Direct and Fluorescence Excitation of the K-Level in Thick Targets of Copper

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The ratio P of the probability of exciting the K shell of copper by direct cathode-ray impact to the probability of exciting the same shell by fluorescence in a thick target, is measured with normally emergent rays over a range of voltages extending from 2 to 17.4 times the K excitation voltage. P is found to vary from 6.63 at $U=2$ to 7.2 at $U=17.4$, where U is the ratio of tube voltage to K excitation voltage. The ratio Q of the probability of a cathode ray ionizing the K shell directly to the probability of a cathode ray exciting a continuous quantum of frequency greater than the K-limit frequency is found to be very nearly constant at 3.5 for the same voltage range. The variation of P with Z is examined theoretically and compared with experiment showing a lack of good agreement.

INTRODUCTION

A NUMBER of measurements have been made of the relative probabilities of exciting the K shell of atoms in a thick target by direct cathode-ray impact and by fluorescence.[1-4] The more quantitative measurements have been made since 1928 and have indicated that for silver and palladium the ratio of the probabilities of exciting the K level by the two methods is practically constant, showing no significant variation with voltage.

The question immediately arises as to whether this ratio will be independent of voltage for other elements and further; what is the variation of this ratio with change in atomic number?

The first attempt to investigate the method of excitation of characteristic x-radiation was made by Beatty.[1] This investigation was made before the development of the Bragg spectrometer and hence the results can be accepted only as tentative. Making use of a copper target this experiment led to the conclusion that practically all the characteristic radiation resulted from direct cathode-ray impact. Beatty likewise examined the subject theoretically and announced that not over ten percent of the radiation could be accounted for by fluorescence.

It was thought expedient to determine the ratio P for Cu by the more accurate methods now available as a step in the more general problem of the variation of P with atomic number.

In the previous work on this subject a curious relationship between line and continuous spectra

was found to persist.[4, 5] The ratio Q defined above was found to be practically constant over a wide range of voltages for the two elements silver and palladium. It is interesting to investigate this ratio further to see whether it will continue to be independent of voltage for other elements, in particular copper.

THEORY OF EXPERIMENT

In previous work recourse was had to a composite target of two elements of adjacent atomic number. The backing block or target proper was made of the one of higher atomic number and the other element was in the form of a foil of sufficient thickness to stop all cathode rays. This insured that all the characteristic radiation from the block was excited by fluorescence. The ratio P was calculated from certain expressions after the ratio of the intensities of the resulting two $K\alpha$ radiations was determined experimentally. This method is quite successful as long as the thickness of the foil necessary to stop the cathode rays is small and the α-line radiation from the block of sufficiently short wave-length. However, as the atomic numbers of the foil and block decrease, the thickness of the foil necessary to stop all cathode rays increases as does also the wave-length of the $K\alpha$ radiation; both factors tending to reduce the intensity of the characteristic x-radiation from the block. The effect of the increase of wave-length of the fluorescence rays from the block on foil absorption more than offsets the effect of the reduction in atomic number. Preliminary calculations showed that for copper such a scheme as outlined above would be impracticable, for the higher voltages at least, as the

[1] R. T. Beatty, Proc. Roy. Soc. A87, 511 (1912).
[2] Mark Balderston, Phys. Rev. 27, 696 (1926).
[3] D. L. Webster, Proc. Nat. Acad. Sci. 13, 445 (1927); 14, 330 (1928).
[4] W. W. Hansen and K. B. Stoddard, Phys. Rev. 43, 701 (1933).
[5] D. L. Webster, Proc. Nat. Acad. Sci. 14, 339 (1928).

intensity of the block characteristic x-radiation would be reduced to a point where it could not be distinguished from the continuous background.

Another method, suggested by Beatty's original experiment, was therefore used. This method makes use of the comparison between the $K\alpha$-line intensity from a copper target and the $K\alpha$-line intensity from the same target covered with an aluminum foil sufficiently thick to stop all cathode rays. The ratio of these two intensities together with certain calculations to be outlined below will enable the determination of the ratio P. It should be noted, parenthetically, that this method using the aluminum foil while enabling one to carry the measurements to a higher voltage than the previously described method, nevertheless eventually is limited. The necessary foil thickness to stop cathode rays, reduces the characteristic x-radiation from the block by increasing both the absorption of the continuous rays going in and the α-line radiation coming out. There is evidently a limiting thickness beyond which no characteristic x-radiation will be observed. A method of overcoming this difficulty by obtaining P from a bare copper target will be discussed below.

