

*Some Measurements on the Absorption of X-Rays of Long Wave-length.* By L. H. MARTIN, M.Sc.,\* Trinity College. (Communicated by Prof. Sir E. RUTHERFORD.)

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§ 1. *Introduction.*

An account is given here of the measurements of the mass absorption coefficients of iron, nickel, copper and aluminium over a wave-length range 0.705 Å.U. to 1.932 Å.U.

As these measurements were made primarily for use in another experiment, described elsewhere, in connection with which a high degree of accuracy was not necessary, they can only be regarded as preliminary. The results, however, show some interesting features which seem to justify their publication.

Measurements in this range of wave-lengths possess a particular interest, as, in virtue of the magnitude of the "total" mass absorption coefficient, the uncertain quantity representing the "scattering" absorption can be neglected in the investigation of the relationship between the "true" or "fluorescent" mass absorption coefficient, and the wave-length of the absorbed radiation. The precise investigation of this relationship has lately acquired an added interest, as the form of the relation deduced in a recent paper by Wentzel† making use of the methods of Schrödinger's wave mechanics, differs somewhat from the well-known forms given by de Broglie‡, deduced semi-empirically, and by Kramers§, deduced from Correspondence Principle considerations.

Measurements have been made previously by Barkla and Sadler|| for these elements over this range of wave-lengths. As sources of radiation, Barkla and Sadler used the *K* characteristic radiations excited in a series of radiators extending from silver to chromium, but as the constituents of such beams differ greatly in wave-length, the absorption coefficients determined differ considerably from the values corresponding to the wave-length of the *K $\alpha$*  radiation.

As the mean absorption coefficient for a heterogeneous beam of X-rays is a function of the thickness of the absorbing material used in their experiments, it is not possible to assign with any certainty "mean" wave-lengths to the series of radiations used.

\* These measurements were made during the period of an International Education Board Fellowship held by the writer.

† Wentzel, *Zeit. f. Phys.* 40, 1926, p. 575.

‡ de Broglie, *Journ. de Phys.* 3, 1922, p. 33.

§ Kramers, *Phil. Mag.* 46, 1923, p. 836.

|| Barkla and Sadler, *Phil. Mag.* 14, 1909, p. 739.

Hewlett\* carried his measurements for aluminium and iron down to 1·006 Å.U.

In the present experiments, the usual direct method for the determination of absorption coefficients has been employed. Homogeneous beams were obtained in the ordinary manner by crystal reflection, and in the case of the longer wave-lengths, suitable targets were employed in the X-ray tube to give characteristic radiations of high intensity. A method was devised which made it possible to employ the same metal foil for absorption measurements on each side of a *K* discontinuity.

In the case of Fe, Ni and Cu, it was found that when the X-rays absorbed lie in the range on the short wave-length side of the respective *K* absorption discontinuities, the mass absorption coefficients are represented with good approximation by a relation of the form

$$\mu/\rho = \text{const. } \lambda^{2.8} \dots\dots\dots(1).$$

On the long wave-length side of the *K* discontinuities and in the case of aluminium throughout the entire wave-length range the absorption coefficients are represented with good approximation by

$$\mu/\rho = \text{const. } \lambda^3 \dots\dots\dots(2).$$

Recently S. J. Allen† has published an account of experiments in which he has been able to extend considerably his previous measurements of the absorption coefficients of a number of elements from carbon to uranium from  $\lambda = 0.709$  Å.U. to 4.0 Å.U. The results of the present paper are later compared with those of Allen, and discussed in relation to the different expressions which have been deduced theoretically relating  $\mu/\rho$  with the atomic number *Z* and the wave-length  $\lambda$  of the absorbed radiation.

## § 2. *Experimental details.*

As intense beams of X-rays of long wave-length were required, it was necessary to use an X-ray tube provided with a thin aluminium window. Different types, including gas tubes, were tried, but the one shown in Fig. 1 was found to be the most satisfactory. This is of the hot cathode type. The electrodes can be withdrawn easily and replaced by the ground-glass joints provided at each end of the tube. The electrodes are surrounded by a brass tube 3 cms. in diameter which is earthed. This protects the glass walls of the X-ray tube from local heating due to scattered electrons, and also has the effect of causing the tube to run "hard." (With the Hyvac alone pumping and the filament off there was no sign of a discharge at 60—75 k.v.) The tube is exhausted continuously

\* Hewlett, *Phys. Rev.* 17, 1921, p. 284.

