

FURTHER STUDIES OF THE ABSOLUTE INTENSITY OF EMISSION OF CHARACTERISTIC X-RADIATION

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Summary

The theory of Metchnik and Tomlin (1963) relating to the characteristic X-ray yield from thick targets bombarded with electrons is further discussed and a simple formula is developed from it. Some selected results from an extensive experimental study of X-ray yield are presented and these are compared with other published results and various theoretical discussions.

I. INTRODUCTION

This paper is a continuation of the earlier work of Metchnik and Tomlin (1963) and presents measurements of the absolute yield of characteristic K radiation from thick targets of light elements (titanium, aluminium, and carbon) together with some further theoretical discussion, particularly of the excitation cross sections, and the development of a relatively simple explicit formula for the yield. These results, both theoretical and experimental, are compared with the work of other authors.

On the theoretical side since the early work of Worthington and Tomlin (1956), which ignored electron scattering in the target, several attempts have been made to allow for this. Archard (1960) assumed two types of idealized electron paths in the target and fixed the disposable parameters by reference to certain experimental results. Brown and Ogilvie (1964) found an explicit expression for the yield by means of an averaging procedure and the introduction of a factor, to account for electron scattering, which had to be determined from a measurement of the yield for each target material. Suoninen (1964) used a semi-empirical discussion of electron penetration from Makhov (1960) to derive an integral expression for the yield.

The theory of Metchnik and Tomlin (1963) is based on a rigorous treatment of multiple scattering in an infinite medium due to Lewis (1950), and, although it is applied to a semi-infinite target, an objection that is less serious for lighter elements, and involves an averaging procedure, it does not depend upon disposable parameters or quantities that must be fixed by measurement.

A simple treatment of the problem, ignoring absorption in the target, was given by Green and Cosslett (1961). This may be expected to be valid for large take-off angles and sufficiently low voltages, and this is shown to be the case in their recent paper (Green and Cosslett 1968).

The most extensive experimental results yet published are those of Campbell (1963), Metchnik and Tomlin (1963), Brown and Ogilvie (1964), Birks *et al.* (1965),

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and Green and Cosslett (1968). Some other results are referred to later. Where direct comparisons are possible there are some discrepancies in reported yield measurements which seem to exceed experimental error, and some specific criticisms of experimental procedure are made in discussing these results.

II. THEORY

(a) Intensity Formulae

Following Metchnik and Tomlin (1963) the intensity of emission, expressed as the number of $K\alpha$ quanta emitted per incident electron per 4π steradian in a direction at an angle ϕ to the target surface when the electron beam is incident at an angle θ to the normal, is given by

$$N_\phi = k \int_{T_0}^{T_K} N Q_K \frac{ds}{dT} \exp(-\mu\rho\langle x \rangle \operatorname{cosec} \phi \cos \theta) dT, \quad (1)$$

where T is the kinetic energy of an electron in the target, T_K the ionization energy of a K-shell electron, N the number of target atoms per unit volume, Q_K the cross section for K excitation, s the distance in which an electron's initial energy T_0 is reduced to T , μ the mass absorption coefficient and ρ the density of the target material, and $\langle x \rangle$ the mean depth of the electrons of energy T . The quantity ds/dT is given by Bethe's (1930) stopping-power formula, while

$$k = Rp\{(P+1)/P\}\omega_K, \quad (2)$$

where R is the rediffusion factor discussed in detail by Tomlin (1964), whose values will be used below. p is the ratio of $K\alpha$ to total K radiation (Williams 1933) and is, of course, unity if the total K intensity is to be calculated. The factor $(P+1)/P$ allows for indirect excitation of K radiation by the continuous spectrum and is unity for light elements with $Z < 30$ (Green and Cosslett 1961; Brown and Ogilvie 1964). The excitation cross section Q and the fluorescence yield ω_K are further discussed in Subsections (b) and (c) below.

