

## A GENERAL QUANTUM THEORY OF THE WAVE-LENGTH OF SCATTERED X-RAYS

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### ABSTRACT

**Corpuscular quantum theory of the scattering of x-rays.**—The theory previously presented<sup>1</sup> gave for the change of wave-length due to scattering, assuming each quantum scattered by a single free electron,  $\delta\lambda_F = (h/mc)$  vers  $\varphi = .0242$  vers  $\varphi$ , where  $\varphi$  is the angle between the primary and scattered ray. This theory is now *extended to scattering by bound electrons*. If the scattering electron is not ejected from the atom, no energy is transferred and no change of wave-length occurs; but if the electron is removed, the change of wave-length must lie between  $\delta\lambda_B = \lambda^2(\lambda_s - \lambda)$  and  $\infty$ , where  $\lambda$  is the incident wave-length and  $\lambda_s$  the critical ionization wave-length for the scattering electron in its original orbit. If we restrict the theory by assuming that the final momentum possessed by the residual atom is that acquired during the absorption from the incident beam, of the energy  $hc/\lambda_s$  required to remove the electron from the atom, and that the electron, now free, receives the impulse resulting from the deflection of the quantum, the resulting change of wave-length is  $\delta\lambda = \delta\lambda_B + \delta\lambda_F$ . *Comparison with experimental results* shows that the restricted theory accounts satisfactorily for scattering by the lighter elements and also for the scattering of tungsten rays by Mo if for heavy elements  $\delta\lambda_F$  is taken to be zero. *Criticism of the "tertiary radiation" hypothesis*, which leads to the expression  $\delta\lambda = \lambda^2(\lambda_s - \lambda)$ , shows that it does not account for the large percentage of polarization, for the large relative intensity and for the homogeneity of the scattered x-rays.

**I**N recent papers the writer<sup>1</sup> and independently P. Debye<sup>2</sup> have developed a quantum theory of the scattering of x-rays by light elements. This theory is based on the idea that an x-ray quantum proceeds in a definite direction, and is scattered by a single electron. The quantum loses energy due to the recoil of the electron, and is thus reduced in frequency. Measurement of the wave-length of scattered x-rays by the writer,<sup>3</sup> Ross<sup>4</sup> and Davis<sup>5</sup> have shown that at least in many cases scattered x-rays occur whose wave-length is changed by the theoretical amount. Moreover, electrons moving in the direction of the primary beam with about the velocity predicted for the recoiling electrons have been found

<sup>1</sup> A. H. Compton, Bulletin Nat. Res. Council, No. 20, p. 19 (1922); Phys. Rev. **21**, 207 and 483 (1923)

<sup>2</sup> P. Debye, Phys. Zeits., Apr. 15, 1923

<sup>3</sup> A. H. Compton, Bulletin N. R. C., No. 20, p. 16; Phys. Rev. **22**, 409 (1923)

<sup>4</sup> P. A. Ross, Proc. Nat. Acad. Sci., July, 1923; Phys. Rev. **22**, 524 (1923)

<sup>5</sup> Bergen Davis, Paper before section B of A. A. A. S., at Cincinnati, December, 1923

by C. T. R. Wilson<sup>6</sup> and by Bothe.<sup>7</sup> There is thus strong evidence that in some cases x-rays are scattered in essentially the manner described by the quantum theory.

Very recently, however, Clark and Duane<sup>8</sup> and Clark, Duane and Stifler<sup>9</sup> have shown the existence of a type of secondary radiation whose wave-length is altered more than this quantum theory demands. The lower frequency limit of this modified radiation is in many cases approximately  $\nu - \nu_s$ , where  $\nu$  is the frequency of the primary ray and  $\nu_s$  is a critical ionization frequency of the radiator. Moreover, when a series of radiators of increasing atomic numbers is used, there is a gradual increase in the displacement of the modified line from about the value given by the quantum formula when radiators of low atomic number are used, to a much larger value for radiators of higher atomic number. This result suggests that the writer's quantum formula for the change of wave-length holds only in the case of scattering electrons which are effectively free,<sup>10</sup> and it becomes important to extend the theory to the scattering of x-ray quanta by electrons that are bound within the atom.

