was no large difference in the absorption in nickel and copper. The magnitude of his experimentally determined absorption coefficients showed that the x-rays were from an element in this region but of atomic number less than that of zinc; since they could not reasonably be from nickel, they must be copper $K\alpha$ radiation.

The soft component in Fig. 6 consists of x-rays and secondary electrons as well as positrons; for this reason it is not possible to give a value for the upper energy limit for the positrons from these data. Barnes and Valley, from cloud-chamber measurements, give a range of 0.20 gram per cm^2 Al, corresponding to an energy of about 0.7 Mev.

The absorption curve in lead shows the gamma-ray to be reduced to half-value by 9.6 grams per cm^2 Pb, which indicates an energy of 1.0 Mev. The fact that the 0.5-Mev annihilation radiation is present to an inappreciable amount lends confirmation to the conclusion that most of the Zn^{65} atoms decay by K electron capture rather than by positron emission.

In an earlier paper[21] one of us reported an activity of 130 days half-life in deuteron-activated copper, after following the decay for three months. Continued observations for two years have shown the period to lengthen out to 222 days, so that the activity is doubtless that of Zn^{65}, formed in the same manner as when reported by Perrier *et al.*, but contaminated with some shorter lived impurity.

This research has been made possible by the generous support of the Research Corporation. We wish also to thank the W.P.A. for valuable assistance.

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The Emission of Secondary Electrons Under High Energy Positive Ion Bombardment

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The yield of secondary electrons from metallic surfaces produced by high energy positive ions has been measured for the energy range 43 to 426 kv. Cold targets of Mo, Pb, Al and Cu were bombarded by protons, H_2^+ ions and He^+ ions. For protons the yield decreased from approximately 4 at low energies to approximately 2 at high energies. For H_2^+ ions the yields were more nearly constant with energy and about equal to 6. No great dependence of the yield on the kind of metal used was detected. The yield of secondaries from Mo under He^+ bombardment was approximately 13 and varied very slowly with the energy. The great majority of the secondaries had energies less than 30 volts in all cases. The electrostatic generator and associated high voltage apparatus is of a type which has been described before. However, several new features in the design and operation are described in detail here. The proton source is of the low voltage, capillary type and delivers 50 μa of focused ions of which approximately 50 percent are protons. This arc has been in operation for a year without replacement.

INTRODUCTION

THE emission of secondary electrons from metallic surfaces bombarded by positive ions has been studied in detail by several investigators in the range below 2000 volts energy. Representative examples are given by the work of Oliphant and Moon[1] in which molybdenum was bombarded by singly charged helium ions, and the work of Healea and Chaffee,[2] in which nickel was bombarded by hydrogen molecular ions. In the region of higher energies there is the work of Linford,[3] who used mercury ions, and the recently announced work of Allen,[4] with protons.

It is the purpose of the present work to investigate the yield of secondary electrons from several

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[1] M. L. E. Oliphant and P. B. Moon, Proc. Roy. Soc. **A127**, 373 (1930).

[2] M. Healea and E. L. Chaffee, Phys. Rev. **49**, 925 (1936).

[3] L. H. Linford, Phys. Rev. **47**, 279 (1935).

[4] J. S. Allen, Bull. Am. Phys. Soc. **13**, No. 5, Abs. No. 17 (1938).

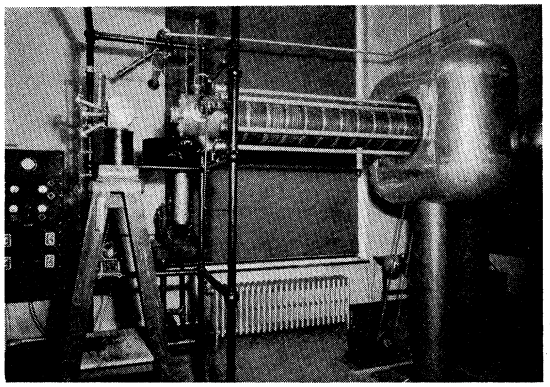

FIG. 1. Assembly.

metals when protons, hydrogen molecular ions, and singly charged helium ions are used as the primary particles at energies up to 426 kilovolts. Since the production of secondaries is an important factor to be considered in the design of high voltage discharge tubes, it was decided to study first the yield of secondaries at various primary energies under reasonably good vacuum conditions, such as obtain in the usual high voltage discharge tube, and leave for the future, work on thoroughly outgassed targets.

The electrostatic generator and associated equipment used in the present work is similar in construction to that which has been described in detail elsewhere.[5] However, several new features have been incorporated in the design, and these will be described in some detail.

