

AN EXPERIMENTAL STUDY OF THE SCATTERING OF APPROXIMATELY HOMOGENEOUS X-RAYS BY POWDERED CRYSTALLINE CARBON, METALLIC LITHIUM, AND LIQUID BENZENE, MESITYLENE, AND OCTANE.

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SYNOPSIS.

Scattering and Absorption of Homogeneous X-Rays by Carbon (Diamond and Graphite), Metallic Lithium, and the Liquids Benzene, Mesitylene and Octane.—

1. *Scattering curves.* The intensity of the radiation scattered by the substance contained in a small capsule placed at the center of the spectrometer was measured by the ionization method for angles from 2° to 165° . While a ZrO_2 screen was used to isolate the K_α line of Mo (0.712 \AA), some radiation of $\lambda 0.445 \text{ \AA}$ was apparently also present. For diamond, graphite and lithium, the maxima agree in position and relative intensity with those found by A. W. Hull by the photographic method. The curves for the liquids, however, are remarkable in that each shows one maximum; 8.5° for benzene, 6.5° for mesitylene and 8.1° for octane, with indications of others unresolved, suggesting that the liquids have a crystal structure. For very small angles the scattering is zero for the solids, and approaches zero for the liquids. 2. *Mass-scattering coefficients* were obtained by integrating the area under the curves, or by experimentally integrating the ionization for all angles of scattering. The coefficients for carbon (diamond and graphite), 0.200, and for lithium, 0.168, agree with Thomson's theoretical values. For benzene, mesitylene, and octane, however, the values 0.238, 0.244, and 0.262 respectively are about ten per cent. larger than those predicted by theory, and give too large specific values for carbon and hydrogen, 0.22 and 0.46 instead of 0.20 and 0.40 respectively. The results are probably accurate to three per cent. 3. *True mass-absorption coefficients*, obtained from the total absorption, the scattering and the density, are found for the solids to be in accord with the theoretical expression $KN^3\lambda^3$, the mean value of K being 4.38×10^{-3} .

Evidence of crystal structure of liquids seems to have been found in the scattering curves for benzene, mesitylene and octane, as suggested above, and the spacings of the planes of atoms responsible for the principal maximum are benzene 4.8 \AA ., mesitylene 6.3 \AA ., and octane 5.0 \AA .

INTRODUCTION.

THE experiments described in the following report are the outgrowth of a former series of experiments made by the writer¹ in which the mass absorption and mass-scattering coefficients of homogeneous x-rays were studied for several substances as a function of the wavelength of the x-rays. The definitions of these coefficients, and a discussion of the expression $KN^3\lambda^3 + (\sigma/\rho)$ which has been proposed to give the value of the total mass absorption coefficient of x-rays of wave-

¹ PHYS. REV., N. S., 17, 1921, p. 284.

length λ in an element whose atomic number is N , is given in the paper referred to above. In this former work the total mass-absorption coefficient was measured by determining the intensity of a beam of homogeneous x-rays before and after passing through a screen of the absorbing element. The mass-scattering coefficient deduced from the above equation was found to depart from the theoretical value deduced by J. J. Thomson,¹ and the value of K to depend on the scattering element. Moreover the curves were not found to be linear with λ^3 as they should have been in the region investigated according to theoretical considerations. In particular there seemed to be something anomalous in the behavior of carbon in the neighborhood of wave-length 0.75 Å. which is near the K_α line of molybdenum 0.712 Å. The scattering of hydrogen calculated from the experiments on water and liquid oxygen, and the absorption coefficient of lithium could not be interpreted in the light of theory. It therefore seemed advisable to measure the mass scattering, and the true mass-absorption coefficients for certain elements and wave-lengths independently of one another. The present work, of which a preliminary report² was made last November before the American Physical Society, consists of measurements of the scattered x-rays as a function of the angle of scattering from graphite, diamond, lithium, and three liquid compounds of carbon and hydrogen. The radiation falling on the scattering material was the x-radiation from a molybdenum anode tube excited at about 30 kv. The radiation was filtered through a screen of zirconium oxide which absorbed very greatly all the radiation except that in the neighborhood of the K_α line of molybdenum.

The general results of the measurements have been expressed in two ways. The intensity of scattering has been plotted as a function of the angle of scattering, and the mass-scattering coefficients and the true mass-absorption coefficients have been tabulated together with other data from which they were directly deduced.

The matter is presented in the following order:

- I. Description of Apparatus and Mounting of the Scattering Material.
- II. Procedure in the Experiments.
- III. Procedure in Calculating the Mass Scattering and the True Mass-Absorption Coefficients.
- IV. Sources of Error.
- V. Results and Discussion.
- VI. Appendix; a Detailed Consideration of a Point arising in III.

¹ Conduction of Electricity through Gases, 2nd edition, p. 325.

² PHYS. REV., N. S., 19, 1922, p. 265.

Description of Apparatus and Mounting of Scattering Material.—The Coolidge tube was mounted on sulphur insulators in a lead-covered wooden box about 2 ft. x 2 ft. x 3 ft. The tube was supplied by a 35 kv. oil-insulated transformer which in turn was supplied by a motor generator set. Regulation of voltage and frequency were obtained by rheostats inserted in the field circuits of the motor generator. A D'Arsonval type milliammeter was inserted in the secondary of the transformer at its midpoint which was grounded. All of the high potential apparatus was enclosed in a wire netting cage and the door into this cage could not be opened without opening the primary circuit of the transformer.

The x-ray spectrometer was constructed in this laboratory, and angular settings could be made with an accuracy within 2' of arc. The ionization chamber was mounted on wheels so that it could rotate about the axis of the spectrometer. The scattering material was mounted in a small celluloid capsule whose axis coincided with that of the spectrometer. The small table holding the scattering material was caused to rotate continuously about the axis of the spectrometer.