In extending the theory it seems desirable to present it first in a very general form; we shall then impose such restrictions as seem to be warranted by the experiments, and shall compare the resulting formulas with the data given by Duane and his collaborators. Finally, Clark and Duane's interpretation of these modified lines as due to "tertiary radiation" excited by the photo-electrons will be briefly discussed.

#### GENERAL THEORY OF SCATTERING BY INDIVIDUAL ELECTRONS

If we retain the conception used in the original theory, that each x-ray quantum is scattered by an individual electron, two cases are to be considered, that in which the electron is not ejected from its atom, and that in which the scattering electron receives an impulse sufficient to eject it from the atom.

In the first case, evidence from x-ray spectra indicates that there is no resting place for the electron within the atom after it has scattered the quantum unless it returns to its original orbit. The final energy of the atomic system is thus the same after the quantum is scattered as it was before (the kinetic energy imparted by the deflected quantum to a body

<sup>6</sup> C. T. R. Wilson, Proc. Roy. Soc. A **104**, 1 (1923)

<sup>7</sup> W. Bothe, Zeits. f. Phys. **16**, 319; **20**, 237 (1923)

<sup>8</sup> G. L. Clark and W. Duane, Proc. Nat. Acad. Sci. **9**, 413, 419 (1923); **10**, 41 (1924)

<sup>9</sup> G. L. Clark, Stifler and W. Duane, paper presented to Am. Phys. Soc. Feb. 23, 1924, privately communicated to the writer (see abstract in Phys. Rev. **23**, 551, 1924)

<sup>10</sup> This was indeed emphasized in the original paper.

as massive as an atom being negligible), implying that the frequency of the scattered ray is unaltered. Scattering by this process would thus give rise to an unmodified line.

In the second case, however, part of the energy of the incident quantum is spent in removing the scattering electron from the atom, part is used in giving the electron and the ionized atom their final motions, and the remainder appears as the scattered ray. If we suppose that the primary beam is propagated along  $OX$ , and if the direction of the scattered ray defines the  $XOY$  plane, the following equations suffice to define the condition of the quantum, the electron and the ionized atom before and after the scattering occurs.

Energy equation,

$$\frac{hc}{\lambda} = \frac{hc}{\lambda'} + \frac{hc}{\lambda_s} + mc^2 \left( \frac{1}{\sqrt{1-\beta^2}} - 1 \right) + \frac{1}{2}MV^2; \quad (1)$$

momentum equations,

$$\frac{h}{\lambda} = \frac{hl_1}{\lambda'} + pl_2 + Pl_3, \quad (2)$$

$$0 = \frac{h}{\lambda'} m_1 + pm_2 + Pm_3, \quad (3)$$

$$0 = 0 + pn_2 + Pn_3; \quad (4)$$

supplementary equations,

$$p = m\beta c / \sqrt{1-\beta^2}, \quad (5)$$

$$l_2^2 + m_2^2 + n_2^2 = 1. \quad (6)$$

In these equations,

$\lambda$  = wave-length of incident x-ray quantum;

$\lambda'$  = wave-length of scattered quantum;

$\lambda_s$  = critical ionizing wave-length for scattering electron;

$h$  = Planck's constant;

$c$  = velocity of light;

$m$  = rest mass of electron;

$\beta c$  = final velocity of recoiling electron;

$p$  = final momentum of electron;

$M$  and  $V$  = mass and final velocity of recoiling atom;

$P = MV$ ;

$l_1, m_1, 0$  = direction cosines of scattered quantum;

$l_2, m_2, n_2$  = direction cosines of  $p$ ;

$l_3, m_3, n_3$  = direction cosines of  $P$ .

