REFRACTION OF X-RAYS BY SMALL PARTICLES

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ABSTRACT

The broadening of a beam of x-rays passed through a mass of small particles is investigated mathematically. It is shown that if the width of the beam, as given by the rocking curve of the second crystal of a double x-ray spectrometer, is w_0 before passing through the refracting material and w afterward, then $w^2-w_0^2=8\delta^2 n_0(\log 2/\delta+1)$, δ being equal to $1-\mu$, the refractive index of the material and n_0 being the average number of particles passed over. The radius of the particles may be found from the expression $R=3\ VD/4n_0$, where V is the volume of refracting material per cm and D the thickness of the material passed through. The above expressions are checked by experiments on graphite as to their dependance on δ, and quantitatively by measurements on graded aluminum powder.

SLACK[1] in the course of his work on the refraction of x-rays by prisms found that the beam was greatly broadened on passage through a prism of graphite. He suggested that the broadening was due to refraction by the individual particles of which the prism was composed, and tested the idea by sending a beam directly through plates of graphite of various thicknesses, finding that the width of the beam, as measured by the rocking curve of the second crystal of the double x-ray spectrometer, increased with increasing thickness of the graphite plate. The present work, which was briefly described at the April 1926 meeting of the American Physical Society, is an attempt to work out mathematically the broadening to be expected, and provides a means of obtaining the size of fine particles which can not be measured by other means.

Distribution of energy from a single particle. For simplicity the particles are assumed to be spheres of radius R and index of refraction $\mu=1-\delta$, where for x-rays δ is of the order of magnitude of 10^{-5}. A parallel beam of x-rays passing through the sphere will be made divergent, for since $\mu<1$, the sphere will act like a negative lens.

A ray striking the sphere at an angle θ (see Fig. 1) is bent so that

$$\sin\phi=(1/\mu)\sin\theta. \tag{1}$$

By symmetry, the ray will be equally bent on emerging from the sphere. Hence the bend produced is $\omega=2(\phi-\theta)$. Then $\phi=\omega/2+\theta$, and

$$\sin\phi=(\sin\omega/2)\cos\theta+(\cos\omega/2)\sin\theta=(\sin\theta)/\mu$$

using (1). Dividing by $\cos\theta$, we get

$$\tan\theta=\frac{\sin\omega/2}{1/\mu-\cos\omega/2} \tag{2}$$

[1] Slack, Phys. Rev. **27**, 691 (1926).

If the energy per unit area per second in the original plane wave is E_0, the energy per second meeting the sphere at an angle between θ and $\theta+d\theta$ is $2\pi R^2E_0 \sin\theta \cos\theta d\theta = dE$ (3), and this energy will be refracted between two cones making angles with their axis equal to ω and $\omega+d\omega$ respectively. From (2) we get

$$d\theta = \frac{(\cos\omega/2)/\mu - 1}{2(1/\mu - \cos\omega/2)^2}\,d\omega.$$

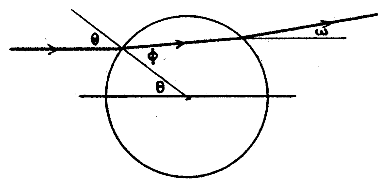

Fig. 1. Path of ray through refracting sphere.

Putting this value with (2) and (3), we get

$$dE = \frac{E_0\pi R^2(\sin\omega/2)\left[(\cos\omega/2)(1/\mu+\mu) - \cos^2\omega/2 - 1\right]}{\mu(1/\mu^2 - (2/\mu)\cos\omega/2 + 1)^2}\,d\omega.$$

Using $\mu = 1-\delta$, $1/\mu = 1+\delta+\delta^2$, $1/\mu^2 = 1+2\delta+3\delta^2$, we get

$$dE = E_0\pi R^2 \frac{(\sin\omega/2)\left[\delta^2 - (\sin^4\omega/2)/4\right]}{(\delta^2+\sin^2\omega/2)^2}\,d\omega.$$

Since, being small, $\sin\omega/2 = \omega/2$, we get

$$dE = E_0\pi R^2 \frac{8\omega\delta^2 - \omega^5/8}{(4\delta^2+\omega^2)^2}\,d\omega$$