† S. J. Allen, *Phys. Rev.* 28, 1926, p. 907.

by a Crawford mercury vapour jet pump (Western Electric Co. pattern) and runs quite steadily when the current is not greater than 5 milliamps. In these experiments the cathode was earthed, and it may be remarked that filaments (5 amp. tungsten wire) then last six months or longer, even with practically continuous running.

A measure of the intensity of the X-ray beam issuing from the tube was obtained by placing a small ionisation chamber between the collimator slits of the spectrometer, and measuring the ionisation produced by means of a Dolezalek electrometer. Since this ionisation chamber measures the total intensity of the beam, it is important to maintain a constant potential across the X-ray tube in order to keep constant the distribution of relative intensities among the various wave-lengths. To this end a voltmeter was placed across the primary of the transformer, and the primary voltage controlled by means of rheostats in series with the primary windings. The tungsten filament was heated from accumulators. The spot from the Dolezalek electrometer, and also that from a Compton electrometer connected with the spectrometer

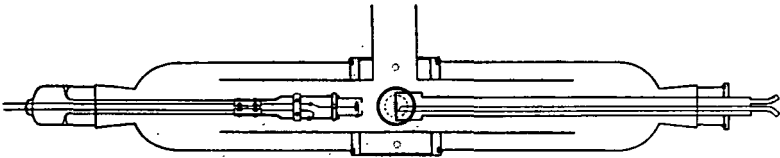


Fig. 1.

ionisation chamber, were arranged to pass across two scales placed one immediately above the other.

For wave-lengths near to and on the short wave-length side of the  $K$  absorption edges of the metals used, a special difficulty arises in obtaining foils which are at once thin enough to transmit a useful percentage of the incident radiation, and sufficiently uniform to justify the determination of the mass per unit area from measurements made on a piece of foil of the order 1 sq. inch in area. (The half value thickness for  $\text{Cu } K_{\alpha_1, \alpha_2}$  in iron is approx. 0.0003 cm.)

Foils of this effective thickness were produced by mounting a thicker foil on the moving arm of a divided circuit. The intensity is measured first with the foil perpendicular to the X-ray beam, and then when the foil is turned through a known angle  $\theta$ . The decrease in intensity is equivalent to that caused by a foil of thickness

$$d = D(\sec \theta - 1),$$

where  $D$  is the actual thickness of foil used.

A typical set of readings is shown in Table 1.

TABLE I.

IRON foil. 0.0117 gm. cm.<sup>-2</sup>  $\lambda = 1.537 \text{ \AA. U.}$   
 Effective mass per unit area 0.0117 (sec 30° - 1) = 0.00180 gm. cm.<sup>-2</sup>.

Intensities		
$I_d$	$I_0$	$I_d$
$\theta = 30^\circ$	$\theta = 0^\circ$	$\theta = 30^\circ$
2.50	4.31	2.20
2.42	4.24	2.29
2.23	4.10	2.14
2.38	4.13	2.44

This method of realising thin foils can be of use only when the initial radiations are of high intensity. In the present experiment, intense homogeneous beams were obtained by reflecting from a calcite crystal the characteristic radiations emitted from different targets placed on the anticathode of the X-ray tube. Quite wide slits may then be used, as the intensity of the general radiation is negligible compared with that of the characteristic radiations. The potential of the tube was controlled so that no second order radiation was reflected. In the case of copper, it was not possible to obtain measurements near the *K* limit on the short wave-length side, as the intensity of the general radiation from the platinum target excited at the low potentials necessary to prevent second order reflection was too small to permit of the method described above for obtaining thin foils. Foils of the required thickness could not be obtained sufficiently uniform.

In order to eliminate as far as possible lack of uniformity in the metal foils, small foils approximately one inch square were used in the determination of the mass per unit area, and in the absorption experiments, different sections of the absorber were placed in turn in the path of the X-ray beam.

The accuracy of the following results is determined by the degree of steadiness which could be maintained in the tube, but principally by the extent of the elimination of lack of uniformity of the foils. In most cases the error is estimated at 3%, but may be greater for values near to and on the short wave-length side of a *K* discontinuity.

### § 3. Results.

The results are collected in Fig. 2, where  $\log \mu/\rho$  is plotted against  $\log \lambda$  and from which the following relations are deduced.

The only results available for comparison are the recent measurements of Allen, and the much earlier ones of Barkla and Sadler. Table 3 shows the different values of  $\mu/\rho$  found for the  $K\alpha_1, \alpha_2$  radiations of the targets Fe, Ni, Cu and Zn.

