THE SPECTRUM OF X-RAYS SCATTERED FROM GRAPHITE STUDIED BY MEANS OF THE DOUBLE CRYSTAL IONIZATION SPECTROMETER

Thesis

by

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ABSTRACT

The scattered spectrum of Graphite is obtained using apparatus consisting of a Hoffmann Electrometer and stationary ionization chamber, (total capacity of insulated system 12 e.s.u.) a specially designed double crystal (calcite) spectrometer, and a specially designed metal x-ray tube fitted with thin windows close to the focal spot so that scattering could be obtained from a graphite scatterer placed 5/8" from the focal spot at an angle of scattering of 165 ± 10°. The tube was operated on the vacuum pumps at 60 KV and 10 m.a. C.P.D.C.

The spectral curves obtained were reproducible and in good agreement with the best work in the field and the following conclusions can be drawn from them. (1) The Compton modified line $MoK\alpha_{1,2}$ scattered from graphite at 165° ± 10° scattering angle is very broad - of the order of 21 X.U. at half maximum height. (2) On account of this excessive breadth there is no discernible separation of the α doublet in the modified spectrum. (3) No fine structure exists in the scattered spectrum of intensity $> 1/5$ of the intensity of the modified radiation. (4) The ratio of intensity of the modified to unmodified scattered radiation determined by measuring the areas under the spectral curves was found to be 7.3 : 1. (5) Viewed in the light of the theory of DuMond for the dependence of modified line breadth on scattering angle, the great breadth of the observed modified line can be interpreted as direct experimental evidence of a dynamic atom. (6) There appears to be no essential difference in the results obtained, whether a spectrum is studied with the single or double crystal spectrometer except in regard to resolution.

INTRODUCTION

The previous work in the field of x-rays on which the present experimental study depends has been done by men of many countries during the past 35 years. In 1895 Roentgen[1] discovered a new type of radiation which he called x-rays. Various other investigators became interested in this new radiation and in 1912 Laue, Friedrick, and Knipping[2] got atomic diffraction patterns from crystals. The symmetry of these patterns was evidence of the existence of a regular crystalline lattice and also gave evidence concerning the type of the basic structure of these lattices. In 1913 the Braggs[3] utilized crystal lattices in their studies of x-ray spectra discovering the Bragg law of x-ray diffraction by crystals:

$$n\lambda = 2d \sin\theta$$

where n is the order number, λ is the wavelength of the incident radiation, d is the lattice constant, and θ is the glancing angle of incidence with the crystal face. Using this relation, the Braggs made very precise determinations of the wavelength of x-rays. Also in 1913, Mosely[4] made a brilliant application of x-ray spectra to the problem of the constitution of matter, discovering the Moseley Law -

$$N \propto \sqrt{\nu}$$

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(1) Roentgen, Sitzungsber der Würzburger Physik-Medic Gesellsch. Jahrg. 1895. (Trans., Stanton, Science 3, 227, 1896)
(2) Laue, Friedrich, and Knipping, Le Radium 10, 47, Feb., 1913.
(3) W. H. and W. L. Bragg, "X-Rays and Crystal Structure".
(4) Moseley, Phil. Mag., 26, 1024, Dec. 1913.

where N is the atomic number, and ν is the frequency of a particular characteristic spectral line for each element. Moseley's discovery makes it fundamentally logical to arrange the elements in a periodic system.

From this time on the application of x-rays as a research tool spread rapidly into many varied fields of science, but the special work in the field of absorption and scattering of x-rays has the most direct bearing on the present study. In October 1922, A. H. Compton[5] applied the laws of conservation of energy and momentum to the interaction of radiation and matter in the case of x-ray scattering by "free" electrons. He used certain quantum concepts of radiation and showed that there should be a loss of energy, when radiation is scattered by "free" electrons, which can be represented as an increase in wavelength, λ, by the well known equation -

$$\lambda' - \lambda = \frac{h}{m_o c}(1 - \cos\phi) = \frac{2h}{m_o c}\sin^2\frac{\phi}{2}$$

where h is Planck's constant, m_o is the rest mass of an electron, c is the velocity of light, λ' is the modified wavelength, λ the unmodified wavelength, and ϕ the angle of scattering. Experimental work in the field by Compton[5]; Ross[6]; Becker, Watson, and Smythe, Brode and Mott-Smith[7], Sharp[8] and several others soon showed that the theory was tenable.

As frequently occurs in physics, several workers get the same results independently about the same time. Ross had got the modified

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(5) Compton, Phys. Rev. 22, Nov., 1923.
(6) Ross, Proc. Natl. Acad. 10, 304, 1924.
(7) Becker, Watson, Smythe, Brode and Mott-Smith, Phys. Rev. 23, 763, 1924.
(8) Sharp, Phys. Rev. 26, 691, 1925.

line experimentally before Compton did his work but did not fully grasp its significance. Again, Debeye[9] got out the theory of the interaction of radiation and "free" electrons about the same time as Compton but his publication of it appeared later. Ross[10] suggested that the breadth of the modified line could be explained as a sort of a Doppler effect due to the randomly oriented velocities of the scattering electrons. Jauncey[11] worked out a theory of this Doppler broadening based on the Bohr model of the atom; and predicted a line structure which was not in good agreement with experiment. Besides this, Jauncey obtained an equation which predicted the relative intensities of modified and unmodified scattering. This part of his work was in good agreement with experiment. In 1929 DuMond[12] published a theory for the dependence of the breadth of the modified line on scattering angle and wavelength. The assumptions on which the theory rests are conservation of Energy and Momentum, loosely bound electrons, with a randomly orientated distribution of velocities, whose velocity is small compared with the velocity of light, and the probability of scattering by a given electron class is proportional to the population of that class. This breadth (for a class of electrons of velocity βc is -

$$\Delta\lambda = 4\beta\lambda^{*}$$

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(9) Debeye, Phys. Zeits 22, pp. 161. Apr. 15, 1923.
(10) Ross, Proc. Nat'l. Acad. Sci. 9, 246, 1923.
(11) Jauncey, Phys. Rev. 25, June 1925.
(12) DuMond, Phys. Rev. 33, 643, 1929.

where $2\lambda^* = \left(\lambda^2 + \lambda'^2 - 2\lambda\lambda' \cos\phi\right)^{\frac{1}{2}}$

and $\beta = \frac{v}{c}$; $\lambda' - \lambda = \frac{h}{m_0 c}(1 - \cos\phi)$

This breadth of course represents the breadth of a particular element of a spectral line. In any real case the observed spectral line would have a shape or structure built up out of a lot of such rectangles, whose breadths depend on the electron velocities and whose areas depend on the number of electrons of a given velocity class. A simplified derivation of this equation for the dependence of the breadth of the modified line on wavelength velocity and scattering angle appears in the Appendix(13). A rigorous derivation of these equations can be found in the literature(14).

DuMond and Kirkpatrick, using the multi-crystal spectrograph, have studied the dependence of breadth on scattering angle and wavelength, and have obtained considerable quantitative evidence for the theory and assumptions. The quantitative verification of this theoretical dependence of breadth on the scattering angle is direct experimental evidence of a dynamic atom !!

It is one of the objects of the present work with the Double Crystal Spectrometer to get further evidence about the breadth of the modified radiation.

By means of this newly developed method of spectroscopy utilizing bi-crystalline reflection, (the Double Crystal Spectrometer(15))

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(13) Appendix p. 27

(14) DuMond and Kirkpatrick, Phys. Rev. 37, No. 2, Jan 15, 1931.

(15) Ross, p. 29, Siegbahn "Spectroscopy of X-Rays".

(15) Davis, Proc. Natl. Acad. of Sci., 13, 419, 1927 (June). Phys, Rev., 17, 608, 1921; Phys. Rev., 27, 18, 1926.