We shall use also the following abbreviations:  $\nu = c/\lambda$  = frequency of incident radiation;  $\varphi$  = angle between incident and scattered quanta;

$\theta$  = angle between incident quantum and  $p$ ;  $\gamma = h/mc = 0.0242 \text{ \AA}$ ;  
 $a = h/mc\lambda$ ;  $s = \lambda/\lambda_s$ ;  $b = p/mc$ ;  $B = P/mc$ ;  $D = \lambda^2/(\lambda_s - \lambda)$ ;  
 $F = \gamma(1 - l_1) = \gamma \text{ vers } \phi$ ;  $A = s(1 + a - \frac{1}{2}as) - B(l_3 - B/2a)$ .

A straightforward solution of these equations, noting that  $\frac{1}{2}MV^2$  is always negligible compared with the other terms of Eq. (1), gives for the change in wave-length,

$$\delta\lambda \equiv \lambda' - \lambda = [\lambda/(1 - A)] [\alpha(1 - l_1) + s(1 - \frac{1}{2}as) + B(l_1l_3 + m_1m_3 - l_3 + B/2a)]. \quad (7)$$

Instead of solving directly for  $\beta$  it is more convenient to calculate the kinetic energy of the recoiling electron, which is given by these equations as

$$E = h\nu \left\{ 1 - \frac{1 - a(l_1s + \frac{1}{2}s^2) + B(l_3 + l_1l_3s + m_1m_3s - B/2a)}{1 + a(1 - l_1 - s) + B(l_1l_3 + m_1m_3)} \right\}. \quad (8)$$

If the scattering electrons are free, the critical ionization wave-length is  $\lambda_s = \infty$ , and the momentum imparted to the atom is  $P = 0$ , in which case these equations reduce to

$$\delta\lambda = F \equiv a\lambda(1 - l_1) = \gamma \text{ vers } \phi, \quad (9)$$

and

$$E_0 = h\nu \left\{ 1 - 1/(1 + a - al_1) \right\} = h\nu \frac{a \text{ vers } \phi}{1 + a \text{ vers } \phi}. \quad (10)$$

These results are identical with those given by Debye and the writer for the scattering by a free electron.

For a definite value of  $s$  the change in wave-length according to Eq. (7) is a minimum when all the impulse from the deflected quantum is absorbed by the atom. In this case

$$\delta\lambda = D \equiv \lambda^2/(\lambda - \lambda_s), \quad (11)$$

which is identical with the minimum wave-length change given by the tertiary radiation theory of Clark and Duane. Eq. (11) may be obtained by substituting the appropriate values of  $B$ ,  $l_3$  and  $m_3$  in Eq. (7), but can be got more easily directly from Eq. (1), noting that the kinetic energy of the scattering electron is zero if the impulse all goes to the ionized atom.

In the general case there is nothing to prevent all of the energy being taken up by the ejected electron, leaving in Eq. (1)  $hc/\lambda' = 0$ , or  $\lambda' = \infty$ . The complete quantum theory of scattering thus gives a possible wave-length range for the scattered ray between  $\lambda + \lambda^2/(\lambda_s - \lambda)$  and  $\infty$ . We thus have precisely the same wave-length range for the secondary radiation as is predicted by the tertiary radiation theory of Clark and Duane, which also assigns a definite lower limit to the wave-length, but supplies no finite upper limit.

## RESTRICTIONS SUGGESTED BY EXPERIMENT

In view of the sharpness of the lines or bands observed in the spectrum of the secondary x-rays, it is clear that the momentum which the atom acquires is defined within rather narrow limits, that is  $B$  in Eq. (7) has a rather definite value. An exact prediction of the value of this momentum requires some knowledge of the atom's internal dynamics and of the mechanism of interaction between the quantum and the electron. Thus a sudden impulse applied to the electron would result in a smaller momentum imparted to the atom than would an interaction lasting for a considerable time interval, just as jerking a sheet of paper from under a book disturbs the book less than removing the paper more slowly. Lacking a sufficient knowledge of this mechanism to make a definite prediction, it is nevertheless of interest to study the result of certain plausible assumptions regarding the impulse imparted to the atom.