The mean depth $\langle x \rangle$ was given by an elaborate expression by Metchnik and Tomlin (1963) and results calculated using this expression will be denoted by MT. The following simpler and very good approximation was obtained later by Tomlin (1966),

$$\langle x \rangle = \frac{8R_B}{Z+9} \left(1 - \frac{(2\alpha+1)(Z+1)}{8\gamma_0} \right) \left[1 - \exp \left\{ -\frac{1}{4}(Z+9) \left(1 - \frac{T}{T_0} \right) \right\} \right], \quad (3)$$

which may be further simplified for present purposes to

$$\langle x \rangle = \frac{8R_B}{Z+9} \left[1 - \exp \left\{ -\frac{1}{4}(Z+9) \left(1 - \frac{T}{T_0} \right) \right\} \right]. \quad (4)$$

In these formulae R_B is the Bethe range for electrons in the target (Worthington and Tomlin 1956), Z is the target atomic number,

$$\gamma_0 = \frac{1}{4}(Z+9) \ln(2T_0/J),$$

where the mean ionization energy J is given by

$$J = 1.602 \times 10^{-12} BZ \quad \text{erg,}$$

with B tabulated by Bakker and Segré (1951), and

$$\alpha = \ln(BZ^{\frac{1}{2}}) - 2.917.$$

The expression (4) relates to electron penetration in an infinite medium. Bishop (1965) showed that the following modification of (4) agreed well with his numerical results for multiple scattering in a semi-infinite target of copper:

$$\langle x \rangle = \frac{8aR_B}{Z+9} \left[1 - \exp \left\{ -\frac{1}{4a} (Z+9) \left(1 - \frac{T}{T_0} \right) \right\} \right], \quad (5)$$

where, for copper, $a = 1.5$. Whether or not this result is generally valid has yet to be established. One might expect that $a \rightarrow 1$ as $Z \rightarrow 0$.

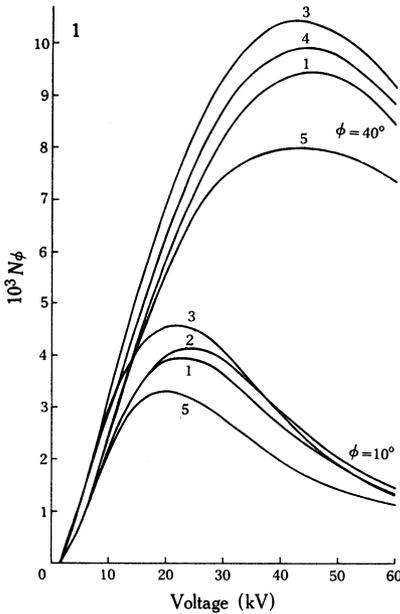


Fig. 2 (below).—Plots of $|Q_K ds/dt|$ as a function of electron energy for copper and aluminium targets.

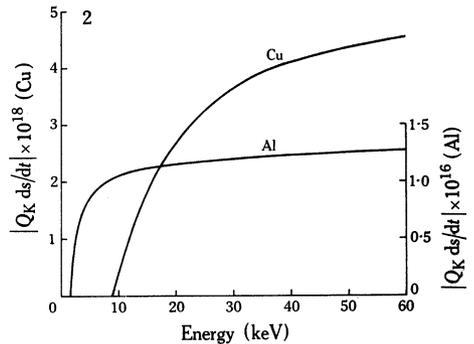


Fig. 1 (left).—Theoretical results for aluminium K radiation intensities: 1, calculated from equations (1) and (4); 2, from the Metchnik–Tomlin (1963) formulae; 3, from the approximate formula (10) omitting the η term; 4, from the complete formula (10); 5, from the formula (5) for a semi-infinite medium, $a = 1.5$. For the angle of emission $\phi = 40^\circ$ curves 1 and 2 were barely distinguishable.

It will appear that quantum yields based on this formula with $a = 1.5$ are somewhat too low, which is consistent with the argument advanced by Metchnik and Tomlin (1963) that their theory involved two opposed sources of error. Thus putting $a = 1$ seems acceptable.

The effects of these different expressions for $\langle x \rangle$ on calculated values of N_ϕ are shown in Figure 1, together with results calculated from an approximate formula derived as follows.

