

Direct and Fluorescence Excitation of the K Level in Thick Targets of Gold

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The ratio P of the number of $K\alpha$ line quanta produced by direct cathode electron impact to the number of quanta produced by fluorescence as observed in the radiation from a thick gold target, is measured with normally emergent x-rays over a range of voltages extending from 1.3 to 2.2 times the K excitation voltage. P is found to vary from 0.195 at $U=1.3$ to 0.27 at $U=2.2$, where U is the ratio of tube voltage to K excitation voltage. The ratio Q of the

probability of a cathode electron ionizing the K shell directly to the probability of a cathode electron exciting a continuous quantum of frequency greater than the K limit frequency is found to vary from 0.07 at $U=1.3$ to 0.11 at $U=2.2$. A discussion of the variation of P with atomic number found experimentally and that to be expected theoretically is given, still indicating a lack of agreement between theory and experiment.

INTRODUCTION

IT has been shown by recent papers¹⁻³ that the number of $K\alpha$ line quanta produced by direct cathode-electron impacts bears very nearly a constant ratio to the number of $K\alpha$ line quanta produced by fluorescence. This ratio, usually called the direct to indirect ratio P , shows a slight increase with voltage tending to become more nearly constant at the higher voltages. A definite idea of the change in P with voltage can be given from the recent work on copper.³ P increases from 6.63 at twice the K excitation voltage for copper to 7.2 at 17.4 times this excitation voltage. In this case the increase of P with voltage was very nearly constant over the entire voltage range. Data on palladium² and silver¹ tend toward a constant P as the voltage increases.

In the work on copper³ a comparison of the variation of P with atomic number, for a constant value of U determined from various experimental data for silver, palladium and copper was made with the theoretical variation of P with Z obtained from an elementary theoretical discussion. The result of the comparison showed a lack of satisfactory agreement between experiment and theory. Theory predicted that P should vary with $Z^{-4.8}$ with constant U , and the empirical data available indicated a variation with $Z^{-2.6}$. This deviation was considered to be outside the experimental limits of error. Any attempt to improve the theory resulted in a slightly wider deviation. The data, however, did

not cover a sufficient number of different elements to make the experimental variation conclusive. It was to clarify this point that the present experiment was started, and in order to obtain as wide a spread of atomic numbers as possible, gold was chosen for the target material.

THEORY OF EXPERIMENT

The same experimental method of measurement of P used for copper³ was used in this investigation. This method makes use of the ratio of the Au $K\alpha$ line intensity from a gold target to the Au $K\alpha$ line intensity from the same target covered with a palladium foil sufficiently thick to stop all the cathode electrons. In the latter case all the K excitation in the Au target is by fluorescence. The ratio P is obtained from the ratio of the two intensities stated above, together with certain calculations to be outlined below. Palladium was chosen for the foil material because the mean depth of production of the continuous rays had been measured previously for the voltage range here considered.²

In the following expressions all intensities are to be defined as energy per unit solid angle, per cathode electron.

The following symbols are defined as follows:

I = intensity of Au $K\alpha$ lines (unresolved) from the bare gold target;

d = part of I produced directly;

i = part of I produced by fluorescence;

i_+ = part of i that is produced at depths greater than \bar{x} , the mean depth of production of the continuous rays;

i_- = part of i produced at depths less than \bar{x} ;

i_0 = intensity of Au $K\alpha$ lines from gold target covered with palladium foil.

The intensity from the bare gold target will be

¹ D. L. Webster, Proc. Nat. Acad. Sci. **14**, 339 (1928).

² W. W. Hansen and K. B. Stoddard, Phys. Rev. **43**, 701 (1933).

³ K. B. Stoddard, Phys. Rev. **46**, 837 (1934).

composed of both direct and fluorescence rays. $I = d + i$ and as $P = d/i$ by definition, $P + 1 = I/i$. The expressions for i_+ , i_- , and i_b are given immediately below.³

$$F(V, 0) = \int_{\nu_k}^{\nu_0} (I(V, \nu)/\nu)(\mu_2/\mu_{\alpha_2}) \log(1 + \mu_{\alpha_2}/\mu_2) d\nu,$$

$$\Phi(V, x) = \int_{\nu_k}^{\nu_0} (I(V, \nu)/\nu)(\mu_2/\mu_{\alpha_2}) \{ [\exp(-\mu_{\alpha_2}x)] [Ei\{(\mu_{\alpha_2} - \mu_2)x\} - \log|(1 - \mu_{\alpha_2}/\mu_2)|] - Ei(-\mu_2x) \} d\nu,$$

