The Radioactivity Induced in Oxygen by Deuteron Bombardment*

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An induced radioactivity of half-life 1.16 minutes has been observed after the bombardment of oxygen or its compounds by 3 MV deuterons. The disintegration particles were observed to be positrons. Chemical tests showed that the active substance was an isotope of fluorine. The probable reactions are therefore:

$$O^{16}+D^2=F^{17}+n^1; \quad F^{17}=O^{17}+e^+.$$

A product of the same properties was already known to be induced in nitrogen under α-particle bombardment. The activation function of oxygen has been measured by collecting radioactive recoil atoms at several points along the path of the deuteron beam when it is passing through an atmosphere of oxygen. The excitation curve drops very rapidly and goes to zero at about 2 MV deuteron energy. This is best accounted for by assuming that energy disappears in this reaction, and that the energy of reaction is the negative of the threshold energy. By correcting for recoil, $Q=-1.8$ MV. To confirm this estimate of the energy of reaction, the maximum angle between the direction of the deuteron beam and the paths of the radioactive recoil atoms has been measured. A piece of optical quartz (SiO_2) was bombarded at a glancing angle by a narrow beam of deuterons, and radioactive recoil atoms were collected on a cylindrical strip of copper which was placed about the target. The maximum angle of recoil was found to be about 30° which corresponds to $Q=-1.3$ MV. The agreement with the value, -1.8 MV found previously, is within the errors of the measurements. In order to check the method the same experiment was performed with a graphite target; the longer half-life of the active material made possible a more accurate measurement. The maximum angle of recoil was found to be 42° for the activation of carbon; this corresponds to $Q=-0.1$ MV for the reaction $C^{12}+D^2=N^{13}+n^1$. This agrees reasonably well with the experimental value of Q found by Tuve and Hafstad and with the value of Q which is deduced from maximum energy of the positrons from N^{13} and the range of the protons which are emitted when carbon is bombarded by deuterons.

INTRODUCTION

NEGATIVE results have been obtained in most of the previous experiments in which oxygen has been bombarded by charged particles. No certain effects have been observed with α-particles or protons of the highest available energies. Oliphant and Rutherford,[1] and Cockcroft and Walton[1] have shown that deuterons lead to the emission of a short proton group, but McMillan and Livingston[2] found no detectable radioactivity when oxygen was bombarded with deuterons of 1.9 MV energy. The reason for this abnormally high stability of the oxygen nucleus has not been clearly understood. The work to be described in this paper shows, however, that in at least one of the cases cited above, the possible reaction did not occur simply because the energy of the bombarding particle was not sufficient to compensate for that lost in the reaction: oxygen may be activated by bombardment with very high energy deuterons, but this activation occurs only above a well-defined threshold. An independent estimate of the energy of this reaction shows an energy loss approximately equal to the threshold energy of the deuteron.

ESTABLISHMENT OF THE REACTION

The first evidence for induced radioactivity in oxygen was found in some experiments on radioactive silicon. When a piece of quartz was bombarded in an atmosphere of oxygen with 3 MV deuterons, an activity was found which was much stronger than any which have been observed in pure silicon. Activities of the same half-life have since been found after the bombardment of iron oxide, water, and gaseous oxygen. A logarithmic plot of the decay curve of the radioactive substance is shown in Fig. 1; measurements of the half-life give a reproducible value of 1.16 minutes.

The tracks of the particles emitted by this substance have been observed in a Wilson cloud chamber.[3] The direction of curvature of the tracks in a magnetic field shows that they are positrons. The probable reactions may be written:

* A preliminary report on these experiments was given at the Los Angeles Meeting, June 10, 1935.

[1] See Cockcroft and Walton, Proc. Roy. Soc. **A144**, 704 (1934).

[2] McMillan and Livingston, Phys. Rev. **47**, 452 (1935).

[3] I am indebted to Dr. Kurie for the use of his apparatus in this experiment.