Figure 2 shows plots of the factor $Q_K ds/dT$ occurring in the integral for N_ϕ . Evidently if $T_0/T_K = V_0/V_K$ (V is the potential corresponding to energy T) is sufficiently large $Q_K ds/dT$ is almost constant over the range of integration and a fairly good approximation for N_ϕ will result if this factor is taken outside the integration sign. A better approximation is obtained by replacing the $Q_K ds/dT$ curve by the tangent drawn at $T = T_0$, that is, by writing

$$Q_K \frac{ds}{dT} = \left(Q_K \frac{ds}{dT} \right)_{T_0} + \left\{ \frac{d}{dT} \left(Q_K \frac{ds}{dT} \right) \right\}_{T_0} (T - T_0). \quad (6)$$

The approximate formulae obtained in this way will clearly be more useful (Fig. 2) for light elements, since for these V_0/V_K may be large for electrons of moderate energy. Writing

$$P = - \left(Q_K \frac{ds}{dT} \right)_{T_0} \quad \text{and} \quad P' = - \left\{ \frac{d}{dT} \left(Q_K \frac{ds}{dT} \right) \right\}_{T_0}$$

and using equation (5) for $\langle x \rangle$ leads to

$$N_\phi = kNe^{-A} \int_{T_K}^{T_0} \{P + P'(T - T_0)\} \exp \left[A \exp \left\{ -\frac{Z+9}{4a} \left(1 - \frac{T}{T_0} \right) \right\} \right] dT, \quad (7)$$

where

$$A = \{8aR_B/(Z+9)\}_{\mu\rho} \operatorname{cosec} \phi \cos \theta.$$

The first of these integrals is readily evaluated in terms of the exponential integral function, and the second by expansion of the integrand in powers of $A \exp[-\{(Z+9)/4a\}(1-T/T_0)]$ to give

$$N_\phi = kNe^{-A} P \left\{ \frac{4aT_0}{Z+9} \left(Ei(A) - Ei(x_K) \right) - \frac{P'}{2P} (T_0 - T_K)^2 - \left(\frac{4aT_0}{Z+9} \right)^2 \frac{P'}{P} S \right\}, \quad (8)$$

where

$$x_K = A \exp \left\{ -\frac{Z+9}{4a} \left(1 - \frac{T_K}{T_0} \right) \right\}$$

and

$$S = \sum \frac{1}{n^2 n!} \left[A^n - \left(1 + n \frac{Z+9}{4a} \left(1 - \frac{T_K}{T_0} \right) \right) x_K^n \right].$$

From Bethe's (1930) formula,

$$-(dT/ds) = (2\pi e^4 NZ/T) \ln(2T/J),$$

and the modified expression for Q_K given by Worthington and Tomlin (1956), namely

$$Q_K = (2\pi e^4 / TT_K) b \ln(4T/B), \quad (9)$$

where

$$B = \{1.65 + 2.35 \exp(1 - T/T_K)\} T_K,$$

we obtain

$$P = \frac{b \ln(4T_0/B_0)}{NZT_K \ln(2T_0/J)}$$

To obtain P'_0 it is sufficient to keep B constant and equal to $4T_K$, since this gives a good approximation for Q_K at low voltages where the first-order correction is most significant. Then

$$\frac{P'}{P} = \frac{1}{T_0} \frac{\ln(2T_K/J)}{\ln(T_0/T_K)\ln(2T_0/J)}$$

Consequently

$$N_\phi = \frac{kb T_0 \ln(4T_0/B_0)}{Z T_K \ln(2T_0/J)} e^{-A} (\xi - \eta), \tag{10}$$

where

$$\xi = \{4a/(Z+9)\}\{Ei(A) - Ei(x_K)\}$$

and

$$\eta = \frac{\ln(2T_K/J)}{\ln(T_0/T_K)\ln(2T_0/J)} \left\{ \frac{1}{2} \left(1 - \frac{T_K}{T_0} \right)^2 + \left(\frac{4a}{Z+9} \right)^2 S \right\}$$

Results for aluminium K radiation calculated from this formula are shown in Figure 1 (curves 3 and 4), from which it appears that omission of the term η results in an overestimate of about 5% when $\phi = 40^\circ$, and a similar error at $\phi = 10^\circ$ for which only the curve with $\eta = 0$ is drawn. In the case of carbon K emission the effect of the η term amounts to only about 2.5%. The theoretical curve is shown in Figure 6(b) together with experimental results.

(b) *Excitation Cross Section*

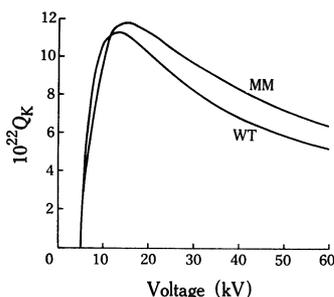


Fig. 3.—A comparison of the MM and WT formulae for the excitation cross section of titanium atoms.