THE HIGH VOLTAGE APPARATUS

The complete assembly is shown in the photograph, Fig. 1. The belt generator is external to the high voltage body and is of the type described by Bramhall.[6] The discharge tube is very similar to the tube described by Van Atta, Van de Graaff and Van Atta[7] and consists of eleven porcelain sections $10\frac{1}{2}$-in. inside diameter by 5 in. long, which contain the steel focusing electrodes. A schematic drawing of the tube is shown in Fig. 2. The power in the high voltage body is supplied by a belt-driven 110-volt 7-amp. alternator which is compact and allows a 50 percent capacity over the usual load requirements. The high voltage is regulated by means of a set of grounded corona points fastened to an adjustable lever arm shown at the top of the photograph.

The tube is evacuated by a steel mercury diffusion pump with a dry ice trap designed to hold 20 lb. of dry ice, which is sufficient for 24 hours operation. The net pumping speed for air at the outlet from the high voltage tube is 75 liters per second. This pump is backed by a glass diffusion pump of the design described by Copley, Simpson, Tenney, and Phipps,[8] which in turn is backed by a Cenco Hyvac. The tube is provided with a separate outlet for pumping the tube down to a rough vacuum by means of a Cenco Hypervac. To avoid the necessity of keeping dry ice on the traps whenever the pumps are not in use, a simple sylphon sealed valve is provided on the high vacuum side of the trap to prevent mercury from entering the high voltage tube. This pumping system has proved itself extremely reliable in operation and does away with the uncertainty regarding tube condition and surface contaminations sometimes attendant upon the use of oil pumps in high voltage work of this kind.

The tube vacuum is measured by a McLeod gauge, and is approximately 1×10^{-6} mm when the ion source is not in use. With the source turned on, the pressure in the tube rises to about 10^{-4} mm of Hg. All joints in the vacuum system are provided with guard rings wherever possible to enable rapid and reliable checking of the lead wires used as vacuum seals. It has been our experience that in a system having a large number of joints, including many which must be changed frequently, it is very desirable to be able to determine quickly and definitely which joint, if any, may be limiting the pressure.

The belt generator terminates in a two-foot sphere supported on a one-foot diameter Textolite column. Unless this column is of the proper conductivity, this type of assembly is characterized by a very high potential gradient at the joint between sphere and column, which gives rise to large corona losses. To reduce this gradient and hence eliminate these losses the following procedure was used very successfully. Four hundred I.

[5] L. C. Van Atta, D. L. Northrup, C. M. Van Atta and R. J. Van de Graaff, Phys. Rev. **49**, 761 (1936).
[6] E. H. Bramhall, Rev. Sci. Inst. **5**, 18 (1934).
[7] C. M. Van Atta, R. J. Van de Graaff, and L. C. Van Atta, Phys. Rev. **51**, 1013(A) (1937).
[8] M. J. Copley, O. C. Simpson, H. M. Tenney and T. E. Phipps, Rev. Sci. Inst. **6**, 265 (1935).

R. C. resistors of 20 meghoms each were soldered together and wrapped in a helix around the column from sphere to ground, roughly 27 turns in all. To space the adjacent turns, two strands of $\frac{1}{2}$-in. cotton rope, thoroughly dried and soaked in boiling beeswax, were wrapped concurrently with the resistance wrapping. The whole was then covered with a plastic mixture of beeswax and Venice turpentine to protect the resistors from corona currents. This method of equalizing the gradient raised the available voltage by approximately 30 percent. Furthermore, when calibrated in terms of a standard high voltage resistance this provided a means of measuring the voltage quickly and accurately. The current through this resistor is about 60 μa at 450 kv.

Up to the present time the maximum operating voltage has been 450 kv. This voltage is obtained with approximately 350 μa of charging current carried by the belt. Under normal conditions there are no visible corona losses to the walls of the room. The maximum voltage is evidently limited by leakage currents along the insulating columns, and can be raised by increasing the charging current. It has been noticed that this current can be increased by 10 to 20 percent by saturating the air in the belt compartment of the generator with carbon tetrachloride.

The room itself is somewhat small for this installation, being 19 ft. long by 13 ft. wide by 16 ft. high, with a minimum distance of 3 ft. from the high voltage body to the walls. When the charging current is raised much in excess of 400 μa corona currents appear, and if the voltage is to be raised still further the insulating properties of the air must be increased. This has been accomplished by saturating the air around the high voltage body with carbon tetrachloride as was done by Joliot, Feldenkrais and Lazard.[9] While this method has not been pushed to its limit, preliminary trials suggest that the limiting voltage obtainable in the future will be well over 500 kv. In order to screen the control part of the room from the carbon tetrachloride vapor, a rubberized canvas partition has been installed coincident with the iron framework shown in the photograph.

