Indirectly and Directly Produced X-Ray Line Radiation*

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A direct comparison has been made of the number of x-ray quanta in the K line spectrum to the number of quanta in the continuous spectrum having energies greater than the critical value for K excitation. From this, one can deduce that, for a copper target operating at low voltages, only about 10% of the line radiation is indirectly produced. A further result is that the ratio of the cross section for direct ionization to the cross section for bremsstrahlung production is about 4/1, a value which is at least an order of magnitude less than theory predicts.

INTRODUCTION

WHEN the existence of line x radiation was established by Kaye,[1] Barkla[2] suggested that the effect was due to the absorption of bremsstrahlung and the subsequent emission of characteristic radiation. Beatty,[3] however, argued that the line emission was virtually all directly produced, and offered experimental evidence based on studies of the intensities from bare and foil-covered targets. Balderston[4] interpreted the data that he obtained from bare-target measurements to show the contrary result, in agreement with Barkla's concept.

Webster and co-workers[5] re-examined the problem by a foil and bare-target method. Their results indicated that for molybdenum, the line radiation observed normally from a target is 35% indirectly produced and 65% directly produced. These figures were constant over a large voltage range. The results of Webster have been generally accepted as being correct.

It should be borne in mind that while the direct ionization represents a primary process, this does not indicate that it must necessarily predominate over the two-stage, indirect ionization process. As will be established, about 60% of the bremsstrahlung having frequencies greater than the critical value will be reabsorbed in the target and the indirect line radiation results from this process. We shall refer to the bremsstrahlung radiation which is capable of producing K excitation as the "effective continuum."

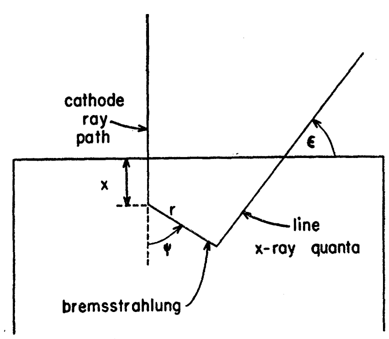

FIG. 1. Geometry for the production of indirect line radiation.

The results of Webster's measurements are at variance with extant theory on the interaction of electrons with matter, although this point has apparently not been stressed in the literature. Webster's data yields the ratio of the probability that a bombarding electron will eject a K electron to the probability that it will radiate a quantum of frequency greater than the critical K absorption frequency; this ratio, which is designated as Q, was measured by Webster *et al.* and found to be 0.85. Theory indicates that in this energy region the value of Q should be several orders of magnitude greater than that reported.

In this paper, a direct comparison will be made of the number of quanta in the K line with the number of quanta in the effective continuum. Under the conditions of the experiment, the results indicate that the direct ionization process is much more probable than the indirect; nevertheless, the indirect process does occur with sufficient likelihood that Q must be considerably smaller than theory indicates.

THEORETICAL

A. Theory of the Experiment

Consider the geometry of Fig. 1. The number of electrons in the beam is assumed to be attenuated according to an exponential law. The energy per electron is considered to be unchanged with depth of penetration, which is essentially correct for the conditions under which the experiment is carried out. The electron is stopped at a depth x below the surface, producing a bremsstrahlung quantum. This quantum is reabsorbed after penetrating a distance r, and K excitation occurs. For the following discussion, the bremsstrahlung is assumed to be sent out isotropically; the obvious error involved in this assumption is of little consequence. Actually most of the cathode electrons are probably proceeding approximately in their original direction, but if one assumes that the bremsstrahlung is all sent out in a horizontal plane, the numerical results do not differ excessively from those obtained under the assumption of isotropic radiation. Webster made the isotropic assumption in obtaining his results. As dis-

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[1] G. W. C. Kaye, Phil. Trans. Roy. Soc. London **A209**, 123 (1909).
[2] C. G. Barkla and C. A. Sadler, Phil. Mag. **17**, 739 (1909).
[3] R. T. Beatty, Proc. Roy. Soc. (London) **87**, 511 (1912).
[4] M. Balderston, Phys. Rev. **27**, 696 (1927).
[5] D. L. Webster, Proc. Natl. Acad. Sci. U. S. **13**, 445 (1927); **14**, 331 (1928). W. W. Hansen and K. P. Stoddard, Phys. Rev. **43**, 701 (1933).

cussed in the preceding paper, the exponential law for the stopping of the electrons conforms adequately to the physical evidence. However, the results obtained from analysis of the data are not particularly sensitive to the form of the actual stopping law employed, and the primary reason for using this particular type of relation is that it yields results in closed form.