The ionization chamber was 1 foot long. The outside brass cylinder was 4 inches in diameter and was earthed. The inside cylinder was $3\frac{3}{4}$ inches in diameter, and was kept charged to a potential of about 250 volts by a battery of dry cells. A horizontal electrode insulated by amber was placed in this inner cylinder about half way between its axis and surface. A fine wire was soldered to this electrode and then passed along the axis of a brass tube leading from the ionization chamber to a point in the axis of the spectrometer about 6 inches above the scattering material. From this point the wire passed down the axis of another tube to one pair of quadrants of an electrometer. The movable and fixed tubes were connected by end pieces, one of which could rotate inside the other. The wire joining the electrode to the electrometer was insulated by amber throughout and was completely shielded, all of the shielding being connected together by soldered wires and grounded. The opening in the ionization chamber was 1.50 cm. in diameter, was situated in the horizontal plane passing through the x-ray beam and scattering material and the distance of this opening from the scattering material was 35.5 cm. The electrodes in the ionization chamber were so placed that none of the x-rays from the scattering material could strike them. A lead screen was placed about half way between the scattering material and the opening in the ionization chamber. An opening in this screen was made just large enough to allow every part of the scattering material to send x-rays to every part of the opening. A lead hood was placed over the space between the ionization chamber and this screen to cut off

possible radiation from other objects than those in the neighborhood of the axis of the ionization chamber. In order to cut off all unnecessary direct x-radiation there were two lead slits between the box and the scattering material, the one nearest the scattering material being about 10 cm. distant.

The other pair of quadrants of the electrometer were connected to the case of the electrometer and to earth. An earthing key was connected through a resistance of about 1,000 ohms to earth. This resistance was connected to a potentiometer so that the key could be charged to any desired potential or earthed at will. This key dipped into a mercury cup connected to the electrode in the ionization chamber. The electrometer needle was charged to a potential of 100 volts, and the sensitivity ranged from 1,000 to 5,000 divisions per volt in the different parts of the work.

The scattering material was mounted in thin cylinders of circular cross section, which were made by cutting sheet celluloid of 1 mil thickness into strips from $\frac{1}{8}$ to $\frac{3}{8}$ inch in width and wrapping them helically on a brass or steel rod, allowing each turn a narrow lap on the preceding one. This helix was then secured at each end and the helical seam cemented with collodion. These capsules were found to absorb less than one per cent. of the x-rays falling on them. Two sizes of capsules with internal diameters of $\frac{3}{16}$ and $\frac{1}{4}$ inch were used.

The material called artificial graphite was a highly purified carbon which was obtained in compressed cakes. It was finely powdered by filing, and the powder packed into the capsule. The natural graphite was originally in chunks. This material was forced through a sieve of 50 meshes to the inch, and then packed into a capsule. The diamond was in the form of five splints. These splints were very thin and their cross-sectional area ranged from 0.010 to 0.001 sq. cm. The lithium, from a very old exhibition sample, of German manufacture, was squirted through a die into a rod of the required dimensions, and immediately enclosed in one of the celluloid capsules. Fortunately none of the three liquids used in these experiments attacked the capsules.

II. PROCEDURE IN THE EXPERIMENTS.

Method of obtaining the data for plotting the curves. The beam of x-rays, which had a rectangular cross section, was adjusted so as to cover the whole width of the capsule of scattering material and to project a short distance on either side. The cross section of the beam in the vicinity of the scattering material was approximately 0.8 cm. square, but this was varied slightly from time to time when different size capsules were used.

The x-ray tube was operated at a potential of about 30 kv. at 60.7 cycles per sec. and the current read on a D'Arsonval type milliammeter was 4 m.a. These conditions of operation were maintained constant to within 1 per cent. throughout the work. A zirconium oxide screen, which reduced the K_{α} line to about 60 per cent. of its intensity without the screen, and reduced the K_{β} line to about 6 per cent. of the intensity of K_{α} line, was placed in the path of the radiation as it emerged from the box containing the tube.

The ionization chamber was set at as small an angle as could be done without allowing any of the direct beam to enter it, the electrometer was charged, the earth connection broken, and the time required for the electrometer deflection to change from one of two definite values to the other was determined. An empty capsule of the same construction as the one containing the scattering material was then substituted for the full capsule, and the time required for the same change in deflection determined. Deducing the rates of deflection, and subtracting that due to the empty from that due to the full capsule, the rate of deflection due to the scattering material was obtained, and this was taken as a measure of the intensity of the scattered radiation entering the ionization chamber. If the intensity was so great that the rate of deflection was no longer an accurate measure of its intensity, the following method was adopted. A lead screen was placed between the scattering material and the ionization chamber, the electrometer charged, the earth connection broken, and the deflection noted. The lead screen was then removed for a definite interval of time and then replaced. When the deflection became constant its value was again noted. Whenever necessary, account was taken of the rate of deflection with the lead screen in place. In this way the intensity was determined from point to point, the steps in most cases being less than the angle subtended by the opening in the ionization chamber at the scattering material, particularly in the region of a pronounced maximum. When once the position of the principal maximum of scattering was known, measurements were made frequently at this setting, to check the constancy of operation. Finally the intensity at the various angles was expressed as percentages of the intensity at the principal maximum.

In order to calculate from the scattering curve the fraction of the incident energy which was scattered it was necessary to know the ratio between the flux of radiation into the ionization chamber at some particular setting and the flux of energy falling on the scattering material; and in addition, the total mass-absorption coefficient of the radiation in the scattering material. These quantities were usually determined at

the beginning and at the end of each series of measurements of the intensity. Two methods were used in determining the above ratio and the total mass absorption coefficient.

