

# CALCULATIONS OF INTENSITIES OF EMISSION OF CHARACTERISTIC X-RADIATION

By S. G. TOMLIN\*

[*Manuscript received July 8, 1964*]

## *Summary*

Intensities of emission of characteristic X-radiation from thick targets of silver, copper, chromium, aluminium, carbon, boron, and beryllium have been calculated using the theory of Metchnik and Tomlin (1963) and the results have been compared with experimental data. The effect of the backscattering of electrons from the target has been examined in detail, and the correcting factor  $k$ , which allows for this and for the effects of fluorescence yield and indirect excitation by the white radiation, has been tabulated.

## INTRODUCTION

In recent years there has been considerable interest in the absolute intensity of emission of characteristic X-radiation from thick targets, especially in connection with the development of the electron probe X-ray analyser. Worthington and Tomlin (1956) presented a theoretical discussion of the problem of calculating the number of  $K\alpha$  quanta emitted from a thick target for each incident electron. A modified theory was given by Archard (1960) and also by Metchnik and Tomlin (1963) who used the multiple electron scattering theory of Lewis (1950) to allow for scattering of the electron beam in the target. These latter authors also compared the results of their theory, and of the earlier theory of Worthington and Tomlin (1956), with experimental measurements on radiation from silver, copper, and chromium targets. They showed that the improved theory of Metchnik and Tomlin (1963) led to reasonably good agreement between calculation and experiment. In any of these theories there appears a factor  $k$  which corrects for the following effects:

- (a) backscattering of electrons from the target,
- (b) fluorescent yield of  $K$  radiation,
- (c) indirect excitation of  $K$  radiation by the continuous radiation always present,
- (d) the fraction of  $K\alpha$  radiation to total  $K$  radiation.

Of these effects, the last three have been carefully discussed by Campbell (1963) in a paper presenting experimental measurement on emission from elements of low atomic numbers. He found a large discrepancy between his measurements on aluminium and the calculations of Worthington and Tomlin (1956). He rightly concluded that this discrepancy was due to an error in the value of  $k$  used in the calculations.

\* Physics Department, University of Adelaide.

It is the purpose of the present paper to give a discussion of the correction for electron backscattering, to make the best possible estimates of the factor  $k$ , and to compare calculated emission intensities with the experimental results of Campbell (1963) and Metchnik and Tomlin (1963) for a number of elements with atomic numbers ranging from 4 to 47.

# ELECTRON BACKSCATTERING

Some of the electrons in a beam falling on a target will be backscattered with energies ranging from zero up to the initial energy. These do not contribute their full quota to the number of emitted quanta. To correct for this, Webster, Clark, and Hansen (1931) used a semi-empirical procedure to calculate what contribution these backscattered electrons would have made by estimating the emission they would produce if allowed to fall on a target. It is proposed to modify their method as follows.

If  $F(W)dW$  is the fraction of the backscattered electrons with energies in the range  $WV_0$  to  $(W + dW)V_0$ , where  $V_0$  is the incident energy (electron-volts),

$$\int_0^1 F(W) dW = 1.$$

If  $I_0$  is the incident electron beam current, the total backscattered current is

$$I_b = bI_0,$$

where  $b$  is the backscattering coefficient discussed by Archard (1961) and by Tomlin (1963).

The backscattered current of electrons with energies in the range  $WV_0$  to  $(W + dW)V_0$  is

$$I_{bW} = bI_0 F(W) dW.$$

If  $V_k$  is the  $K$ -ionization energy of the target atoms, then electrons backscattered with energies in the range  $V_0 - V_k$  to  $V_0$  cannot have excited a  $K$  electron to produce a photon and should, therefore, be subtracted from the incident beam. Putting

$$W_k = V_k/V_0,$$

the current to be subtracted is

$$bI_0 \int_{1-W_k}^1 F(W) dW.$$

To correct for the remaining backscattered electrons, we calculate how many photons they would produce if allowed to fall upon a second target without further backscatter.