$$f(V, x) = \int_{\nu_k}^{\nu_0} (I(V, \nu)/\nu)(\mu_2/\mu_{\alpha_2}) \{ \exp(a\mu_{\alpha_2}x) \} Ei\{-(a\mu_{\alpha_2} + \mu_1)x\} - Ei(-\mu_1x) \} d\nu,$$

$$R = \frac{1}{2} u_{\alpha} \nu_{\alpha} (\mu_k/\mu_2),$$

where $\mu_1 = 6.13 \times 10^{27} c^3 \nu^{-3} + 12$ cm⁻¹ absorption coefficient for Pd;⁴

$\mu_2 = 4.31 \times 10^{28} c^3 \nu^{-3} + 16.4$ cm⁻¹ absorption coefficient⁴ for Au;

$\mu_{\alpha_1} = 49.5$ cm⁻¹ absorption coefficient of Pd for Au $K\alpha$ rays;⁴

$\mu_{\alpha_2} = 73.2$ cm⁻¹ absorption coefficient of Au for Au $K\alpha$ rays;⁴

$a = \mu_1/\mu_2$;

$\mu_K/\mu_2 = 0.82$ ratio of K absorption to total absorption in Au;

u_{α} = number of K electrons ejected that reappear as $K\alpha$ quanta;

ν_K = frequency of K discontinuity;

ν_0 = high frequency limit;

ν_{α} = Au α line frequency;

\bar{x}_1 and \bar{x}_2 are the mean depths of production of the continuous rays in Pd and Au, respectively;

x_1 thickness of Pd foil;

$$I_1(V, \nu) = \delta k[(\nu_0 - \nu) - bZ_{Pd}\nu_0];$$

$$I_2(V, \nu) = k[(\nu_0 - \nu) - bZ_{Au}\nu_0];$$

$$b = 1.2 \times 10^{-3} \text{ sec}^{-1.5}$$

The above expressions for i_+ , i_- and i_b are evaluated by numerical integration. The constant δ in $I_1(V, \nu)$ is evaluated for each voltage under the criterion that $I_1(V, \nu_{\alpha}) = I_2(V, \nu_{\alpha})$; that is, the continuous spectrum intensities are matched at ν_{α} .

The expression for $P + 1$ is

$$P + 1 = I/i = (I/i_b)[i_b/(i_+ + i_-)],$$

where I/i_b is the ratio of the intensity of the Au α lines without the foil to the intensity with the Pd foil, measured in the same units. The α line intensity produced directly is a fraction $P/P + 1$ of the total.

The ratio Q of the probability of a cathode electron ejecting a K electron directly to the

$$i_+ = R[\exp(-\mu_{\alpha_2}\bar{x}_2)]F(V, 0); \quad i_- = R\Phi(V, \bar{x}_2);$$

$$i_b = R[\exp(-\mu_{\alpha_1}x_1)]f(V, x_1 - \bar{x}_1),$$

where

probability of a cathode electron exciting a continuous quantum of frequency greater than the K limit frequency as determined from a thick target, may be immediately evaluated from P with a small calculation.⁶

$$Q = iP \exp(\mu_{\alpha_2}\bar{x}_2) / [\mu_{\alpha}\nu_{\alpha} \int_{\nu_k}^{\nu_0} (I_2(V, \nu)/\nu) d\nu].$$

The results of the calculations obtained by numerical integration, together with the measurements and values for P and Q are given in Table I.

TABLE I. Calculations and measurements leading to the values for P and Q .

$U = V/V_K$	1.3	1.6	1.9	2.2
Calculated quantities				
i_+/D^*	0.0545	0.168	0.319	0.49
i_-/D	0.0098	0.038	0.089	0.167
i/D	0.0643	0.206	0.408	0.657
i_b/D	0.0351	0.113	0.225	0.365
Measured quantities				
$\bar{x}_1 \cdot 10^4$ in cm	3.5	5.3	7.4	10.5
$\bar{x}_2 \cdot 10^4$ in cm	2.8	4.3	6.0	8.5
I/i_b	2.19	2.23	2.27	2.28
Results				
P	0.195	0.225	0.255	0.27
$P/P + 1$	0.163	0.184	0.203	0.212
Q	0.07	0.088	0.103	0.11

* $D = Rk\nu_K$.