(15) DuMond and Hoyt, Phys, Rev. 36, Dec. 15, 1930.

(15) Allison and Williams, Phys. Rev., 35, 149, 1930.

one can study the Compton effect to advantage for large scattering angles. The double crystal spectrometer has very high resolution (if the crystals approach perfection) and gives a very fine grained study of the spectrum. Some investigators[16] using the double crystal spectrometer have reported fine structure in the modified scattered spectrum. Some investigators find indication of separation of the two components of Mo K $\alpha_{1,2}$ scattered from carbon[17]; and Ross[18] (using a single crystal spectrographic method) published a photograph which seemed to show the separation of the K α doublet. All of these reported experiments are tested experimentally, as sort of a by product of the present study.

X-RAY SCATTERING

When a beam of x-rays strikes matter a complicated set of corpuscular and radiation phenomena occur. These phenomena may be listed as -

Corpuscular	Radiation
Photo electrons (if $\nu > \nu_k$)	Fluorescent X-Radiation (if $\nu > \nu_k$)
. . . . .	Transmitted x-ray beam
Recoil atom	Unmodified scattered x-rays
Recoil electron	Modified scattered x-rays

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(16) Davis and Mitchell, Phys. Rev., 32, 331, 1928.
(17) Bearden, Phys. Rev., 36, 791, 1930.
(17) Gingrich, Phys. Rev., 36, 1050, 1930.
(18) Ross, Proc. Nat'l. Acad., 10, 304, 1924.

where ν is the incident frequency and ν_1 is the excitation frequency of any level in an atom of the material which the x-rays strike. The scattered x-rays are the part of this interaction which is of particular interest for us.

The unmodified scattering from different atoms is coherent (both with itself and the primary beam) since this is the type of scattering that gives Bragg (reflection) diffraction from crystal lattices where there is a regular spacing of atomic planes and hence an associated constant phase relation between the parts of the same x-ray wave train diffracted from the different atomic planes in the crystal. However, the modified scattering from different atoms is incoherent with the primary beam as shown by the fact that, in the case of diffraction by a crystal, only the initial wavelengths and no modified wavelengths appear in the process of Bragg reflection.

The theory of modified scattering as given by Compton deals with "free" or loosely bound electrons, hence one would expect to observe a greater intensity for the modified radiation where the binding energy of the electrons is small (that is for elements of small atomic number). Conversely where the binding energy of the electrons is large the modified scattering would be small and most of the scattering would be unmodified. Or one can say that the condition necessary to obtain modified scattering is that the energy, $h\nu$, of the primary radiation must be large compared with the binding energy of the scattering electrons and, further, for very short waves such as the γ rays from radium one would expect all the scattered radia-

tion to be modified. These qualitative predictions are very beautifully shown in the work of Compton and Woo[19]. The effect of absorption also bears on the matter in that the absorption must be small if the atoms inside the scatterer are to be effective in giving scattered intensity. To get the maximum scattered intensity one must make use of the scattering from the maximum number of electrons. Hence, since when the atomic number is small, the absorption is also small, one can get the effect of scattering by a considerable volume of scatterer; while for material of high atomic number the scattering is due to a relatively superficial layer of atoms.

A further consideration of importance in getting experimental data on scattering phenomena is the great loss of intensity inherent in the scattering process. In fact in the process of scattering only about 1/1000 of the initial intensity survives, hence in studying scattering phenomena one must arrange for a very intense source of x-rays. This can be done by using higher excitation voltages, higher currents, and putting the scatterer close to the focal spot. The voltage can not be too high or the continuous spectrum of the half wavelength is reflected in the second order and produces background in the region of interest. The practical limit on voltage is about 3 times the excitation voltage for the spectral line used. The only limits on the current arise in the tube and power supply and sometimes the cooling system of the target can only dissipate a limited quantity of heat. Also one can not gain without limit by reducing

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(19) Compton and Woo. "X-Rays and Electrons", p. 268.

the distance from the scattering block to the focal spot, because as this distance is decreased the inhomogeneity of scattering angle increases and gives a false broadening which masks the real broadening which is of course the very effect of interest for this experiment.

APPARATUS

Target Material. To have something definite to measure it is usual to study some intense characteristic spectral line. Mo $K\alpha_{1,2}$ is used in this experimental work (Silver $K\alpha_{1,2}$ is also frequently used). The essential requirements for the spectral line radiation are: That it must penetrate the scatterer well, reflect at reasonable angles from the crystals of the spectrometer, not suffer too much loss by absorption in air, x-ray tube windows, and ionization chamber window, and must be quite totally absorbed in a gas filled ionization chamber of reasonable length. (Some heavy gas such as Methyl Bromide, Ethyl Bromide, Sulfur Dioxide or Methyl Iodide is usually used.) Mo $K\alpha$ radiation fulfills all these requirements quite well. For silver radiation one would have to make the ionization chamber longer than for Molybdenum radiation.

Tube and Scatterer. A specially designed metal x-ray tube was used at 50 KV and 10 to 15 MA to excite a graphite scattering block placed so that its front face was 5/8" from the focal spot and outside of the x-ray tube vacuum. The x-ray tube was fitted with two very thin (.003") aluminum windows. Both the filament and target could be

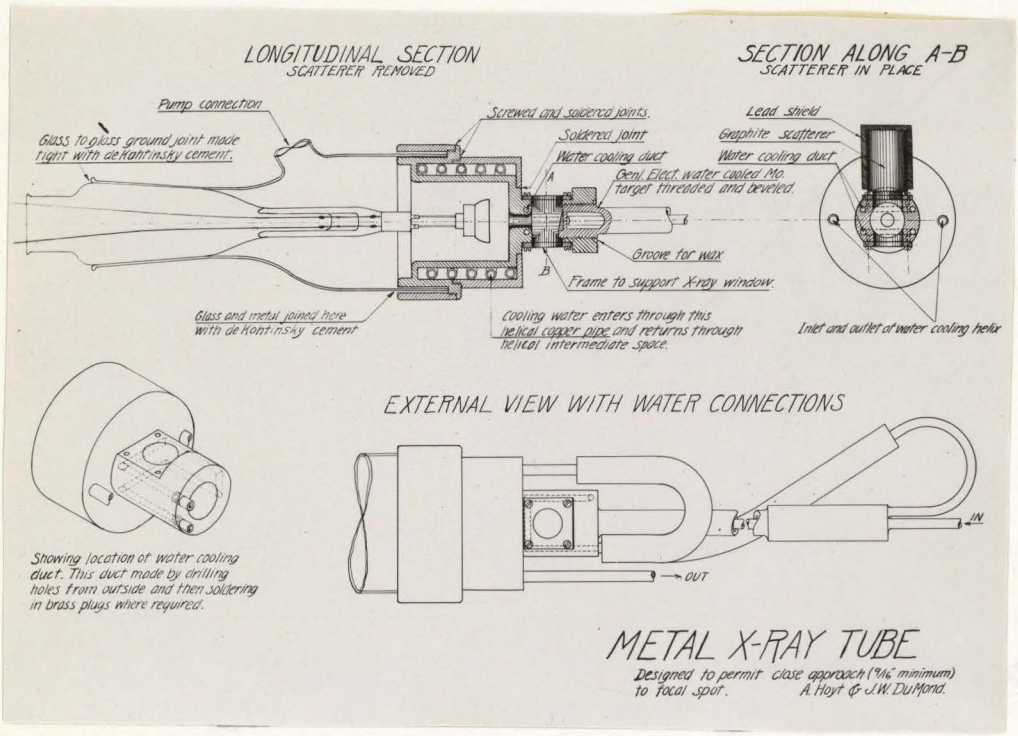

Fig. 1. Metal x-ray tube and scatterer used to study the Compton effect at large scattering angles. Spectral curves, fig. 12 and 13, were obtained using this tube.

easily removed and the target and all the wax joints were water cooled. The tube was operated on the pumps and DeKohtinsky high temperature wax was used to seal the various parts of the tube together. One will have no trouble in operating an x-ray tube "on the pumps" as long as two conditions are fulfilled, (1) Large size vacuum connections to the mercury diffusion pumps. (2) A high speed pumping system. (The size of the vacuum connection is a more important factor than the absence of bends). The scatterer used was a cylinder of Acheson Graphite 1" in diameter and 2" high. It was inclosed in a close fitting lead box to prevent leakage of x-rays through the scatterer into the room. A diagram of the x-ray tube and scatterer is shown in Figure 1.

