THE SCATTERING OF X-RAYS FROM GASES

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Abstract

Intensity of the scattering of molybdenum x-rays from carbon dioxide, argon, oxygen, nitrogen, helium, and hydrogen at scattering angles from 20° to 155°.— All except hydrogen show excess scattering (with helium it may, however, be due to nitrogen present as an impurity): the excess scattering extends to larger angles from the primary beam as the atomic number of the gas increases and as the wavelength increases. This excess scattering appears to be mostly, if not wholly, due to interference arising within the atom. Destructive interference is prominent with argon, and seems probable (though it is very weak) with oxygen, carbon dioxide, and nitrogen. Mark and Schocken's results, which indicated scattering by gases follows the $(1+\cos^2\theta)$ law, are not confirmed. Hydrogen follows within experimental error the quantum theory prediction of Breit, Dirac, Gordon, and Waller. Even when the K_α rays of molybdenum are used the scattering from hydrogen does not show the irregularities predicted by Debye's interference theory.

Introduction

THE intensity of x-rays scattered in different directions by an atom has been calculated on the basis of electromagnetic theory for a number of different atomic models by Darwin,[1] Debye,[2] Schott,[3] Compton,[4] Glocker and Kaupp,[5] and Glocker.[6] Interference in the x-rays scattered from the groups of electrons postulated in these models should cause a deviation from the distribution of intensity calculated for an electron scattering independently $(1+\cos^2\theta)$. In addition to this interference arising within the atom, there may exist interference due to a regular spacing of the atoms in a molecule, and also that due to a preferred spacing of molecules among themselves. The phenomenon of excess scattering may be the result of all these causes when solids and liquids are used as the scattering material. Debye[7] has calculated that the latter cause may be eliminated by using a perfect gas as the scatterer. Therefore excess scattering from a monatomic perfect gas can be ascribed only to the structure of the atom. Jauncey[8] has obtained excess scattering in the case of the diffuse scattering from a crystal. To reduce this scattering to the scattering from a single molecule requires the use of Debye's temperature factor. Jauncey concludes from his experiments that this theoretical factor of Debye's is greatly in error. It is hardly possible

[1] C. G. Darwin, Phil. Mag. **27,** 315 (1914).
[2] P. Debye, Ann. d. Physik **46,** 809 (1915).
[3] G. A. Schott, Proc. Roy. Soc. **A96,** 395 (1920).
[4] A. H. Compton, Wash. Univ. Studies **8,** 99 (1921).
[5] R. Glocker and M. Kaupp, Ann. d. Physik **64,** 541 (1921).
[6] R. Glocker, Zeits. f. Physik **5,** 54 (1921).
[7] P. Debye, Mathematical Journal of Physics **4,** 133 (1925); also Phys. Zeits. **28,** 135 (1927).
[8] G. E. M. Jauncey, Phys. Rev. **20,** 405 (1922).

at present to determine definitely how a single atom scatters by experiments of the type Jauncey performed, while it is possible to do so by scattering from gases.

This paper is a report of an experimental determination of the distribution of x-rays scattered from hydrogen, helium, nitrogen, oxygen, argon, and carbon dioxide. In spite of numerous suggestions by Debye, Schott, and Compton that experiments of this type should give important information on the nature of atoms and radiation, the only previous attempt seems to be that of Mark and Schocken.[9] They scattered Mo $K\alpha$ rays (crystal reflected) from carbon dioxide and argon, and found very nearly a $(1+\cos^2\theta)$ distribution of intensity, or scattering function as it has been called, for both gases. No evidence was found for excess scattering from an individual atom of argon and practically none from an individual molecule of carbon dioxide. They report the scattering of carbon dioxide as slightly greater than argon from 30° to 50°, and slightly less than argon from 140° to 160°.