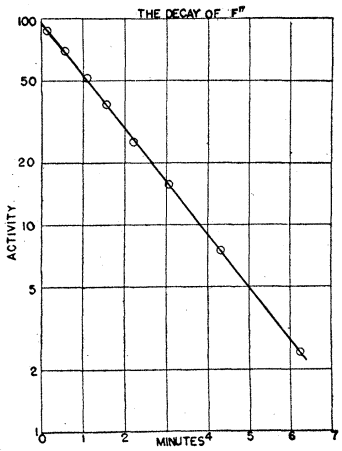

FIG. 1. A logarithmic plot of the decay of F^{17}; the half-life of this substance is found to be 1.16 minutes.

$$O^{16}+D^2=F^{17}+n^1, \quad (1)$$

$$F^{17}=O^{17}+e^+. \quad (1a)$$

The identity of the active substance was established by a chemical separation. A small amount of water was bombarded behind an aluminum window of 4 mm stopping power. The water was then rinsed into a boiling solution of potassium fluoride which also contained small amounts of aluminum chloride and nitric acid. Calcium chloride was then added and the precipitate, calcium fluoride, filtered out; active aluminum from the window remained in the acid solution. The precipitate showed a large activity which decayed with the expected period.

A radioactivity of the same properties has been observed by Wertenstein[4] and others after the bombardment of nitrogen by α-particles. In this case the probable reactions are

$$N^{14}+He^4=F^{17}+n^1, \quad (2)$$

$$F^{17}=O^{17}+e^+. \quad (2a)$$

THE EXCITATION FUNCTION OF OXYGEN

It will be observed that the reaction giving rise to F^{17} is analogous to the well-known reactions:[2]

$$C^{12}+D^2=N^{13}+n^1, \quad (3)$$

$$N^{14}+D^2=O^{15}+n^1. \quad (4)$$

However, the excitation function for oxygen must be quite different from those of the neighboring elements since the above reactions will go at very low bombarding energies while Livingston and McMillan could not activate oxygen with 1.9 MV deuterons. It would seem, then, that a study of this abnormal excitation should be of considerable interest.

The excitation curve for oxygen has been determined with some care while approximate curves have been obtained for carbon and nitrogen for purposes of comparison. The measurements were made by passing the deuteron beam through a gas containing the element to be studied and collecting radioactive recoil atoms at various points along the path of the beam. The activity was collected on a series of large mesh copper screens which were placed perpendicularly to the beam so that it passed through each one of them in turn. The screens were spaced about 6 mm apart. The activity collected on a screen should be proportional to the excitation of the element in question by deuterons of the energy which they have as they pass the screen. The experiment was carried out in a space about 3 cm long while the total range of the beam was more than 10 cm; since the experiments were run at atmospheric pressure, it was necessary to replace some of the earlier screens by foils of known stopping power in order to carry the measurements to lower energies.

The activities deposited on the screens were high enough to allow the exposure and measurement of about six screens at one time. The decay of the activity on each screen was determined, and, in this way, activities due to contaminations could be eliminated. In Fig. 2, the results of these measurements on carbon and oxygen are shown. A few points on the nitrogen excitation curve were obtained, and it was shown to be similar to that of carbon. The gases used were carbon dioxide and free nitrogen and oxygen. The short period activity which would arise from the oxygen in carbon dioxide was allowed to die out before the longer lived activity from carbon was measured.

The most serious error in these measurements is caused by the interference of the screens with the beam; if any large fraction of the beam is stopped by the screens, the excitation curve will

[4] Wertenstein, Nature **133**, 564 (1934); Ellis and Henderson, Proc. Roy. Soc. **A146**, 206 (1934).

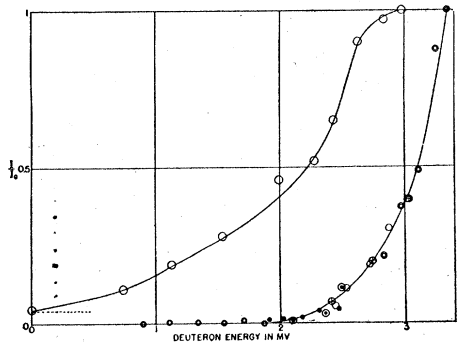

FIG. 2. The excitation curves of oxygen and carbon. In the oxygen curve (right) the five points along the axis shown as heavy circles are those for which diffusion has been eliminated. Diffused activity has not been eliminated on any of the points on the carbon curve (left) and there is consequently a background, shown as a dotted line, which must be subtracted from the curve.