Although the integrations are performed over the limits zero to infinity, the region involved is very small ($x<r<10^{-4}$ cm), and for a remote detector it is permissible to regard all effects as occurring at a point.

Let N_D be the total number of K-line radiation quanta produced directly that are sent out per unit solid angle per unit time under a constant cathode-ray flux. Then, using b as the absorption coefficient for electrons,

$$\int_{x=0}^{x=\infty} dN_D=\int_0^\infty N_D b e^{-bx}dx=N_D. \qquad (1)$$

Note that N_D is not the number of K excitations produced directly. That number would be N_D/ω, where ω is the fluorescent efficiency.

The number of directly produced line quanta which are observed will be

$$N_{0D}=\int_0^\infty N_D b e^{-bx}\exp(-\mu_\alpha x\csc\epsilon)dx = bN_D/(b+\mu_\alpha\csc\epsilon)=gN_D, \qquad (2)$$

where μ_α is actually the effective absorption coefficient for all K-line radiation.

Similarly, let N_c be the total number of quanta in the effective continuum that are sent out per unit solid angle per unit time. Then

$$\int_{\nu_K}^{\nu_0}\int_{x=0}^{x=\infty} dN_c=\int_{\nu_K}^{\nu_0}\int_0^\infty N_c b e^{-bx}dxd\nu=N_c. \qquad (3)$$

The number of these quanta which will emerge making an angle ϵ with the target surface will be

$$\begin{aligned} N_{0c}&=\int_{\nu_K}^{\nu_0}\int_0^\infty N_c b e^{-bx}\exp(-\bar{\mu}x\csc\epsilon)dxd\nu \\ &=fN_c=\frac{b}{b+\bar{\mu}\csc\epsilon}N_c. \end{aligned} \qquad (4)$$

Here $\bar{\mu}$ is the average value of the absorption coefficient for radiation in the effective continuum, and the summing of the number of quanta in the effective continuum is represented by the integration between ν_0 and ν_K. These frequencies represent the Duane-Hunt short-wavelength limiting frequency and the critical frequency for K excitation, respectively. The number of indirectly produced line quanta will be given by

$$\begin{aligned} N_I=&\int_{\nu_K}^{\nu_0}\int^{\Delta\nu}\int_0^\infty \tfrac{1}{2}u\omega N_c b e^{-bx}dx\int_0^{x\sec\Psi}\bar{\mu}\exp(-\bar{\mu}r)dr\int_0^{\pi/2}\sin\Psi d\Psi d\nu d\nu' \\ &+\int_{\nu_K}^{\nu_0}\int^{\Delta\nu}\int_0^\infty \tfrac{1}{2}u\omega N_c b e^{-bx}dx\int_0^\infty\bar{\mu}\exp(-\bar{\mu}r)dr\int_0^{\pi/2}\sin\Psi d\Psi d\nu d\nu'. \end{aligned} \qquad (5)$$

The integration over $\Delta\nu$ refers to the process of counting the number of quanta in the lines, and u is the percentage of absorption due to K ionization. Upon integration, one obtains

$$N_I=u\omega lN_c=u\omega[\tfrac{1}{2}+\tfrac{1}{2}(\bar{\mu}/b)\ln(1+b/\bar{\mu})]N_c. \qquad (6)$$

The number of these indirectly produced line quanta which will be emergent at the angle ϵ will be

$$\begin{aligned} N_{0I}=&\int_{\nu_K}^{\nu_0}\int^{\Delta\nu}\int_0^\infty \tfrac{1}{2}u\omega N_c b e^{-bx}dx\int_0^{x\sec\Psi}\bar{\mu}\exp(-\bar{\mu}r)dr\int_0^{\pi/2}\exp[-\mu_\alpha(x-r\cos\Psi)\csc\epsilon]\sin\Psi d\Psi d\nu d\nu' \\ &+\int_{\nu_K}^{\nu_0}\int^{\Delta\nu}\int_0^\infty \tfrac{1}{2}u\omega N_c b e^{-bx}dx\int_0^\infty\bar{\mu}\exp(-\bar{\mu}r)dr\int_0^{\pi/2}\exp[-\mu_\alpha(x+r\cos\Psi)\csc\epsilon]\sin\Psi d\Psi d\nu d\nu' \\ &=u\omega hN_c=\frac{u\omega}{2}\frac{N_c\bar{\mu}}{b+\mu_\alpha\csc\epsilon}\left[\ln\left(1+\frac{b}{\bar{\mu}}\right)+\frac{b}{\mu_\alpha\csc\epsilon}\ln\left[1+\frac{\mu_\alpha\csc\epsilon}{\bar{\mu}}\right]\right]. \end{aligned} \qquad (7)$$

