SCATTERING OF X-RAYS FROM GASES

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Abstract

Measurement of the intensity of scattering of x-rays by various gases has been made for scattering angles between 10° and 90°. Soller slits were used to get a well-defined scattering angle and matched filters of ZrO_2 and SrO were used to isolate the K_α lines of molybdenum. Intensity measurements have been made for hydrogen, helium, oxygen, neon and argon. All the measurements were put on the same scale by comparison of the intensity of scattering from the various gases with the scattering from hydrogen at 90°. This is equivalent to putting the intensities on an absolute scale since the scattering from hydrogen at 90° can be calculated with considerable certainty. Values of the structure factors for neon and argon were calculated and comparison made with those determined from wave mechanics for similar atoms.

Introduction

IN VIEW of Compton's[1] recent theory for determining the electron distributions in atoms from the angular distribution of intensity of x-rays scattered from monatomic gases it was thought desirable to get more complete experimental data.

Barrett[2] in 1928 published data for the scattering of x-rays from most of the gases included in this paper. He measured the intensity as a function of angle but no attempt was made to get absolute intensity measurements.

In the work reported here a more definite scattering angle has been obtained by the use of Soller slits, x-rays made homogeneous by the use of balanced filters have been used wherever the intensity permitted, and all the measurements have been put on the same scale by comparing the intensities from the various gases with that from hydrogen. This is equivalent to putting the intensity measurements on an absolute scale since the scattering from hydrogen at 90° can be calculated with considerable certainty.

Apparatus and Procedure

The set-up is shown in the diagram Fig. 1. A molybdenum target x-ray tube was operated at 40 kv peak and 35 m.a. with full-wave rectification. The tube was immersed in oil in a lead box fitted with a lead glass front. A small celluloid window within 2 mm of the tube transmitted the primary beam with only a small reduction in intensity due to the 2 mm of oil. This arrangement

[1] A. H. Compton, Phys. Rev. **35,** 925 (1930).

[2] C. S. Barrett, Phys. Rev. **32,** 22 (1928). Measurements of the relative scattering of hydrogen and Argon have been made by Herzog but not with homogeneous radiation. G. Herzog Helvetica Phys. Acta **2,** 169 (1929).

allowed the tube to be brought very near the spectrometer and the intensity was considerably greater than would have been possible with the tube in air.

The intensities were measured with an ionization chamber and a Compton electrometer with which currents of 5×10^{-15} amperes could be measured with a reproducibility of about five percent.

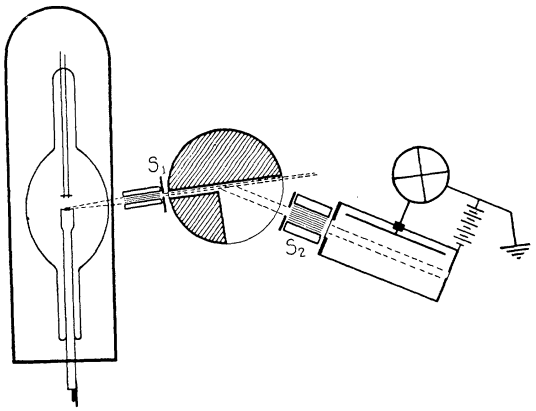

Fig. 1. Arrangement of apparatus.

The scattering chamber is shown in Fig. 2. It was made from a brass tube 5.5 inches in diameter, 3 inches high and having 1/8 inch walls. The top and bottom were 0.5 inch steel plates grooved and fitted with rubber gaskets and

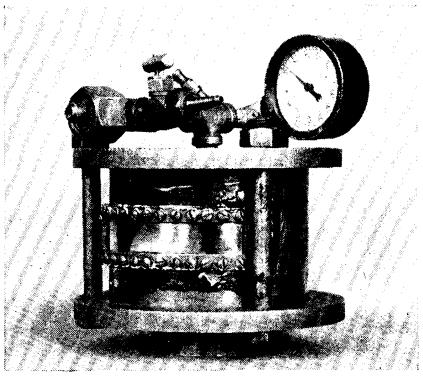

Fig. 2. Scattering chamber.

held in position by three bolts as shown. The windows were of 0.7 mm celluloid 2 cm high. They were screwed down on rubber gaskets and picein wax used where it was found necessary. The base was fitted to the spectrometer table so it could be removed and replaced without affecting the readings. The top contains a gauge, an oxygen tank connection, a needle valve for the chamber and a side tube for evacuating the chamber and the leads. The chamber

was partially filled with sealing wax as shown in Fig. 1, and lined with lead. This cut the volume to about 250 cc and acted as a shield for the diffusely scattered x-rays.