drop too rapidly. The screens were lined up as well as possible to prevent any screen except the first from stopping deuterons, but, since the deuteron beam was curved by the strong magnetic field of the ion accelerator, the alignment could not be very precise. However, the importance of the errors arising from this cause may be determined by changing the spacing of the screens. If the experiment is performed twice under the same conditions except that all six screens are in place in one case and only the first and last screens are in place in the other, the total number of deuterons stopped by the screens should be much higher in the former case; the weakening of the beam may be measured by plotting the results of the two experiments together. If the two curves coincide at all points after they have been fitted at one point, the error under investigation is unimportant. In the actual measurements, the effective spacing was changed by introducing foils between two of the earlier screens. Many exposures were made with oxygen with great differences in the effective spacing. It will be seen in Fig. 2 that all of the several runs follow the same curve independent of the spacing. The activity found on the first screen in each run was found to be reproducible but evidently too high. This is to be expected since this screen certainly stops 10 percent of the beam. For this reason, the activity of the first screen is not shown in the curve.

A correction for the variation of the range of the recoil particles with the deuteron energy might be applied since this range determines the effective target thickness in these experiments. However, the correction is not important for the oxygen curve.

It was shown very simply that diffusion of the active gas from one region to another had little effect on the curve where the activity was large. The activity of a screen was measured on both sides; the side of the screen which was struck by the beam was found to be twice as active as the other. This is about the difference in intensity to be expected if a sheet of the same thickness as the wires of the screen were placed over the active substance. It is evident, then, that nearly all of the activity on the screen is on the "front" side. Since the recoil atoms in both the carbon and the oxygen reactions are driven only in the forward direction (see the later experiments) nearly all of the activity must be deposited on the screens by recoil. However, where the activities are very low, diffusion of the activity undoubtedly becomes important. Below 2 MV on the oxygen curve, the very small activities which are found show no definite trend and are probably due entirely to diffusion. In order to eliminate diffused activity, the five points at the lowest energy were taken with an aluminum foil of 49 mm stopping power absorbing most of the energy of the deuterons; it will be noticed that the activities of this set of points are decidedly lower than the next higher set where most of the stopping power was oxygen, and there was, consequently, more activity to diffuse. It seems safe to say, then, that there is no detectable excitation of oxygen by deuterons of less than 2 MV energy.

The excitation curve of carbon which is shown in Fig. 2 was obtained from a single set of measurements and is by no means precise. However, the general trend of the curve has been checked by an independent method. Since this curve was obtained only for comparison with that of oxygen, the corrections mentioned previously were not made. Since diffusion was not eliminated, there is a background due to it which must be subtracted from the curve. A few points on excitation curve of nitrogen have been obtained; they follow the carbon curve very closely.

The Energy of Reaction and the Mass of F^{17}

The obvious explanation for the very great difference between the excitation curve of oxygen and those of carbon and nitrogen is that a large amount of energy is lost in the oxygen reaction so that the energy deficiency must be made up by the deuterons before any excitation occurs. There can be no question that the deuterons are entering the oxygen nucleus, since disintegration protons have been obtained from oxygen under bombardment by deuterons of much less than 2 MV energy.[5] How accurately this energy loss may be estimated from the excitation curve is a difficult question. In the last section it was shown that the excitation curve of the reaction shows an experimental threshold in the neighborhood of 2 MV bombarding energy. However, the real threshold which corresponds to the loss of energy in the reaction will lie somewhat below this energy. In the case of the excitation of carbon, which is closely analogous to that of oxygen, the real threshold (i.e., the energy of reaction) is about zero (see later experiments) while the experimental threshold as found by Hafstad and Tuve[6] and by Cockcroft, Gilbert and Walton[7] is below 0.4 MV. This difference is due, at least in part, to the potential barrier of the nucleus which is penetrated with great difficulty by charged particles of low energy. In the case of oxygen, it is evident that, in the neighborhood of the experimental threshold, the potential barrier is being penetrated with relative ease since the reaction $O^{16}+D^2=O^{17}+H^1$ occurs at comparatively low bombarding energies; it would be expected then that the difference between the real and experimental thresholds would be less than for carbon, because of the relative ineffectiveness of the potential barrier. If then we take the real threshold as equal to 2 MV, we are certainly not high by more than 0.4 MV. Correcting for recoil, the energy of disintegration, $Q=(8/9)2.0=-1.8$ MV.