The x-rays which get to the spectrograph must have traversed the thin aluminum windows 3 times. The loss by absorption entailed in this triple transit amounts to 30% for the wavelength of Molybdenum K α_1 . The remaining 70% was found sufficient for measurement.

Attempts were made to get results with two commercial x-ray tubes but were unsuccessful because of absorption in the glass of the tubes and because of the great distance from the focal spot to the scatterer. In three experimental tubes which had a large scattering block inside the vacuum, difficulty was encountered because it was not possible to properly bake out the scattering materials (Carbon and Beryllium) and the pumping system was not quite fast enough to handle the gas that was given off. This condition made the experimental curves so jagged they could not be interpreted with certainty. All of the tubes made could be operated with moderate power but in

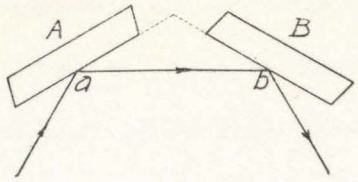

Fig. 3. Only the symmetrical ray is reflected.

A and B are Calcite Crystals.
ab is a symmetrical ray.

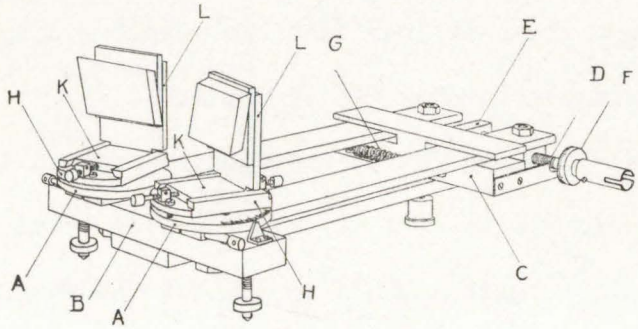

Fig. 2. Double Crystal Spectrometer, providing equal and opposite rotation for both crystals with respect to the line joining the pivot centers.

B and C are part of the bed block.
A and A are the rotating tables on which crystals are mounted.
Crystal Mountings L, K slide, perpendicular to the rotation axes, in the grooves H.
G is a spring to take up any backlash.
E is a thrust bearing for the right and left hand screw D.
F is a drum calibrated in 10" divisions with 1' of arc corresponding to a complete revolution.

studying scattered radiation x-ray tubes must be run very severely in order to make the scattered intensity sufficient to measure.

Spectrometer. The special design of Double Crystal spectrometer used is shown in perspective in figure 2. When used as an instrument for analysing spectra the instrument is used in the anti-parallel position. ((n,n) in the notation of Allison and Williams[20])

The nature of bi-crystalline reflection is such that only the symmetrical ray is reflected. The Bragg angle which determines the wavelength reflected is 1/2 the supplement of the angle between the crystal faces. (See Fig. 3)

If we neglect the phenomena of "vertical divergence" or cross-fire for the moment, since only the symmetrical ray is reflected, it follows that the radiation that gets through the spectrometer is both very nearly parallel and very nearly monochromatic. This very high resolution obtained is a resolution in direction, not a resolution in space. And further, since the emergent beam is parallel and has suffered two successive crystalline reflections, its cross-section is equivalent to the projected area of the last reflecting crystal face (1/4" x 1 1/4" approx.). Hence from an extended x-ray source such as a scattering body, one can and does gain intensity by using large crystals in the spectrometer. There is nothing about the spectrometer which requires fine slits. All that is necessary is a judicious disposition of large stops, edges, and baffles.

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(20) Allison and Williams. Phys. Rev. 35, 149. 1930.

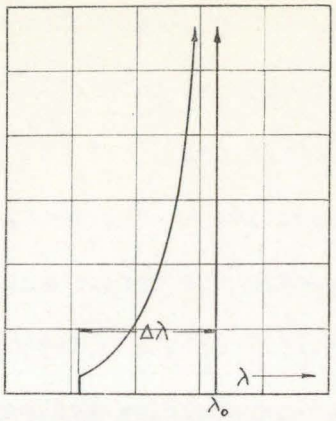

Fig. 4. Spectral Intensity Distribution selected by the geometry of bicrystalline reflection.

The equation of the curve, calculated on the assumption that the x-ray intensity is uniformly distributed over the permitted range of vertical divergence, is: -

$$y = \frac{k}{\left[2\lambda_0(\lambda_0 - \lambda_\alpha)\right]^{\frac{1}{2}}}$$

where

$$\lambda_0\left(1 - \frac{\alpha^2}{2}\right) < \lambda_\alpha < \lambda_0$$

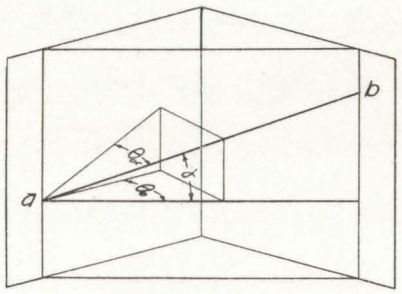

Fig. 5. Geometrical dependence of glancing angle on the vertical divergence, α.

The Spectrometer as a Band Pass Filter. The "filter band" which the spectrometer passes (neglecting crossfire) is a very narrow wavelength band with an infinite ordinate. When the effect of "crossfire" is considered the band broadens on the short wavelength side, the ordinate falling off approximately as a parabola and terminating abruptly at the limit of the "vertical divergence". As a function of wavelength this is shown in Figure 4. The width (at the base) of this "filter band" can be got as a function of the angle of cross fire in several ways. It can be found quite generally as in a publication by Schwarzschild[21] or in a simple way as outlined below[22]. In Figure 5 the geometry of the bicrystalline reflection is shown. θ_0 is the maximum Bragg angle, θ_α is the minimum Bragg angle due to "cross fire" of a vertical divergence angle α.

From Figure 5 we have

$$\sin\theta_\alpha = \sin\theta_0 \cos\alpha$$

but $$\frac{n\lambda}{2d} = \sin\theta$$

this gives $$\lambda_\alpha = \lambda_0 \cos\alpha$$

and $$\Delta\lambda \equiv (\lambda_0 - \lambda_\alpha) = \lambda_0(1-\cos\alpha) = 2\lambda_0 \sin^2\frac{\alpha}{2} \cong 2\lambda_0\frac{\alpha^2}{4}$$

or $$\Delta\lambda \cong \frac{1}{2}\lambda_0\alpha^2$$

which gives the width with good approximation ($< 1\%$) as long as α is less than 10° of arc.

This "filter band" is displaced bodily to longer wavelengths as the angle between the crystals is increased. When this band passes over a sharp spectral line the shape of the line obtained is roughly

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(21) Schwarzschild, Phys. Rev. 32, Aug. 1928.
(22) DuMond and Hoyt, Phys. Rev. 36, Dec. 15, 1930.