The chamber was found to be very satisfactory as pressures of 200 pounds were used with a leak of only a few percent per week and the corrections for scattering from the evacuated chamber were almost negligible.

Reference to the diagram will show the arrangement of the chamber and the slit system. Soller slits were used to collimate the primary as well as the scattered beam. These slits allowed a maximum angular divergence of less than 2° in the horizontal plane. Slit S_1 was 3 mm wide and 1 cm high. Slit S_2 was 1 cm high and could be varied in width to 1.5 cm. Besides defining the

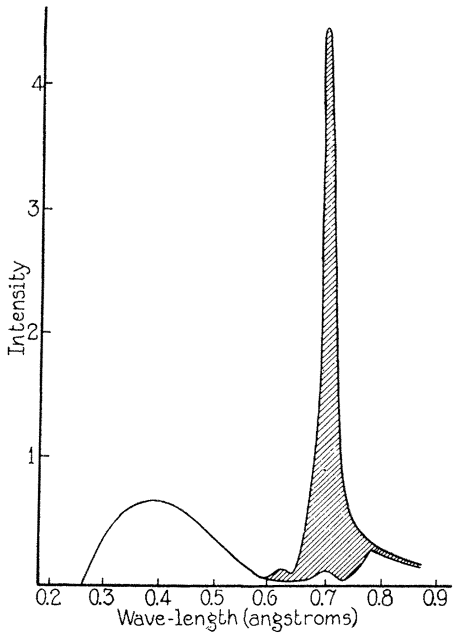

Fig. 3. Transmission through ZrO_2 and SrO filters.

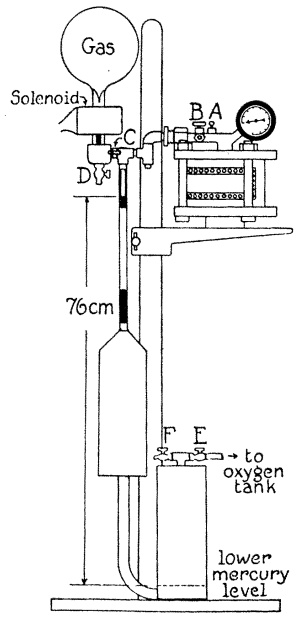

Fig. 4. High pressure mercury pump.

scattering angle the second slit served the additional purpose of determining the scattering volume. With this slit remaining constant the scattering volume was proportional to sin ϕ. The height of the slits S_1 and S_2 made the actual scattering angle slightly larger than indicated on the spectrometer circle. It was necessary to make correction for this on the readings between 10° and 30°.

The $K\alpha$ lines of molybdenum were separated out by the balanced filter method.[3] A filter of ZrO_2 containing about 0.025 gm per cm^2 was balanced by a filter of SrO so that less than five percent of the radiation corresponding to the difference of these filters lay outside of the range between the K critical absorption edges of Zr and Sr. A curve showing the transmission by these

[3] P. A. Ross, Phys. Rev. **28,** 425 (1926).

filters is shown in Fig. 3. The shaded portion represents the difference of the transmission by the two filters. The filters were additionally tested by measuring the absorption coefficient of aluminum which gave a straight line when log (I_0/I) was plotted against thickness. It is very important that these filters be well balanced so that reliable corrections for absorption can be made when elements like argon are compared with oxygen.

Since some of the gases were originally at atmospheric pressure it was necessary to construct a pump to put them into the chamber at the desired pressure. Fig. 4 shows the construction of the pump. The cylinders were of seamless steel tubing with welded bottoms and top and connecting tube. The rest was constructed mainly from iron pipes and connections. The vertical pipe was cut through at two places and a glass tube waxed inside to make the mercury level visible. With valve B closed and all the others open the system

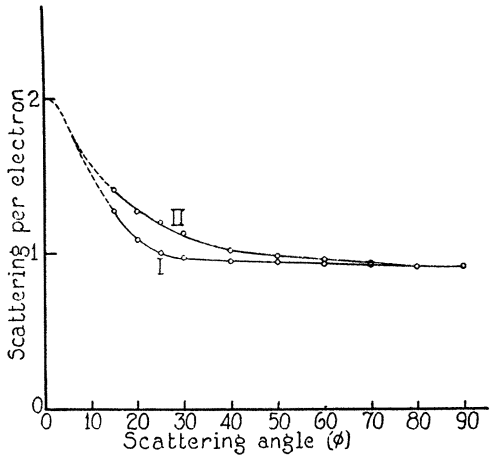

Fig. 5. Curve I hydrogen, Curve II helium.