The fraction of the energy striking the sphere, or the probability of a "ray" striking the sphere, being refracted through an angle lying between ω and $\omega+d\omega$ is then

$$P'_\omega = \frac{8\delta^2\omega - \omega^5/8}{(4\delta^2+\omega^2)^2}\,d\omega.$$

This equation holds from $\omega=0$ to $\omega = 2\sqrt{2\delta}$, beyond which total reflection sets in. At the critical angle we have $\phi=\pi/2$, $\omega=\pi-2\theta$, $d\omega=-2d\theta$, $\sin\omega = \sin(\pi-2\theta) = -\sin 2\theta$, $dE = 2\pi R^2E_0 \sin\theta\cos\theta d\theta = \pi R^2E_0 \sin 2\theta d\theta = (\pi R^2E_0/2)\sin\omega d\omega$, or, for the probability of a ray being totally reflected through an angle lying between ω and $\omega=d\omega$ we have

$$P_\omega'' = (\omega/2)d\omega$$

Hence, for the total probability of a ray being bent to between ω and $\omega+d\omega$ we have

$$P_\omega = P_\omega' + P_\omega'' = [((8\delta^2\omega - \omega^5/8)/(4\delta^2+\omega^2)^2) + \omega/2]d\omega.$$

The integral of P_ω over its range of application, 0 to $2\sqrt{2}\delta$, is $1+\delta$, which, because of the approximations made, differs from the proper value of 1, but only to a negligible amount.

Effect of many spheres. Suppose there are N spheres per unit volume. The total area for a depth D presented by these spheres to a beam one cm^2 in cross section is $N\pi R^2 D$. Therefore the average number of spheres crossed by a single ray is $N\pi R^2 D$, and hence the average length of a "jump" between "impacts" is $1/N\pi R^2 D = \lambda$. It may be compared with the mean free path in kinetic theory, the light rays being regarded as molecules of zero radius and mass, and the spheres as molecules of finite mass and radius R. The kinetic theory expression (see Jeans, Dynamical Theory of Gases, p. 268) gives then the same result. We can then apply the line of reasoning adopted by Jeans to find the probability of deviations from this average value. The probability of a single jump having a length lying between l and $l+dl$ is (p. 274) $(e^{-l/\lambda}/\lambda)dl$ where λ is the average value given above.

The probability that the total distance for two jumps lie between D and $D+dD$ is $=\int_{L=0}^{L=D}$ (probability of getting to between L and $L+dL$ in one jump)$\times$(probability of getting from L to between D and $D+dD$ in one jump)

$$= dD\int_{L=0}^{L=D} (e^{-L/\lambda}/\lambda)dL(e^{-(D-L)/\lambda}/\lambda) = dD\int_0^D (e^{-D/\lambda}/\lambda^2)dL$$
$$= (De^{-D/\lambda}/\lambda^2)dD.$$

The chance of getting to between F and $F+dF$ in three jumps is $=\int_0^F$ (probability of getting to between D and $D+dD$ in two jumps)$\times$(probability of getting from D to between F and $F+dF$ in one jump)

$$= dF\int_0^F (De^{-D/\lambda}/\lambda^2)dD(e^{-(F-D)/\lambda}/\lambda)dD$$

$$= dF\int_0^F (De^{-F/\lambda}/\lambda^3)dD = (F^2 e^{-F/\lambda}/2\lambda^3)dF.$$

By induction the chance of getting to between L and $L+dL$ in m jumps is

$$P_L = \frac{L^{m-1}\,e^{-L/\lambda}}{(m-1)!\lambda^m}\,dL.$$

The probability of taking m jumps to go a distance D equals the probability of going *at least* D in m jumps, but not in $m-1$ jumps which equals (the probability of going any $L(<D)$ in $m-1$ jumps)$\times$(probability of going at least $D-L$ in one jump)

$$= \int_0^D \frac{L^{m-2}e^{-L/\lambda}}{(m-2)!\,\lambda^{m-1}}\left(\int_{D-L}^{\infty} \frac{e^{-l/\lambda}}{\lambda}\,dl\right)dL = P_m.$$