Because of the presence of the Compton effect, there is an increasing amount of modified radiation as the atomic weight of the scatterer decreases. Woo[10] predicts, on the basis of his experiments, the scattering from hydrogen and helium to be almost completely modified. The scattering function for these elements should therefore be that predicted by the quantum theories (unless possibly the modified ray is not truly incoherent). The older forms of quantum theory do not give a unique prediction for this function. Perhaps the most satisfactory of these is Compton's.[11] Breit,[12] on the basis of the correspondence principle; Dirac,[13] with the matrix mechanics of Heisenberg and Born; Gordon[14] and Waller[15] from the standpoint of the deBroglie-Shroedinger wave mechanics, all agree in predicting this function to be

$$I_\theta = \frac{Ine^4(1+\cos^2\theta)}{2m^2r^2c^4(1+\alpha \operatorname{vers}\theta)^3}, \qquad \alpha = \frac{h\nu}{mc^2}. \tag{1}$$

where I_θ is the intensity of the scattered rays at the angle θ from the primary beam, and at the distance r from the scatterer, which is composed of n electrons of charge e and mass m. The primary beam, of intensity I, is assumed unpolarized.

A test of the old vs. the new quantum theories was recently performed by Dr. Bearden and the author,[16] based on the fact that the latter predicts the angle at which x-rays become polarized by scattering to be 90°, while the

[9] H. Mark and K. Schocken, Naturwiss. **15,** 139 (1927).

[10] Y. H. Woo, Phys. Rev. **27,** 119 (1926).

[11] A. H. Compton, Phys. Rev. **21,** 491 (1923). (A summary of these theories is given in Compton's "X-Rays and Electrons," D. Van Nostrand Co., New York City (1926), pp. 296–305.)

[12] G. Breit, Phys. Rev. **27,** 362 (1926).

[13] P. A. M. Dirac, Proc. Roy. Soc. **A111,** 405 (1926).

[14] W. Gordon, Zeits. f. Physik **39,** 117 (1926).

[15] Ivar Waller, Phil. Mag. **4,** 1228 (1927).

[16] C. S. Barrett and J. A. Bearden, Phys. Rev. **29,** 352 (1927).

older theories give a polarizing angle dependent upon the wave-length and less than 90°. Our experiment gave 90° independent of the wave-length, within experimental error, and confirmed the theory on which Eq. (1) is based. Thus it appears that hydrogen and helium should scatter according to Eq. (1) except at small angles where some unmodified scattering may occur. With heavier gases there may be angles at which the electrons of an atom scatter almost independently and yet not give rise to completely modified scattering. One would expect in this case a scattering function intermediate between Eq. (1) and the classical formula for independent scattering of unpolarized x-rays by electrons. The latter is

$$I_\theta = \frac{Ine^4(1+\cos^2\theta)}{2m^2r^2c^4} \tag{2}$$

Apparatus

The apparatus used is drawn to scale in Fig. 1. A water-cooled molybdenum-target x-ray tube was operated at currents up to 60 milliamperes and voltages up to 63 kv while connected to mercury pumps. The radiation from this was passed through various types of filters at F, through slit S_1, and entered a metal chamber C through a window of thin celluloid. The gas to be studied was admitted to C at pressures ranging from less than one millimeter to several atmospheres. Secondary radiation from a small volume of this gas passed through slits S_2 and S_3 into an ionization chamber. The ionization current produced in this chamber was taken as a measure of the intensity of the scattered radiation at the different angles θ from the direct beam.

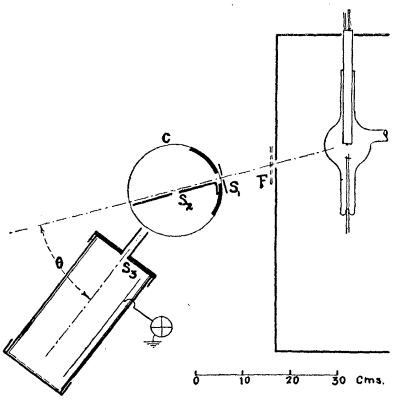

Fig. 1. Diagram of apparatus.

The x-ray tube was operated from kenotron-rectified a.c., and a balanced circuit was used to minimize fluctuations in current and voltage.[17] By having a voltmeter and milliameter constantly in sight and controls convenient, the error due to power-line fluctuations was kept low. Most of the readings were taken between 2:00 and 5:00 A.M., when power-line fluctuations were at a minimum.