Let us suppose for example that in removing the electron from the atom all of the work is done by the incident ray. The energy absorbed in this process is  $hc/\lambda_s$ , and since the absorption of this energy leaves the electron at rest outside the atom, the whole impulse,  $h/\lambda_s$ , accompanying the absorption of the energy, must be imparted to the remainder of the atom. Since the electron is now free, any further action of the radiation on the electron will not affect the atom. At the end of the process, the atom therefore retains the momentum  $P = h/\lambda_s$  in the direction of the primary beam.

In Eq. (7) we have, therefore,  $B = P/mc = h/mc\lambda_s = \alpha s$ ,  $l_3 = 1$ , and  $m_3 = 0$ . On substituting these values, Eq. (7) reduces to

$$\begin{aligned} \delta\lambda &= [\lambda/(1-s)] [\alpha(1-s)(1-l_1) + s] \\ &= \lambda^2(\lambda_s - \lambda) + \gamma(1-l_1) = D + F \\ &= \lambda^2(\lambda_s - \lambda) + (h/mc) \text{ vers } \varphi = \lambda^2(\lambda_s - \lambda) - .0242 \text{ vers } \varphi. \end{aligned} \quad (12)$$

From Eq. (8) we find that the kinetic energy of the recoil electron is

$$E = h\nu \cdot \frac{\alpha(1-s)^2 \text{ vers } \varphi}{1 + \alpha(1-s) \text{ vers } \varphi}. \quad (13)$$

Solving the original equations, we find that the angle  $\theta$  between the primary beam and the motion of the electron which recoils from a quantum scattered at an angle  $\varphi$  is given by

$$\tan \theta = - \frac{\cot \frac{1}{2}\varphi}{1 + \alpha - \alpha s}. \quad (14)$$

It will be seen that according to these equations the motion of the recoil electrons is not much affected by the constraining forces until  $s = \lambda/\lambda_s$ .

becomes comparable with 1, in which case their energy is reduced. This result is supported qualitatively by the recent experiments of Bothe<sup>11</sup> on the ranges of the recoil electrons ejected from different substances.

#### EXPERIMENTAL TEST

The experiments of Clark and Duane<sup>8</sup> and of Clark, Duane and Stifler<sup>9</sup> have shown that the modified line or band excited by electrons of the *s* group always occurs at wave-lengths greater than  $\lambda + \lambda^2/(\lambda_s - \lambda)$ . This is in complete accord with our general Eq. (7). It will be of interest, however, to compare also the displacement of the peak of the modified line with the value predicted by Eq. (12).

Perhaps the most precise experimental data referring to the scattering of x-rays by electrons which are loosely bound are those<sup>12</sup> for the scattering of molybdenum  $K\alpha$  rays by carbon. In these experiments, for the rays scattered at 45°, 90° and 135°, the peak of the modified line was displaced (0.0242 vers  $\varphi$ ) Å within a probable error of about  $\pm 0.001$  Å. This is exactly the displacement *F* predicted by Eqs. (7) and (12) for free electrons.

The extensive data of Clark, Duane and Stifler are presented in Table I. This table includes only those lines for which the experimental curves are available and for which the positions of the peaks of the modified lines can accordingly be determined. Clark and Duane<sup>8</sup> have published also the short wave-length limits of certain other modified lines, which agree very well with Eq. (11). In the column describing the origin of the modified line, the symbol MoK $\alpha$ -f indicates that the line is due to molybdenum  $K\alpha$  rays scattered by free electrons, the symbol MoK $\alpha$ -AlK indicates that molybdenum  $K\alpha$  rays are scattered by electrons in the K energy level of aluminium, etc.  $\delta\lambda(\text{obs})$  is in every case the observed wave-length difference between the peaks of the modified line and of the unmodified line excited by the same primary ray.