THEORETICAL

In the following development all intensities are to be defined as energy per unit solid angle, per cathode electron.[3,4]

The following symbols are defined as follows:

Let I = intensity of Cu $K\alpha$ lines (unresolved) from the bare copper target.

d = fraction of I excited directly.

i = fraction of I excited by fluorescence.

i_+ = part of i that is excited at depths greater than the mean depth of production of the continuous rays $\bar{x}$.

i_- = part of i excited at depths less than $\bar{x}$.

i_b = intensity of Cu $K\alpha$ lines from Cu target covered with Al foil.

The intensity from the bare copper target will be composed of both direct and fluorescence rays. $I = d + i$ and as $P = d/i$ by definition, $P + 1 = I/i$ where $i = i_+ + i_-$. The expressions for i_+, i_- and i_b are given immediately below. The reader is referred to Webster's[4] original paper for the development of the expressions for i_+ and i_-. The expression i_b is obtained in substantially the same way, taking into account the Al foil on the copper target.

$$i_+ = R \exp\left(-\mu_{\alpha 2}\bar{x}_2\right)F(V, 0); \qquad i_- = R\Phi(V, \bar{x}_2); \qquad i_b = R \exp\left(-\mu_{\alpha 1}x_1\right)f(V, x_1 - \bar{x}_1)$$

where

$$F(V, 0) = \int_{\nu_k}^{\nu_0} \frac{I(V, \nu)}{\nu} \frac{\mu_2}{\mu_{\alpha 2}} \log\left(1 + \frac{\mu_{\alpha 2}}{\mu_2}\right)d\nu$$

$$\Phi(V, x) = \int_{\nu_k}^{\nu_0} \frac{I(V, \nu)}{\nu} \frac{\mu_2}{\mu_{\alpha 2}}\left\{\exp\left(-\mu_{\alpha 2}x\right)\left[Ei\{(\mu_{\alpha 2} - \mu_2)x\} - \log\left|\left(1 - \frac{\mu_{\alpha 2}}{\mu_2}\right)\right|\right] - Ei(-\mu_2 x)\right\}d\nu$$

$$f(V, x) = \int_{\nu_k}^{\nu_0} \frac{I(V, \nu)}{\nu} \frac{\mu_2}{\mu_{\alpha 2}}\left[\exp\left(a\mu_{\alpha 2}x\right)Ei\{-(a\mu_{\alpha 2} + \mu_1)x\} - Ei(-\mu_1 x)\right]d\nu$$

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

where $\mu_1 = 3.86 \times 10^{24}c^{2.88}\nu^{-2.88}$ cm^{-1} = absorption coeff. for Al[6]

$\mu_2 = 1.165 \times 10^{25.4}c^{2.8}\nu^{2.8}$ cm^{-1} = absorption coeff. for Cu[6]

$\mu_{\alpha 1} = 133.7$ cm^{-1} = absorption coeff. of Al for Cu $K\alpha$ -rays

$\mu_{\alpha 2} = 460$ cm^{-1} = absorption coeff. of Cu for Cu $K\alpha$ -rays

$a = \mu_1/\mu_2$

$\mu_k/\mu_2 = 0.88$ = ratio of K absorption to total absorption in Cu[6]

u_α = α-line fluorescence efficiency in Cu

ν_k = frequency of K discontinuity

ν_0 = high frequency limit

ν_α = α-line frequency

$\bar{x}_1$ and $\bar{x}_2$ are the mean depths of production of the continuous rays in Al and Cu, respectively

x_1 = thickness of Al foil.

[6] Martin and Lang, Proc. Roy. Soc. A137, 199 (1932).

$I(V, \nu)$, the intensity of the continuous radiation per unit frequency interval at the mean depth of production $\bar{x}$, is approximated as $I(V, \nu) \cong k(\nu_0 - \nu)$. This could be improved somewhat for voltages less than twice the excitation voltage for Cu $K\alpha$-rays but any better approximation for higher voltages is unknown at present, so this form is used in all the numerical calculations.