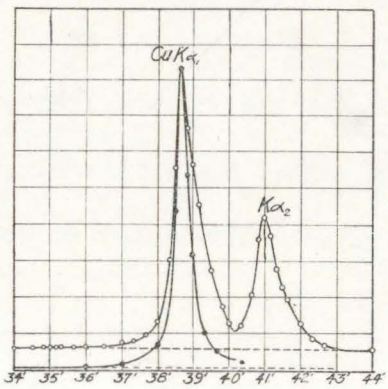

Fig. 6. Showing the dependence of the shape of the spectral line on the vertical divergence. (Abscissae are glancing angles from Calcite in minutes of arc + a constant.)

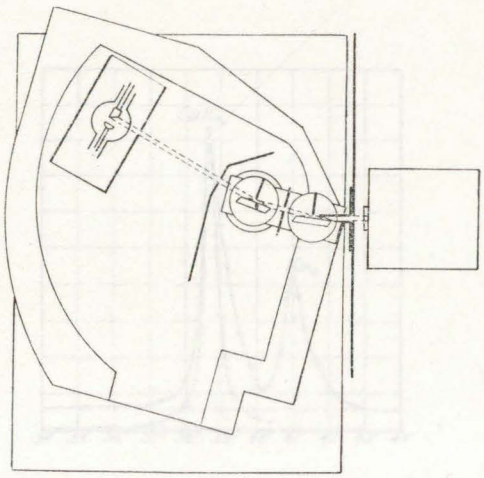

Fig. 7. Showing the distribution of x-ray shielding.

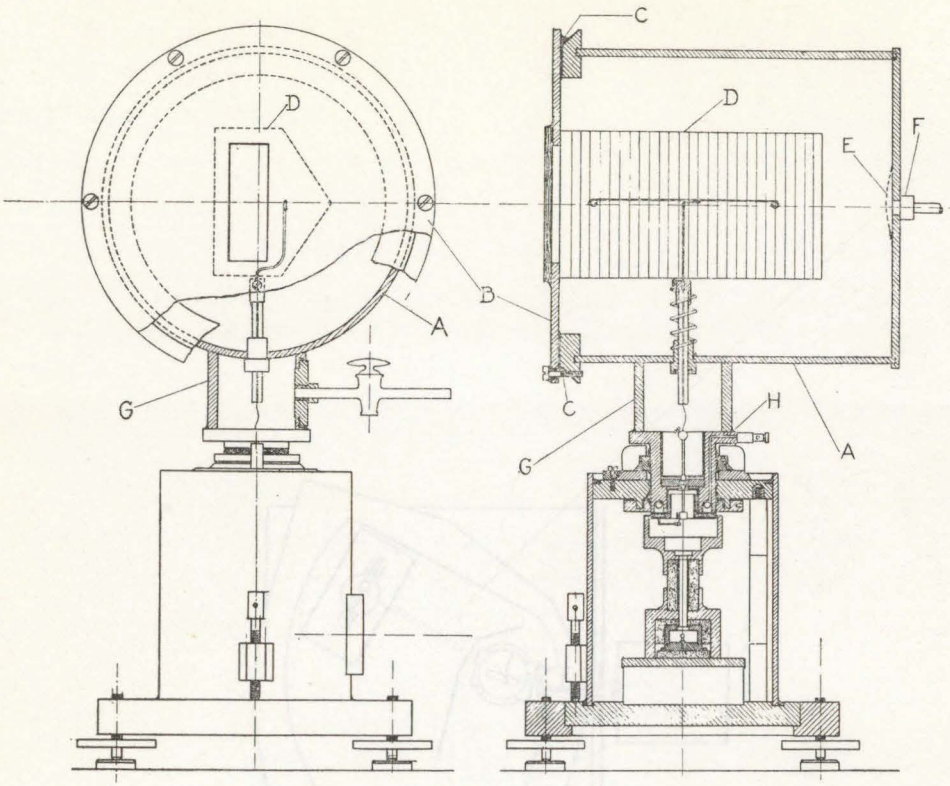

Fig. 8. Complete Cross Section of Ionization Chamber and Hoffman Vacuum Electrometer.

A - Ion chamber
B - Insulated front plate supporting Internal Grid
C - Wax seal and insulation gasket
D - Internal grid at +120 volts
E - Fine meshed nickel screen over side tube connection
F - Bushing connection to desicator, manometer, and vacuum pump.
G - Evacuated shielding sleeve
H - Wax seal to electrometer

the reverse of the shape of the filter band which the spectrometer transmits. If the vertical divergence is limited with suitable stops the width $\Delta\lambda$ of the filter band can be made as narrow as we please so that the spectral lines obtained experimentally will become symmetrical. This is shown in Fig. 6.

To insure that all the radiation which actually gets to the ionization chamber has suffered two successive crystal reflections all other possible paths which radiation might take are carefully blocked off by stops, baffles, edges, shields, snouts, etc., all made of lead or leaded rubber. The x-ray tube itself is carefully wrapped up in leaded rubber in case any x-ray leakage appears to come through the sides and ends of the tube. Figure 7 shows the distribution of x-ray shielding in a particular case.

The details of the initial adjustment of the spectrometer are described quite fully in the literature (23).

Ionization Chamber and Electrometer - Since the monochromatic beam of radiation passed by the spectrometer has a large extended cross section it is necessary to measure this x-ray intensity by means of an ionization chamber. The complete assembly of the ionization chamber and electrometer is shown in Figure 8. The ion chamber is filled with dry Methyl Bromide gas at a pressure of one atmosphere. This gas is used because it is a gas, not a saturated vapor, so that it can surely be kept dry with a phosphorous pentoxide desicator..

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(23) DuMond and Hoyt, Phys. Rev. 36, Dec. 1930.

A further reason for using Methyl Bromide is that the K absorption edge of Bromine is near and on the long wavelength side of the spectral region of interest. This gas will absorb 97% of (λ = 708 X.U.) the x-rays used in 17 cm of path at 1 atmosphere pressure. (To accomplish the same result with an air filled chamber would require one 24 meters long!) The ionization chamber has a large paraffin impregnated balsa wood window (2" x $3\frac{1}{2}$") stuck onto the front brass plate with beeswax and coated on the inside with a very thin (.0002") sheet of Berylium Aluminum Alloy to render it conducting. It is extremely important that the whole interior of the ion chamber be conducting so that the sensitive element will be electrically shielded, since stray varying charges collect on the glass side tubes and induce other varying charges on their ends which project into the inside of the ion chamber. Internal nickel screens were spot welded over all the openings in the ion chamber that connected to such things outside as pumps, manometer, desicator, etc. Inside the large shell of the ion chamber was placed a grid of very fine wires which defined the volume from which the ions were collected. The main purpose of this was to cut down the effect of alpha particles from the walls of the ion chamber (so as to reduce the natural leak) by cutting down on the actual wall area. The distance from the outside wall to the internal grid was greater than the α particle range in the gas. The collector was set to one side of the chamber

so as not to be in the main x-ray beam. The grid was held at +120 volts so that the positive ions were collected. The ionization chamber was always used at saturation current because experiment showed that saturation first occurred at about 45 volts. The ion chamber was connected directly to the electrometer by an evacuated shield, and everything near the ion chamber and electrometer was carefully grounded to a shielded ground wire of large size.