If  $N_{\phi 0}(V)$  is the number of photons produced per electron per  $4\pi$  steradian, in the direction making an angle  $\phi$  with the target surface for an incident energy  $V$ , the number of quanta generated in the secondary target is

$$\int_0^{1-W_k} bF(W) N_{\phi 0}(WV_0) dW.$$

Hence if  $N_{\phi 0}$  is the calculated number of photons emitted from a target per electron per  $4\pi$  steradian, neglecting the backscatter, the true number emitted is given by

$$N_{\phi} = RN_{\phi 0},$$

where

$$R = 1 - b \left\{ \int_{1-W_k}^1 F(W) dW + \int_0^{1-W_k} F(W) \frac{N_{\phi 0}(WV_0)}{N_{\phi 0}(V_0)} dW \right\}. \quad (1)$$

$R$  is the correcting factor for electron rediffusion or backscattering. To evaluate  $R$  one needs an expression for  $F(W)$ , or experimental curves may be used if available. We use one of the empirical expressions suggested by Webster, Clark, and Hansen (1931), which, when normalized, reads

$$F(W) = \frac{g^2}{1 - (g+1)e^{-g}} (1-W)e^{-g(1-W)}, \quad (2)$$

where  $g$  is a parameter to be found for a given target.

From  $F(W)$  the first moment of  $W$  is

$$\overline{W} = \frac{(g-2) + (g+2)e^{-g}}{g[1 - (g+1)e^{-g}]}, \quad (3)$$

and according to Sternglass (1954) the results of his experimental studies of backscattering led to the relation

$$\overline{W} = 0.45 + 2 \times 10^{-3}Z, \quad (4)$$

where  $Z$  is the atomic number of the target atoms.

From a graphical solution of these equations one finds that, for  $Z$  in the range 4–70,  $g$  is a very nearly linear function of  $Z$  and may be written

$$g = 0.032Z + 1.90. \quad (5)$$

This equation has been used to find  $g$  and hence the function  $F(W)$  for a given target.

Figure 1 shows distribution functions  $F(W)$  calculated in this way, together with normalized experimental curves obtained from the results of Sternglass (1954). Although the agreement is not good, it is adequate for estimating the correcting factor  $R$  because the deviations are reduced in the integrations over  $W$ . For example, for 40 kV electrons incident normally on a copper target, the above method of calculation gives  $R = 0.86$  for radiation emitted at  $\phi = 50^\circ$ . If one assumes that the true curve  $F(W)$  for copper would be very close to that of Sternglass (1954) for iron and uses this experimental result for  $F(W)$ , one finds  $R = 0.89$ .

In evaluating the expression for  $R$  (equation (1)),  $b$  may be obtained from the empirical formula given by Tomlin (1963) or from experimental measurements referred to in the same paper. In some of the following calculations, in cases where the electrons are incident at  $45^\circ$  to the target surface, the experimental results of Campbell (1963) have been used. The integrals were evaluated numerically and, in the second one of

equation (1), the ratio  $N_{\phi 0}(WV_0)/N_{\phi 0}(V_0)$  was taken from experimental curves. If such curves are not available, theoretical curves calculated with  $R = 1$  may be used to estimate this term and an improved value of  $R$  then obtained. If necessary, the process may be repeated for a better approximation. Some calculated values of  $R$  are given in Table 1.