In the first method a thin lead sheet through which had been bored a circular hole about 0.10 cm. in diameter was placed immediately behind the capsule so that the beam of x-rays through the hole passed through the axis of the capsule. A lead disc mounted on the axle of a small electric motor was placed between the capsule and the ionization chamber with its face perpendicular to the beam of x-rays. The axis of the ionization chamber was set to coincide with that of the beam of x-rays. There was a narrow radial slit in the disc so that when the disc was in rotation the small beam passing through the circular hole entered the ionization chamber for a short interval once during each revolution of the disc. By this means it was found possible to secure a conveniently measurable fraction of the energy flux falling on the capsule. This fraction of the energy flux was calculated from the cross section of the x-ray beam striking the capsule when unobstructed by the lead sheet, the area of the hole, and the angular width of the slit in the lead disc. The cross section of the x-ray beam falling on the capsule was deduced from an x-ray shadow picture made by the x-ray beam on a photographic film placed perpendicular to the x-ray beam and in contact with the full capsule, the beam passing first through the capsule, then striking the film. The width of the shadow of the capsule checked with the diameter of the capsule. The diameter of the hole was measured with a micrometer microscope. The total mass-absorption coefficient of the radiation in the scattering material was determined from measurements of the energy flux through the small hole and rotating disc, determined alternately when the x-rays passed through the full and the empty capsule.

There are two objections to the method outlined above; namely, a flash of x-rays entering the ionization chamber is not representative of the average intensity of the beam, but is a very small section of the radiation emitted during one cycle of the alternating current. The number of these flashes received by the chamber during a determination is very large, however, and unless the disc should have a speed bearing a simple relation to the frequency of the alternating current from the transformer, this consideration should not lead to undesirable results. On the other hand, the thin lead screen with the small hole had to be adjusted very accurately perpendicular to the x-ray beam, for it had a thickness comparable with the diameter of the hole. This consideration led to the adoption of another method for determining the total mass-absorption coefficient and the ratio of the energy flux falling on the capsule to that entering the ionization chamber at a definite setting.

The following method was designed to allow the total energy flux falling on the capsule to pass into the ionization chamber for the time of one complete cycle of the alternating current from the transformer. A lead disc carrying an angular slit of adjustable width was mounted on a horizontal axle, so that in rotating, the beam of x-rays was allowed to pass through the slit when the latter was vertical. This disc and axle was controlled by a stiff steel helical spring in the same manner as the balance wheel of a watch. Two stops with trigger action were arranged so that one of them could hold the disc displaced from its vertical position by a definite amount, and upon release, the other stop would catch and hold the disc at the end of its first swing. By successive adjustments of the stiffness of the spring, the width of the slit, and the height of the stops, the time for the slit to sweep across a point in the x-ray beam was made equal to the period of the alternating current to within 1 per cent. In order to determine when the adjustment was completed, the face of the disc was covered with a thin film of smoke and a vibrating reed operated by the alternating current supplying the transformer traced a curve on the disc as it swung. The disc was mounted close to the box containing the tube, where the cross section of the beam was a minimum. Obviously it makes no difference what part of the cycle the exposure begins if it lasts one complete cycle.

A lead slit open at the lower end and having a width equal to that of the capsule was placed on the capsule when making measurements of the direct beam, so that only the portion of the beam striking the capsule could pass on to the ionization chamber. The opening in the ionization chamber was large enough to receive all of the radiation which passed through the capsule, and the exact position of the chamber was secured with the aid of a fluorescent screen.

The procedure was as follows: with the empty capsule in place the lead disc was secured by one of the stops. The electrometer was charged, the earth connection broken, and the electrometer deflection noted. The stop was tripped, allowing the disc to swing. The deflection of the electrometer changed rapidly, and became constant again in about one minute, at which time its value was again noted. From the change in deflection the charge given to the insulated system was computed, and this was taken as a measure of the radiation falling on the capsule during one cycle. The full capsule was then substituted for the empty one, and the procedure repeated, from which was computed the radiation transmitted through the scattering material during one cycle. A number of such determinations were made at each experiment, those with the empty and full capsules being distributed in such a manner that the mean

values of each corresponded to the same epoch. The intensity of the scattered radiation for the principal maximum of the scattering material was next determined in the manner already described.

By multiplying the ordinates of a scattering curve by the sine of the angle of scattering, and by a factor to take account of the size of the angular opening of the ionization chamber measured from the scattering material, a new curve was obtained, the area under which gave the total amount of scattered radiation in terms of the radiation falling on the scattering material. This procedure rests upon the assumption that an element of the scattering material containing a large number of small crystals would scatter radiation equally in all directions making a given angle with the direction of the direct beam.

The following method was devised so that the procedure of calculation from the scattering curve just described would be done automatically by the method of measuring the scattering. Having first measured the intensity of the direct beam through the empty and full capsules, the ionization chamber was set close to the direct beam. With a lead screen in front of the chamber the electrometer was charged, the earth connection broken, and the deflection noted. At a definite instant, which was recorded beforehand, the lead screen was removed and the ionization chamber moved along from degree to degree, leaving it on each degree for an interval of time proportional to $\sin \theta$, where θ was the angular setting of the chamber from the direct beam. The schedule for moving the chamber was prepared beforehand, and was so designed that the amount of radiation entering the ionization chamber during its tour around the circle was equal to the total amount of radiation scattered in 1 second.

The total charge taken by the electrometer during its tour around the circle was then proportional to the total x-rays scattered during 1 second. This total charge was deduced from the total change in deflection of the electrometer. This procedure was carried through for the empty capsule and for the full capsule in such a manner that the results for each corresponded to the same epoch. At the end, the intensity of the direct beam was again measured through the empty and full capsules.

III. PROCEDURE IN CALCULATING THE MASS-SCATTERING COEFFICIENT AND THE TRUE MASS-ABSORPTION COEFFICIENT.

1. Calculation of the total mass-absorption coefficient $(\mu + \sigma)/\rho$. The densities of the materials in the capsules were determined, in the case of the solids, by dividing the mass of the material packed in the capsule by the volume of the capsule; in the case of the liquids by finding the loss in weight of a sinker of known volume when suspended in them.

When the small hole and rotating disc were used for measuring the total mass-absorption coefficient, this quantity was given by the expression

$$d = e^{-\frac{\mu + \sigma}{\rho} t},$$

where t is the average length of path of the x-rays through the scattering material, and d is the ratio of the intensity of the beam through the full capsule to that through the empty capsule.