ALUMINIUM foils. Mass per unit area 0.1222 gm. cm.<sup>-2</sup>  
 " " " " 0.09152 gm. cm.<sup>-2</sup>.  
 Values corrected for 0.31% iron content.

Wave-length Å. v.	Slit width mm.	$\mu/\rho$
0.705	.15	4.98
0.799	.15	7.17
0.965	.15	12.1
1.100	.15	18.2
1.293	.08	30.3
1.433	.08	40.4
1.537	.08	51.4
1.655	.08	63.9
1.932	.08	91.8

IRON foil, obtained by electrolytic deposition.  
 Mass per unit area 0.0117 gm. cm.<sup>-2</sup>

Wave-length Å. v.	Slit width mm.	$\mu/\rho$
0.705	.15	36.7
0.799	.15	53.1
0.965	.15	87.1
1.100	.20	130
1.293	.35	200
1.433	.30	265
1.537	.30	316
1.655	.30	416
1.932	.08	68.8

NICKEL foil, obtained by electrolytic deposition.  
 Mass per unit area 0.01362 gm. cm.<sup>-2</sup>  
 " " " " 0.01142 gm. cm.<sup>-2</sup>

Wave-length Å. v.	Slit width mm.	$\mu/\rho$
0.705	.15	42.1
0.799	.15	58.8
0.965	.15	103
1.100	.20	145
1.389	.30	272
1.432	.08	298
1.537	.08	43.9
1.655	.08	53.8
1.932	.08	87.8

COPPER foils. Mass per unit area 0.01508 gm. cm.<sup>-2</sup>  
 " " " " 0.01252 gm. cm.<sup>-2</sup>

Wave-length Å. v.	Slit width mm.	$\mu/\rho$
0.705	.15	49.6
0.799	.15	72.4
0.965	.15	118
1.432	.08	41.8
1.537	.08	50.1
1.655	.08	62.4
1.932	.08	95.3

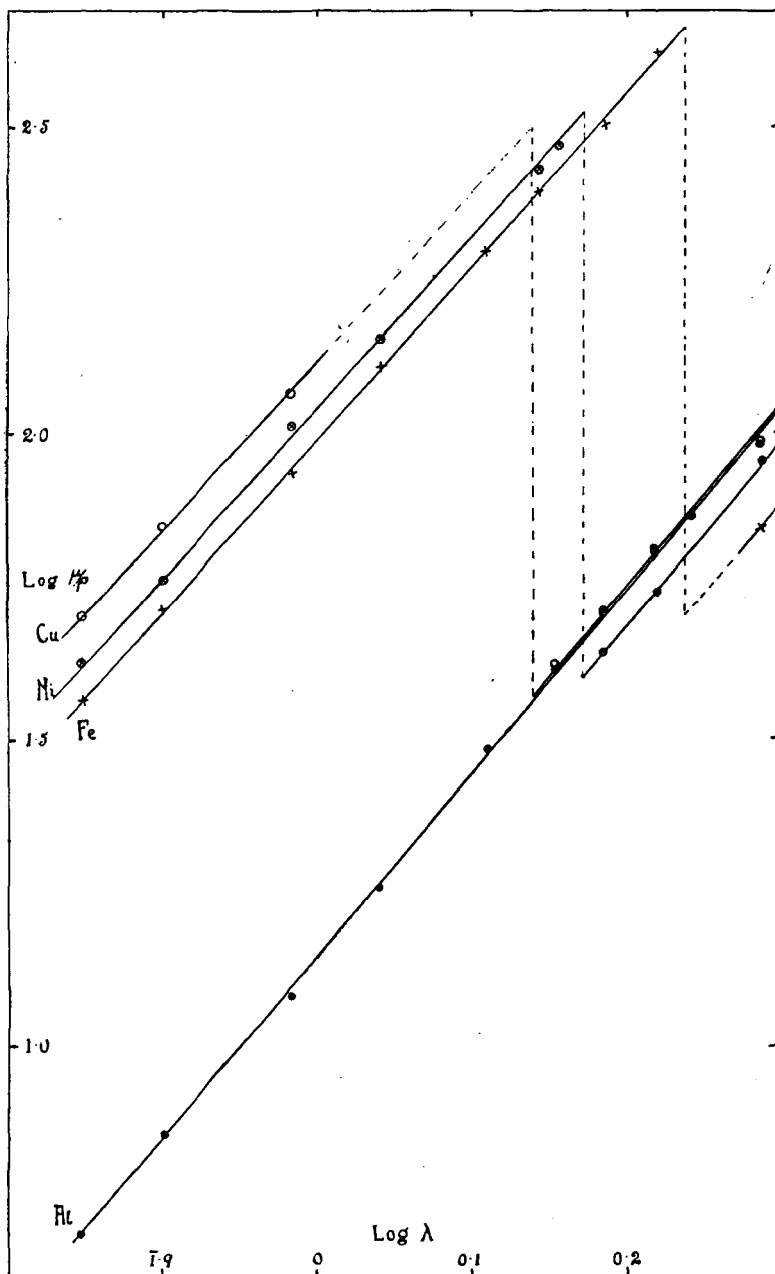


Fig. 2.