The excitation curve for the reaction: $N^{14}+He^4=F^{17}+n^1$ has been determined by Haxel.[8] A threshold was found in this excitation curve at an α-particle energy of 6 MV. This threshold may be taken as a measure of the energy of reaction by the same arguments as are used above. The α-particles are known to be entering the nucleus below the threshold since protons are emitted at considerably lower bombarding energies. Then from the threshold energy $Q=-(7/9)6=-4.7$ MV. The same kind of an error is made here as in the previous case.

The consistency of these assumptions may be checked by calculating the mass of F^{17} from the two reactions. From the oxygen-deuteron reaction:

$$F^{17}=16.0000+2.0142-1.0083+0.0019=17.0078. \quad (1')$$

From the nitrogen-α-particle reaction:

$$F^{17}=14.0080+4.0034-1.0083+0.0050=17.0081. \quad (2')$$

The check is well within the error in the masses. The masses are taken from the paper of Oliphant, Kempton and Rutherford[9] except for N^{14} for which the old Aston value is used. The use of the new masses of Aston[10] or those generally accepted last year give satisfactory checks although the absolute values obtained differ considerably. The agreement of the two calculated values is independent of the mass of the neutron and of a systematic error in the estimates of the energies of reaction. The check does show, however, that the interpretation of the experiments is consistent.

The Angle of Recoil of Active Atoms

An attempt was made to detect the neutrons given off during the excitation of oxygen by measuring the neutron induced activity in silver. However, no effect was found which was large enough to show against the high neutron background of the apparatus. The value of the energy of reaction obtained from the excitation curve cannot, then, be checked by studying hydrogen recoils of the neutrons, but it is possible to study the radioactive recoil nuclei which are formed at the same time as the neutrons. Since these neutrons are not very energetic, they are not able to drive the recoil nuclei in the backward direc-

[5] Cf. 1. The bombarding energy used by these workers was less than 1 MV.
[6] Hafstad and Tuve, Phys. Rev. **47**, 506 (1935).
[7] Cockcroft, Gilbert and Walton, Proc. Roy. Soc. **A148**, 225 (1935).
[8] Haxel, Zeits. f. Physik **93**, 400 (1935).
[9] Oliphant, Kempton and Rutherford, Proc. Roy. Soc. **A150**, 241 (1935).
[10] Aston, Nature **135**, 541 (1935).

tion but simply deviate them from the path of the deuteron beam by an angle dependent on their energy.

The velocity of the recoil nucleus, V_R, may be found from the equations of conservation of energy and momentum:

$$V_R=\frac{1}{M_n+M_R}M_D V_D\cos\theta\pm\frac{M_n}{M_n+M_R}\left\{\frac{2M_D}{M_n^2}E_D\cos^2\theta+2\frac{M_n+M_R}{M_R M_n}\left[Q+E_D\left(1-\frac{M_D}{M_n}\right)\right]\right\}^{\frac{1}{2}}. \quad (5)$$

M, V and E are mass, velocity and energy, respectively, and the subscripts D, n and R refer, respectively, to the bombarding particle, the emitted particle, and the recoil nucleus; θ is the angle between the path of the deuteron and that of the recoil atom; and Q is the kinetic energy which appears in the reaction. The expression under the radical reduces to the form:

$$(1-\beta^2\sin^2\theta)^{\frac{1}{2}},$$

where $\beta^2=\dfrac{2M_R E}{(M_R+1)\{[(M_R-1)/(M_R+1)]E+Q\}}$.

If $\beta^2>1$ there will be an angle between 0° and 90° above which this term becomes imaginary. For reactions in which $Q<E$, approximately, there will be a maximum angle of deflection given by the equation

$$\sin^2\theta_{\max}=1/\beta^2$$

or
$$Q=E\left(\frac{2M_R}{M_R+1}\sin^2\theta_{\max}-\frac{M_R-1}{M_R+1}\right). \quad (6)$$

This maximum angle should be sharply defined and accurately measurable; the value of Q obtained from it is independent of range velocity data which are not very accurately known for heavy particles. The result is, also, largely independent of the number of charges on the recoil atoms and of their velocity after emerging from the target.

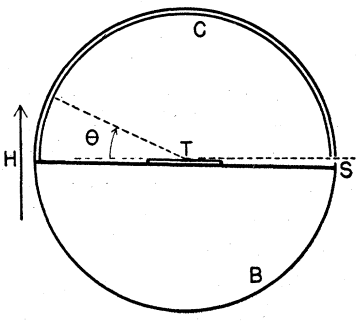

FIG. 3. Apparatus for measuring the angle of recoil of radioactive atoms. The tube is 4 cm in diameter.