In earlier work the semi-empirical expression for Q_K (equation (9)) was used as a convenient form for numerical calculation, and it is a manageable form for the derivation of the relatively simple formula (10). However, it is derived from approximation to a very elaborate formula discussed by Massey and Mohr (1933), Burhop (1940), and Mott and Massey (1965). With the availability of high speed computers the use of this more reliable formula becomes feasible (Fong 1967). Figure 3 shows a comparison of this formula (curve labelled MM) with equation (9), for which the curve is labelled WT. This curve for titanium is typical of those for

other elements, showing that the WT (Worthington and Tomlin 1956) formula tends to underestimate the cross section at higher energies.

The effects of these two expressions on calculated X-ray yields are shown in Figures 4-6.

(c) Fluorescence Yield

A comprehensive review of the atomic fluorescence yield has been given by Fink *et al.* (1966), from which it appears that Callan's (1963, cited after Fink *et al.*) theoretical results for ω_K in the range of $Z > 20$ are in good agreement with observations, although the latter show a rather wide spread. From $Z = 20$ to 16 Callan's curve is also in fairly good agreement with increasingly uncertain experimental values, but it is not at all clear that his curve could be reliably extrapolated to $Z = 13$ for aluminium, and for this and lighter elements the experimental data are very discrepant. For such elements we have therefore chosen values of ω_K to make our theoretical yields agree well with the experimental results, i.e. we have used our measurements of yield as a means of determining ω_K .

III. EXPERIMENTAL

The X-ray tube was that described by Metchnik and Tomlin (1963) except that the long slit window was replaced by a number of fixed ports to which gas-flow proportional counters could be attached with only a single window, and no air path, between detector and source. Two counters were constructed. Counter A had a cylindrical active volume of diameter 0.264 in. and length 0.882 in., and a central tungsten wire of 0.0005 in. diameter. Counter B had diameter 0.5 in., length 5.0 in., and a central platinum wire of 0.002 in. diameter. The gas used was a 3% carbon dioxide-argon mixture with a flow rate of 1.0-1.5 ft³ hr⁻¹. Aluminized Mylar windows were used for aluminium K radiation for which the measured transmission ratio was 0.42. For the softer carbon K radiation a thin film of Parlodion (cellulose nitrate) was applied to a brass block in which were six small holes subtending a total solid angle at the target of 1.89×10^{-5} steradian. The counters were used with standard commercial counting equipment.

A detailed account of the design and properties of these counters is given by Fong (1967), who carefully investigated all the factors relevant to their performance as accurate absolute detectors of X-ray quanta. This involved verification of linearity and proportionality, a study of the escape peak and resolution, and determination by absorption measurements of the quantum counting efficiency.

To eliminate spurious counts that might arise from back-scattering electrons striking the counter window, it was necessary to place a small magnet, producing a transverse field of 700 Oe, in front of the counter to deflect such electrons from the window.

For each intensity measurement the pulse height discriminator adjustments were made after recording a differential counting curve. A total number of counts in excess of 10^4 was registered, and the counting rate was kept below the predetermined limit where resolution-time losses became significant. These conditions could be met with an electron beam current of about 10^{-7} A and a counting time of 50 or 100 sec. Contamination of the target surface, which was remote from the diffusion pump, was never obvious, but the surface was frequently cleaned and polished. There was no sign of any decrease in emission with time for any of the K radiations studied.

An important consideration was the error arising from the counting of that part of the continuous spectrum accepted by the pulse height discriminator. For aluminium K and shorter wavelength radiation the correction was readily made by observing the trend of the differential curve due to the bremsstrahlung radiation alone, on either side of the characteristic peak, and drawing a line below the peak to separate continuous and characteristic radiation. Since the correction for the wavelengths mentioned was small a straight line was an adequate approximation for the dividing line. The correction was then made by measuring the areas under the curves. Also allowance could be made for the "tails" of the characteristic peak beyond the discriminator window by estimating the appropriate areas under the differential curve. The correction required was very small.

For wavelengths larger than aluminium K radiation the resolution of the proportional counters did not permit an accurate graphical procedure. In this case the method of Campbell (1963) was followed using a computer for the curve fitting procedure involved (Fong 1967).