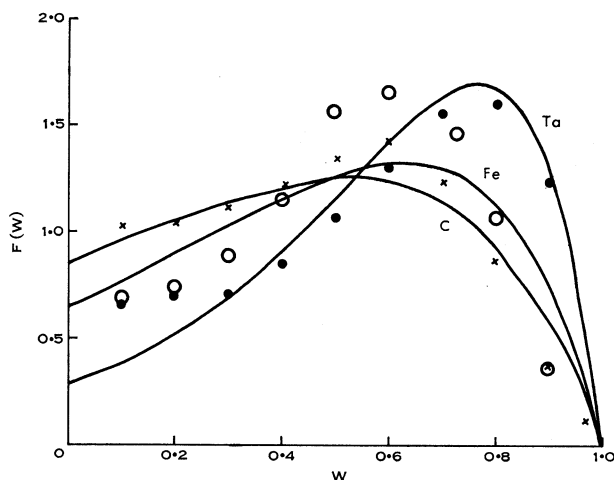


Fig. 1.—The function  $F(W)$  compared with experimental points from the normalized curves of Sternglass (1954). Experimental points for tantalum ●, for iron ○, for carbon ×.

TABLE 1

VALUES OF  $R$  FOR ELECTRONS INCIDENT NORMALLY ON Ag, Cu, AND Cr, AND AT AN ANGLE OF  $45^\circ$  ON Al, C, B, AND Be

$\phi$  is the angle of emission with respect to the target surface

Target	$\phi^\circ$	Voltage (kV)								
		5	10	15	20	30	40	50	60	70
Ag	40					0.65	0.71		0.81	0.80
	1					0.65	0.71		0.78	
Cu	40				0.84	0.86	0.88	0.86	0.87	
	5				0.83		0.86		0.82	
Cr	40				0.90		0.90		0.87	
	5				0.89		0.84		0.78	
Al	45		0.88		0.85	0.85				
C	45	0.92	0.90		0.88	0.84				
B	45	0.91	0.89		0.86	0.83				
Be	45	0.94	0.91	0.88	0.80	0.73				

### INTENSITY CALCULATIONS

The theoretical results reported here were obtained using the theory and notation of Metchnik and Tomlin (1963). According to this theory the number of  $K\alpha$  photons emitted per incident electron per  $4\pi$  steradian, in a direction at an angle  $\phi$

to the target face, when the beam is incident at an angle  $\theta$  to the normal, is given by the following integral, which must be evaluated numerically,

$$N_{\phi} = k \int_{T_0}^{T_k} NQ \frac{ds}{dT} \exp(-\mu\rho\langle x \rangle \operatorname{cosec} \phi \cos \theta) dT, \quad (6)$$

where

$$k = Rp \left( \frac{P+1}{P} \right) \omega_k,$$

$T$  is the kinetic energy of an electron,  $N$  is the number of atoms per unit volume of the target,  $Q$  is the cross section for  $K$  excitation,  $s$  is the distance in which an electron's initial energy  $T_0$  is reduced to  $T$ ,  $\mu$  is the mass absorption coefficient and  $\rho$  the density of the target material, and  $\langle x \rangle$  is the mean depth of electrons of energy  $T$ .

TABLE 2

VALUES OF  $k \times 10^3$  FOR  $K\alpha$  RADIATION FROM Ag, Cu, AND Cr WITH ELECTRONS INCIDENT NORMALLY, AND FOR TOTAL  $K$  RADIATION FROM Al, C, B, AND Be WITH ELECTRONS INCIDENT AT  $45^\circ$

$\phi$  is the angle of emission with respect to the target surface

Target	$\phi^\circ$	Voltage (kV)								
		5	10	15	20	30	40	50	60	70
Ag	40						710	760	800	800
Cu	40				270	270	280	270	280	
	5				270		270		260	
Cr	40				170		170		160	
	5				170		160		150	
Al	45		24		23	23				
C	45	1.32	1.30		1.27	1.21				
B	45	0.63	0.62		0.60	0.58	0.56			
Be	45	0.28	0.26	0.25	0.23					

In the factor  $k$ ,  $R$  is the rediffusion factor discussed above,  $p$  is the ratio of  $K\alpha$  to total  $K$  radiation and is tabulated by Williams (1933). When the total number of  $K$  photons is to be calculated,  $p$  is, of course, unity. According to Green and Cosslett (1961) the factor  $(P+1)/P$ , which allows for indirect excitation of  $K\alpha$  radiation by the continuous radiation, is not appreciably different from unity for  $Z < 30$ . This is the case for all the targets considered here except silver, for which Webster's (1928) experimental values have been used.