The maximum angle of recoil was measured with the apparatus shown in Fig. 3. The deuteron beam enters the brass tube, B, through the slit, S, and strikes the target, T, at a glancing angle. The radioactive recoil atoms are collected by a metal strip, C, which lines the inside of the tube B. This arrangement collects only those atoms of which the component of velocity perpendicular to the beam is approximately parallel to the strong magnetic field H which must be present in this type of ion accelerator. The field cannot therefore change the angle θ greatly although very slow or highly charged recoil atoms are considerably bent by the magnetic field. In performing the experiment, the target was bombarded in vacuum for several half-lives of the substance. The metal strip was removed, cut into small slices perpendicular to its long side, and the activity of each piece was measured on the electroscope. The decay of some of the pieces was checked to be sure that the activity was due to the element studied.

THE ENERGY OF REACTION IN THE ACTIVATION OF CARBON

Because of the convenient half-life (10.4 minutes) and the great intensity of the activity of carbon, the first experiments were performed on a graphite target. Fig. 4 shows a histogram of the distribution of activity between 12° and 100°. The distribution between 80° and 180° was studied carefully in another experiment and was found to be constant within the accuracy of the measurements. This small activity might be ascribed to a weak group of neutrons of energy greater than $2E$, but the lack of any apparent dependence of the distribution on the angle leads one to suspect that the activity is due to some other cause. Since the total activity found in this

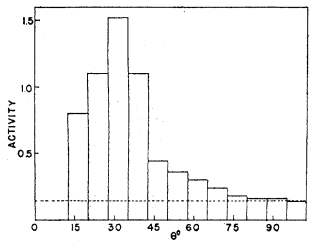

FIG. 4.

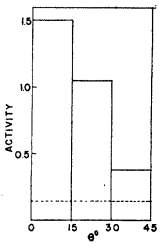

FIG. 5.

FIG. 4. The angular distribution of the recoil atoms from carbon. The dotted line is a background probably due to diffused activity.

FIG. 5. The angular distribution of the recoil atoms from carbon. The dotted line is again a background.

region is less than one percent of that of the graphite target, it is probable that enough charged atoms could escape from the target at thermal velocities and stick to the surrounding surfaces to account for the observed activity. The dotted line in Fig. 4 shows the magnitude of this background which should be subtracted from the observed activities at smaller angles. After correcting for the background, a "tail" of low intensity remains between 42° and 80°. Since cloud chamber experience shows that about ten percent of the observed tracks of heavy atoms are perceptibly bent, it seems likely that the tail is composed of atoms which have been scattered from the intense beam which is found at smaller angles. Scattering is nearly as probable in this experiment as in a cloud chamber since most of the recoil atoms must travel an appreciable distance below the surface of the target before they emerge. The rate of fall of the curve in this region is not inconsistent with the assumption that the active atoms have been scattered.

Taking the maximum angle of recoil as the high angle side of the region of intense activity, $\theta_{\max}=42°$, and $Q=-0.1$ MV. This value of Q may be compared with one calculated from the reactions:

$$C^{12}+D^2=C^{13}+H^1+Q_1, \tag{7}$$

$$N^{13}=C^{13}+e^+ +(e^-)+Q_2; \tag{7a}$$

combining these with the reaction

$$C^{12}+D^2=N^{13}+n^1+Q \tag{3}$$

we obtain $$Q=H^1-n^1-e^+-(e^-)+Q_1-Q_2. \tag{8}$$

Q_1 may be calculated from the data of Henderson, Livingston and Lawrence;[11] if it is assumed that no γ-rays accompany the protons, $Q_1=2.65$ MV. Q_2 may be taken as 1.3 MV, the maximum energy of the positrons from N^{13}, as measured by a number of workers.[12] Using the same masses as before we obtain $Q=0.1$ which is in excellent agreement with the measured value. The two values of Q obtained here are in reasonably good agreement with the measurements of Tuve and Hafstad[13] who find $Q=0.25$ MV for this reaction. The agreement of these three values of Q seems to be a good check on the reliability of this method; however, the measurements described here leave open the possibility of very weak groups of higher energy neutrons. Evidence for such groups has been found by Cockcroft, Gilbert and Walton, although the activities reported were very small. The measurements do show, however, that $Q\cong 0$ for the principal group of neutrons which agrees with the conclusions of Tuve and Hafstad.