TABLE 2.

Element	$\cdot 705 \text{ \AA. U.} \rightarrow \lambda_K$	$\lambda_K \rightarrow 1.932 \text{ \AA. U.}$
	$\frac{\mu}{\rho} (K+L+\dots)$	$\frac{\mu}{\rho} (L+M+\dots)$
Fe	$98\lambda^{2.80}$	—
Ni	$110\lambda^{2.78}$	$12.4\lambda^3$
Cu	$130\lambda^{2.80}$	$14.1\lambda^3$
Al	$\frac{\mu}{\rho} (K+L+\dots)$ $13.9\lambda^{2.98}$	

TABLE 3. Values of  $\mu/\rho$ .

Absorber	Al			Fe			Ni			Cu		
Observer	B. & S. A.	M.		B. & S. A.	M.		B. & S. A.	M.		B. & S. A.	M.	
Radiation												
Fe $K\alpha_1 \alpha_2$	88.5	94.0	91.8	66.1	71	68.8	83.8	90	87.8	95.1	99	95
Ni $K\alpha_1 \alpha_2$	59.1	61.5	63.9	314	430	416	56.3	60	53.8	61.8	64	62.4
Cu $K\alpha_1 \alpha_2$	47.7	48.5	51.4	268	330	316	(62.7)	47.5	43.9	53.0	50	50.1
Zn $K\alpha_1 \alpha_2$	39.4	40.3	40.4	221	290	265	265	323	298	(55.5)	42.0	41.8

Those readings of Barkla and Sadler which are enclosed in brackets refer to cases in which the  $\beta_{12}$  constituents of the  $K$  radiations are of shorter wave-length than the  $K$  limit of the absorbing material, and therefore lead to values of  $\mu/\rho$  which are too high.

In the case of aluminium, the agreement shown is quite satisfactory. The smaller values of Barkla and Sadler would be expected as the weak short wave components  $K\beta_{12}$  are present in the beams of X-rays used by them. The agreement between the values of Allen and those given here for aluminium persists throughout the whole range of wave-lengths, and indeed, Allen finds that in the wave-length range  $\lambda 0.08 \text{ \AA. U.}$  to  $\lambda 4.0 \text{ \AA. U.}$  the absorption coefficients of aluminium are given with some accuracy by the formula

$$\mu/\rho = (13.95 \pm 0.05) \lambda^{2.92} + (0.16 \pm 0.02),$$

a relation to which that given here (Table 2) is equivalent within the limits of experimental error.

A satisfactory agreement is found too in the case of Fe (see Table 3) and Cu, but in the case of Ni, the values found here are in general from 5 % to 10 % smaller than those given by Allen.

It is difficult to account for this discrepancy, particularly as the general variation of  $\mu/\rho$  with  $\lambda$  is the same in both experiments. With regard to the present experiments possible errors introduced by lack of uniformity of the foil used or even in the determination of the mass per unit area would be reduced to a minimum by the use of two separate specimens of nickel. As mentioned above, care was taken to prevent any second order radiation being present.

The following values were found for the *K* jump, i.e. the ratio of the absorption coefficient on the short wave-length side to that on the long wave-length side at the *K* limit. It must be pointed out that the higher value refers to  $\lambda = 0.7 \text{ \AA.U.}$  and is deduced on the assumption that the *L* absorption obeys a  $\lambda^3$  law over the range,  $\lambda_K \rightarrow 0.7 \text{ \AA.U.}$  Allen finds as the result of his more extensive measurements that the *K* jump for these elements reaches its maximum value in the neighbourhood of  $\lambda = 0.6 \text{ \AA.U.}$ , when the exponent of  $\lambda$  for the *K* absorption approximates to that for the *L* absorption on the long wave-length side of the *K* limit.

The smaller value given for the *K* jump is its value at the *K* limit.

TABLE 4.