It will be seen that every observed line except one is accounted for by this form of the quantum scattering theory. The one exception is found in the case of rock-salt, where a line ( $\lambda = 0.823$  Å) occurs between the theoretical positions of the MoK $\alpha$ -NaK and the MoK $\alpha$ -AlK lines. The fact that this line does not alter its position as the angle of scattering is changes from 90° to 135° in the manner characteristic of the modified lines indicates that it is not a true modified line but has some other origin.

When the lighter elements are used as radiators, it will be found from this table that Eq. (12),  $\delta\lambda = D + F$ , predicts the peak of the modified line

<sup>11</sup> W. Bothe, *Zeits. f. Phys.* **20**, 237 (1923)

<sup>12</sup> A. H. Compton, *Phys. Rev.* **22**, 409 (1923)

within experimental error. The only serious departure from this value is observed in the case of the tungsten K rays scattered by molybdenum. In this case the displacement approaches the value  $\delta\lambda = D$ , given by Eq. (11). As we have seen, this means that when the scattering electron is ejected from an atom in which it is tightly bound, most of the impulse

TABLE I  
Wave-length change of modified lines  
(Data of Clark, Duane and Stifler)<sup>8,9</sup>

Radi-ator	Angle	Origin	$\delta\lambda$ (obs) peak	$\delta\lambda$ (calc) peak( $D+F$ ); limit $D$	Nature of line	
Li	135°	MoK $\alpha$ -f	.035A	.041	0	unresolved from K $\alpha$
C	90	MoK $\alpha$ -f	.030	.024	0	partially resolved
Ice	90	MoK $\alpha$ -f	.025	.024	0	partially resolved
Al	90	MoK $\alpha$ -AlK	.094	.093	.069	rather sharp
Al	90	MoK $\beta$ -AlK	.067	.078	.054	unresolved from K $\alpha$
NaCl	90	MoK $\alpha$ -NaK	.058	.070	.046	unresolved doublet
		MoK $\beta$ -ClK	.137	.130	.106	
NaCl	135	MoK $\alpha$ -NaK	.073	.087	.046	unresolved doublet
		MoK $\beta$ -ClK	.152	.147	.106	
NaCl	90	unknown				$\lambda = .823A^*$
NaCl	135	unknown				$\lambda = .823A^*$
C	90	WK $\alpha$ -f	.023	.024	0	faint, unresolved
Cu	90	WK $\alpha$ -f	.028	.024	0	rather broad line
		WK $\beta$ -CuK	.053	.051	.027	
Mo	90	WK $\alpha$ -f	.024	.024	0	unresolved
Mo	90	WK $\alpha$ -MoK	.106	.130	.106	rather broad
Mo	90	WK $\beta$ -MoK	.081	.103	.079	rather broad
La	90	WK $\alpha$ -f	.021	.024	0	faint, unresolved

\*Wave-length unaltered as  $\varphi$  changed.

is transferred to the atom before the electron escapes—a result which might have been anticipated. The quantum theory of scattering in its general form is therefore adequate to account completely for the wave-lengths of the modified lines observed from the heavier as well as the lighter elements.

#### THE "TERTIARY RADIATION" HYPOTHESIS

To account for the wave-length of the modified lines which they observed, Clark and Duane have suggested<sup>13</sup> the hypothesis, mentioned above, that these rays are produced by the collision with the surrounding atoms of the photo-electrons ejected by the primary x-rays. A similar

<sup>13</sup> G. L. Clark and W. Duane, Proc. Nat. Acad. Sci. **9**, 422 (1923)

view was at one time defended also by the writer.<sup>14</sup> Since the kinetic energy of the photo-electron is  $h(\nu - \nu_s)$ , the maximum frequency of the x-rays which they can excite is  $\nu' = \nu - \nu_s$ . This corresponds to a minimum wave-length greater than the primary by  $D \equiv \lambda^2 / (\lambda_s - \lambda)$ , identical with that given by Eq. (11).