When one cycle of the x-rays falling on the capsule was allowed to enter the ionization chamber the total mass-absorption coefficient was calculated from the following expression:

$$d = 1 - \frac{\pi}{4} b + \frac{1}{3} b^2 - \frac{\pi}{32} b^3 + \frac{1}{45} b^4 - \dots,$$

where

$$b = \frac{\mu + \sigma}{\rho} D.$$

D is the diameter of the capsule.

2. Calculation of the fraction of the x-rays falling on the scattering material that was scattered. When using the scattering curves the first procedure was to multiply the ordinates of the scattering curve by the factor

$$\frac{2 \sin \delta}{1 - \cos \delta} \sin \theta,$$

where δ is half the plane angle subtended at the scattering material by the opening in the ionization chamber, and θ is the angle from the direct beam at which the scattering was measured. The area under this new curve was then found, using 2δ as the unit abscissa and 100 per cent. as the unit ordinate. This area is equal to the total measured scattered x-rays in terms of that measured at the angle of maximum scattering. The number for the total measured x-rays was then corrected by estimation for the scattering between 165° and 180° inaccessible to the ionization chamber. This correction usually amounted to 1 or 2 per cent. of the total measured x-rays. Next the correction for absorption in the scattering material (see appendix) was applied, giving the total scattered x-rays in terms of that measured at the angle of maximum scattering. Finally, this number was divided by the ratio of the energy flux falling on the scattering material to that entering the ionization chamber at the angle of maximum scattering, giving the fraction of incident x-rays that was scattered.

When the scattering was measured by rotating the ionization chamber

from degree to degree the total measured scattered x-rays for the empty capsule were subtracted from that for the full capsule, and the corrections for the region inaccessible to the ionization chamber and for the absorption in the scattering material were applied. This gave the total x-rays scattered in 1 second which was then divided by the x-rays falling on the scattering material in 1 second.

3. Calculation of the mass-scattering and the true mass-absorption coefficients. When one cycle of the radiation falling on the capsule was allowed to enter the ionization chamber, the amount of the direct beam taken out by the scattering material is given directly by $1 - d$. When the small hole and rotating disc was used the fraction of the direct beam of x-rays taken out by the scattering material was calculated from the expression

$$\frac{\pi}{4}b - \frac{1}{3}b^2 + \frac{\pi}{32}b^3 - \frac{1}{45}b^4 + \dots,$$

where

$$b = \frac{\mu + \sigma}{\rho} D.$$

Let this fraction be denoted by T , while S denotes the fraction of the incident beam scattered. Then since scattering and absorption take place together

$$\frac{S}{T} = \frac{\sigma}{\mu + \sigma},$$

so that the mass-scattering coefficient is given by

$$\frac{\sigma}{\rho} = \frac{S}{T} \cdot \frac{\mu + \sigma}{\rho}.$$

Each of the three quantities on the right were measured so that σ/ρ could be calculated. The true mass-absorption coefficient was then obtained by subtracting the mass-scattering coefficient from the total mass-absorption coefficient.

IV. SOURCES OF ERROR.

1. On account of the finite size of the scattering material there is some indefiniteness as to its centroid. The volume of the scattering material was about 0.10 cm.³ and 0.20 cm.³ for the smaller and larger capsules respectively.

2. The x-rays were not parallel at the scattering material, so that the total mass-absorption coefficients as calculated would be slightly in error. The distance from the source of x-rays to the scattering material was 1 meter.

3. Saturation voltage in the ionization chamber was secured by trial, and then twice that voltage allowed as a factor of safety.

4. The electrometer scale was not absolutely uniform, but the proper corrections were applied wherever necessary.

5. The variation of voltage, frequency, and thermionic current through the tube would introduce serious errors in work of this kind if not carefully controlled. All of these were carefully regulated by hand, frequently one person giving this his entire attention.

6. Variation in the effective amount of methyl bromide in the ionization chamber during a series of experiments. Although this was at times rather large, standardizing measurements were made at the beginning and end of each series, and frequently during the series, so that it was possible to allow by interpolation for uniform changes such as the above.

7. Non-uniformity of the density of the scattering material in the capsules. In the case of the liquids this consideration does not enter, and in the case of the diamond splints it is safe to conclude that the effect is negligible. With the two kinds of graphite every precaution was taken to insure uniformity of distribution by careful packing, and the observations were made allowing the x-rays to fall on different sections of the capsule. In the case of the lithium there is a possibility (hardly probable) of blow holes. These blow holes might even be filled with some of the oil in which the lithium had been kept.

8. The adjustment of the small hole behind the scattering material in the one method for measuring the absorption coefficient and the adjustment of the slit on the scattering material in the other method were made with great care as a small displacement of these limiting devices would lead to a relatively large error in the results.

All of these main sources of error and several other minor ones have been carefully considered and it is estimated that the final results are not in error by more than 3 per cent. on account of them.

V. RESULTS AND DISCUSSION.

Table I. gives the results on the mass-scattering and the true mass-absorption coefficients of the substances given in the first column. The second column gives the method used in the determination, the number in brackets showing the number of separate determinations from which the values in a given line were deduced.

Method *A* signifies that the calculations were made from the scattering curve for the material. The comparison with the direct beam of x-rays in these calculations was determined by allowing one cycle of the x-rays

falling on the capsule to enter the ionization chamber. In method *B* the ionization chamber was moved from degree to degree allowing the charge on the electrometer to accumulate. The comparison with the direct beam of x-rays in these calculations was determined by the small hole and rotating disc method. Method *C* differed from method *B* in that the comparison with the direct beam was as in method *A*. The third column gives the percentage *S*, of the x-rays falling on the scattering material, which was scattered. The fourth column gives the fraction *F*, of the x-rays falling on the scattering material, which was removed by the combined processes of absorption and scattering. The fifth, sixth, and seventh columns give the total mass-absorption coefficient, the mass-scattering coefficient, and the true mass-absorption coefficient respectively. The eighth column gives the means of the mass-scattering coefficient for each substance. The ninth column gives the value of *K* calculated from the expression $\mu/\rho = KN^3\lambda^3$ where μ/ρ is the true mass-absorption coefficient.