The other quantities involved in the calculation of  $N_{\phi}$  are discussed for each target material in turn and the resulting values for  $k$  are given in Table 2.

*Silver.*—Calculations of the intensity of  $K\alpha$  radiation were carried out for normal incidence of electrons. The experimental results of Webster, Hansen, and Duveneck (1933) for the cross section  $Q$  were used. The mass absorption coefficient  $\mu$  was taken from Allen's (1947) tables. The fluorescence yield  $\omega_k$  was taken as 0.80. The formula of Laberrigue-Frolow and Radvanyi (1956) gives 0.802, Burhop's (1952) formula gives 0.794 and Stephenson's (1937) experimental value is 0.81. Using  $R$  values from Table 1 the resulting values of  $k$  are very little different from those used by Metchnik

and Tomlin (1963), except at low voltages, and their intensity curves are not much affected by the use of the new  $k$  factor.

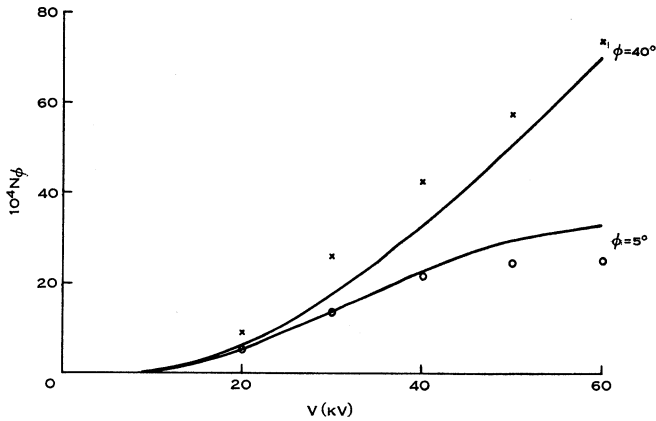


Fig. 2.—Theoretical curves and experimental points for copper  $K\alpha$  intensities. Electrons incident normally.

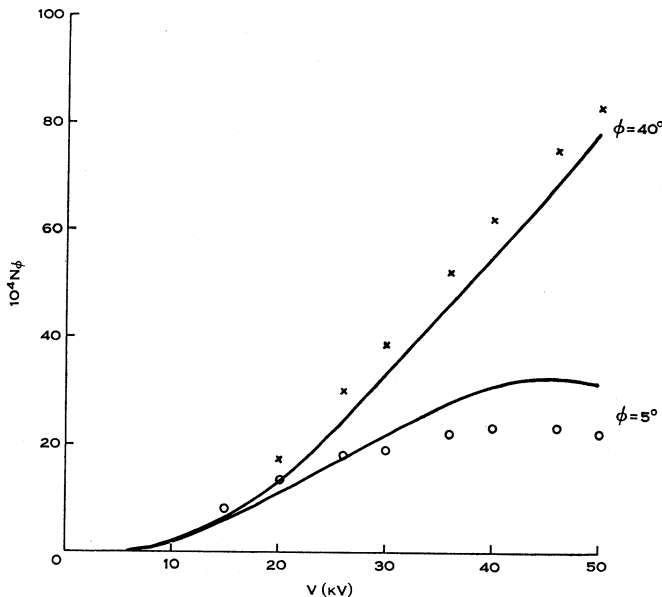


Fig. 3.—Theoretical curves and experimental points for chromium  $K\alpha$  intensities. Electrons incident normally.