$P+1 = I/i$ can be found without the foil measurements, if u_α and k are known. u_α may be found from various data now available, but k is unknown for the present work. However, by making use of i_b

$$P+1 = \frac{I}{i} = \frac{I}{i_b}\left[\frac{\exp(-\mu_{\alpha_1} x_1)f(V, x_1 - \bar{x}_1)}{\exp(-\mu_{\alpha_2}\bar{x}_2)f(V, 0) + \Phi(V, \bar{x})}\right]$$

where I/i_b is the ratio of the intensity of the Cu α-lines without the foil to the intensity with the Al foil measured in the same units. This is accomplished by comparing each line intensity with the continuous intensity at the mean depth of production $\bar{x}$ for each case and then reducing these to the same intensity unit. The remainder of the expression for $P+1$ is readily calculated. The unknown or uncertain factors such as k and u_α appear in both numerator and denominator and are eliminated. The α-line intensity produced directly is a fraction $P/P+1$ of the total.

When this method fails the unknown factors in i may be obtained from values of P measured by the Al foil method at lower voltages; allowing the evaluation of P for higher voltages from the bare copper target measurements of the ratio of line ordinate to continuous ordinate.

$$P+1 = \epsilon I(V, \nu_\alpha)/i = \gamma(\nu_0 - \nu_\alpha)\delta/S,$$

where ϵ is the ratio of K line area to continuous ordinate at λ_α; γ is the ratio of line ordinate to continuous ordinate;

$$S = \tfrac{1}{2}\nu_\alpha\nu_K[\exp(-\mu_{\alpha_2}\bar{x}_2)F(V, 0) + \Phi(V, \bar{x}_2)];$$

and δ, a factor to be found from P measured by the Al foil method at lower voltages, the ratio of line area to line ordinate being constant.[3]

The ratio Q of the probability of a cathode ray ejecting a K electron directly to the probability of a cathode ray exciting a continuous quantum frequency greater than the K-limit frequency, as
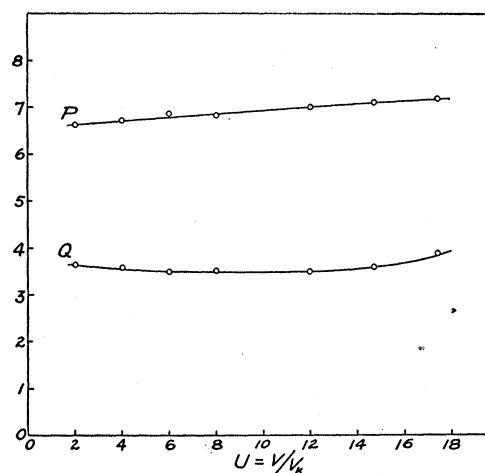

FIG. 1. P and Q are plotted as functions of U.

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

$U = V/V_k$	2	4	6	8	12	14.7	17.4
Calculated quantities							
i_+/D^*	0.317	1.795	3.50	5.18	8.64	10.6	11.7
i_-/D	0.043	0.486	1.30	2.67	5.70	8.4	12.2
i/D	0.360	2.281	4.80	7.85	14.34	19.0	23.9
i_b/D	0.122	0.640	1.27	1.94	—	—	—
$\delta \times 10^{-15}$	1.01	0.97	1.00	1.06	1.15†	1.22†	1.28†
Measured quantities							
$\bar{x}_1 \cdot 10^4$ cm	0.59	2.14	3.95	5.85	—	—	—
$\bar{x}_1 \cdot 10^4$ cm	0.18	0.65	1.20	1.78	3.2	4.4	6.6
I/i_b	22.4	27.4	29.7	31.6	—	—	—
γ	49.2	117	146	161	178	181	183
Results							
P	6.63	6.71	6.87	6.83	7.0	7.1	7.2
$P/P+1$	0.869	0.870	0.873	0.872	0.875	0.877	0.878
Q	3.67	3.58	3.50	3.50	3.5	3.6	3.9

* $D = Rk\nu_k$; † extrapolated values.

determined from a thick target, may be immediately evaluated from P with a small calculation.

$$Q = \mu_2 iP \exp(\mu_{\alpha_2}\bar{x}_2) \Big/ \mu_k R \int_{\nu_k}^{\nu_0} \frac{I(V, \nu)}{\nu} d\nu.$$

The results of the calculations obtained by numerical integration, together with the measurements and values for P and Q are given in Table I. The results are shown graphically in Fig. 1.