A Hoffmann vacuum, binant, Electrometer(24) was used. The capacity of the electrometer was measured, with the aid of an auxiliary known capacity, to be 3 e.s.u. When the ion chamber was added the capacity of the insulated system was again measured and found to be about 12 e.s.u. The small capacity, and extremely good insulation of the electrometer makes the instrument very sensitive to small charges. The sensitivity to charge is quite unrelated to the voltage sensitivity, since the electrometer needle is insulated while it is deflecting; and as charges accumulate, the needle deflects more and more over one of the permanently charged "binants" so that the charges on the needle are continually redistributing themselves. During this redistribution it is difficult to say what the potential does, while in measuring the voltage sensitivity the needle is always connected to a battery so that the potential is held constant. In case the charge redistribution can be

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(24) Hoffmann, Ann. der Physik. 52.7 p. 665, Aug. 16, 1917.

neglected (which of course it can't) the capacity would be the factor which would relate the voltage sensitivity to the charge sensitivity. However, in this particular research the voltage sensitivity was about 700 mm/volt. This sensitivity could be increased by introducing a weaker suspension*. In the set up as used it was found possible to detect 10^{-16} amps. and hence to measure 10^{-14} amps. with a precision of 1% in a single reading of a time duration approximately 2 minutes. The Hoffmann Electrometer was found to be an extremely satisfactory instrument with a uniform scale and a stable zero. Sometimes there is difficulty with the initial adjustment but once set up it requires no attention over long periods of time (1 - 2 yrs.). The ionization currents of course are measured in terms of the rate of drift of the electrometer needle, care being taken to eliminate any effects due to the natural period of the suspension.

The suspension used has a free mechanical period of oscillation of 7 sec. A period of say 30 sec or more would be desirable in case the electrometer were to be used to collect negatives or positives in a vacuum. (Such a suspension would be critically damped at a few mm pressure in the electrometer.) Such a suspension is possible but in the present case is out of place because the natural leak due to cosmic rays, etc., makes a large limiting background leak in the ion chamber which it is not easy to eliminate completely. In particular, if residual radiation makes 10 ions/cc/sec. in a chamber of 10^3 cc, this is a current of $4.77 \times 10^{-10} \times 10^4 \times \frac{1}{3} \times 10^{-9} = 1.6 \times 10^{-15}$ amps. for a basic background leak in a 1 liter ion chamber. This could be made smaller by a factor of 10 by reducing the volume of the chamber. It can be balanced out by such things as a photo electric leak, etc., etc.; but these devices usually increase the capacity of the system.

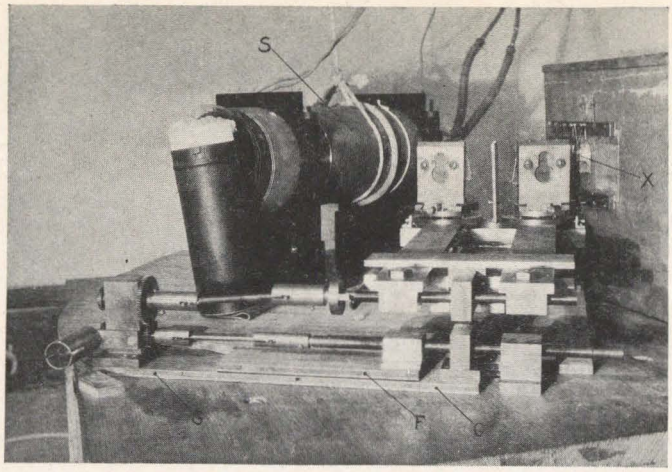


Fig. 9. Showing Gear Mechanism for turning crystals, tube, and spectrometer at the proper speeds.

G and F - false table tops
X - lead snout, S - leaded rubber shield

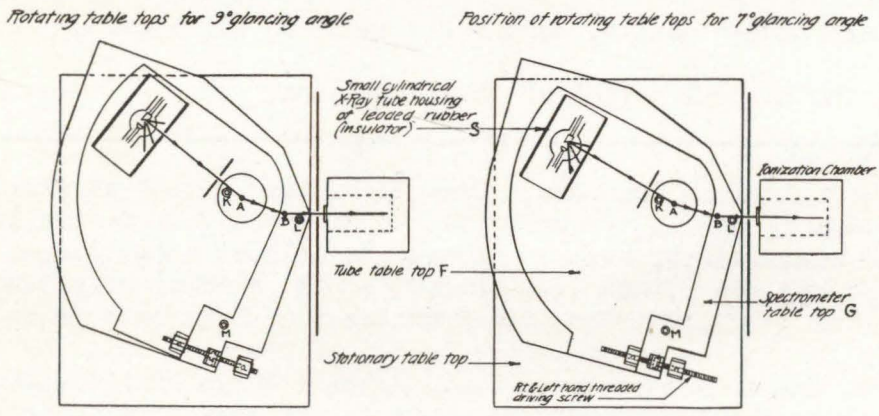


Fig. 10. Schematic diagram of the table tops.

Table top F turns about large pivot A.
Table top G turns about small pivot B.
Spectrometer leveling screws rest in base plates at K, L, and M on G.

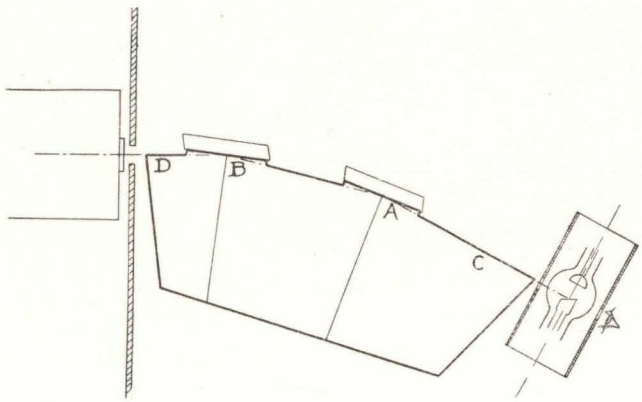


Fig. 11. Showing method of aligning x-ray source.

CABD is the path of the specular radiation.

Miscellaneous adjustments. The crystals were turned equally and in opposite directions with respect to the spectrometer frame by means of a right and left hand screw and a thrust bearing fixed to the frame. A similar mechanism geared to the spectrometer screw by a 2:1 gear and universal joints kept the slits, tubing, ionization chamber and spectrometer always in line as the glancing angle was varied. This mechanism is shown in Fig. 9, which is a picture of an actual set up. The way that the mechanism works in conjunction with the two false rotatable table tops is shown in Fig. 10. The reason for this somewhat elaborate mechanism is that, in order to reduce the capacity of the detecting system, the ionization chamber was mounted directly on the electrometer. Obviously it was not convenient to move the electrometer and so all the rest of the set up had to be moved (at proper speeds) to eliminate the following sources of uncertainty -

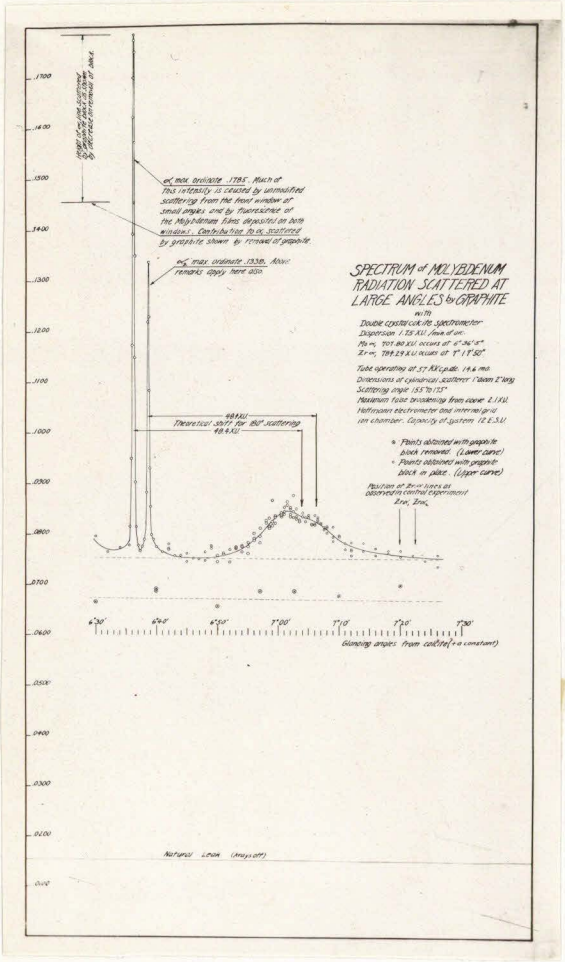
(1) X-ray beam walking across the crystal face as the angle was changed and hence being reflected from a different part of the crystal at different times.