THE ENERGY OF REACTION IN THE ACTIVATION OF OXYGEN

The angle of recoil of F^{17} was measured in order to check the estimate of the energy of activation of oxygen as obtained from its excitation curve. Because of the short half-life of F^{17}, this was a much more difficult experiment than the corresponding measurement on carbon. A piece of optical quartz was used as a target and the angle of incidence of the beam was reduced to less than 5°. It was necessary to check the decay of each section of the collector strip in order to eliminate activities due to silicon and to carbon contamination; only three sections could be measured in the available time.

The angular distribution so obtained is shown as a histogram in Fig. 5. This may be interpreted only by comparison with the corresponding distribution from carbon. Enough activity was found between 45° and 180° to account for a background of about the same intensity as that found in the carbon distribution; this background is shown as a dotted line in Fig. 5. If the back-

[11] Unpublished.

[12] Neddermeyer and Anderson, Phys. Rev. **45**, 653 (1934); Alichanow, Alichanian and Dzelepow, Nature **133**, 950 (1934); Cockcroft, Gilbert and Walton, cf. 7.

[13] Tuve and Hafstad, Phys. Rev. **48**, 106 (1935).

ground is subtracted from the curve, only a small activity remains in the region 30°–45°; this activity is of about the same intensity as that previously ascribed to scattering and it is probably due to the same cause. In short, the distribution found for oxygen would be nearly the same as that for carbon if the carbon curve were shifted about 15° toward smaller angles. The maximum angle of projection may, then, be taken as approximately 30° and from Eq. (6) $Q=-1.3$ MV. The agreement with the value $Q=-1.8$ MV as obtained with the excitation curve is as good as could be expected in an experiment of such low resolving power.

In conclusion, the author wishes to express his thanks to Professor Lawrence and to other workers in the laboratory for their assistance and advice in this work. The financial support of the Research Corporation and the Chemical Foundation is gratefully acknowledged.

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Rotational Structure of the Schumann-Runge Bands of Oxygen in the Vacuum Region

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Absorption bands of the Schumann-Runge system of oxygen have been photographed with a 3-meter vacuum spectrograph in the region λ1760–λ1925. A rotational analysis yields constants for the upper states which are new in the range $v'=8$ to 15 inclusive. Origins of the bands in this progression are represented within the limits of accuracy of the observations by the following equation:

$$\nu_{(v',\,0)}=49{,}014.93+700.360(v+\tfrac{1}{2})-8.0023(v+\tfrac{1}{2})^2-0.37535(v+\tfrac{1}{2})^3.$$

The energy of dissociation, D_0'', is 5.05 volts.

INTRODUCTION

A ROTATIONAL analysis of absorption bands of the Schumann-Runge system of O_2 obtainable in air at atmospheric pressure has been made by Curry and Herzberg,[1] who obtained data on the upper vibrational levels $v'=1$ to 7 inclusive. In the course of some experiments on absorption in the vacuum region of diatomic gases at low pressures, we obtained clearly resolved rotational structure of further members of the system, and have been able to extend the analysis to $v'=15$. The emission bands of this system all lie on the long wave-length side of the origin of the system and involve no upper levels with v' greater than 2; hence the absorption bands furnish the only available information about the higher levels of the upper electronic state, and we are reporting briefly the results of our analysis.

EXPERIMENTAL

The spectrograph used was designed by Professor H. E. White and built in the shops of the Physics Department. The design is similar to that of the 2-meter instrument described by Jeppesen,[2] except for the enlarged dimensions required to accommodate the 3-meter grating. The grating was ruled with about 14,000 lines per inch, at the National Physical Laboratory, in England. Light from the slit falls on the grating at a grazing angle of about 7 degrees, and the plateholder covers the range 0–3100A in the first order. Two oil diffusion pumps in series, backed by a Megavac pump, exhaust the 180-liter receiver to 10^{-5} mm in about one hour. During the absorption experiments, tank oxygen was admitted directly to the receiver at pressures ranging from 0.02 to 20 mm. At higher pressures in the receiver, the flow of gas through the slit to the discharge tube was too rapid to be balanced

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[1] Curry and Herzberg, Ann. d. Physik **19**, 800 (1934).
[2] C. R. Jeppesen, Phys. Rev. **44**, 165 (1933).