IV. RESULTS AND DISCUSSION

A series of measurements of copper and chromium K radiation was made in order to test pulse height discrimination as a method for isolating characteristic radiations and measuring the absolute intensities. Some of the earlier measurements (Metchnik and Tomlin 1963) of copper K emission using balanced filters, to isolate the K line, with scintillation and proportional counters were repeated and compared with corresponding measurements made with the gas flow counters and pulse height discriminator. These results were also compared with Metchnik's (1961) measurements made with various detectors and a crystal monochromator (Fong 1967). The very satisfactory agreement of all these observations indicated that the pulse height discriminator method was a sound technique. The greatest discrepancy in all the measurements of copper $K\alpha$ yield was 20% and generally the agreement was within about 10%.

Experimental results are presented for titanium, aluminium, and copper K radiations and are compared with theoretical results calculated from the Metchnik-Tomlin (1963) formulae and those of other authors where feasible. The values of ω_K used in our calculations are given in the captions to the appropriate figures and the values of k were obtained from Tomlin's (1964) tables with modifications, for new choices of ω_K and, in some cases, for angle of incidence which affects the back-scattering coefficient. Theoretical curves were calculated using both the MM and WT formulae for the excitation cross sections.

(a) *Titanium K Radiation*

Figure 4 shows the results of measurements of emission at angles of 40° and 5° with the electron beam incident normally, together with the results of calculations from the Metchnik-Tomlin formulae. The experimental results fall about 25% below the MM curve calculated with Callan's (1963) value of $\omega_K = 0.213$. There is good agreement between theory and experimental results for larger ϕ , where the target absorption is small, if $\omega_K = 0.155$, which is Burhop's (1952) value. Assuming that

our theory and measurements are reliable for such conditions, we conclude that the above value is a reasonably accurate one of ω_K for titanium.

Other measurements of titanium K radiation yield have been given by Brown and Ogilvie (1964) and Birks *et al.* (1965). Green and Cosslett (1968) have published experimental values of X-ray generation efficiency, i.e. the number of quanta generated within the target. For comparison purposes these values for titanium, aluminium, and carbon, have been converted to observed yields, N_ϕ , by means of the absorption function of Green (1964). Our measurements, made with the same geometry as used by Brown and Ogilvie, are about 10–15% higher than theirs, which is reasonably satisfactory. The results of Birks *et al.* are some 60–70% higher than ours, but these authors apparently measured the target-to-ground current and do not state that they allowed for secondary electron emission. If they did not, it might well account for this and the similar discrepancy that also exists between their measurements and ours for copper K radiation.

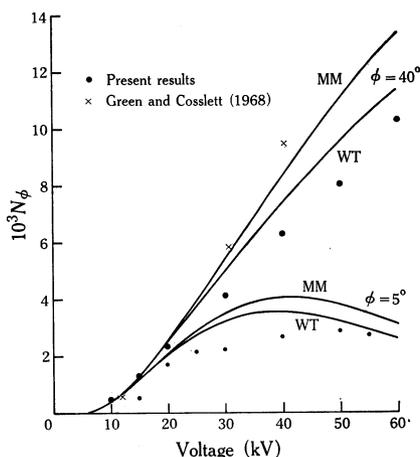


Fig. 4.—Intensities of titanium K emissions. The curves labelled MM and WT were calculated from the corresponding excitation cross section formulae. Experimental points are also shown, where the values of Green and Cosslett (1968) have been converted to observed intensities.

The observed yields obtained by Green and Cosslett (1968), which are also plotted in Figure 4, are higher than ours: about 50% greater at 40 kV and about 12% above our theoretical curve calculated for $\omega_K = 0.213$. However, our result for $V_0/V_K = 6$ falls on their lower theoretical curve (Fig. 9 of their paper) while their experimental point is appreciably above their upper theoretical value.

(b) Aluminium K Radiation

Measurements were made at $\phi = 5^\circ, 10^\circ, 20^\circ, 32^\circ$, and 38° for normal incidence of electrons, and also with the target inclined at 45° to the electron beam and a take-off angle of 45° . This was the geometry used by Campbell (1963), and Figure 5(a) shows his results, a theoretical curve (dashed) due to Archard (1960), and our experimental results and theoretical curves. The latter were calculated with $\omega_K = 0.045$ as determined by Bertrand, Charpak, and Suzor (1959). Since Campbell's results were about 65% of ours, we repeated the measurements with a plain Mylar window in the counter to eliminate any possibility of excitation of aluminium K radiation

in this window. There was no significant change in the results, and the two counters A and B previously described also yielded consistent measurements.