(2) X-ray beam walking across the ion chamber window (which might conceivably vary in transmission).

(3) X-ray beam walking across the focal spot of the x-ray tube while the spectrum was being explored.

In the relatively rough alignment required to place the focal spot or scattering source, the angles were set approximately by means of a bristol board template. The process is shown in Figure 11.

Direct radiation was used at first to locate the spectral line



Fig, 13. Spectrum Mo Kα Scattered from Graphite.

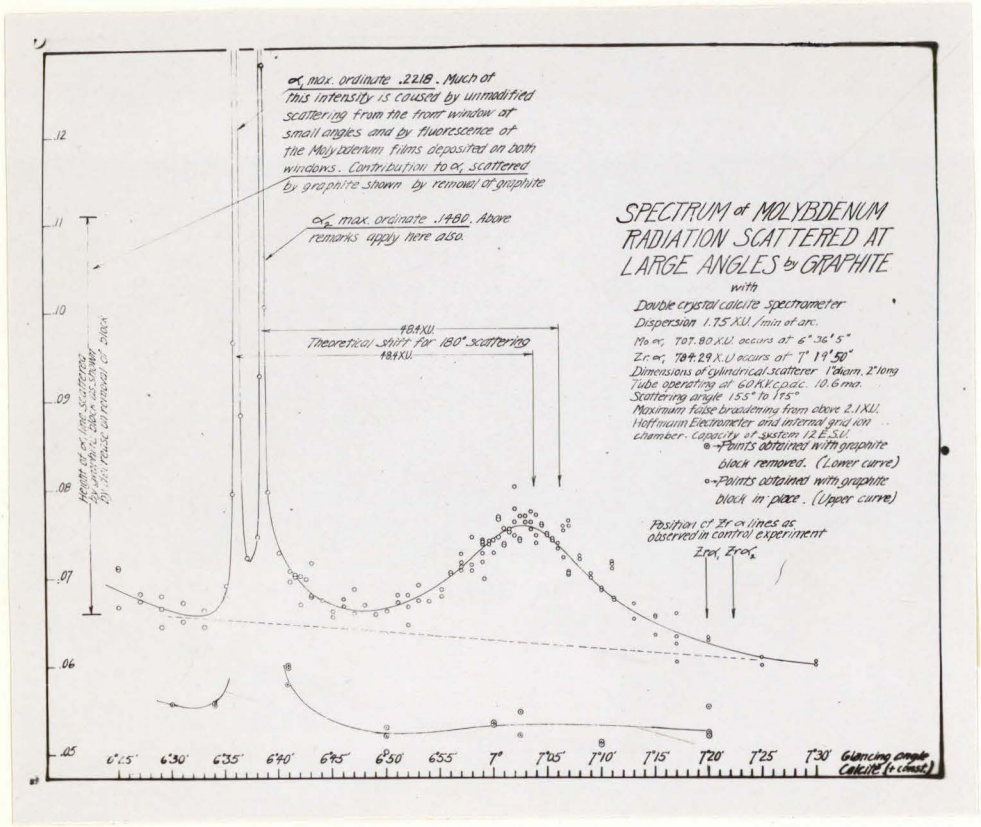


Fig. 12. Spectrum, Mo K$\alpha_{1,2}$ Scattered from Graphite.

and make sure that the spectrograph was in adjustment. This verified, the primary source was replaced by a scattering source.

The dispersion of the spectrometer was measured as 1.75 U.X. per minute of arc. This agrees with the value published by Sieg\bahn[25]. The method used to obtain this dispersion was to calculate it from the observed positions of Mo K$\alpha_{1,2}$ (both direct and scattered) and of Zr. K $\alpha_{1,2}$(Fluorescent) and the known wavelengths of these lines. The agreement of the dispersion of the spectrometer with the known dispersion for calcite is a good check of the accuracy of the calibrated angle scale marked on the spectrometer screw drum.

EXPERIMENTAL RESULTS

The spectral curves shown in Figures 12 and 13 represent the scattered spectrum of Molybdenum Kα_1 and α_2 in the first order. Acheson Graphite was used as a scatterer and the scattering angle was 155° to 175°. The x-ray tube was operated at about 60 KV c.p.d.c. and 10 MA. The time required to obtain one of these curves was from 6 - 8 hours. The work had to be done at night when the transient demands on the power line were at a minimum. The ordinates represent x-ray intensity on an arbitrary scale and the abscissae represent the glancing angle with the calcite crystal in minutes of arc (which is nearly proportional to wavelength).

In obtaining the spectral curves shown several readings were

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(25) Sieg\bahn, Spectroscopy of X-Rays, p. 251.

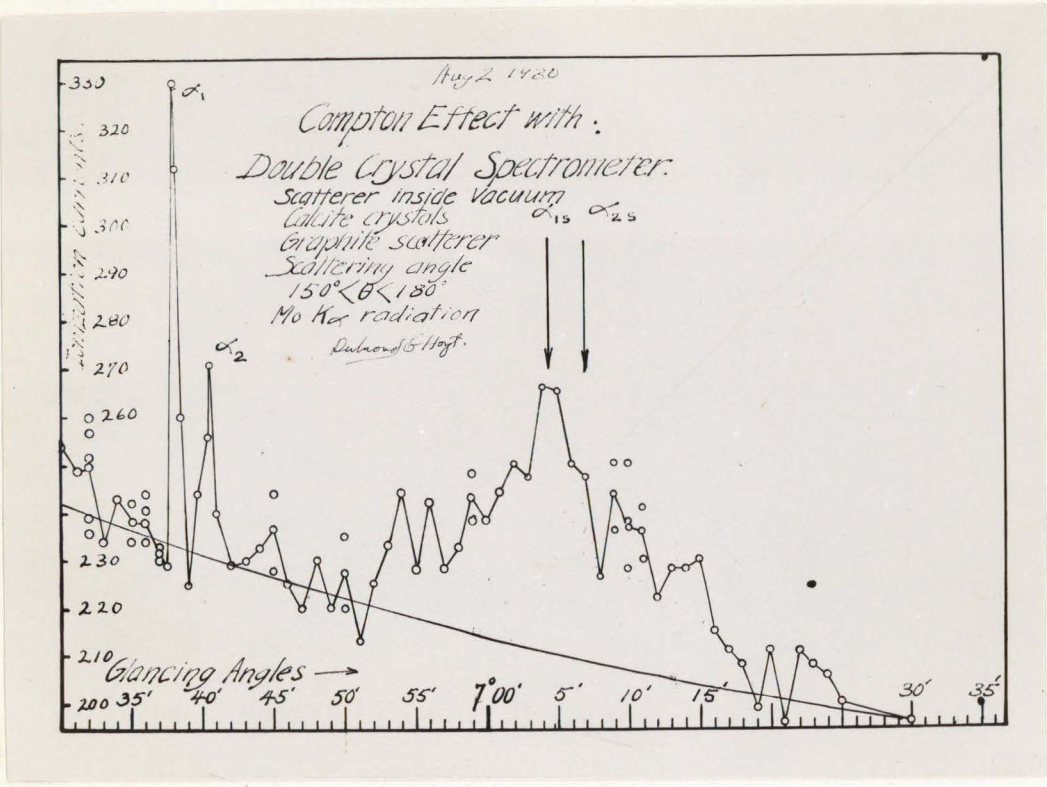

Fig. 14. Spectrum Mo K Scattered from Graphite.