The gas chamber C, 20 cms in diameter, 11 cms deep, was lined with lead foil and had a vertical lead partition extending across it parallel to the primary beam. The partition contained a rectangular slit S_2 (1.2 cms wide by 2.4 cms high) opposite the center of the chamber. A window 2.4 cms high was cut most of the way around the chamber and covered with clear cellu-

[17] J. A. Bearden, Phys. Rev. **29**, 20 (1927).

loid 0.55 mm thick, through which the x-rays could enter and leave the chamber with negligible absorption. When evacuated and disconnected from the oil pump, the leak in this chamber was of the order of a centimeter of pressure per hour. Slit S_1 was rectangular, 8 mms wide and 2.1 cms high and was placed so that about 5 mms intervened between the edge of the primary beam of x-rays and the partition carrying S_2. The slit S_1 was 11 cms from the center of the chamber. Slit S_3 at the ionization chamber was rectangular, 1.3 cms wide by 2.3 cms high, and was 18 cms from S_2. A lead tube of the same cross section extended from this slit toward S_2 to shield the ionization chamber from spurious radiation.

The ionization chamber was cylindrical, 12 cms in diameter and 30 cms long and was filled with methyl bromide. A voltage of about 350 was applied to it, and the ionization currents were measured with a Compton electrometer by the rate of deflection method. Measurements in the range 20°–100° (i.e. those plotted in Fig. 7) were made with the tube and its lead box as shown; the focal spot was 38 cms from the center of the scattering chamber. Later measurements, extending from 20° to 155° (Figs. 2 to 6) were made with the focal spot moved to 40 cms. The partition for these measurements was placed on the opposite side of the primary beam from that shown in Fig. 1, and the ionization chamber also used on that side, but the dimensions of the apparatus remained practically unchanged.

Some experiments were done with the total unfiltered radiation from the tube. Operating at 63 kv and 24 ma, the value of the effective wavelength (determined by absorbtion in 0.65 mm aluminum) was 0.48A; operating at 45 kv (approx) and 35 ma, it was 0.50A. With a 1.3 mm aluminum filter at F, Fig. 1, the x-rays at 63 kv, 24 ma had an effective wavelength of 0.39A. These conditions are the ones used in obtaining Figs. 2 to 7. For experiments where a monochromatic beam was desired, balanced filters of zirconium oxide and strontium oxide were used according to a method suggested by Professor P. A. Ross.[18] Each filter was adjusted to such thickness that it absorbed the same amount of the K_β radiation. They were then found to be well matched at all wave-lengths except those between their K absorption limits. Placing first one then the other in the primary beam and taking the difference of the intensity transmitted, a highly monochromatic spectrum was obtained.[19] With the large cross section of the beam available by this method, the energy used was much greater than could be obtained with the usual crystal reflection method. The accuracy was impaired, however, by the fact that it was necessary to deal with differences of readings rather than with the readings themselves.

The gases used were from tanks: the hydrogen was said by the manufacturers to be electrolytic and to contain only 0.03 percent oxygen as an impurity; the oxygen, 0.1 percent impurity. The purity of the helium (obtained from the U. S. Government) was not known, but it undoubtedly contained several percent of nitrogen. The removal of this was attempted

[18] P. A. Ross, Phys. Rev. **28,** 425 (1926).
[19] C. S. Barrett, Proc. Nat. Acad. Sci. **14,** 20 (1928).

by passing it slowly through two traps in series containing cocoanut charcoal and immersed in liquid air. It was necessary to pass the helium through these traps slowly while the chamber was below atmospheric pressure, so the leakage of air into the chamber may have contaminated the purified helium. The argon was about 95 percent pure, and the nitrogen and carbon dioxide probably somewhat better than this. All gases were admitted to the scattering chamber through a dehydrating tube and plugs of glass wool. In the case of every gas except helium, then, the scattering from impurities should have been negligible. The ratio of the intensities of the scattering from different gases at the same pressure and angle support this conclusion. The gas pressures in the scattering chamber were measured with a mercury manometer and were kept constant to one percent during readings.