EXPERIMENTAL

The type of tube used in this research is not sufficiently different from other tubes used in this

laboratory and described elsewhere[7] to warrant a detailed description.

The target is a block of copper; the angle between the normal to the target face and the cathode-ray stream being 60 degrees. The target is surrounded by the equivalent of a Faraday cylinder to insure this definite angle of incidence for the cathode rays for all positions of the target. This is important for the measurement of the mean depth of production of the continuous rays by the method discussed below.

For the measurement of the mean depth of production $\bar{x}$ of the continuous rays a modification of Kulenkampff's[8] method was used. The tube was arranged so that the cathode-ray stream was parallel to the slits of the spectrometer. The intensity of the continuous spectrum at some wave-length less than the K-limit wave-length was observed for a number of positions of the target; the target being rotated about the cathode-ray stream as an axis, to these positions. Then if φ is the angle of rotation about the axis measured from the position $\varphi = 0$ illustrated in Fig. 2,

$$\log I_\varphi = \log I_{\bar{x}} - \bar{x} \sec 30° \sec \varphi$$

provided all the continuous rays are assumed to originate at the depth $\bar{x}$.

If now $\log I_\varphi$ is plotted against $\sec \varphi$, $\bar{x}$ may be obtained from the slope of this curve at $\sec \varphi = 1$.

This method worked admirably with copper but when tried with aluminum, I_φ did not differ measurably from $I_{\varphi=0}$ except for φ very close to 90 degrees. φ could not be determined accurately enough for the rapid variation of $\sec \varphi$ in this region, so for aluminum this method was abandoned. The values for $\bar{x}$ were finally calculated from the Cu $\bar{x}$ measurements by assuming that $\bar{x}$ is approximately inversely proportional to the number of electrons per unit volume.[9] This extrapolation is decidedly uncertain but fortunately no great accuracy is needed as i_b for which $\bar{x}_1$ is needed changes very slowly with variation in $\bar{x}_1$.

In the calculations the assumption is made that not only the continuous rays for a given wave-length are all produced at a depth $\bar{x}$ but
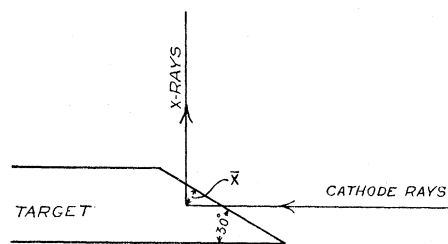
[7] D. L. Webster, W. W. Hansen and F. B. Duveneck, Rev. Sci. Inst. **3**, 729 (1932).
[8] H. Kulenkampff, Ann. d. Physik **69**, 548 (1922).
[9] H. M. Terrill, Phys. Rev. **21**, 476 (1923); **22**, 107 (1923).

FIG. 2.

also that all wave-lengths from λ_k to λ_0 are also produced at this depth. A special investigation, rather rough to be sure, indicated no significant variation in $\bar{x}$ with λ except very near the quantum limit, substantiating to some extent the latter assumption.

In measuring the ratio I/i_b the first step was to measure the ratio of the line ordinate to the continuous ordinate for the bare Cu target. The same ratio was obtained for the Cu target covered with the Al foil. Since the ratio I/i_b really is obtained by matching the continuous spectrum intensities in the two cases, it is essential that this be done at the mean depth of production in each case.

Therefore

$$I/i_b = \frac{I_\alpha/I_{c\alpha} \exp (\mu_{\alpha_1}\bar{x}_1 - \mu_{\alpha_2}\bar{x}_2)}{i_\alpha/i_{c\alpha}}.$$

In the measurement of γ, the ratio of the line ordinate to the continuous ordinate, the continuous spectrum intensity was corrected for second and higher order reflections from calcite by using Kirkpatrick's method. The continuous spectrum intensity at λ_α was obtained analytically by a Taylor series expansion about λ_α.

$$I(\lambda_\alpha + \Delta\lambda) = I(\lambda_\alpha) + C_1\Delta\lambda_\alpha + C_2(\Delta\lambda_\alpha)^2 + \cdots.$$

Terms involving the third and higher powers in $\Delta\lambda$ were neglected. $I(\lambda_\alpha + \Delta\lambda)$ was measured for at least three different $\Delta\lambda$'s.