To obtain physically important quantities from the x-ray data, relations must be developed which relate measured intensities to P and Q, where P is defined as

$$P=\frac{\text{directly produced line radiation}}{\text{indirectly produced line radiation}}=\frac{N_D}{N_I}, \qquad (8)$$

and Q, as indicated previously, is given by

$$\begin{aligned} Q&=\frac{\text{cross section for direct } K \text{ excitation}}{\text{cross section for bremsstrahlung production}} \\ &=\frac{\text{probability of direct ionization}}{\text{probability of bremsstrahlung production}} \\ &=\frac{(N_D/\omega)}{N_c}. \end{aligned} \qquad (9)$$

Let us define R as the measured ratio of the integrated number of quanta observed in the lines to the integrated number of quanta observed in the effective continuum. In this treatment we shall consider our detectors to be 100% efficient; in the treatment of the raw data it will be necessary to correct for this effect. By definition,

$$R=(N_{0D}+N_{0I})/N_{0c}. \tag{10}$$

Substituting our derived expressions, we obtain

$$R=(gN_D+u\omega hN_c)/fN_c. \tag{11}$$

From our previous definition of P and Q, the equations may be readily manipulated into the following forms:

$$P=(fR/glu\omega)-(h/gl), \tag{12}$$

and

$$Q=(fR/g\omega)-(hu/g). \tag{13}$$

B. Theoretical Value of Q

The so-called stopping power, dE/dx, of a medium being traversed by a beam of electrons may be expressed in the following fashion:

$$-dE/dx=nE\phi, \tag{14}$$

where n is the number of atoms/cm^3 and ϕ is the cross section per atom. For loss of energy by radiation, it was shown by Racah[6] that

$$\phi_{\mathrm{rad}}=16/3(e^4/m^2c^4)(Z^2/137), \tag{15}$$

where the symbols have their conventional meanings. This equation is applicable for all energies less than approximately 0.5 Mev.[7] An expression similar to (14) may be written for the stopping of the cathode electrons by inelastic collisions with K electrons. We have available the Bohr expression[8]

$$-\frac{dE}{dx}=\frac{4\pi e^4 n}{mv^2}Z\ln\left[\frac{mv^2}{2I}\left(\frac{e}{2}\right)^{\frac{1}{2}}\right], \tag{16}$$

where I is the ionization potential. This yields

$$\phi_{\mathrm{direct}}=(4\pi e^4/E_0^2)\ln(1.16E_0/E_K). \tag{17}$$

The ratio Q will be given by

$$Q=\tfrac{3}{4}\pi[(mc^2)^2/E_0^2](137/Z^2)\ln(1.16E_0/E_K). \tag{18}$$

For the conditions of this experiment, the quantity Q equals approximately 300. If the energy of the incoming cathode electron is averaged over the range E_K to E_0, the numerical figure is approximately 155.

Another expression for the collision cross section commonly used in x-ray studies is given by the Rosseland ionization function,[9]

$$\frac{di}{dx}=\frac{2\pi e^4 n}{E_0}\left(\frac{1}{E_K}-\frac{1}{E_0}\right). \tag{19}$$

The indicated cross section becomes

$$(\phi')_{\mathrm{direct}}=\frac{2\pi e^4}{E_0}\left(\frac{1}{E_K}-\frac{1}{E_0}\right). \tag{20}$$

Here the value of Q is

$$\frac{3\pi}{8}\frac{(mc^2)^2}{E_0^2}\frac{137}{Z^2}\frac{(E_0-E_K)}{E_K}. \tag{21}$$

The numerical value is somewhat smaller, being approximately 110, and the result if averaged over the range of energies will be about 65.