A comparison of the position of the maxima in the scattering curves for the solid substances shows good agreement with those obtained by Hull¹ using the photographic method. The relative intensities of the

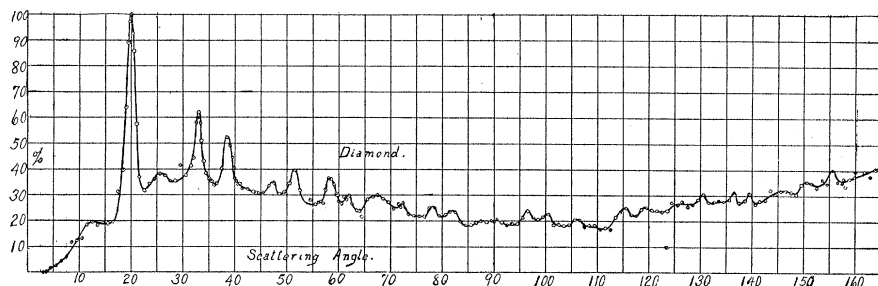


Fig. 1.

various maxima agree as well as might be expected considering the degree of uncertainty that lies in estimating relative intensities from blackening of photographic plates.

A novel point of interest shown by the curves and previously reported by the writer² is that for the solid substances the scattering becomes zero for very small angles from the direct beam, and approaches zero for the liquids. A special experiment was carried out to check this fact. A lead slit about 3 mm. wide was placed over the opening in the ionization chamber and it was found that at 2° from the direct beam the liquids

¹ PHYS. REV., N. S., 10, 1917, p. 661.

² PHYS. REV., N. S., 17, 1921, p. 284.

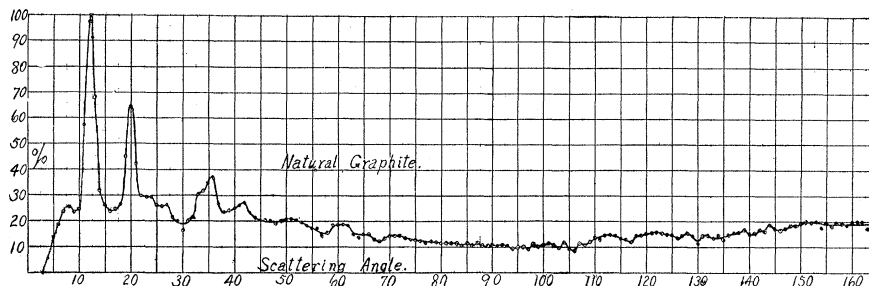


Fig. 2.

scattered less than 10 per cent. as much as at the angle of maximum

TABLE I.

1.	2.	3.	4.	5.	6.	7.	8.	9.
Substance.	Method.	S%.	F%.	$\frac{\mu + \sigma}{\rho}$	$\frac{\sigma}{\rho}$	$\frac{\mu}{\rho}$	Mean $\frac{\sigma}{\rho}$	K.
Diamond Splints.....	A(1)	11.86	26.1	.428	.195	.233	.199	4.40×10^{-3}
Diamond Splints.....	B(10)	12.16	31.7	.515	.198	.317		
Diamond Splints.....	C(2)	12.19	29.6	.497	.205	.292		
Natural Graphite.....	A(1)	6.55	18.6	.553	.195	.358	.201	4.33
Artificial Graphite....	A(1)	6.33	17.4	.551	.200	.351		
Artificial Graphite....	B(7)	6.49	17.4	.551	.206	.345		
Artificial Graphite....	C(3)	6.35	15.2	.477	.199	.278	.168	4.35
Metallic Lithium.....	A(1)	4.21	5.78	.217	.158	.059		
Metallic Lithium.....	C(4)	4.14	4.80	.206	.178	.028		
Benzene.....	A(1)	9.28	18.6	.478	.238	.240	.238	4.36
Benzene.....	B(3)	6.90	14.7	.497	.233	.264		
Benzene.....	C(3)	6.69	13.8	.468	.227	.240		
Benzene.....	C(1)	9.70	21.6	.567	.255	.312	.244	
Mesitylene.....	A(1)	9.39	18.4	.478	.244	.234		
Mesitylene.....	B(2)	6.83	14.4	.489	.232	.257		
Mesitylene.....	C(2)	7.02	12.8	.431	.236	.195	.262	
Mesitylene.....	C(1)	9.89	21.3	.564	.262	.302		
Octane.....	A(1)	8.05	15.5	.496	.258	.238		
Octane.....	C(1)	8.29	16.3	.524	.266	.258		

scattering, while for graphite and diamond the scattering was zero at angles of 5° and 6° respectively. Since this fact was published, A. H. Compton¹ has given a theoretical discussion in which he has shown that the scattering would be expected to become zero for small angles in the case of crystalline material. A quantitative explanation of this behavior for the liquids is as yet not forthcoming. It will be noticed that there is a prominent maximum which occurs for the solid substances at a smaller

¹ Not yet published.

angle than the principal maximum which corresponds to a wave-length 0.712 \AA . Assuming that this maximum at the smaller angle is due to

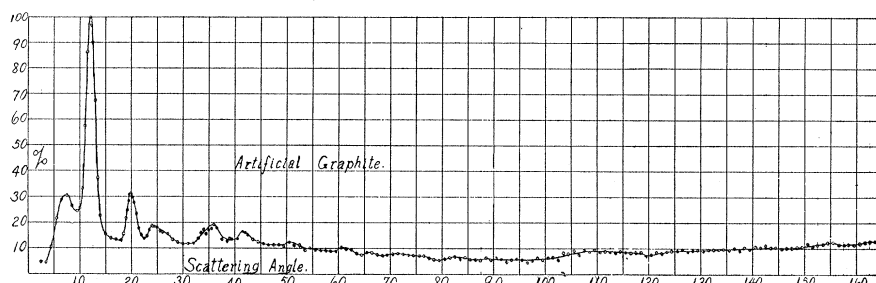


Fig. 3.

the short wave-length radiation which gets through the Zr filter, the effective wave-length was calculated from the data on diamond graphite and lithium supposing that this maximum was due to reflection from the set of planes giving the principal maximum in each case. This gave the effective wave-length of this radiation as 0.446 , 0.441 , and 0.449 \AA .