could be evacuated through valve D. Closing valve D and breaking the tip in the glass container by means of the electromagnet, the gas expanded into the upper cylinder of the pump. By closing valves C and F the gas could be forced into the scattering chamber by applying the oxygen tank pressure at valve E. With repeated pumping it was necessary to draw the mercury back into the lower container by applying the vacuum pump at F.

Hydrogen

The hydrogen used in this experiment came from a tank especially prepared and analysed by Burdett Oxygen Company and found to have less than 0.1 percent oxygen and CO_2 and 0.07 percent water vapor by volume. After scattering measurements had been made on the gas the density was measured by weighing and no additional impurities were found to be present. The scattering chamber connected directly to the hydrogen tank, was evacuated together with all the leads, flushed out several times and filled to about 200 pounds pressure. It was then placed in position on the spectrometer table

and the scattered intensity measured from 10° to 90°. Even at 200 pounds pressure, however, it was necessary to use general radiation in place of the balanced filters. Curve I, Fig. 5 shows the scattering per electron corrected to $\lambda = 0.71$A plotted against ϕ. On the basis of the classical theory of scattering the value of $S \equiv I_s/ZI_e$ for hydrogen would be unity at 90°. The fact that practically all the intensity scattered at 90° from hydrogen is modified will make the experimental value at this angle slightly lower than unity. The Breit-Dirac relation may then be used

$$S_{class} = S_{mod}\{1 + \gamma \text{ vers } \phi\}^3. \quad (1)$$

Placing for 90° $S_{class.} = 1$, we have thus, $S \equiv S_{mod.} = (1+\gamma \text{ vers } 90°)^{-3}$. For $\lambda = 0.71$ A, γ vers $90° = 0.0243$, whence $S = 0.91$.

The data included in table I are all based on a value of S for hydrogen equal to 0.91 at 90°. To get the classical values of S Eq. (1) must be used.*

TABLE I. *Scattering per electron.* $\lambda = 0.71A$.

Angle	Hydrogen	Helium	Oxygen	Neon	Argon
10°			8.05	8.55	13.90
15°	1.27	1.45	4.90	6.87	10.65
20°	1.10	1.34	2.96	5.16	8.50
25°	1.03	1.27	2.00	3.85	6.35
30°	1.00	1.20	1.72	3.11	5.23
40°	0.98	1.08	1.43	2.04	4.15
50°	0.97	1.01	1.15	1.53	3.32
60°	0.95	0.97	1.04	1.32	2.74
70°	0.93	0.95	1.02	1.24	2.28
80°	0.92	0.92	1.01	1.18	2.10
90°	0.91	0.91	0.96	1.06	1.95

In so far as the scattering per electron from hydrogen is known, the scattering from other gases can be put on an absolute basis by comparison with it. Since the intensity of scattering from hydrogen is weak and difficult to measure it was first carefully compared with oxygen which was then used as a secondary standard of comparison.

COMPARISON OF OXYGEN AND HYDROGEN

Oxygen and hydrogen were compared several times at 90° using general radiation and pressures of about 200 lb/sq. in. for both gases. This involves a large correction for absorption for an unknown wave-length. To make this correction the intensity of scattering was plotted as a function of pressure. The decrease in scattering per gm at 200 lb/sq. in. over that at small pressures was taken as the absorption correction.

Comparison of hydrogen at 200 pounds pressure with oxygen at 40 pounds pressure, taking account of the difference in pressure and making only a very slight absorption correction, gave results in very close agreement with those of the first method.

* Equation (1) gives the classical value only when all the scattered radiation is modified. In the case of oxygen, neon and argon the unmodified radiation must also be considered. Compton (ref. 1) has shown how the classical value of S can be gotten in this case.

The hydrogen curve was corrected to a wave-length of 0.71A by a method shown by the curves of Fig. 6. The curves are: I, hydrogen with general radiation; II, oxygen with general radiation; and III, oxygen taken with $\lambda = 0.71$A and arbitrarily fitted to the other oxygen curve at 90°. From these curves the intensity of scattering from hydrogen for $\lambda = 0.71$A was determined.