*Copper.*—The incident electrons were assumed normal to the target.  $Q$  was obtained from the experimental curve for nickel (Pockman *et al.* 1947) and  $\mu$  was taken from Allen's tables. From Burhop's (1952) formula  $\omega_k = 0.36$ , and from that of Davidson and Wyckoff (1962)  $\omega_k = 0.365$ . Hence the values for  $k$  in Table 2 were derived. These are a little different from the values used by Metchnik and Tomlin (1963), and intensity curves for copper are redrawn and compared with experimental results in Figure 2.

*Chromium.*—The calculations were carried out for normally incident electrons.  $Q$  was obtained as for copper, and  $\mu$  from Allen's tables.  $\omega_k$  was taken to be 0.21, the value from Burhop's (1952) formula. Davidson and Wyckoff's expression gives  $\omega_k = 0.212$  and Laberrigue-Frolow and Radvanyi's (1956) gives  $\omega_k = 0.249$ . The former figures led to the values of  $k$  in Table 2 and to the intensity curves of Figure 3.

*Aluminium.*—For comparison with the experimental data of Campbell (1963), the total  $K$  emission from aluminium, carbon, boron, and beryllium has been calculated for electrons incident on the target at  $45^\circ$ , and for radiation emitted at  $45^\circ$  to the target surface. In view of the limitations in Metchnik and Tomlin's (1963) theory,

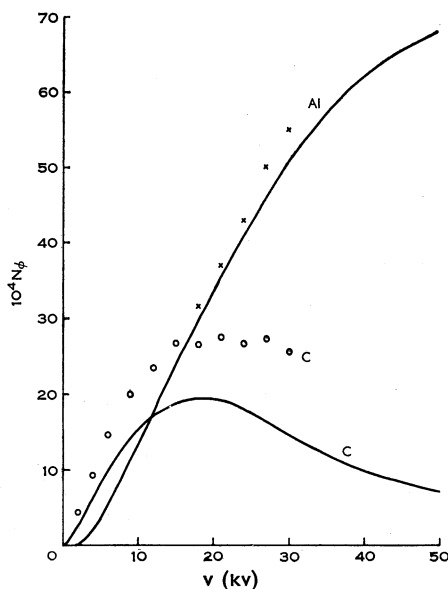


Fig. 4.—Theoretical curves and experimental points (Campbell 1963) for total  $K$  radiation intensities from aluminium and carbon targets. Electrons incident at  $45^\circ$ .

pointed out by them, its application to the case of an inclined target might be expected to be less reliable than for normal incidence. However, in the case of light elements there will be less scattering through large angles, and the validity of the theory should not be seriously impaired for such targets.

The mass absorption coefficient of aluminium for its own  $K$  radiation is given by Allen (1947) as 330, by Henke, White, and Lundberg (1957) as 322.6, and by recent measurements of Cooke *et al.* (1962) as 348.9 for the  $K\alpha$  radiation and 310 for  $K\beta$ . In the present calculations I have used  $\mu = 349$ , having verified that the use of two exponential terms in the integral (6), weighted in accordance with an estimated ratio of  $K\alpha$  to  $K\beta$  intensities, makes a negligible difference.

For aluminium and the lighter elements considered below,  $Q$  was obtained from the formula of Worthington and Tomlin (1956). Burhop's (1952) formula gives  $\omega_k = 0.0234$ , and that of Laberrigue-Frolow and Radvanyi (1956) gives  $\omega_k = 0.0267$ .

The experimental value of Davidson and Wyckoff (1962) is 0.027 and using this, together with  $R$  from Table 1, the results for  $k$  in Table 2 were obtained. Figure 4 shows a comparison of experimental and theoretical intensity curves. It may be remarked that Kulenkampff and Spyra (1954) found a back-reflection coefficient  $b = 0.26$  which is appreciably less than Campbell's (1963) value. This makes the values of  $R$  greater than those of Table 1 by about 5%.