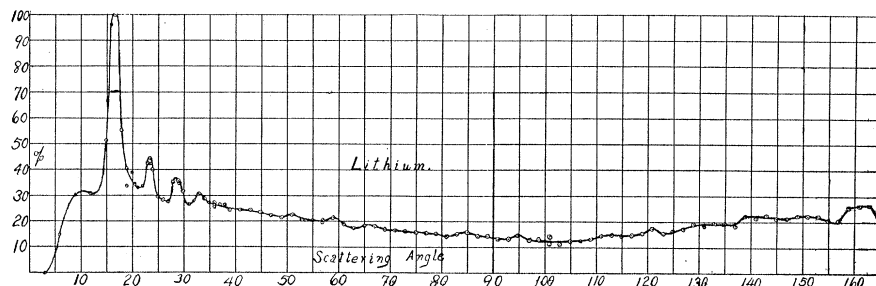


Fig. 4.

for the three substances in the order named. The mean value for this wave-length is then 0.445 \AA . This explains the appearance of the maximum occurring in the neighborhood of 25° in the curve for diamond, for this is the correct position for second-order reflection of wave-length 0.445 \AA . from the III planes.

The appearance of a very pronounced maximum at a small angle for each of the liquids investigated points very strongly toward the view that these liquids have something of a crystal structure. These maxima occur at rather small angles, and consequently the maxima due to the first two or three orders of reflection for the wave-length 0.445 \AA . would overlap the first two due to the wave-length 0.712 \AA . It will be seen

that this idea readily lends itself to explain the relatively complicated structure of the maxima, and the slightly irregular course of the scattering

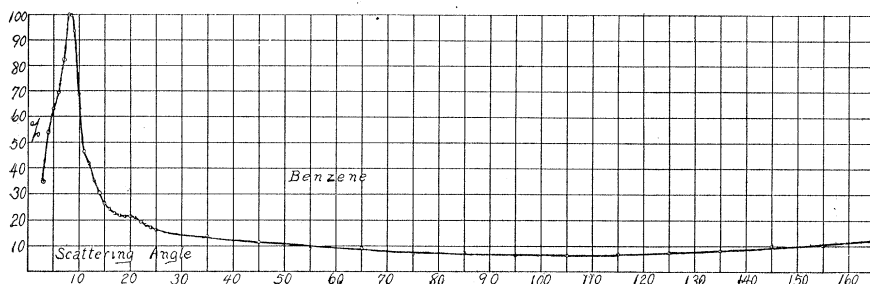


Fig. 5.

curves for the liquids between 10° and 25° scattering angle. The capsules containing the liquids from which these curves were obtained were 0.25 inch in diameter, so that the overlapping would be greater than with the smaller capsules (0.187 inch in diameter) which were used for graphite and diamond. The scattering curves for these liquids for the first 25° have been studied very carefully, using capsules 0.187 inch in diameter, and a slit 3 mm. wide across the opening in the ionization chamber. With this arrangement the complicated structure of the maxima shown in the curves in this paper have been resolved into separate maxima, and the position of the principal maximum due to

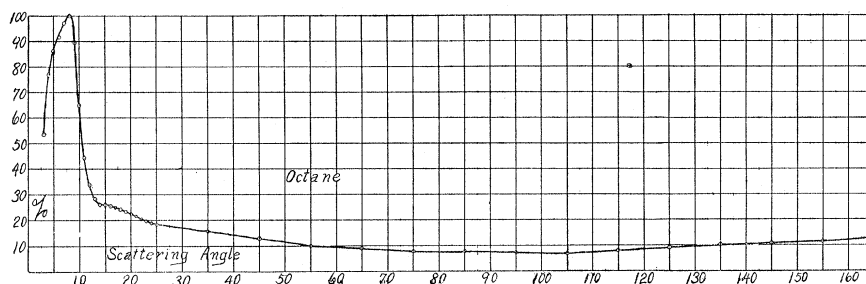


Fig. 6.

wave-length 0.712 \AA . found as follows: benzene $8^\circ.5$, mesitylene $6^\circ.5$, octane $8^\circ.1$. The distances between the planes of atoms responsible for these maxima are as follows: benzene 4.8 \AA ., mesitylene 6.3 \AA ., octane 5.0 \AA .

Very little information can be drawn from the curves in regard to the intensity of reflection from the crystal planes in the successive orders, for the number of sets of planes coöperating at a given angle of scattering

in the case of powdered crystals is too great. Since there are an infinite number of sets of planes in any given crystal lattice, the number of coöperating sets at any given angle of scattering would be very great, and would be greater the smaller the individual crystals of the material. This point is illustrated by a comparison of the curves for natural and artificial graphite. The artificial graphite was more finely powdered than the natural graphite, and the maxima for the latter stand out more prominently than for the former. The curves indicate, however, that the reflections in the successive orders decrease at least as rapidly as the inverse square of the order of reflection.