In an apparatus of this sort, where both the space and power available at high voltage are limited, the problem of a suitable ion source is particularly important. The source should give as high an ion purity as possible, should be small, reliable, and easy to start and operate, and should be as efficient in power consumption as possible. In addition, it should deliver the ions in a state which makes for ease of focusing, since auxiliary power supplies for probe and accelerating electrodes are necessarily bulky and power consuming. The type chosen for use in the present installation was the Pyrex capillary source described by Lamar, Buechner, and Compton,[10] and was constructed as shown in Fig. 3.

[9] F. Joliot, M. Feldenkrais, and A. Lazard, Comptes rendus **202**, 291 (1936).

[10] E. S. Lamar, W. W. Buechner, and K. T. Compton, Phys. Rev. **51**, 936 (1937).

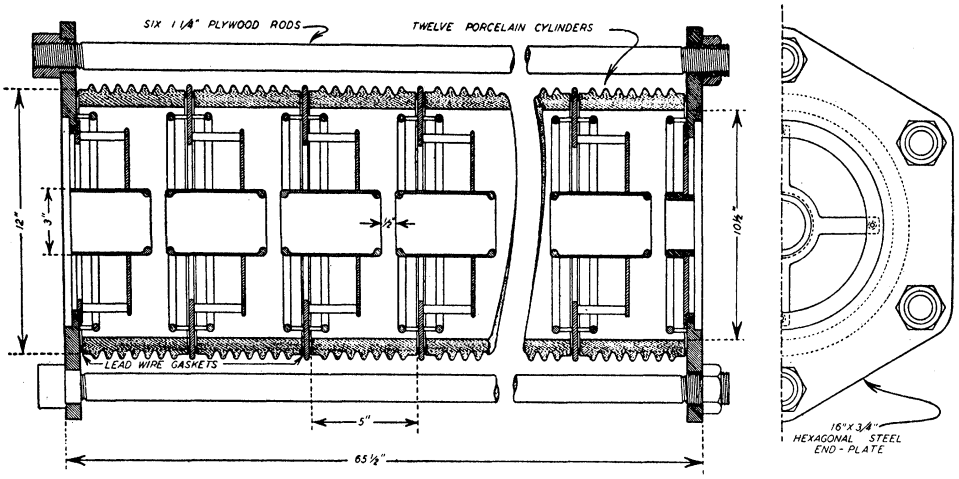

FIG. 2. Cross section of high voltage discharge tube.

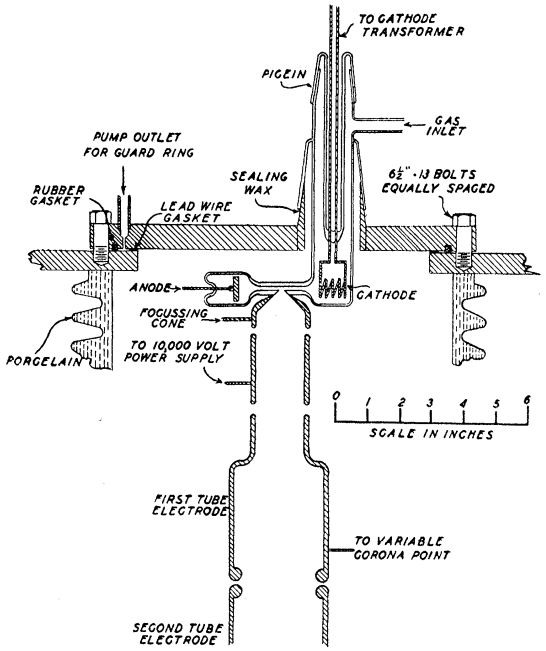

FIG. 3. Schematic diagram of ion source.

The capillary was constructed of heavy wall Pyrex capillary with an inside diameter of 3.5 mm having an outlet hole 0.5 mm in diameter in the center. This hole was beveled so that the hole itself had "zero thickness." Onto this capillary were blown chambers for the anode and cathode and the whole assembly was supported on a single glass grind, as shown. To facilitate replacement of cathodes the cathode leads were brought out on a grind sealed with Picein.

The source so constructed has been exceedingly reliable, and after approximately 12 months of steady operation the original assembly has never been removed. In operation the arc is usually run at 0.2 amp. with an arc drop of approximately 120 volts d.c., and under these conditions will deliver 60 μa of focused ions. The hydrogen pressure in the arc is kept at about 0.1 mm by means of a variable needle valve in the gas line. Tank hydrogen is used, the tank being at ground potential, and fed to the needle valve and source at slightly above atmospheric pressure through Pyrex tubing.

With a source of this type, the cathode is the main obstacle to steady and reliable operation. After some experimentation, we found that commercial uncoated ribbon as used in 866 tubes[11] wound into a helix and then coated with R-500 solution, were, when used at 2.5 v and 5 amp. very reliable and trouble free. (The R-500 solution is a barium carbonate-strontium carbonate suspension made by the Baker Chemical Company.) Cathodes of this type have useful lives of over 500 hours and, when they fail, a cleaning with HNO_3 and recoating is usually sufficient to restore emission properties.