Brown and Ogilvie (1964) obtained yields at $\phi = 15.5^\circ$ that are about 75% of our values measured at $\phi = 10^\circ$. Figure 5(b) shows our measurements for $\phi = 32^\circ$ compared with those of Dolby (1960), and those of Green and Cosslett (1968) converted to observed values, together with theoretical curves due to Suoninen (1964) and those from the Metchnik-Tomlin (1963) theory. There is good agreement between the experimental results shown.

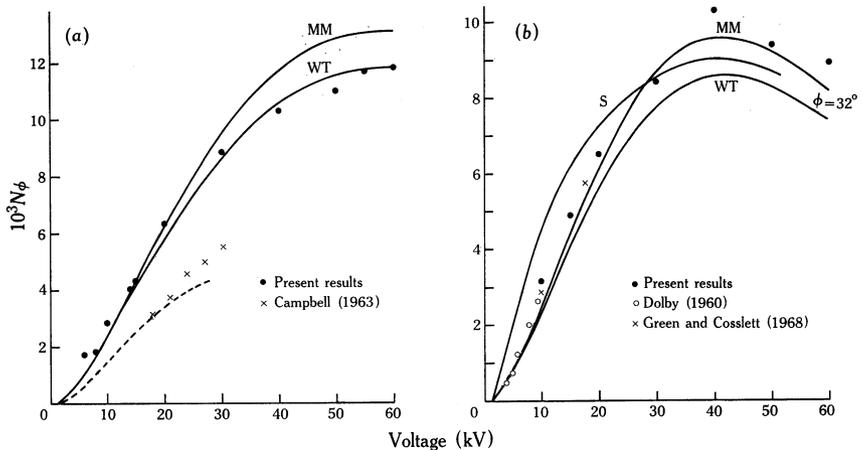


Fig. 5.—Intensities of aluminium K emissions for (a) oblique geometry, i.e. electrons incident on the target at 45° and observations at right angles to the electron beam, and (b) electrons incident normally. The curves labelled MM and WT are our theoretical curves for $\omega_K = 0.045$, and experimental points are also shown. In (a) the dashed curve is from Archard's (1960) theory, after Campbell (1963), with $\omega_K = 0.023$. In (b) the curve S is from Suoninen (1964) for $\phi = 30^\circ$ and $\omega_K = 0.028$, while the experimental points of Dolby are for $\phi = 33^\circ$ and those of Green and Cosslett have been converted to observed intensities.

Suoninen's theory, for a given value of ω_K , gives considerably higher yields than the Metchnik-Tomlin theory, as is evident from Figure 5(b) where Suoninen's curve is plotted for $\omega_K = 0.028$ and the others for $\omega_K = 0.045$. This, together with the relatively large differences in the theoretical curves at lower voltages, suggests that Makhov's (1960) theory gives a very different picture of electron penetration as compared with the discussion of Metchnik and Tomlin (1963) based on Lewis's (1950) theory of multiple scattering.

(c) Carbon K Radiation

Figure 6(a) shows experimental results for the target inclined at 45° to the electron beam and a take-off angle of 45° . Campbell's (1963) results obtained with this same geometry are also shown together with Metchnik-Tomlin theoretical curves and also a curve (dashed) from Archard's (1960) theory as presented by Campbell (1963). Our theoretical curves were obtained using a value of $\omega_K = 2.00 \times 10^{-3}$, which gives good agreement with the experimental data. This was

done since there are no experimental values and it seemed doubtful that any existing theories were adequate for such light elements. Green and Cosslett (1968) quote some values for the parameter appearing in Burhop's formula with which one obtains $\omega_K = 1.7 \times 10^{-3}$.