It should be mentioned that the shape of the scattering curves depends to some extent on the dimensions of the scattering material and the opening in the ionization chamber; the changes in the curves becoming more abrupt the smaller these dimensions. The relative heights of the maxima and minima would also be slightly dependent on these considerations. The radiation falling on the scattering material contains, besides the K_{α} line of molybdenum, an appreciable amount of shorter wave-length radiation, which fact in itself complicates the interpretation of the curves to a certain extent. With different thicknesses of filtering material the scattering curves would be different. The more homogeneous the radiation the more prominent, and the sharper would be the maxima of the curves. But even with constant filter thickness, dimensions of apparatus, and conditions of tube operation, there is still a factor

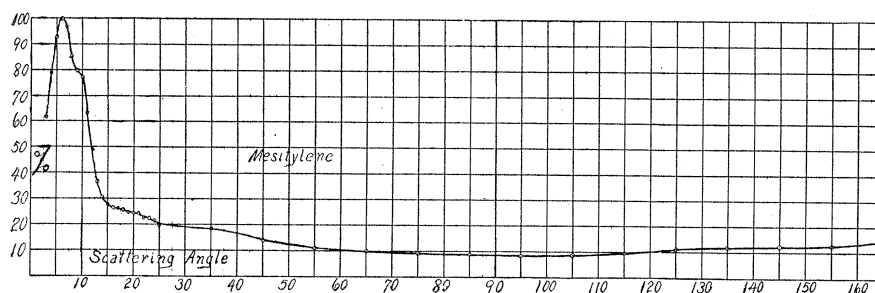


Fig. 7.

which affects the experimental scattering curves. The ionization chamber is not long enough to absorb all the radiation which enters it; especially is this true for the short wave-length radiation. When the ionization chamber had been freshly filled with methyl-bromide gas the short wave-length radiation was absorbed to a much greater extent by the gas than a few days later when the gas had become less susceptible to ionization. The longer wave-length radiation of the K_{α} line was

absorbed almost to the same extent when the chamber was freshly filled, and for a few days thereafter. It may readily be seen that the experimental scattering curve, with a given distribution of radiation falling on the scattering material, will depend upon the density of methyl-bromide gas in the ionization chamber; the maxima will be sharper and more pronounced, the less the density of the gas. For the sake of uniformity and in order to have large ionization currents which could be measured with accuracy, the curves shown in this paper were determined in the interval between freshly filling the ionization chamber and a time when the ionization current had fallen to half value with a constant intensity of x-rays. In the other part of the work where the ionization chamber was rotated continuously so as to automatically integrate the scattered radiation, the density of the gas in the ionization chamber varied widely. It is noticeable in this connection that although the total and true mass-absorption coefficients varied through a wide range for a given substance the mass-scattering coefficient was nearly constant. This is to be explained by the variation in the amount of methyl-bromide gas in the ionization chamber. Since there is both short and long wave-length radiation present the ionization chamber will give a relatively greater importance to the short wave-length radiation when freshly filled than some time later, so that the effective wave-length for ionization will be shorter the greater the density of methyl bromide in the chamber. According to theoretical considerations the mass-scattering coefficient is independent of the wave-length (over certain regions of wave-length this is not true) while the true mass-absorption coefficient in this region of wave-length should be proportional to the cube of the wave-length.

In order to test the above proportionality, the factor of proportionality was calculated for diamond, graphite, and lithium from the equation $\mu/\rho = KN^3\lambda^3$. The value of λ to be substituted in this equation was determined for diamond and graphite by reading off from the absorption curve for carbon previously published by the writer the wave-length corresponding to the value for the total mass-absorption coefficient. The value of the wave-length for lithium was taken to be the same as that for natural graphite as the conditions of operation were about the same in the two cases. It is noticeable that K is nearly the same for diamond, graphite, and lithium, its mean value being 4.38×10^{-3} . This contradicts the writer's former work on lithium where K and the absorption coefficient were found to be abnormally high. The lithium used in the former experiments must have been impure. At least it is known that potassium chloride was mixed with the lithium chloride in the electrolytic separation of the lithium.

From the data given in the table it is seen that the mass-scattering coefficient for carbon agrees with the theoretical value calculated by J. J. Thomson,¹ the average values for graphite and for diamond agreeing with one another and with the theoretical value, 0.200, within the limit of experimental error. The mass-scattering coefficient is thus seen to be independent of the crystal structure. The mean value for lithium 0.168 comes within 3.5 per cent. of the theoretical value 0.174. The mass-scattering coefficients found for the liquids varied somewhat erratically, moreover they are greater than the theoretical values assuming the mass-scattering coefficient of carbon and hydrogen to be 0.200 and 0.399 respectively. The theoretical values for benzene, mesitylene, and octane are 0.215, 0.220, and 0.231 respectively, while the means of the experimental values are 0.238, 0.244, and 0.248 respectively. Assuming that these experimental mass-scattering coefficients are each made up of two parts, one due to carbon and one due to hydrogen, each part in proportion to the mass of carbon and hydrogen present respectively, the mass-scattering coefficients of carbon and hydrogen were calculated to be 0.228 and 0.375 respectively. The value thus found for hydrogen comes within 6 per cent. of the theoretical value 0.399, while the value for carbon is 14 per cent. higher than the value found for graphite and diamond. The mass-scattering coefficients of the three liquids found by method *A* furnish probably a more just basis for determining the separate coefficients for carbon and hydrogen than the above calculation, for the three curves were determined simultaneously point for point. The three equations for the three substances are:

$$\begin{array}{ll} \text{For benzene} & 12C + H = 3.100, \\ \text{For mesitylene} & 9C + H = 2.440, \\ \text{For octane} & 16C + 3H = 4.893, \end{array}$$

where C and H represent the mass-scattering coefficients of carbon and hydrogen respectively. These equations give three independent values for C and H.

	C.	H.
	0.220	0.488
	0.221	0.441
	<u>0.220</u>	<u>0.460</u>
Means,	0.220	0.463

It is seen that both the values of C and H are considerably larger than the theoretical values. If these liquids contained very small percentages of some element of moderately large atomic weight, such as bromine,

¹ Conduction of Electricity through Gases, 2nd edition, p. 325.

some of the absorbed x-rays reëmitted as characteristic x-radiation would be of sufficient penetrating power to reach the ionization chamber where it would add to the effect of the scattered radiation. At least this hypothesis is plausible in the case of the octane, for bromine was one of the constituents of one of the compounds from which it was prepared. The origin of the benzene and mesitylene is not known, but their boiling points and densities indicate that they are relatively pure. One might look for some such effect as proposed above by the method devised by A. H. Compton.² This was tried at the angle of maximum scattering for benzene with a negative result.