The Q values which would be predicted for the conditions of Webster's experiments are of this order of magnitude and would not be constant, in contrast to Webster's fixed value of approximately 1.

EXPERIMENTAL

The general techniques employed were outlined in the preceding paper. All measurements were taken with the Economy two-crystal spectrometer and recorded with a ratemeter and chart recorder. The resolution was that characteristic of two-crystal spectrometers, the crystals themselves having $(1, -1)$ widths of about 1 v at the Cu $K\alpha$ line. The effect of gear noise was not observable.

The target take-off angles employed were 20° and 7.5° in the continuously pumped tube and 4° in the commercial Philips diffraction tube. The results in the latter case are always open to query about the extent and effect of target pitting. Furthermore, the value of $\csc\epsilon$ is sensitive in this region to small errors in measuring ϵ; it is probably impossible to definitely establish the angle to an accuracy of better than 0.5°.

The fundamental principle of this experiment involves the comparison of the number of quanta radiated in the K emission lines with the number of quanta in the effective continuum. The areas in the lines were obtained by running the recorders at much higher speeds and smaller time constants than in the measurement of the continuum. The quantities which were directly compared were the areas in the effective continuum and the area in the $K\beta$ line. The ratio of the total emission in the $K\alpha_1$-$K\alpha_2$ complex to the emission in the $K\beta$ lines was obtained by repeated measurements. These ratios are listed in Table I. The ratios at the

[6] G. Racah, Nuovo cimento **11**, 461 (1934).

[7] W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, New York, 1950), 2nd ed., p. 173.

[8] H. A. Bethe and J. Ashkin, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), p. 253.

[9] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1951), 2nd ed., p. 71.

TABLE I. The values of R, P, Q, and ratio of number of quanta in $K\beta$ and $K\alpha$ complex. Both line intensity ratios are corrected for counter sensitivity and air absorption, but the third column is also corrected for target absorption.

Target angle ϵ	$K\beta/(K\alpha+K\beta)$	$K\beta/(K\alpha+K\beta)$ corrected	R	P	Q
4°	0.123	0.118	3.04	8.9	4.6
7.5°			2.61	9.0	4.6
20°	0.106	0.105	2.20	8.8	4.5

angles measured are not in complete agreement (see column 2); the effect of angle is to tend to produce a discrepancy in the direction observed but it is not of sufficient magnitude to account for it entirely. Surface pitting and contamination may provide the explanation, or the difficulty may merely be due to insufficient statistical accuracy. The ratio measured at 20° was used for the 7.5° target because pitting could scarcely have been involved in these two arrangements. The tabulated results are in good agreement with those of Williams[10] who obtained 0.107. The raw data obtained must be corrected for the effect of counter efficiency, absorption in the air, and also for target self absorption and crystal reflectivity where necessary.

All measurements were made at an x-ray tube voltage of 11.8 kv. The target material was copper. The value of the various absorption coefficients are listed with Table II. The value of $\bar{\mu}$ listed was taken at the wavelength for which one observes half of the effective continuum at longer wavelengths and the other half at shorter wavelengths. A more detailed averaging process was utilized for getting $\bar{\mu}$ for the 4° target, but its use would have produced no significant effect on the results.

TABLE II. Absorption coefficients at angles employed.

Angle ϵ	$\mu_2 \csc\epsilon$	$\mu_1 \csc\epsilon$	$\bar{\mu} \csc\epsilon$	b
4°	39 200	4740	34 500	38 500
7.5°	21 000	2540	18 600	38 500
20°	7960	964	7050	38 500

A value of b was obtained in the preceding paper for the 4° target which in principle should be applicable to the 7.5° and 20° targets. Unfortunately, a rather serious discrepancy exists which can only be resolved by assuming anode defects. An r_E of 1.22 was obtained experimentally for the 20° target and a value of 1.45 for the 7.5° target. The latter of these two is considered to be the more reliable measurement, but it is the most difficult to reconcile with the value of 2.10 found at 4°. A fairly satisfactory compromise seems to be to assume a value of $b=38\,500$/cm, which will produce r_E values of 1.15 (which is within the experimental error), 1.45, and 1.80 (for which one must assume an effect of pitting).

[10] J. H. Williams, Phys. Rev. 44, 146 (1933).