The best curve from a group of partial failures. X-ray tube used to obtain this curve had a large scattering block of graphite in the vacuum.

taken at each angle setting of the spectrometer crystals. This method of observation strikes an average at the time the observations are made and gives a very definite idea of the reliability of the data. In the curves plotted all the observations are plotted and no averages are taken. The data is apparently reproducible to about 1/6 of the quantity of interest and the average would be somewhat better than this.

As a check to make sure that the spectral curve obtained was really produced by the presence of the scatterer, the scatterer alone was removed (absolutely everything else about the set up was left unchanged) and further points were taken throughout this spectral region and the part of the curve which is due to carbon scattering was hence definitely established. The background in the check run (scatterer removed) was due to amorphous background which arises from fluorescence in the crystals, scattering in the crystals, direct leakage, scattering from stops and windows, natural leak of the electrometer (residual rays), fluorescence from Molybdenum films sputtered on the windows, and air scattering.

As an example of a series of curves that were too jagged to interpret with sufficient accuracy we have Fig. 14. This curve was obtained using a special x-ray tube which had the scatterer in the vacuum. Of course in the light of more recent work some weight can be ascribed to the breadth of the modified line shown in Fig. 14.

INTERPRETATION OF EXPERIMENTAL RESULTS

In analysing the experimental curves more closely one must consid-

er several things. Plotting a continuous spectral curve of a group of readings extending over 6 hours time, presupposes a constant initial x-ray intensity over the whole time of operation. To obtain this, manual adjustment of the current and voltage was used. With an open scale milliameter and a telescope the x-ray tube current was easily held to .1 ma in a total current measured of 10 ma or more; hence the tube current was held steady to 1%. The dependence of x-ray line intensity on voltage goes as the 3/2 power of the voltage (instead of as the 1st power, in the case of the current). The total energy of the continuous spectrum varies as the square of the voltage and therefore the background ordinate due to an element of continuous spectrum $(\Delta\lambda)_b$ would be proportional to the 1st power. So the voltage applied to the x-ray tube is the more essential thing to hold constant. Considerable difficulty arose with this factor because all sources of A. C. power available showed 2% variation during the day. The simplest and cheapest solution was to work at night when the transient demands on the power line were a minimum. It was found that at night from 12 to 6 A.M. the voltage could be maintained constant to 2/3% of the chosen value. These sources of uncertainty are something under 3%.

The resolution of the spectrometer affects the shape of the experimental curve especially in the neighborhood of sharp peaks. The resolution of the spectrometer is of the order of

$$\text{Resolution} = \frac{\Delta\nu}{\nu} = \frac{1}{1500}$$

where $\Delta\nu$ is the width of a spectral line at half maximum. This reso-

lution is much better than that of the usual x-ray spectrograph so no corrections need be applied to the experimental curves.

In the modified spectrum there enters a false broadening due to the inhomogeneity of scattering angle causing a spurious uncertainty in shift. The effect of this inhomogeneity in scattering angle on the shift is a minimum at 0 and 180 degrees as can be seen from the derivative of Compton's equation. It is not convenient to work near 0° scattering as there is no way to separate the direct transmitted primary beam from the scattered beamsand besides there is no shift (and no breadth is expected). If the scattering occurs near 180° the experimental arrangement can be very nicely effected. In the present experiment the inhomogeneity was caused by a variation in scattering angle from $\phi_1 = 155^\circ$ to $\phi_2 = 175^\circ$ for the extreme possible limits. The maximum possible false broadening that this can give is

$$\Delta\lambda_{\phi_1} - \Delta\lambda_{\phi_2} = \frac{h}{m_o c}(1 - \cos\phi_1 - 1 + \cos\phi_2) = \frac{h}{m_o c}(\cos\phi_2 - \cos\phi)$$

$$= 24.2\ (.9962 - .9062) = 2.3 \text{ X.U.} \quad \text{false broadening.}$$

This is roughly half of the separation of the Kα doublet and should not conceal any separation of this doublet in the modified region if it should exist. As a matter of fact the effective false broadening is apt to be somewhat less than this maximum false broadening calculated, since the scattering from the extreme limits of the scattering block is weaker because of the geometrical restrictions of the set up. An estimate of of the probable effective false broadening is about 1.5 X.U. and this quantity can be subtracted from the observed breadth of the modified line.

It would appear desirable to reduce this inhomogeneity of scatter-

ing angle; but the consequence of reducing this is a loss of intensity of the scattered radiation to be measured. Since the modified scattered intensity is already rather small compared with background intensity, it is not wise to reduce this scattering angle inhomogeneity too far.

In getting useful experimental data contrast is the all important requisite. In this case it is necessary to make the spectral lines stand out by themselves as much as possible. This means it is necessary to reduce the background. Although the spectral lines stand on top of whatever background there is, each reading of the ionization current includes the background as well as the line. Hence it is obvious that a weak line would be lost against a large background since it would show in the ionization currents observed as small fluctuation in a large quantity. The causes of background are -

(1) Continuous spectrum

(2) Natural leak of electrometer and ion chamber

(3) Amorphous or non-selective crystal scattering and fluorescence

(4) X-ray leakage in which x-rays reach the chamber without suffering bicrystalline reflection in the prescribed orders.

The methods of eliminating the background due to these causes are -

(1) Regulation of the vertical divergence

(2) Internal grid, crass volume of ion chamber, dry shielded insulators, evacuated lead connections

(3) Selective filtration of the x-ray beam so as to cut out the wavelengths which are harder than the region being studied. (For Mo Kα a Zr filter of the proper thickness is used. For Cu Kα a Ni filter of the proper thickness is used.)

(4) Lead shields, stops, knife edges, and, most important of all, a snout at the ion chamber window.

In this particular experiment it was thought best not to use a Zr filter as the fluorescent zirconium α lines come rather close to the shifted scattered Mo α lines and thus might distort the background in an undesirable way. With judicious shielding it was possible to reduce the background until it was about 4 times the heigth of the modified line for one of the best curves, and 7 times this for the other. As can be seen from Figures 12 and 13 this is not too disparate.

Another source of uncertainty in background arises in the faintness of the radiation measured. The ionization currents measured were of the order of 10 times the ionization due to cosmic rays (in the volume of the ion chamber). The natural leak of the electrometer is about twice the effect of cosmic rays alone, the excess being due to γ rays from radioactive substances in the earth's crust, and α and β particles from the walls of the ion chamber. The wall effect has been reduced by a special chamber design (internal grid), but there is an uncertainty of about 3% in the measurement of the remainder of the natural leak. This would be an uncer-

tainty of 1% in the total ionization current usually measured in a spectral curve. This uncertainty is unavoidable and must be reckoned with the other uncertainties. Altogether the various uncertainties add up to about 4% and to do better than this one has to take several readings at each crystal setting of the spectrometer and hope that the average will be about twice as good as the individual observations. Referring to the curves one can see that 4% is the order of the observational uncertainties.

In the special design of tube used there was unmodified scattering from the thin aluminum windows and Molybdenum fluorescent radiation from a sputtered film on the windows. These united with the true unmodified scattering from the graphite to rear up excessively high peaks at the position of the unmodified line. Since there were readings taken with the scatterer removed, this can be corrected for. To determine what proportion of the actual peak is unmodified scattering from carbon, when the crystals were set to reflect the peak of K α_1 the scatterer alone was removed without disturbing the crystals. The amount by which the peak was reduced ~~then~~, represents how much of the observed peak in the region of unmodified scattering was due to the presence of the graphite scatterer. This quantity is shown in Figures 12 and 13.

CONCLUSIONS

The order of reliability of the data is apparent in the dispersion of points plotted. In case the average points were used it

is obvious that the curves would pass quite smoothly thru these points. However, even by drawing the most unfavorable curve thru the experimental points shown, the following conclusions will still remain:

(1) The Compton Modified line (Mo $K\alpha_{1,2}$) scattered from graphite at 155-175° scattering angle is very broad, of the order of 21X.U. at half maximum.