VI. APPENDIX. CORRECTION FOR ABSORPTION OF THE SCATTERED RADIATION WITHIN THE SCATTERING MATERIAL.

Attempts to arrive at a complete solution of this problem have not succeeded.

Making the usual assumption of exponential absorption of the x-rays, let $(\mu + \sigma)$ be the linear absorption coefficient, where μ represents that part of the coefficient due to true absorption, and σ that due to scattering. Let us suppose that the distance of the source of x-rays from the scattering material is great enough so that the x-rays are parallel in passing through. This condition is sufficiently fulfilled in the present work. It may then be shown that of a beam of x-rays striking a circular cylinder of material perpendicular to its axis, a fraction is scattered per unit solid angle in the direction making an angle θ with the beam, given by the following expression:

$$S_{\theta} = f(\theta) \left(1 - \frac{4}{3\pi} a + \frac{1}{8} a^2 - \frac{4}{45\pi} a^3 + \frac{1}{192} a^4 - \dots \right),$$

where $a = (\mu + \sigma)D$ and D is the diameter of the cylinder $f(\theta)$ is a factor depending on the crystal structure, expressing the angular distribution of the scattered radiation. Not all of this scattered radiation gets out of the scattering material because it is absorbed and scattered on its way out. If it assumed that none of S_{θ} which is rescattered gets out, we arrive at the following expression for the fraction of the original beam getting out of the cylinder per unit solid angle in the direction making an angle θ with the beam

$$S_{\theta}' = f(\theta) \int_{-R}^{+R} \int_{-\sqrt{R^2-x^2}}^{+\sqrt{R^2-x^2}} e^{-(\mu+\sigma)[x+\sqrt{R^2-y^2}+\sqrt{R^2-(x\sin\theta+y\cos\theta)^2-x\cos\theta+y\sin\theta}]} dx dy,$$

where R is the radius of the cylinder. The origin of coördinates is taken

² PHYS. REV., N. S., 18, 1921, p. 96.

on the axis of the cylinder. The above assumption is not justifiable, and consequently S_θ' given above is too small. The first four terms of the expansion in powers of a , of the above integral, which were found for me by Prof. E. W. Chittenden, of the department of mathematics at this university, are as follows:

$$S_\theta' = f(\theta) \left[1 - \frac{8}{3\pi} a + \frac{1}{4} \left(1 - \frac{\cos \theta}{4} + \frac{\gamma(\pi - \gamma)}{2\pi \sin \gamma} + \frac{\pi - 2\gamma}{4\pi} \cos \gamma + \frac{\sin \gamma}{\pi} \right) a^2 - \frac{24}{45\pi} \left(1 - \frac{7}{22} \cos \theta + \frac{1}{22} \cos^2 \theta - \frac{1}{22} \cos^3 \theta + \frac{1}{22} \cos^4 \theta \right) a^3 \right],$$

$$a = (\mu + \sigma)D$$

and

$$\begin{aligned} \gamma = \theta & \quad \text{if} \quad 0 \leq \theta \leq \frac{\pi}{2}, \\ \gamma = -\theta & \quad \text{if} \quad -\frac{\pi}{2} \leq \theta \leq 0, \\ \gamma = \pi - \theta & \quad \text{if} \quad \frac{\pi}{2} < \theta \leq \pi, \\ \gamma & \quad \cdot \\ \gamma = \pi + \theta & \quad \text{if} \quad -\pi \leq \theta < -\frac{\pi}{2}. \end{aligned}$$

The ratio S_θ/S_θ' gives the correction factor for a given angle θ , that is the number by which the measured scattered radiation must be multiplied to give the actual amount scattered. This factor will be too large, however, because, as mentioned above, S_θ' is too small. The diamond splints being the substance requiring the largest correction factor, calculations were made for it for nine values of θ from 0 to π . The factor has its largest value 1.228 at 0 and continuously decreases to its smallest value 1.200 at π , the value 1.216 at $\pi/2$ being almost the mean of the other two.

Now we may get another estimate of the correction factor by assuming that all of the originally scattered radiation which is rescattered gets out. With this assumption the fraction of the original beam getting out of the cylinder per unit solid angle in the direction making an angle θ with the beam is given by the following expression:

$$S_\theta'' = f(\theta) \times \int_{-R}^{+R} \int_{-\sqrt{R^2-x^2}}^{+\sqrt{R^2-x^2}} e^{-\mu [x + \sqrt{R^2-y^2} + \sqrt{R^2-(x \sin \theta + y \cos \theta)^2 - x \cos \theta + y \sin \theta} - \sigma \sqrt{R^2-y^2+x}] dx dy}.$$

Since the previous integral when solved varied so little with the angle θ it was deemed sufficient to solve the above expression for $\theta = 0$. Now

this expression will give too large a value for the scattered radiation getting out of the cylinder, and too small a value for the correction factor for some of the rescattered radiation will be absorbed. The above integral when solved for $\theta = 0$ gives

$$S_{\theta}'' = f(\theta) \left(1 - \frac{4}{3\pi} \frac{b^2 - a^2}{b - a} + \frac{1}{8} \frac{b^3 - a^3}{b - a} - \frac{4}{45\pi} \frac{b^4 - a^4}{b - a} + \dots \right),$$

where $a = \mu D$; $b = (\mu + \sigma)D$.

The values of S_{θ}/S_{θ}' and S_{θ}/S_{θ}'' for $\theta = 0$ for the diamond capsule were respectively 1.228 and 1.130. The mean value of this is 1.179, and since the value 1.228 for $\theta = 0$ was about 1 per cent. larger than the mean value, the value 1.17 was taken as the average correction factor for the diamond capsule. In the case of the other materials the difference between the two estimates of the correction factor were much smaller. For instance for the artificial graphite the two values were 1.11 and 1.07, giving a mean value of 1.09.

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