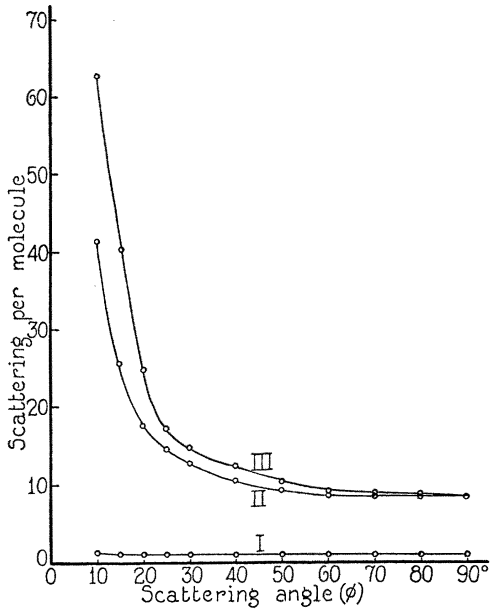

Fig. 6. Curve I hydrogen (general radiation), Curve II oxygen (general radiation), Curve III oxygen (filtered radiation, $\lambda = 0.71$A).

Helium

The helium came in liter containers at atmospheric pressure and was introduced into the chamber by means of the mercury pump described above. The pressure used was about 115 lb/sq. in. A number of sets of readings were taken and the intensity compared directly with hydrogen. The intensities of scattering from hydrogen and helium at 90° were found to be equal within the experimental error. Curve II, Fig. 5 shows the scattering per electron from helium corrected to $\lambda = 0.71$A in the same manner as for hydrogen.

In introducing the helium into the chamber about 0.9 percent of air by volume entered as an impurity. This was found to be the case by measurement of the density after the readings had been taken. Correction was of course made for this and would not lead to any appreciable error because the scattering from air could easily be measured. The data for hydrogen and helium are in very close agreement with the results obtained by Barrett. Although he obtained no excess scattering from hydrogen his data only extended to what corresponds to $\phi = 30°$ on the above curve.

Neon

Neon was also obtained in liter containers at atmospheric pressure and was put into the scattering chamber with the aid of the mercury pump.

The intensity measurements were made with filtered radiation and a number of sets of readings taken. In order not to have to evacuate and refill the chamber repeatedly, a sheet of celluloid was used for an intermediate comparison. Comparisons of the intensity from neon were made with the celluloid scatterer and then the chamber was evacuated and filled with oxygen and the comparisons with the celluloid scatterer again made. Corrections were made for the different absorption coefficients, and the results are shown in curve IV,

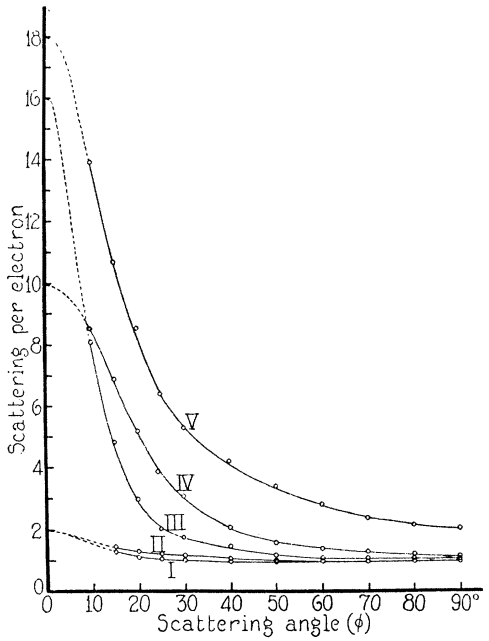

Fig. 7. $\lambda = 0.71$A. Curve I hydrogen, Curve II helium, Curve III oxygen, Curve IV neon and Curve V argon.

Fig. 7. The scattering per electron from neon at 90° is very nearly that from oxygen. The neon curve begins to rise at larger angles than does the oxygen curve, a difference similar to that between hydrogen and helium.

Argon

The argon used in these measurements was obtained in a tank with about 0.3 percent impurity. The chamber could be readily filled to any desired pressure directly from the tank. This gas, however, presented greater difficulties than the other gases since its absorption coefficient is so much larger. Fig. 8 shows the relation between intensity and pressure for argon and oxygen. Curves I and II represent respectively the measured intensity from argon and oxygen; curves IV and III represent the values after correction for absorption

has been made. To make the corrections as small as possible and still get sufficient intensity from both oxygen and argon pressures of about 40 to 50 lb/sq. in. were used.