(2) There is no discernible separation of the two components α_1, α_2 in the modified spectrum since the lines are so broadened by the scattering at large angles.

(3) No fine structure exists in the scattered spectrum of intensity $> 1/5$ the modified radiation.

(4) The ratio of intensity of modified to unmodified radiation can be determined by measuring the proper areas under the curves. This ratio is about 7.3:1

(5) Viewed in the light of the Theory of DuMond [26] for the dependence of the modified line breadth on scattering angle, the great breadth of the modified line observed can be interpreted as experimental evidence for a dynamic atom.

(6) There appears to be no essential difference in the results obtained with the single and double crystal methods of studying the spectrum.

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(26) Simplified theory in Appendix,--- Complete theory, Phys. Rev. 37 Jan. 15,1931.

AGREEMENT WITH OTHER WORKERS

The work reported here is in complete agreement with the work of DuMond and Kirkpatrick[27] using the Multi-crystal Spectrograph. It is in qualitative agreement with the work of Bearden[28]. It is in disagreement with the work reported by Davis[29] and Collaborators*. It is also in disagreement with the work of Gingrich.[30]

Further work can be done with the double crystal spectrometer in this field in attempting to study the scattering from other scattering materials such as Berylium, Aluminum, Lithium, and Paraffine; and other wave lengths than Mo K α can be used. However the studies will be limited to the region near 180° scattering angle, so perhaps this work can be done more thoroughly with the multicrystal spectrograph as it is not restricted in this way.

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* In a letter from Davis received by DuMond recently, Davis states that they have been unable to verify their published experiments.

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(27) DuMond and Kirkpatrick, Phys. Rev. 37, Jan. 15, 1931.
(28) Bearden, Phys. Rev. 791, 1930.
(29) Davis, Phys. Rev. 32, 331. 1928.
(30) Gingrich, Phys. Rev. 36, 1050, 1930.

APPENDIX

Simplified derivation of the dependance of the breadth* of the Compton line on scattering angle.

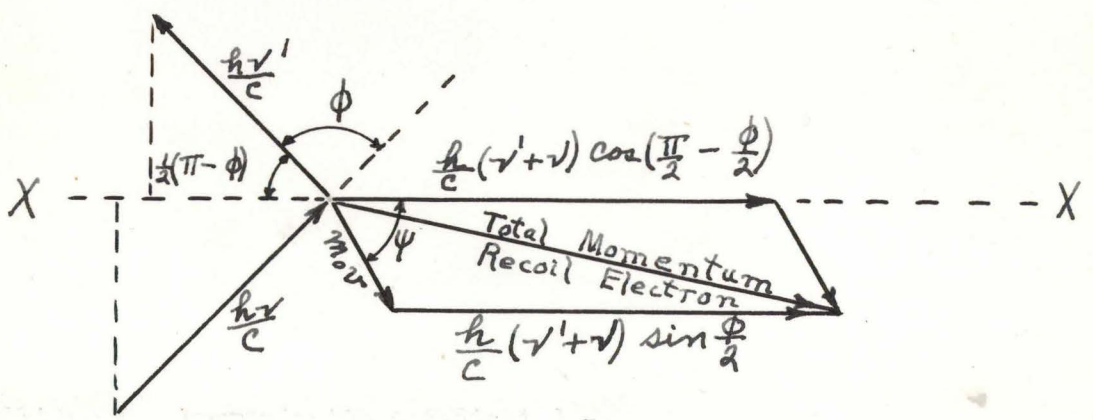
Ross[31] suggested that the breadth of the Compton line was a sort of a Doppler effect in which the randomly oriented velocities of the scattering electrons caused the broadening. If one chooses a symmetric set of coordinates as indicated in the figure the problem of representing the interaction analytically takes on a certain complete axial symmetry and hence can be treated as a problem in a plane without loss of generality. ν is the incident frequency, ν' the modified frequency, ϕ the angle of scattering, $m_0 v$ the initial electron momentum, and ψ its angle of orientation with respect to the symmetric axis XX.

In this derivation the assumptions and approximations are: -

(1) Neglect relatively or $v << c$.

(2) $\left.\begin{matrix} \nu + \nu' \cong 2\nu \\ \lambda + \lambda' \cong 2\lambda \end{matrix}\right\}$ and $\lambda\lambda' \cong \lambda^2$

(For MoK$\alpha_{1,2}$ these approximations are always closer than 3%)



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* Essentially as given by DuMond (unpublished)

(31) Ross, Proc. Nat'l Acad. Sci., 9, 246, 1923.

Making the approximations indicated -

$$(1)\quad \left\{\begin{matrix}\text{Total Recoil}\\ \text{Electron Momentum}\end{matrix}\right\}^2 = (m_0 v')^2 \cong m_0^2 v^2 + \left(\frac{2h\nu}{c}\sin\frac{\phi}{2}\right)^2 + + 2m_0 v \frac{2h\nu}{c}\sin\frac{\phi}{2}\cos\psi$$

Since the initial energy of the electron is $\frac{m_0 v^2}{2}$ and the final energy can be got by multiplying (1) by $\frac{1}{2m_0}$ we have the gain in kinetic energy for the scattering electron (which is equal to the loss in energy of the radiation); as,

$$(2)\quad \left(\frac{m_0 v'^2}{2} - \frac{m_0 v^2}{2}\right) = h(\nu - \nu') \cong \frac{h^2\nu^2}{m_0 c^2} 2\sin^2\frac{\phi}{2} + \frac{v}{c} 2h\nu \sin\frac{\phi}{2}\cos\psi$$

using $\nu\lambda = c$; $\lambda\lambda' = \lambda^2$; and $\beta = \frac{v}{c}$;

$$(3)\quad hc\frac{(\lambda' - \lambda)}{\lambda^2} \cong \frac{h}{m_0\lambda^2} 2\sin^2\frac{\phi}{2} + 2\beta\frac{hc}{\lambda}\sin\frac{\phi}{2}\cos\psi$$

Multiplying (3) by $\frac{\lambda^2}{hc}$ we get,

$$(4)\quad \lambda' - \lambda \cong \frac{2h}{m_0 c}\sin^2\frac{\phi}{2} + 2\beta\left(\lambda\sin\frac{\phi}{2}\right)\cos\psi$$

which expresses the change in wavelength which occurs in scattering (with an accuracy always better than 3% in the case of $\lambda \cong 700$ X.U.). Since $0 < \psi < 180$ the amount of shift varies and is uncertain by an amount $\Delta\lambda$ where $-2\beta\left(\lambda\sin\frac{\phi}{2}\right) < \Delta\lambda < +2\beta\left(\lambda\sin\frac{\phi}{2}\right)$. and the whole uncertainty in shift is

$$(5)\quad \Delta\lambda \cong 4\beta\lambda\sin\frac{\phi}{2}$$

A more accurate theory can be found in the literature[32] which gives

$$-2\beta\lambda^* < \Delta\lambda < 2\beta\lambda^*$$

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(32) DuMond and Kirkpatrick, Phys. Rev., 37, Jan. 15, 1931.

where $2\lambda^* = (\lambda'^2 + \lambda^2 - 2\lambda\lambda' \cos\phi)^{\frac{1}{2}}$

Making the approximations $\lambda'\lambda \cong \lambda'^2 \cong \lambda^2$ we get,

$$2\lambda^* \cong \sqrt{2}\cdot\lambda(1-\cos\phi)^{\frac{1}{2}} \cong 2\lambda \sin\frac{\phi}{2}.$$

Checking the approximate uncertainty in shift obtained in (5).

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