The thickness x_1 of the aluminum foil necessary to stop all cathode rays was calculated from Williams's[8] expression for cathode-ray retardation in the form

$$x = \frac{V_k^{1.56}(U^{1.56} - 1)A}{7.65 \times 10^4 Z\rho},$$

where x is the penetration in cm measured along the path of cathode ray, A the atomic weight, Z the atomic number, ρ the density, V_k the excitation voltage for Cu $K\alpha$-rays, and $U = V/V_k$. Since the direct range is the important thing here rather than the total range Williams's[10] estimate $x_{\text{direct}}/x = 1/1.37$ for Al obtained from Schonland's[11] work, is used. Parenthetically it might be pointed out that $\bar{x}$ as measured by target absorption methods is roughly $1/10$ of x calculated from Williams's formula. This relation holds more accurately for the measurements of $\bar{x}$ in palladium[4] than for copper. In connection with any use of this expression it should be remembered that $\bar{x}$ is measured in a direction perpendicular to the target face, which is inclined 60° to the cathode-ray stream.

DISCUSSION OF RESULTS

The first topic to be discussed in this section is the important one of limit of error. The random error in the determination of I/i_b is about 2 percent. It is practically impossible to determine the overall error in P beyond a rough estimate because of the numerous assumptions involved in the calculation of the fluorescence intensity. It is thought, however, that P and Q are determined with a probable overall error less than 20 percent. For the three highest voltages the error is probably somewhat greater due to the uncertainty in determining δ by extrapolation from the lower voltage values. The error in $P/P+1$, the fraction of the total α-line radiation that is directly produced, is of course very much less than 20 percent.

The results tabulated in Table I are in general accord with other determinations[3, 4] of P and Q, (except that of Wisshak[12]) in that both P and Q show little variation with voltage. It is interesting to note at this point that Beatty's original estimate of the indirectly produced x-radiation in copper is exceptionally good considering the necessarily rough nature of his experiment. It should be remembered that he estimated that not more than 10 percent of the characteristic radiation in copper could be accounted for by fluores-

cence; the present experiment showing that about 13 percent is produced indirectly.

In palladium it was found that Q was about 0.9, showing that it is more probable for a cathode ray to excite a continuous quantum of frequency greater than the K-limit frequency than for it to eject a K electron directly. It is to be observed that for copper the probability of direct ionization is about 3.5 times that of its continuous spectrum equivalent. A general variation of this type is to be expected from the change of characteristic and continuous thick target intensities with atomic number. As to the value of Q to be expected beyond the limits of the voltage range examined here, the reader is referred to the discussion of this point elsewhere.[4]

VARIATION OF P WITH Z

Now that quantitative measurements of P are available for at least three atomic numbers it will be of interest to investigate the variation of P with Z. In order to do this one examines separately the variation of d and of i with Z.

The $K\alpha$-line intensity produced directly from a thick target may be obtained as follows:

$$d = \exp\left(-\mu_\alpha \bar{x}_d\right) \int_0^{U_0} i(U)(ds/dU)dU,$$

where $i(U)$ is the α-line intensity per unit solid angle, per unit cathode-ray path, and per cathode ray of energy U; U_0 is the initial energy of the cathode ray; dU/ds, the cathode-ray retardation expressed as a function of U.

In the above expression all electrons are assumed to possess an energy U after traversing a distance s in the target; also the number of cathode rays losing a large amount of energy in any one interaction producing either characteristic or continuous x-radiation is small compared to the total number. Rediffusion effects are neglected. Correction for target absorption is made by the insertion of the attenuation factor $\exp\left(-\mu_\alpha \bar{x}_d\right)$ on the assumption that the average depth of production of the directly produced α-line x-rays is $\bar{x}_d$. The approximate expression, $U_s{}^n = U_s{}^n - as$ is used for cathode-ray retardation, in which $n = 2$ for the Thomson-Widdington law and about 1.6 to agree with the more recent work of Williams[10] and Webster, Hansen and

[10] E. J. Williams, Proc. Roy. Soc. **A130**, 310 (1931).
[11] B. F. J. Schonland, Proc. Roy. Soc. **A108**, 187 (1925).
[12] F. Wisshak, Ann. d. Physik **5**, 507 (1930).