In the case of argon, the comparison of intensity with oxygen was done directly by taking a number of readings with one gas and then refilling the chamber with the other gas with the x-ray tube in operation during the process. Several of these comparisons were made and the pressures varied somewhat to make the absorption corrections more certain.

It is noticeable that in argon the scattering per electron is considerably larger than for the other gases. This would correspond to a greater electron density near the center of the argon atom.

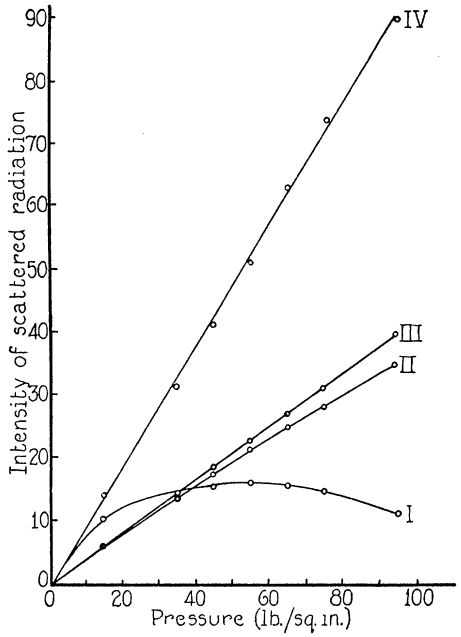

Fig. 8. Curves I and II represent respectively the measured intensity from argon and oxygen curves. Curves IV and III represent the values after correction for absorption has been made.

Errors

The largest errors involved in measuring the scattering per electron would probably be those of comparing the scattering from one gas with that of another. In the case of the comparison of the intensity of scattering from hydrogen with that from oxygen the probable error from the mean was about 1 percent. It is likely that there are other errors such as uncertainty of wavelength, measurement of pressure and corrections for absorption which would also amount to about 1 percent. This would mean that the scattering from oxygen would probably be correct to about 2 percent. In comparing neon and oxygen, considering the probable error already mentioned, the uncertainty

in the neon data would be of the order of 3 percent. In argon the large correction for absorption would probably increase the uncertainty to about 4 or 5 percent. In the case of hydrogen and helium the probable error of any point of the curve was about 2 percent.

Structure Factor

Calculation of the structure factors for neon and argon have been made from the relation given by Compton

$$F = Z\left\{\frac{S-1}{Z-1}\right\}^{1/2} \tag{2}$$

where S is the classical value of the scattering per electron, Z is the atomic number and F the structure factor. The values thus calculated are given in Table II. In order to make comparisons between these data and the structure

Table II. *Structure factor.*

ϕ	$\left(\frac{\sin \phi/2}{\lambda}\right)$	Neon		Argon	
		S_{class}	F	S_{class}	F
10°	0.123	8.55	9.16	13.90	15.63
15°	.190	6.87	8.08	10.65	13.53
20°	.245	5.16	6.80	8.50	11.93
25°	.306	3.86	5.64	6.36	10.10
30°	.365	3.12	4.86	5.26	9.00
40°	.482	2.06	3.43	4.17	7.77
50°	.596	1.56	2.49	3.35	6.69
60°	.704	1.37	2.02	2.79	5.84
70°	.808	1.30	1.80	2.34	5.06
80°	.905	1.26	1.70	2.18	4.72
90°	.995	1.17	1.37	2.05	4.49

factors calculated from crystalline reflection it is necessary to correct the latter for the effect of thermal agitation which does not affect the intensity of x-rays scattered from gases.

Some very interesting comparisons have been made by James and others[4,5] between the crystal reflection data at low temperatures and the structure factors calculated from Hartree's wave mechanics solution of the charge distribution in the K^+, Cl^- and Na^+ ions. Making temperature corrections and assuming the existence of zero point energy they get structure factor curves which are in good agreement with the theoretical curves.

The values of F from Table II are plotted in Figs. 9 and 10 together with F values from wave mechanics taken from the above mentioned papers.

In Fig. 9 the solid line represents the F curve for Na^+ calculated from wave mechanics and the broken line represents the F values for neon. From a consideration of equation (2) the reason for the apparently large inaccuracies in

[4] R. W. James and Brindley, Proc. Roy. Soc. **A121,** 155 (1928).
[5] R. W. James, I. Waller and D. R. Hartree, Proc. Roy. Soc. **A118,** 343 (1928).

