The true base line occurs at 0.05 mm per sec., found by moving the ionization chamber from its correct position for reflection from the crystal. Lines from the L_I and L_{II} levels appear in the graph but their relative intensities, with respect to the lines from the L_{III} level, cannot be compared with that from primary x-rays from a U target. The ratio of the peak intensities of the α_{12} to β_2 read from the graph is 2.22. Both the
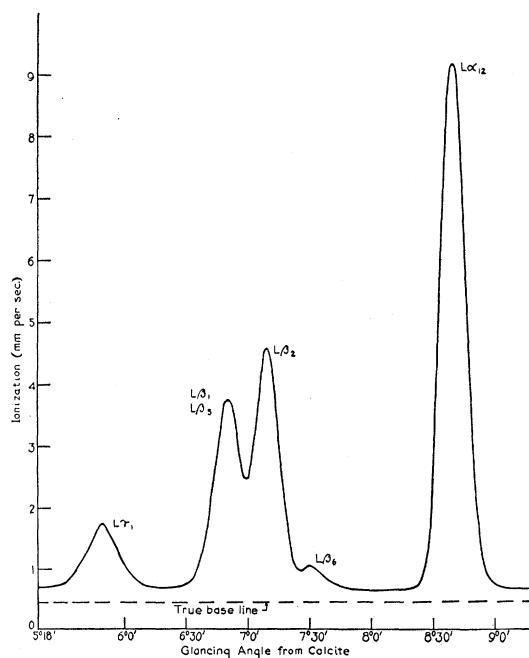

FIG. 4. Graph of intensity against crystal angle for the fluorescence spectrum from the L_{III} level of uranium.

[15] Allison, Phys. Rev. 41, 1 (1932).

α and β-lines are of shorter wave-length than the K limit of bromine, so that the bromine K radiation will be excited in the ionization chamber and part of this will escape to the walls of the chamber. The method of correction for this follows that given by Allison and Andrew,[8] and introduces a factor of 1.09.

The other corrections follow as in the previous experiment: (1) correction for absorption in the air and mica window, a factor of 1.053; (2) correction for relative absorption in the U, a factor of 1.62; (3) correction for relative reflection from the calcite crystal, a factor of 0.903. This gives a final result of 3.73 for the relative intensities of the U $L\alpha_{12}L\beta_2$, in the fluorescence spectrum.

The relative intensities of these lines in the characteristic spectrum were determined by Allison[16] and his results give the value of 3.78. Thus it would appear that the same relative intensities are obtained for the lines from the L_{III} level in the characteristic spectrum as in the fluorescence spectrum.

CALCULATION OF THE FLUORESCENCE YIELD FROM THE URANIUM L_{III} LEVEL

The diagram of the apparatus used is shown in Fig. 1. The fluorescence molybdenum radiation passed through two circular holes 1 cm in diameter and 20 cm apart, and was visible on a fluorescent screen. The ionization chamber was 10.7 cm in diameter, and 13.2 cm long, and the windows at the front and back were made so that none of the uranium radiation struck the walls or the back of the chamber.

A difficulty was encountered which was due to the radioactivity of the uranium, or rather to the β-particles given out by the radioactive products UX_1, UX_2. The ionization caused by the β-particles was eliminated by interposing a piece of paraffin 1.485 cm thick between the uranium and the ionization chamber. This of course reduced the intensity of the fluorescence beam from the uranium; but not too seriously.

The intensity of the direct beam from the molybdenum was reduced to measurable values by using absorbing screens of silver of known thickness. Silver was chosen because it can be obtained pure, and also no silver K radiation will be

excited. The absorption coefficient determined in this manner will be less than the correct absorption coefficient for silver since some of the scattered radiation can enter the ionization chamber.

In a similar manner the absorption coefficient for paraffin cannot be taken directly from tables, but was found approximately by finding the absorption coefficient of paraffin for the direct molybdenum beam. The value found in this experiment for $\lambda = 0.698A$, which is weighted mean wave-length from Mo, is 0.384. From data given by Allen[17] the absorption coefficient for paraffin for $\lambda = 0.698A$ is 0.523. It was therefore assumed that the absorption coefficient for $\lambda = 0.876A$, which is the weighted mean wave-length from the uranium, should be, for this experimental arrangement $0.384 \times 0.901 / 0.523 = 0.661$ where 0.901 is the value given by Allen for $\lambda = 0.876$. There may be here a large source of error, for the scattering of the molybdenum and the uranium beams will not be the same, but this is some extent taken up by the difference in the geometry of the two beams. The molybdenum beam is more or less a parallel cylindrical one, while the uranium beam is a divergent one.

The intensities observed for the uranium fluorescence beam must then be multiplied by $e^{0.982}$ or 2.67.

Five separate determinations were made with different voltages and currents through the x-ray tube, and different pressures of methyl iodide in the ionization chamber. Sufficient intensity was found when the tube was operated at 30 kv and 10 milliamperes.

From Eq. (3) the contribution from the Mo $K\alpha_1$ and U $L\alpha_1$ is given by substituting $0.56\mu'$ for τ_L'. Similarly, the contribution from the Mo $K\alpha_1$ and U $L\alpha_2$ will be given by replacing $100/149.8$ by $11/149.8$ in Eq. (3) and letting μ'' be the absorption coefficient of U $L\alpha_2$ in U. Similarly for all the lines. The distance r from the center of the uranium to the center of the diaphragm S_2 was 9.54 cm, and A'' the area of S_2 was 20.27 cm². The intensity I'' of the fluorescence radiation from the uranium will be given by

$$I'' = w_L(S_1 + S_2 + S_3 + S_4)I'A''/4\pi r^2$$

where S_1, S_2, S_3, S_4 represent the contributions

[16] Allison, Phys. Rev. 32, 907 (1926).

[17] Allen, Phys. Rev. 28, 907 (1926)

made by the Mo $K\alpha_1$, $K\alpha_2$, $K\beta_1$, $K\beta_2$ lines, respectively. Typical values of these contributions are $S_1 = 0.210$, $S_2 = 0.1065$, $S_3 = 0.047$ and $S_4 = 0.008$. These values include the corrections for the absorption of the various wave-lengths in the methyl iodide. Hence

$$I'' = 0.371 w_L I'/4\pi r^2.$$

I'' and I' are proportional to the ionization currents observed for the uranium fluorescence beam and the direct molybdenum beam, respectively, after each has been corrected for absorption. After correction for absorption in the paraffin, $I'' = 25.6$ mm per sec. After correction for absorption in the silver, $I' = 5822$ mm per sec. Thus

$$w_L = \frac{25.6}{5822} \frac{1}{0.371} \frac{4\pi r^2}{A''} = 0.67.$$

The value given above was also the average of all the readings.

Another method of calculation, which is considerably simpler than the one given above is that of taking average values for the wave-lengths and absorption coefficients. The weighted mean average of the wave-length of the molybdenum fluorescence beam is $\lambda' = 0.698$A, and for the uranium beam is $\lambda'' = 0.876$A. These values are taken from the fluorescence spectrum reported in this paper. The values of μ' and μ'', the atomic absorption coefficients of the molybdenum and uranium beams, respectively, in uranium are 3.65×10^{-20}, 2.92×10^{-20}. The calculation then gives $w_L = 0.66$.

The fact that these two calculations give values so close to one another, may be taken to show that the intensities of the lines in the characteristic and fluorescence spectra, used here, are very nearly the same. It should be noted even if Meyer's value of β_2 is as much as 5 percent too low, the change in the result for the final value for w_L would not be more than 0.5 percent.