Although we have seen that this value of the minimum wave-length is in satisfactory accord with experiment, there are other considerations which seem to make the hypothesis untenable.

(1) *Polarization of the secondary x-rays.* Under the best conditions, for the rays emitted at  $90^\circ$  with the motion of the impinging electron, experiment<sup>15</sup> shows that not more than about 25 per cent of the x-rays produced when a cathode electron traverses matter are polarized. Since the photo-electrons responsible for the tertiary radiation are ejected through a wide range of angles, the x-rays which they excite should be even less strongly polarized. It is found, however, that the secondary x-rays from light elements at right angles with the primary beam contain not more than perhaps 2 per cent of unpolarized x-rays.<sup>16</sup> Since the spectrum of the secondary rays from these substances shows a large fraction of the energy in the modified ray, it follows that the modified ray at right angles with the incident beam is nearly completely polarized. This fact is not consistent with the hypothesis of tertiary radiation.

(2) *Energy of the secondary x-rays.* In the case of the secondary x-rays from carbon, excited by the  $K\alpha$  line from molybdenum, the spectra show that about  $2/3$  of the energy lies in the modified ray. Hewlett has shown,<sup>17</sup> however, that in this case the total energy in the secondary beam corresponds to a mass scattering coefficient of 0.20, in accurate agreement with the simple electron theory of scattering. Since the mass absorption coefficient of the molybdenum  $K\alpha$  line in carbon is about .055, this means that 36 per cent of the energy removed from the primary beam reappears as secondary x-rays, or about 24 per cent as modified rays. Even if we suppose that all of the energy removed from the primary beam is initially transformed into photo-electrons, this means that the efficiency of production of x-rays by the impact of these photo-electrons must be 24 per cent. Experiment shows,<sup>18</sup> however, that the efficiency of production of x-rays by electrons traversing carbon

<sup>14</sup> A. H. Compton, *Phil. Mag.* **41**, 762 (1921)

<sup>15</sup> C. G. Barkla, *Phil. Trans.* **204**, 467 (1905); et al.

<sup>16</sup> A. H. Compton and C. F. Hagenow, *Phys. Rev.* **18**, 97 (1921); *Journ. Opt. Soc. Am.*, April 1924

<sup>17</sup> C. W. Hewlett, *Phys. Rev.* **20**, 688 (1922)

<sup>18</sup> Cf. summary by Bergen Davis, *Bull. Nat. Research Council No. 7*, p. 415 (1920)



with the velocity of these photo-electrons is considerably less than 0.1 per cent. It follows that no appreciable fraction of the modified rays in this case can be due to tertiary radiation. On the other hand, the agreement of Hewlett's experiments with the theoretical scattering coefficient loses its significance unless the modified as well as the unmodified rays are truly scattered.

(3) *Width of the modified lines.* In Ross's spectrum<sup>19</sup> of the  $K\alpha$  line of molybdenum as scattered by paraffine at  $55^\circ$ , the modified  $\alpha_1$  and  $\alpha_2$  can be distinguished, though they differ in wave-length by only .004 A. This implies a sharpness of the modified line which is quite inconsistent with the view that the peak is merely the maximum of a band of general radiation produced by the impact of cathode rays.

These considerations are, however, all in agreement with the idea that the modified line is a type of truly scattered rays. In view of the fact that the wave-length of these lines can be satisfactorily accounted for by the quantum theory of scattering, and especially in light of the experimental evidence for the existence of the recoil electrons, it is very difficult to avoid the conclusion that the modified rays observed in the spectra of secondary x-rays result from the scattering of whole quanta by individual electrons.

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<sup>19</sup> Reference is made to an unpublished spectrum shown before the A. A. S. and Am. Phys. Soc. in December, 1923; see Phys. Rev. **23**, 290 (1924)