*Carbon.*—There is a dearth of information about the X-ray properties of elements lighter than aluminium. There appears to be no accurate measurement of the mass absorption coefficient of carbon for its own  $K$  radiation. An entry in Allen's tables in the "Handbook of Chemistry and Physics" is not consistent with the tables as

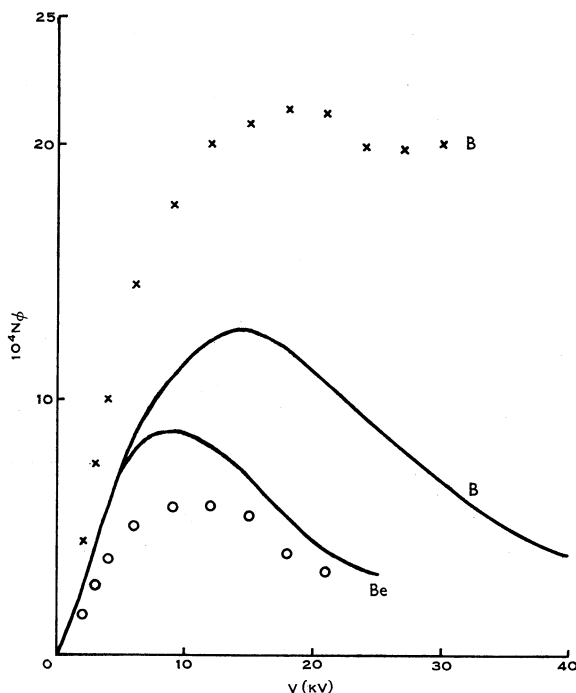


Fig. 5.—Theoretical curves and experimental points (Campbell 1963) for total  $K$  radiation intensities from boron and beryllium targets. Electrons incident at  $45^\circ$ .

given by Compton and Allison (1935). From a graphical extrapolation of the figures given by Henke, White, and Lundberg (1957) one finds  $\mu = 2200$ . Alternatively, an estimate may be made by extrapolation of a  $\log \mu$ – $\log Z$  plot (Fig. 6). From this graph  $\mu = 2150$  for carbon, and the following calculations were made with  $\mu = 2200$ .

There are no experimental measurements of  $\omega_k$  for light elements, and it is doubtful if any of the suggested formulae are very reliable (Campbell 1963). Burhop's (1952) formula gives  $\omega_k = 1.44 \times 10^{-3}$  for carbon and this was used to produce the values of  $k$  in Table 2. Intensity curves are given in Figure 4.

*Boron.*—The  $k$  values of Table 2 were obtained using  $\mu = 3400$ , from Figure 6, and  $\omega_k = 6.9 \times 10^{-4}$ , from Burhop's (1952) formula. Experimental and theoretical intensity curves are shown in Figure 5.



*Beryllium*.—According to Figure 6,  $\mu = 5900$  and from Burhop's formula  $\omega_k = 2.84 \times 10^{-4}$  from which the tabulated values of  $k$  were calculated. Using these figures the theoretical curve of Figure 5 was obtained.

### DISCUSSION

For aluminium and the heavier elements considered, the agreement between theory and experiment is reasonably good over the ranges covered by the experimental data. In these cases the absorption factors and values of  $k$  may be regarded as fairly reliable and the discrepancies are perhaps mainly due to the limitations of the theory. The calculated results for carbon and boron are considerably less than the experimental ones but there the values of  $\mu$  and  $\omega_k$  may be suspect. The positions of the maxima in the emission curves agree reasonably well with those observed and, since these depend mainly upon the absorption term in the integral for  $N_\phi$ , it appears that the values of  $\mu$  used were not grossly in error. However, the fluorescence yield