DISCUSSION

The values of f, g, h, and l are given in Table III. By using the theory previously developed, one obtains the values of P and Q listed in Table I. The common value of b is used throughout. If the 4° target data were corrected for the assumed pitting, the R value and consequently the P and Q values would be lower than those given. Table III also gives P' as a function of angle where we define P' as the ratio of the observed direct line intensity to the observed indirect line intensity. From the defining equations, it can easily be shown that

$$P'=(gl/h)P. \tag{22}$$

We see that for small angles ϵ the indirect radiation becomes relatively small.

The values of P and Q obtained at each of the target angles are found in Table I. The agreement is probably better than is warranted considering the uncertainties about the surface effects. However, one can say that compared to Webster's data the results of this experiment indicate a larger ratio of the directly produced line radiation to that produced indirectly. The conditions of this experiment are somewhat different from the Webster experiments, but it seems unlikely that the difference in results can be accounted for in this fashion.

On the other hand, these results corroborate the generally low value of Q, the ratio of the probability of direct ionization to bremsstrahlung emission, despite the fact that the actual numbers are considerably different. As we pointed out earlier, this result is not in agreement with the theoretical relation that indicates a value of Q of the order of 100.

It should be emphasized that the results are little dependent on the proper law for stopping the electrons; nor is the result sensitive to the choice of the absorption coefficients b and μ. It does not depend strongly on the distribution of bremsstrahlung for the accelerated electron. It is only important to recognize that the electrons are stopped close to the surface. Furthermore, the bremsstrahlung rays in the effective continuum are soon attenuated, while the line radiation is comparatively little effected. Thus all effects come from a small region of space and the following order of magnitude argument

TABLE III. Geometric factors relating directly and indirectly produced line intensities.

ϵ (degrees)	f	g	h	l	P'/P
4	0.537	0.858	0.282	0.585	1.77
7.5	0.683	0.918	0.356	0.585	1.51
10	0.742	0.938	0.399	0.585	1.37
20	0.850	0.968	0.469	0.585	1.21
30	0.893	0.979	0.500	0.585	1.15
40	0.914	0.982	0.517	0.585	1.11
50	0.927	0.985	0.527	0.585	1.095
60	0.935	0.987	0.531	0.585	1.09
90	0.946	0.990	0.538	0.585	1.08

is valid. One observes experimentally that the intensity in the lines is two or three times that in the effective continuum. Whatever the distribution in space of the bremsstrahlung, the percentage of the effective continuum which produces K excitation can only be expected to be between 45 and 90%. Most probably it will be about 50%. Further, only two-fifths of the indirectly excited atoms produce line radiation, and one sees that the value of P must lie close to the value of 8.9 we obtained. Since the direct and indirect radiation were subject to the same Auger probabilities, P and Q will only differ due to purely geometric factors. For example, assuming the value of 50% just given for the efficiency of conversion of bremsstrahlung to K excitation, the value of Q will be less than P by this factor.

The one experimental fact that leads to this unexpected result is the ratio of the number of quanta in the line to the number in the effective continuum, and there is little likelihood that the values given for R are grossly in error. The data were taken under conditions where experimental errors could be minimized. The voltage value of 11.8 kv represents a practical optimum. For lower voltage the intensity in the continuum becomes small and there is loss of statistical accuracy; for higher voltage, the continuum cutoff is difficult to establish because of shielding problems, and large corrections must be made for counter sensitivity.

To illustrate the lack of sensitivity of the results to anything but the measured value of R, calculations were made under the assumption that all x radiation was produced at the particular depth X_c calculated in the previous paper from the jump ratio. The integrations must be carried out numerically and they were only done on the 7.5° target. We obtained $P=9.1$ and $Q=4.65$. Values of $b=60\,000$ cm^{-1} and $20\,000$ cm^{-1} were also used in the expressions derived previously. We consider these values to be beyond probability. For $b=60\,000$, we obtained $P=11.0$ and $Q=5.1$, while for $b=20\,000$ we found $P=6.9$ and $Q=3.8$. Calculations for various distributions of the bremsstrahlung leave the P and Q value in the same general region.

Because of the simplicity and directness of the experiment it is difficult to escape the conclusion that the ratio of theoretical cross sections for direct K excitation and bremsstrahlung production is in error by at least an order of magnitude. There are obvious weaknesses in the theoretical considerations which underlie the derivations for both cross sections, and it is difficult to ascertain at present what is the principal cause of the discrepancy.