*K* jumps.

$$\frac{\mu}{\rho} (K+L+\dots) / \frac{\mu}{\rho} (L+M+\dots)$$

Element	Allen	Martin
Fe	9.5—10	9.2—10
Ni	8.8—9.8	8.5—9.6
Cu	9.1—9.8	8.6—10

§ 4. *Discussion of results.*

Several expressions have been deduced theoretically relating the “fluorescent” absorption coefficient  $\tau$  with the fundamental quantities *Z*, the atomic number of the absorbing element, and  $\lambda$ , the wave-length of the absorbed radiation.

De Broglie assumes that the laws of black body radiation can be extended to the case of X-ray absorption, and deduces semi-empirically the following expression for the atomic absorption coefficient

$$\tau_A = \text{const. } \lambda^3 \sum_n \frac{b_n}{\lambda_n^2} \dots\dots\dots(1),$$

where  $b_n$  is the number of electrons in the  $n$  quantum group and  $\lambda_n$  is the wave-length of the corresponding absorption discontinuity.



Kramers, by means of the application of the Correspondence Principle to a "hydrogen-like" atom, deduced the following expression

$$\tau_A = \text{const. } \lambda^3 Z^4 \sum_n \frac{b_n}{a_n n^2} \dots\dots\dots(2),$$

where  $a_n$  is the statistical weight of the electrons in the  $n$  quantum group.

In both (1) and (2) the summation is to be extended only over those groups of electrons whose critical frequency is less than that of the radiation considered. Recently Bothe\* has considered the effect of those electron groups which correspond to critical frequencies which are greater than that of the absorbed radiation. He finds the following relation for the atomic absorption coefficient when  $\lambda \ll \lambda_n$

$$\mu_A = \frac{2\pi\rho^2}{mc^2} \lambda^3 \sum_n \frac{b_n}{\lambda_n^2} \dots\dots\dots(3).$$

When  $\lambda > \lambda_n$  the ratio of the absorption coefficient  $\mu_n$  to that at  $\lambda_n$  on the short wave-length side of the discontinuity,  $\mu_n^0$ , is given by

$$\frac{\mu_n}{\mu_n^0} = \frac{3.74 \times 10^{-5}}{\lambda_n} \left[ \frac{2r^2 - 1}{r^2 - 1} - 2r^2 \log \frac{r^2}{r^2 - 1} \right],$$

where

$$r = \lambda/\lambda_n.$$

As such an absorption cannot be associated with the ejection of photoelectrons, Bothe concludes that at any discontinuity a large increase in the scattering coefficient should be found on the long wave-length side.

In the present experiments the scattering coefficient is so small compared with the fluorescent coefficient, that even an increase in the scattering to several times its normal value would have escaped detection.

Recently Wentzel (*l.c.*) has applied the methods of Schrödinger's wave mechanics for a "hydrogen-like" atom to obtain an expression for the  $K$  absorption coefficient which differs considerably from equations (1), (2) and (3), viz.

$$\frac{\mu}{\rho} \mathbf{A} = 1.82 \times 10^{12} \left( \frac{\lambda}{\lambda_K} \right)^{5/2} [3(A)\lambda^{1/2} + (B)\lambda_K^{1/2}]^2 \dots(4),$$

or, replacing  $\lambda_K$  by a constant multiple of  $Z^{-2}$ ,

$$\frac{\mu}{\rho} \mathbf{A} = aZ^2 \lambda^{7/2} + bZ^4 \lambda^3 + cZ^2 \lambda^{5/2} \dots\dots\dots(5),$$

\* W. Bothe, *Zeit. f. Phys.* 1927, p. 653.

where  $a, b, c$  are constants, and  $A$  is the atomic weight. ( $A$ ) and ( $B$ ) are in the present state of the theory indeterminate, but limiting values of  $\mu/\rho$  can be obtained. ( $A$ ) and ( $B$ ) are related to a phase angle  $\alpha$  by the relations

$$\cos \alpha = (A), \quad \sin \alpha = (B);$$

if  $\alpha = 0, \pi; \quad \frac{\mu A}{\rho} = \text{const. } Z^3 \lambda^{7/2};$

$$\alpha = \frac{\pi}{2}, \frac{3\pi}{2}; \quad \frac{\mu A}{\rho} = \text{const. } Z^3 \lambda^{5/2} \dots\dots\dots(6).$$

Unfortunately, owing to the presence of the factor  $3^2$  in equation (4), the limits are rather wide near the  $K$  absorption edge.