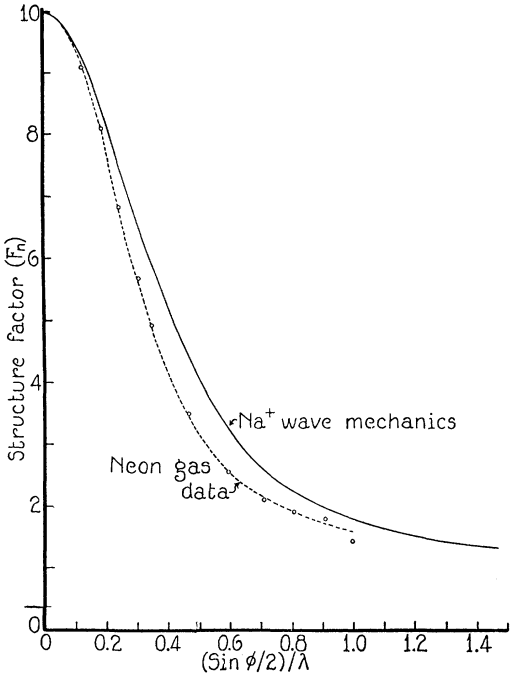
Structure factor (F_n)
Na^+ wave mechanics
Neon gas data
$(\sin \phi/2)/\lambda$

Fig. 9.

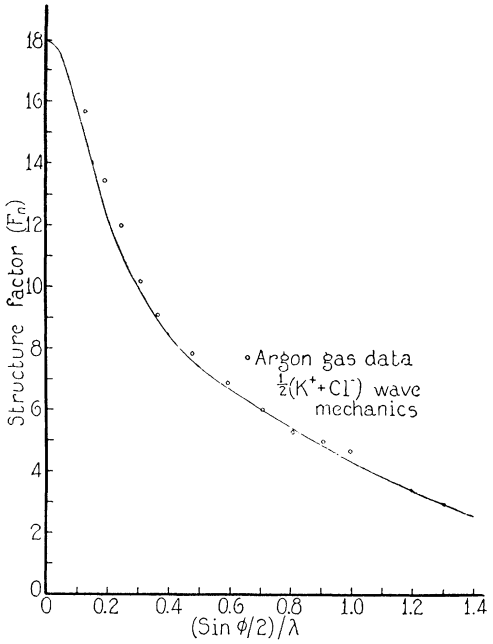
Structure factor (F_n)
Argon gas data
$\frac{1}{2}(K^+ + Cl^-)$ wave mechanics
$(\sin \phi/2)/\lambda$

Fig. 10.

the values of F when S is nearly equal to unity can be seen. For neon at $(\sin \phi/2)/\lambda = 1.00$ an error of 2 percent in S gives rise to an error of 9 percent in F.

A more significant comparison between theory and experiment is shown in Fig. 10. The curve represents the average of the F values for K^+ and Cl^- determined from wave mechanics. The circles give the F values from Table II for argon. The agreement is seen to be very satisfactory.

As has recently been shown by Compton[6] a comparison of these data with that determined from reflection from a crystal of KCl and corrected to 0°K gives a good experimental check of the existence of zero point energy.

Conclusion

These data furnish a significant check of Compton's recent theory of the intensity of x-rays scattered from gases and the use of these data for the determination of electron distribution in atoms. The agreement between the F curves of argon and those determined from wave mechanics and also from crystal reflection should give more confidence in the use of these methods for determining the distribution of charge in the atom.

A comparison of the F curves determined by these three methods is also a further check on the existence of zero point energy.

Calculation of the electron distribution curves from these data are now in progress.

In conclusion the author wishes to express his appreciation to Professor A. H. Compton, who suggested this problem, for his continued assistance in its solution.

[6] A. H. Compton, Phys. Rev. **37**, 104 (1931).

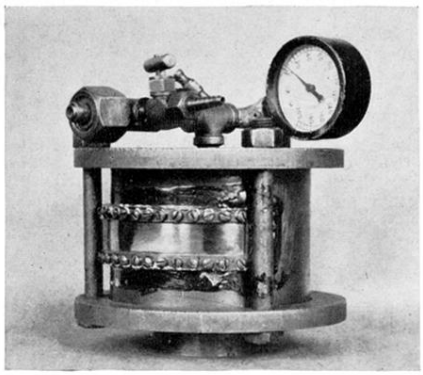

Fig. 2. Scattering chamber.