EXPERIMENTAL DISCUSSION

In this experiment as in previous ones¹⁻³ the x-rays leave the target perpendicularly. The angle between the incident cathode electrons and the observed emergent x-rays is 60 degrees.

⁶ The expression for Q given in reference 3 is incorrect. The numerical values for Q , however, are correct.

⁴ F. K. Richtmyer, Phys. Rev. 27, 1 (1926).

⁵ Int. Crit. Tab. 6, 46 (1929).

Dispersion of the x-ray beam was obtained by a Bragg spectrometer.

The mean depth of production \bar{x}_2 of the continuous rays in gold was measured by the method used by Webster and Hennings.⁷ The mean depth \bar{x}_1 in palladium was obtained from a previous paper.² It is assumed in this work as before that the continuous rays of frequency greater than the K excitation frequency are all produced at the same average depth \bar{x} . The justification for this assumption has been discussed before.³

In obtaining the ratio I/i_b the first step was to measure the ratio of the line ordinate to the continuous ordinate I_α/I_{c_α} for the bare gold target. By keeping the spectrometer set-up unchanged, the corresponding ratio i_α/i_{c_α} was then obtained for the gold target covered with the palladium foil. Since the ratio I/i_b is really obtained by matching the continuous spectrum intensities in the two cases, it is essential that this be done for the continuous intensities correcting for target absorption.

Therefore

$$I/i_b = [(I_\alpha/I_{c_\alpha}) \exp(\mu_{\alpha 1}\bar{x}_1 - \mu_{\alpha 2}\bar{x}_2)] / (i_\alpha/i_{c_\alpha}).$$

When the slits on the spectrometer were set for a reasonable exposure time for the thin foil measurements, the resolution was such that it was impossible to obtain continuous spectrum points between the $K\alpha$ and $K\beta$ lines. Analytical interpolation was therefore resorted to, to obtain I_{c_α} and i_{c_α} . The continuous spectrum was assumed to approximate a fourth power parabola. This form was chosen after examining the continuous spectrum curves used in measuring \bar{x}_2 .

DISCUSSION OF RESULTS

Before discussing the results certain errors involved in the measurement of P should be specifically pointed out. The error in the determination of I/i_b is approximately four percent. The major uncertainty here is due to the method of interpolation. Expansion in a Taylor's series about λ_α to obtain the continuous intensity at λ_α was used. Because of the symmetry of the continuous spectrum the cubic term was neglected

and fifth and higher power terms discarded. It is unfortunate that a continuous point could not be obtained between the $K\alpha$ and $K\beta$ lines. This would have increased the accuracy of I_{c_α} materially. Various other methods of interpolation such as approximate graphical ones, assuming the form of the continuous spectrum to approximate a cubic, etc., gave values for I_{c_α} that differed from the average by as much as two percent. It is observed that the same uncertainty occurs in i_{c_α} hence the influence on I_α/i_α is less. The intensity for the measured continuous points was determined with an accuracy of better than one percent by repeated measurements on each point.

Little accuracy can be claimed for the \bar{x} measurements in Au. The sharp decline of the continuous spectrum intensity due to excessive transmission of the ionization chamber near the Au K limit made the extrapolation necessary in the determination of \bar{x} very uncertain. This uncertainty led to a special investigation of the variation of $P+1$ with change in \bar{x}_2 . It was found that an error of 15 percent (the probable error in the determination of the mean depth) in \bar{x}_2 , caused an error of 2.3 percent in $P+1$. As long as $P \gg 1$, the influence on P of such an error in \bar{x}_2 is not important with reference to other errors in the experiment, but when $P < 1$, as in the present case, it becomes relatively more important. For example although $P+1$ changes by 2.3 percent for a 15 percent change in \bar{x}_2 , P changes by very nearly 12 percent. It should be pointed out that this line of argument also applies to the error in $P+1$ due to probable error in I/i_b . For an error of four percent in I/i_b the error in P is approximately 18 percent. These two sources of error and the way in which their effect is amplified indicates that the results for gold are relatively less accurate than the P and Q determinations for the other elements.¹⁻³

Numerous assumptions are made in the development and evaluation of the expressions for i_+ , and i_- and i_b . It is practically impossible to estimate the influence of errors introduced by these assumptions. It is thought, however, that a probable overall error of 30 percent in the determination of P will include all these uncertainties. A similar overall error is to be understood for the values for Q .