The volume of the gas effective in scattering at different angles was not the same. To correct for this, the path of the primary beam was filled with light solid substances such as cotton batting, small waxed paper cylinders, and bran. The scattering at each angle was then obtained. These substances were then made into narrow vertical cylinders and placed in the primary beam with the scattering chamber removed, and with the x-ray tube operating under the same conditions as before. The ratio of the scattering of a substance outside the chamber to its scattering at the same angle inside gave a correction factor which, multiplied by the original gas readings for that angle, reduced them to the value that would be obtained from a small cylinder of gas in the primary beam. Corrections for spurious scattering from the walls, ionization leak, etc., had to be made in all measurements, and amounted on the average to five or ten percent of the total current. The materials used in this calibration were of course only approximately uniform in density when placed in the chamber. Though precautions were taken against errors due to this fact, nevertheless the accuracy of the gas readings was considerably lowered by multiplication by this factor. The relative scattering of two gases at the same pressure and angle could be determined with a probable error of one to two percent when the intensities due to them were nearly the same. Because of the difficulty in obtaining the correction factors for 20° and 155° they are somewhat less reliable than those for the other angles. After reduction by this correction factor, the probable error was 3 to 5 percent, though of course the accuracy of the *ratio* of the two remained the same.

Procedure and Results

Fig. 2 shows the ionization current obtained when the ionization chamber was set at a particular angle and the pressure of oxygen in the scattering chamber was varied. It shows clearly that, over the whole range of angles, the scattering was proportional to the gas pressure. Thus taking the scattering of a gas in the chamber at a certain pressure and subtracting the scattering at zero pressure, one has left only the true scattering from the gas.

The curve R in Fig. 3 is the volume correction factor mentioned previously. The data in Fig. 2 are multiplied by the appropriate values from

this R curve and plotted as the solid curve of Fig. 3. The classical theory for the scattering from a single electron, Eq. (2), is plotted as the broken curve of Fig. 3 marked C, and the quantum theory, Eq. (1), is the broken curve Q. All oxygen values have been multiplied by an arbitrary factor (the same for all angles) to make the solid curve fall somewhere between C and Q

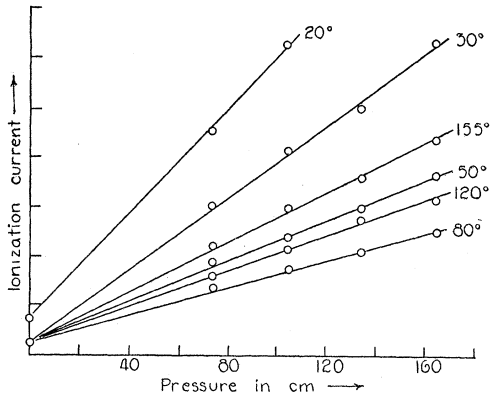

Fig. 2. Scattering from oxygen at different pressures.

at the larger angles. As there is both modified and unmodified radiation present at these angles in unknown proportions, one does not know exactly where in this range the solid curve should fall. In Fig. 3 excess scattering is evident below about 60°. The lowness of the 80° scattering possibly indicates that there is destructive interference in this neighborhood.

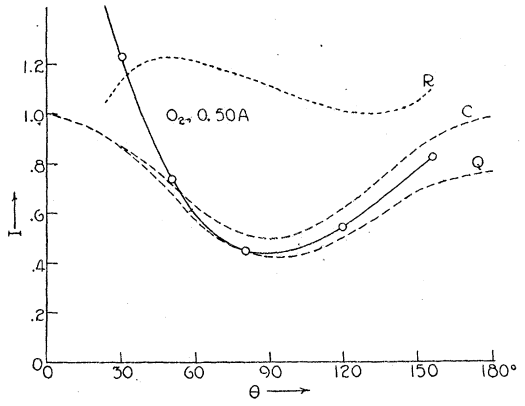

Fig. 3. Scattering from oxygen.

Figs. 4 and 5 represent data obtained in a similar manner for the gases oxygen, argon, nitrogen, and carbon dioxide at two different wave-lengths. The same pressure was used for each gas, and the intensity of the primary beam remained the same, for all the data in each figure. In these figures the scattering of each gas is divided by its number of electrons per molecule;

i.e., the curves represent the scattering per electron. The evidence is convincing for the presence of both constructive and destructive interference in x-rays scattered from argon. Remembering that the proportion of modified radiation present increases with the angle of scattering, one would expect the curve representing absence of interference to lie relatively nearer the curve Q, in the region between C and Q, at large angles than at small angles.