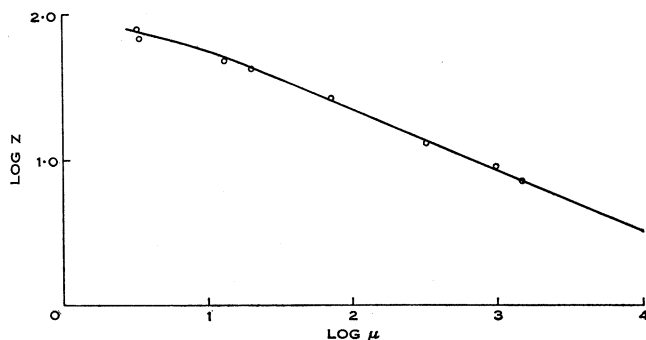


Fig. 6.—A log-log plot of the mass absorption coefficient for an element's own  $K$  radiation against atomic number.

for these very light elements is considerably in doubt. The better agreement of the curves for beryllium is somewhat surprising and suggests that either the variation of  $\omega_k$  with  $Z$  is complicated for the lightest elements, or that the experimental measurements, which are difficult for the soft beryllium radiation, may be in error. Evidently data for soft X-rays are very inadequate.

### ACKNOWLEDGMENT

The author is grateful to Miss Heather Barrow for carrying out a great deal of numerical work.

### REFERENCES

- ALLEN, S. J. M. (1947).—"Handbook of Chemistry and Physics." (Chemical Rubber Publishing Co.: Cleveland, Ohio.)
- ARCHARD, G. D. (1960).—Proc. 2nd Int. Symp. on X-ray microscopy and X-ray microanalysis. (Elsevier: Amsterdam.)
- ARCHARD, G. D. (1961).—*J. Appl. Phys.* **32**: 1505.
- BURHOP, E. H. S. (1952).—"The Auger Effect." (C.U.P.)
- CAMPBELL, A. J. (1963).—*Proc. Roy. Soc. A* **274**: 319.

- COMPTON, A. H., and ALLISON, S. K. (1935).—"X-Rays in Theory and Experiment." (Van Nostrand: New York.)
- COOKE, B. A., POUNDS, K. A., RUSSELL, P. C., and STEWARDSON, E. A. (1962).—*Proc. Phys. Soc. Lond.* **79**: 883.
- DAVIDSON, F. D., and WYCKOFF, W. G. (1962).—*J. Appl. Phys.* **33**: 3528.
- GREEN, M., and COSSLETT, V. E. (1961).—*Proc. Phys. Soc. Lond.* **78**: 1206.
- HENKE, B. L., WHITE, R., and LUNDBERG, B. (1957).—*J. Appl. Phys.* **28**: 98.
- KULENKAMPPF, H., and SPYRA, W. (1954).—*Z. Phys.* **137**: 416.
- LABERRIGUE-FROLOW, J., and RADVANYI, P. (1956).—*J. Phys. Rad.* **17**: 1944.
- LEWIS, H. W. (1950).—*Phys. Rev.* **78**: 526.
- METCHNIK, V., and TOMLIN, S. G. (1963).—*Proc. Phys. Soc. Lond.* **81**: 956.
- POCKMAN, L. T., WEBSTER, D. L., KIRKPATRICK, P., and HARWORTH, K. (1947).—*Phys. Rev.* **71**: 330.
- STEPHENSON, R. J. (1937).—*Phys. Rev.* **51**: 637.
- STERNGLASS, E. J. (1954).—*Phys. Rev.* **95**: 345.
- TOMLIN, S. G. (1963).—*Proc. Phys. Soc. Lond.* **82**: 465.
- WEBSTER, D. L. (1928).—*Proc. Nat. Acad. Sci.* **14**: 337.
- WEBSTER, D. L., CLARK, H., and HANSEN, W. W. (1931).—*Phys. Rev.* **37**: 115.
- WEBSTER, D. L., HANSEN, W. W., and DUVECK, F. B. (1933).—*Phys. Rev.* **43**: 839.
- WILLIAMS, J. H. (1933).—*Phys. Rev.* **44**: 146.
- WORTHINGTON, C. R., and TOMLIN, S. G. (1956).—*Proc. Phys. Soc. Lond.* **69**: 401.