⁷ D. L. Webster and A. E. Hennings, Phys. Rev. 21, 301 (1923).

The results tabulated in Table I are in accord with other determinations¹⁻³ of P and Q . It is seen that the gradual increase of P and Q with U and the tendency toward a constant value for the higher voltages is also observed for gold.

In gold it is approximately four times as probable for the K shell to be ionized by fluorescence as for it to be ionized by cathode electron impact, while the probability of a cathode electron producing a K quantum is only about 1/10 of the probability of its producing a continuous quantum of frequency greater than the K limit frequency.

VARIATION OF P AND Q AND Z

In a previous paper³ the writer gave a simple theoretical argument indicating that P should vary as $Z^{-4.8}$ for a constant value of U . The empirical data then available indicated that P varied as $Z^{-2.6}$. The collected data for Ag, Pd, Cu and Au for several values of U are given in Table II. In Fig. 1 $\log P$ is plotted against $\log Z$

TABLE II. P tabulated for various elements, for several values of U .

U	1.5	2.0	2.5
P_{Cu}	6.60*	6.63	6.65
P_{Pd}	1.93*	1.97	1.99
P_{Ag}	1.85	1.89	1.92
P_{Au}	0.22	0.26	0.28*

* Extrapolated values.

for $U=2$. It is seen that no constant power of Z will fit the data.

If it is assumed that P should vary with Z to a constant power then the straight line A in Fig. 1 having an average slope of -2.78 for the three values of U given in Table II is the best fit. This assumes that the data for P are all of the same accuracy. Actually the data on Au are not as accurate as the other data and hence should be weighted less. This would decrease the absolute value of the slope slightly.

On the other hand the only reason for expecting P to vary with a constant power of Z is obtained from the theory given in the paper on copper.³ This theory is incomplete. There are a number of factors which vary in a minor fashion with Z such as the rediffusion of cathode electrons, cathode electron retardation, μ_k/μ_0 , etc. These were assumed to be independent of Z . If these assumptions are not made, it is evident

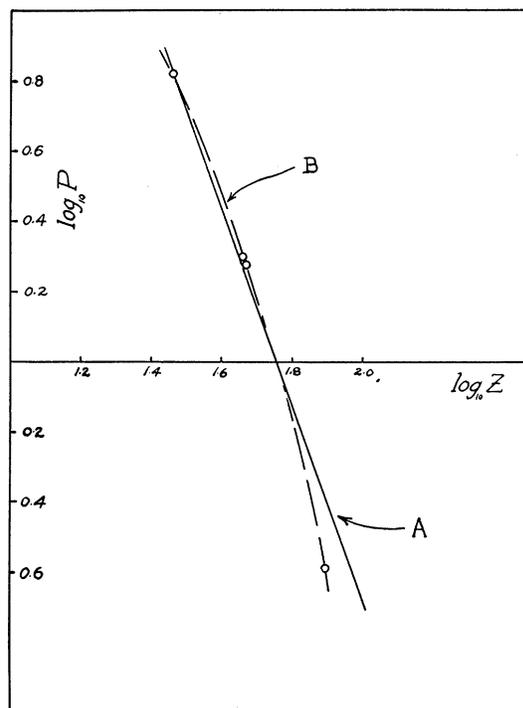


FIG. 1. $\log_{10} P$ plotted as a function of $\log_{10} Z$ for $U=2$.

that P will not vary with a constant power of Z . It so happens that the influence of these factors is very small in comparison with the factor $Z^{-4.8}$, and unless there is some factor omitted in the theoretical discussion or some assumption involved in the calculation of P that is not justified, the agreement between theory and experiment must be considered unsatisfactory. Discarding the notion that P should vary with a constant power of Z , one is no longer restricted to a straight line for the $\log P$ vs. $\log Z$ curve. The points in Fig. 1 are distributed in such a fashion that they do not indicate a unique curve. A parabola passed through the points of Fig. 1 shown by curve B will serve for the empirical variation of P with Z for interpolation purposes until more data are obtained. Curve B corresponds to the formula

$$\log_{10} P = -2.71 + \log_{10} Z(6.76 - 2.97 \log_{10} Z).$$

Interpolation according to this formula will be well within the experimental error.

The foregoing discussion applies equally well to Q as the variation with Z should be approximately the same as that for P .