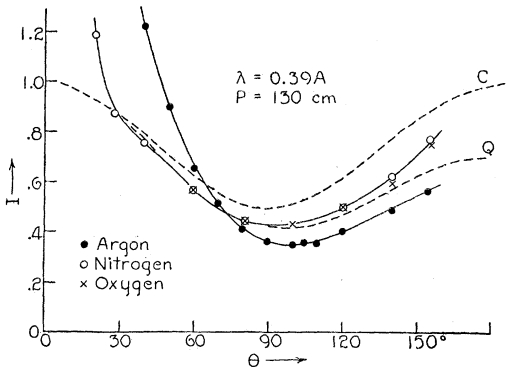

Fig. 4. Scattering from argon, nitrogen, and oxygen.

In the light of this, Figs. 3, 4, and 5 may indicate the presence of a little destructive interference for the gases oxygen, nitrogen, and carbon dioxide in the region around 60°–90°.

Further evidence that there is interference in x-rays scattered from argon is the variation with wave-length of the scattering from this gas illustrated

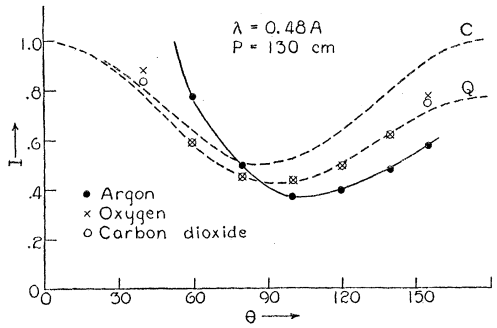

Fig. 5. Scattering from argon, oxygen, and carbon dioxide.

in Fig. 6. The curves for three different effective wave-lengths, $\lambda_1 > \lambda_2 > \lambda_3$, are arbitrarily reduced to the same value at 100°. In this figure the volume correction factor has not been used. Another test of the same type was made with two different wave-lengths, over the range 30°–150°. In all cases the excess scattering increased and extended to larger angles with increase in wave-length, as predicted by the classical theory.

Another series of experiments, giving the scattering per electron for hydrogen, helium, nitrogen, oxygen, and argon, is summarized in Fig. 7. Constructive and destructive interference from argon is again prominent. The fact that excess scattering becomes concentrated nearer the direct beam as the atomic number of the scatterer decreases may be interpreted as evidence of increasing size of the atom with decreasing atomic number. The helium curve shows excess scattering; but due to the possible presence of nitrogen as an impurity, one hesitates to ascribe this excess solely to the helium itself. Helium and hydrogen scatter equally from 60° to 100°. Hydrogen follows the broken line of Fig. 7, which represents Eq. (1) indicating completely modified radiation.

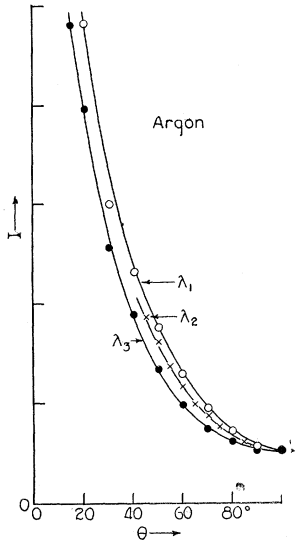

Fig. 6. Scattering from argon as a function of wave-length ($\lambda_1 > \lambda_2 > \lambda_3$).

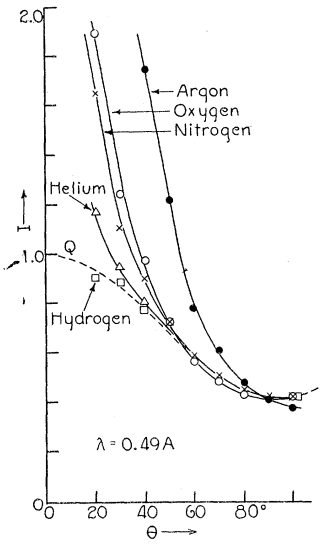

Fig. 7. Scattering of argon, oxygen, nitrogen, helium, and hydrogen.

Previous experiments[19] over the range 20°–90° with oxygen, carbon dioxide, and hydrogen showed oxygen (and also carbon dioxide, whose values could not be distinguished from those of oxygen) to have excess scattering below 60° which varied with the wave-length, while the scattering from hydrogen was practically independent of the wave-length. The accuracy of these experiments was not as great as that of the curves shown here, but in the case of hydrogen it was sufficient to show the absence of interference effects of the type predicted by Debye and others.

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