$$P_m = \int_0^D \frac{L^{m-2}e^{-D/\lambda}}{(m-2)!\,\lambda^{m-1}}\,dL$$

$$P_m = \frac{(D/\lambda)^{m-1}e^{-D/\lambda}}{(m-1)!}$$

The probability of making n impacts is the probability of making $n+1$ jumps. Hence

$$P_n = \frac{(D/\lambda)^n e^{-D/\lambda}}{n!}\,.$$

The average value of P is, of course, D/λ. The root mean square deviation is $(D/\lambda)^{1/2}$. If D/λ is large, the fractional root mean square deviation is small and hence no appreciable error will be introduced by assuming that all rays make D/λ impacts in passing through the refracting material. Even if D/λ is as small as 5, calculation shows that the above assumption yields results which differ only very slightly from those obtained by using the expression for P_n. In what follows, n will be assumed equal to $n_0 = D/\lambda = N\pi R^2 D$ for all rays.

If the volume of the spheres present per cm^3 is V we have $V = 4N\pi R^3/3$ or $N = 3V/4\pi R^3$. Hence the average number of impacts is $n_0 = 3VD/4R$.

The number of impacts per ray giving rise to bends lying between ω and $\omega + d\omega$ is $n_0 P_\omega$. The angles are so small that they may be added like displacements. These $n_0 P_\omega$ displacements will be distributed at random as to direction, and hence their total effect (see Rayleigh, Scientific Papers, Vol. I, p. 491) will on the average be proportional to the square root of the number. Hence they will produce a bend equal to $(n_0 P_\omega)^{1/2}\omega$. There is a similar expression for every ω. To add the effects due to all the ω's in such a way as to take into account their random distribution in direction we take the square root of the sum of the squares, or rather, the square root of the integral of the squares. That is, the average total bend of a ray will be

$$\alpha_0 = \left(\int_0^{2\sqrt{2\delta}} \omega^2\left(\frac{\omega}{2} + \frac{8\delta^2\omega - \omega^5/8}{(4\delta^2+\omega^2)^2}\right)d\omega\right)^{1/2}.$$

$$\alpha_0 = \left(4\delta^2 n(\log \frac{2}{\delta} + 1)\right)^{1/2}.$$

The distribution of energy around this average value can be investigated by the methods of the theory of probabilities as given by Bachelier in "Calculs des Probibilités." It leads to the following result, also given in Rayleigh, loc. cit. The fraction of the original energy which will be bent to an angle lying between α and $(\alpha+d\alpha)$ is

$$P_\alpha = (2\alpha e^{-\alpha^2/\alpha_0{}^2}/\alpha_0{}^2)d\alpha$$

In the derivation of the expression for P_α the expression for P_ω is effective only through its effect on the "fonction d'instabilité," which is simply α_0 above. That is, the expression for P_α would have been obtained from any expression for P_ω having the same mean square value, including the case that all rays were bent by a single sphere an amount $\alpha_0/\sqrt{n_0}$, but with random directions. Physically this means that the form of P_α depends on the variations introduced in the total bend of the different rays by the random directions of their bends from the individual particles, rather than on the fact that some paths may have a larger proportion, say, of small bends than others. This is equivalent to saying that in all paths there will be equal numbers of individual bends of equal amounts. Since the thickness of the material passed through in a given sphere is a function of the bend produced, both depending on the angle at which the ray strikes the sphere, what has been said means that all rays may be regarded as having passed through equal amounts of material and thus as having been absorbed equally. Hence absorption will not alter the shape of the distribution given by P_α, but merely reduce by a constant ratio the amount of energy at any α. As we are interested only in the width of the distribution curve we may therefore disregard absorption.