Duveneck.[13] $i(U)$ is obtained from Bethe's[14] expression for the ionization cross section for K electrons, although the exact dependence on U is not essential here.

$$i(U) = \frac{c_1 u_\alpha \nu_\alpha b}{Z^4 U} \log \frac{4U}{B} \sim \frac{c_2 u_\alpha b}{Z^2} f(U),$$

in which quantities not previously defined are the constants c_1 and c_2; b, a factor changing very slowly with Z, and $B \cong 1$ except for very low atomic numbers.

Integration then gives

$$d \sim \text{const. } u_\alpha b \exp\left(-\mu_\alpha \bar{x}_d\right) Z^{0.2} F_d(U_0),$$

$$\text{assuming } n = 1.6.$$

This expression for d together with i from the section on theory expressed in a more suitable fashion

$$i \sim \text{const. } \mu_k Z^5 \exp\left(-\mu_\alpha \bar{x}_i\right) F_i(U_0)/\mu$$

gives $P \sim A\beta Z^{-4.8}\Psi(U_0)$. Here A is a constant, β is a factor including a number of factors such as μ_k/μ, rediffusion,[15] etc., that show such small variations with Z that this dependence may be neglected in this rough consideration. The assumption is also made that the mean depth of production of the directly produced rays $\bar{x}_d$ is equal to the mean depth of the fluorescence rays $\bar{x}_i$, which is sufficiently good for the purpose here.

The comparison of P for different elements keeping U constant (assuming the form of $\Psi(U_0)$ remains the same), should show a direct variation roughly as $Z^{-4.8}$. Referring to Table II it is seen that this is not confirmed experimentally, in fact the experimental dependence insofar as it can be estimated from the experimental data at hand is about $Z^{-2.6}$.

The estimated error in both the palladium and

[13] Webster, Hansen and Duveneck, Phys. Rev. **44**, 258 (1933).

[14] H. Bethe, Ann. d. Physik **5**, 325 (1930).

[15] A special calculation showed that rediffusion effects would introduce a multiplication factor of 1.08 into the comparison of the P's for palladium and copper. The variation of μ_k/μ for the same comparison is negligible.

TABLE II. *Comparison of P for different elements keeping U constant. The ratio varies more nearly as $Z^{-2.6}$ than as $Z^{-4.8}$.*

U	$\dfrac{P_{Cu}}{P_{Pd}}$	$\dfrac{P_{Cu}}{P_{Ag}}$	U	$\dfrac{P_{Cu}}{P_{Pd}}$	$\dfrac{P_{Cu}}{P_{Ag}}$
1.5		3.57	4.0	3.24	
2.0	3.37	3.51	6.0	3.26	
2.5	3.34	3.46	Ave.	3.30	3.49
3.0	3.31	3.41	$(Z_{Cu}/Z_{Pd})^{-4.8}$	9.10	10.06
			$(Z_{Cu}/Z_{Ag})^{-2.6}$	3.31	3.50

copper P measurements is not in excess of 20 percent. Assuming that the error is 20 percent and of opposite sign in the two experiments, which seems exceedingly improbable, the dependence of P on Z would be about $Z^{-3.3}$, still leaving a factor of $Z^{-1.5}$ to be accounted for if the theoretical prediction is correct. Of course a number of simplifying assumptions were made in the theoretical expression for the variation of P with Z but all of them together do not seem to be of sufficient importance to account for the divergence of the theoretical and experimental results. In fact any improvement in the theory such as taking into account rediffusion, etc., seems to make the divergence slightly greater.

In spite of the fact that the theory does not predict the variation of P with Z quantitatively it shows several qualitative facts quite clearly. First, it predicts a rapid decrease in P with increasing Z; and second, it shows the increasing relative importance of the fluorescence method of excitation over the direct excitation with increasing Z. The directly produced x-rays increasing with Z to a positive power less than 1 and the fluorescence rays increasing very much faster, about as Z^5.

The variation of Q with Z theoretically as predicted by a line of reasoning similar to that used for P is the same as for P, namely, an inverse variation with $Z^{4.8}$. The experimental variation is the same as for P so the same discrepancy between prediction and observation is observed for Q as for P.