For the purpose of comparison, the values of  $\mu/\rho$  calculated for Fe from equation (4) and the experimentally determined values are given in Table 5. Equation (4) represents  $K$  absorption only, so that the calculated values have been increased by a factor 1.125 corresponding to a  $K$  jump 9.

The agreement between the calculated and observed values corresponding to  $\alpha = 0$  or  $\pi$  is very striking, since  $\alpha$  is the only constant in (4) which can be adjusted. The variation of  $\mu/\rho$  with  $\lambda$  found experimentally is however much slower than  $\lambda^{7/2}$ . Experiments by Richtmeyer and Warburton\* and Allen† in a wave-length range which is much further removed from the  $K$  discontinuities find the following relations which are in good agreement with the theoretical relations (1), (2) and (3).

Element	Richtmeyer & Warburton	Allen
Fe	$110\lambda^3 + \sigma/\rho$	$104\lambda^{2.92} + \sigma/\rho$
Ni	$145\lambda^3 + \sigma/\rho$	$128\lambda^{2.92} + \sigma/\rho$
Cu	$153\lambda^3 + \sigma/\rho$	$142\lambda^{2.92} + \sigma/\rho$

TABLE 5.

Wave-length Å.U.	$\mu/\rho$ calculated		$\mu/\rho$ observed	
	$\alpha = 0, \pi$	$\alpha = \frac{\pi}{2}, \frac{3\pi}{2}$	Allen	Martin
1.655	495	57	430	416
1.537	380	47	330	316
1.433	295	39	290	265
1.293	205	31	213	200
1.100	117	20	132	130
0.965	75.7	14	91.8	87.1
0.799	38.9	10	53.8	53.1
0.705	24.6	6.1	38.4	36.7

It has already been pointed out that the  $\lambda^3$  law holds for aluminium in a range of wave-lengths 0.1 Å.U. to 1.932 Å.U.

\* Richtmeyer and Warburton, *Phys. Rev.* 22, 1923, p. 539.

† Allen, *Phys. Rev.* March 1926, p. 266.

A somewhat similar breakdown in the  $\lambda^3$  law near a  $K$  limit has been observed by Richtmeyer in the case of Mo, and by Stoner and Martin in the case of Sn, Ag and Pd. Stoner and Martin found that from  $\lambda = 0.3 \text{ \AA.U.}$  to the  $K$  jump, the rate of increase of  $\mu/\rho$  with wave-length is slightly greater than  $\lambda^{3/2}$ , while on the long wave-length side of the  $K$  limit, the  $\lambda^3$  law was found to hold with good approximation.

These results, however, are in complete disagreement with some precise measurements made recently by Richtmeyer and Bishop\* near the  $K$  absorption limit of silver. They find that after correcting for the finite width of the slits, the  $\lambda^3$  law holds on each side of the  $K$  limit.

In the present experiments, however, it must be mentioned that although wide slits have been used near the  $K$  limit, no appreciable error can have entered on this account, because the radiations used are homogeneous and of high intensity relative to the general radiation. These experiments show that in the case of iron, nickel and copper, there is a definite departure from the  $\lambda^3$  law near the  $K$  limits.

The extent to which the  $Z^4$  law holds is shown in the following table:

TABLE 6.

Absorber	$\left(\frac{\text{Atomic Nos.}}{26}\right)^4$	Relative values of $\mu/\rho \times \text{Atomic weight}$			
		$\lambda = 1.932 \text{ \AA.U.}$		$\lambda = .965 \text{ \AA.U.}$	
		Allen	Martin	Allen	Martin
Fe	1	1	1	1	1
Ni	1.35	1.37	1.38	1.27	1.28
Cu	1.55	1.50	1.50	1.43	1.46

Table 6 indicates the existence of a considerable departure from a  $Z^4$  relation on the short wave-length side of the  $K$  limit, and it would appear that a generalised expression similar to equation (5) in which the factors  $a$ ,  $b$  and  $c$  are functions of  $Z$  and  $\lambda$  seems necessary to represent the values of  $\mu/\rho$  in an extended wave-length range. Wentzel points out that  $a$  depends ultimately on  $\lambda$  and  $Z$  and it is possible that when, as a result of the development of wave mechanics,  $a$  can be evaluated, a form of (5) will be found to fit experimental results.

In conclusion, the author wishes to express his gratitude to Professor Sir Ernest Rutherford for the continued advice and assistance received from him during the course of this experiment.

\* Richtmeyer and Bishop, *Phys. Rev.* 27, 1926, p. 294.