Since measurements are actually made on the horizontal distribution resulting after passing the beam from a vertical slit through the refracting material, we will seek an expression for the fraction of the energy lying between a horizontal angle β and one $\beta+d\beta$ with the original direction. This is found to be

$$P_\beta = (e^{-\beta^2/\alpha_0^2}/\sqrt{\pi\alpha_0})d\beta$$

So far the original beam has been assumed to have been parallel. Actually it is found that the energy is distributed around the central direction giving a rocking curve which may be closely represented by an equation of the form $y = ae^{-bx^2}$. If tangents be drawn to the curve at its two points of inflection, and the height of their intersection be called h and the width of the curve at $y=h/2$ be called w_0, then the equation of the curve becomes

$$y=(h\sqrt{e}/2)e^{-2x^2/w_0^2}.$$

Each part of this curve will be distributed according to the expression for P_β. By integration the fraction of the original energy to be found after passage through the refracting material, between γ and $\gamma+d\gamma$ may be shown to be

$$P_\gamma=(w_0h\sqrt{e}/2\sqrt{2\alpha_0^2+w_0^2})e^{-2\gamma^2/(2\alpha_0^2+w_0^2)}$$

This is the equation of a curve having a width (as above defined) of $w=(w_0^2+2\alpha_0^2)^{1/2}$.

Hence, if the width of the rocking curve of the second crystal of a double x-ray spectrometer be measured before and after the interposition of a layer of refracting substance, we have $\alpha_0^2=(w^2-w_0^2)/2$. Knowing α_0^2, δ for the refracting material, and D, we may compute n_0, the average number of particles passed through by a ray by the equation $n_0=\alpha_0^2/4\delta^2$ $(\log 2/\delta+1)$, and if the volume per cm^3 of refracting material is known, the radius of the particles may be determined by $R=3VD/4n_0$.

Table I gives the results of the measurements of Slack on graphite. These are exhibited graphically in Fig. 2. $w^2-w_0^2$ should be proportional to D, and, as is shown in the graph, this was found to be the case, within the rather large limits of error of this experiment. Also the size of the particles as calculated from the data on the two wave-lengths used comes out approximately the same.

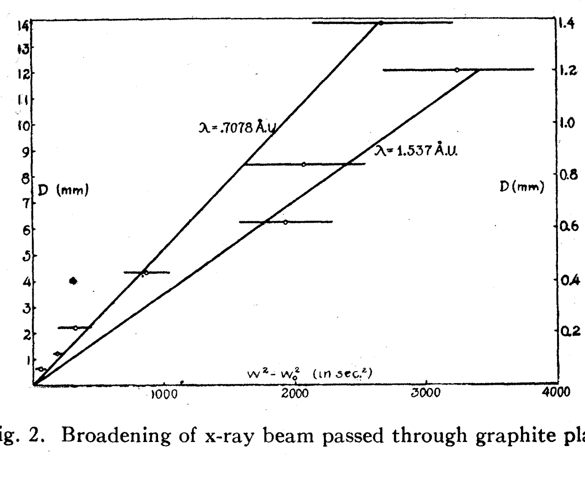

Fig. 2. Broadening of x-ray beam passed through graphite plates.

TABLE I

Graphite

Thickness in mm	Width in seconds
$\lambda = .708$A; $\delta = 1.23\times 10^{-6}$	
.0	6
.62	10 ± 2
1.2	15 ± 2
4.3	30 ± 3
8.4	45 ± 5
13.8	52 ± 5
$\lambda = 1.537$A; $\delta = 4.7\times 10^{-6}$	
.0	10.8
.62	45 ± 4
1.2	58 ± 5

TABLE II

Aluminum, 250–300 *mesh*

Thickness of container in cm	1.30	.400
Density of powder	1.35	1.31
Wave-length $\times 10^8$	.528	.708
$\delta\times 10^6$	.95	1.70
w_o	8.2″	8.7″
w	11.6″	24.6″
Diameter in cm (calc.)	.006	.0069
Dimensions in cm (obs.)	(.0114 × .0055)	

A further test of the theory was made by passing the beam of x-rays through small brass boxes provided with paper walls, containing aluminum powder which had been graded between screens of 250 and 300 mesh. The results are shown in Table II together with the actual average dimensions of the particles, as measured by examination with a compound microscope provided with a calibrated eyepiece. The calculated dimensions are in excellent agreement with those observed, particularly considering that the particles were far from the spherical shape assumed by the theory.

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