The agreement between Campbell's measurements and ours is good but the results of Green and Cosslett, converted as described above, are about twice our measurements for normally incident electrons and a take-off angle of $\phi = 40^\circ$ as shown in Figure 6(b), where again the Metchnik-Tomlin theoretical curves are plotted with $\omega_K = 2.00 \times 10^{-3}$. Such a discrepancy would appear to be well outside the

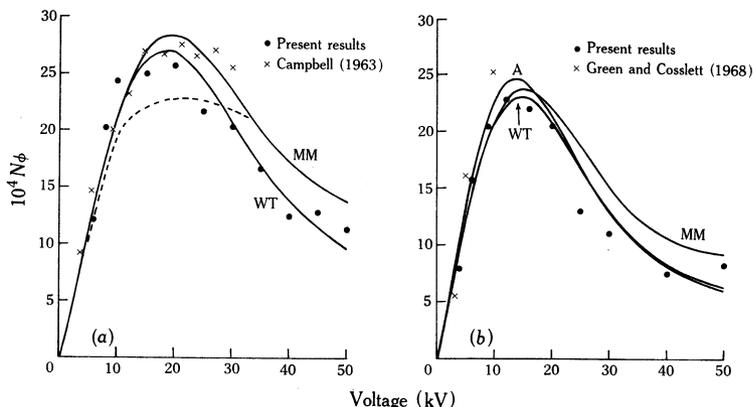


Fig. 6.—Intensities of carbon K emissions for (a) electrons incident at 45° and observation at right angles to the electron beam, and (b) electrons incident normally and a take-off angle $\phi = 40^\circ$. The curves labelled MM and WT are our theoretical results with $\omega_K = 2.00 \times 10^{-3}$, and experimental points are also shown. In (a) the dashed curve is from Archard's (1960) theory, after Campbell (1963), with $\omega_K = 1.44 \times 10^{-3}$. In (b) curve A is from the approximate formula (10) and the experimental results of Green and Cosslett have been converted to observed intensities and then divided by two.

limits of experimental error. Green and Cosslett used a window in their counter which is much more highly absorbing for carbon K radiation than the very thin nitrocellulose windows used by Campbell and by us. Such a window would have a large absorption correction, and possibly carbon K radiation may have been indirectly excited in it. Dolby (1960) using the same sort of counter also obtained much higher yields than those of Figure 6(b).

V. CONCLUSIONS

The discrepancies among the experimental results of different workers appear to be rather greater than experimental error. In the case of titanium K radiation our results are a little higher than those of Brown and Ogilvie (1964) but somewhat lower than those of Green and Cosslett (1968). For aluminium K radiation Campbell's (1963) values are less than ours, which agree well with the measurements of Dolby (1960) and of Green and Cosslett. Our measurements for carbon agree well with those of Campbell but are much less than those of Green and Cosslett. For each of these targets at least one other investigation has yielded results very close to ours.

On the theoretical side, apart from the theory of Archard (1960), which depends upon adjustment of disposable parameters, and of Brown and Ogilvie (1964), which requires a measurement with each target material to fix a parameter, the most complete attempts to produce satisfactory theories are those of Green and Cosslett (1961), Suoninen (1964), and Metchnik and Tomlin (1963) together with the further modifications given here. Green and Cosslett's treatment neglects X-ray absorption in the target, so that the emission is independent of angle, and is therefore valid only for a limited range of incident electron energy. Within these limitations it appears to be satisfactory. The remaining two theories attempt to take account of absorption in the target and do so with some success. However, Suoninen's theory appears to overestimate the emission as compared with that of Metchnik and Tomlin (1963), although both put the peak of the emission curve at the same voltage. Apart from the absolute values of N_ϕ , complicated by uncertainty about values of ω_K for the light elements, the shapes of the curves from the Metchnik-Tomlin theory, or from the simpler form (equation (10)) suitable for light elements (see Fig. 6(b)), fit the experimental data over the whole of the nonrelativistic voltage range very well.

By choosing the values of ω_K to secure good agreement between our theoretical and experimental results we arrive at the following values for fluorescent yield, which may be compared with the values from Burhop's (1952) formula after Green and Cosslett (1968):

Target element	C	Al	Ti	Cr	Cu
$\omega_K \times 10^3$ (present paper)	2.0	0.045	0.16	0.23	0.37
$\omega_K \times 10^3$ (Green and Cosslett)	1.7	0.028	0.18	0.24	0.40

The value of $\omega_K = 0.045$ for aluminium may seem high, but reference to the review of Fink *et al.* (1966) shows that it is not inconsistent with the values given for carbon and titanium.

VI. ACKNOWLEDGMENTS

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