As pointed out by Lamar, Samson and Compton,[12] there are numerous advantages to allowing the ions to drift out of the arc into a region of relatively high vacuum before applying a focusing voltage. This is particularly true with a nonmetallic source. In the present installation the first few high voltage electrodes were constructed as in Fig. 3. To provide a focusing field around the source outlet, a steel cone was placed as shown. The first cylinder was connected to a 10,000-v power supply Variac controlled in the high voltage body and the second cylinder was held at some definite unmeasured potential by means of an external corona point. The adjustment of the potential on the first two cylinders could be made from the ground end of the apparatus, thus enabling the operator to focus the beam to any desired size, without changing the tube voltage. With this arrangement the spot could be varied in diameter from 1 mm to about 2.5 cm without appreciable change in ion current. The spot size was determined by inserting a quartz plate into the line of the beam, by means of an external switch acting through a sylphon. Once the desired voltage and spot size have been set the three variables, voltage, current and spot size, remain very constant; no appreciable change was detected over periods of 15 minutes and more.

PROCEDURE FOR MEASURING SECONDARY EMISSION

The apparatus for measuring the yield of secondary electrons is shown schematically in Fig. 4. The positive ions which have been accelerated down the main vacuum tube are brought through the narrow tube, A, into a

[11] Obtained through the courtesy of Raytheon.
[12] E. S. Lamar, E. W. Samson and K. T. Compton, Phys. Rev. **48**, 886 (1935).

magnetic field chamber, B, for analysis. The ions of the desired mass are selected and passed through the tube, C, into the collector chamber. In this chamber are situated the target, P, set at 90° to the positive ion beam, a collecting cylinder, S, and an auxiliary collecting plate, D, of circular cross section with a hole through the center slightly larger than the diameter of the tube, C.

The magnet used was similar in design to the Cenco 79650 with 8-cm pole pieces producing a fairly constant field up to 7500 gauss across a 1.6-cm gap. The ion stream down the main vacuum tube was analyzed by means of this magnet, and under normal operating conditions roughly 50 percent of the total current was due to protons.

With the target at ground potential practically all the secondaries will be collected by the cylinder with a potential of +45 volts or above, as can be seen in Fig. 6. In measuring the yield of secondaries the following procedure was used. With the collector, S, at −90 volts the current to the target was measured. Under this condition all secondary electrons were returned to the target and a true positive ion current, I_p was measured. Then the collector potential was changed to +90 volts and the secondary electron current, I_s, to the collector was determined. As a check the current to the target, with +90 volts on the collector was then determined and this, as it should, equalled the sum of the positive ion current to the target and the secondary negative current leaving the target, I_p+I_s.

The purpose of the auxiliary collector, D, is to detect the presence of any secondary negative current from the tube, C, or any secondary currents from the target of sufficient energy to escape the collector. In all cases the currents to this auxiliary collector were much less than one percent of the positive ion current or the secondary currents. With a negative potential of 90 volts on the collector a minute positive current to the collector was noticed, but was sufficiently small to be neglected, i.e., about 0.1 percent of the target current. These positive currents may be reflected positive ions from the target or photoelectrons caused by soft x-rays striking the collector.

On installing a new target, pumping down the tube and beginning measurements it was found that the yield initially for any voltage was relatively high. After about two hours under high vacuum followed by bombardment for half an hour at 400 kv, the yields dropped as much as 50 percent to steady values which thereafter were quite reproducible. Evidently the preliminary bombardment serves to free the surface of some impurities the presence of which makes the initial readings unreliable. The results quoted below are actually average values of a large number of readings. The spread in values is roughly five percent of the average, providing the procedure mentioned above is followed.

Since the heating or degassing should be higher at higher energies we tried to minimize any cumulative heating effect by varying the voltage at random. This produced no noticeable change in the results. The readings for both H_2^+ ions and protons were taken concurrently, that is, with the tube voltage fixed the magnetic field current was changed to bring in the desired particle. The positive ion currents used were usually about 5 μa, with the focusing adjusted so that the beam was spread over a square cm area.

The targets were prepared by acid cleansing followed by polishing with emery, and were then washed in alcohol and distilled water. Several molybdenum targets were prepared in different ways, but in no case did this affect the results appreciably.

No especial effort was made to adapt the proton source for use with helium. Under the same conditions already described for protons the arc behaved very satisfactorily as a He^+ ion source giving currents of the same order of magnitude as for hydrogen. Commercial helium was used, which we attempted to purify by means of a misch-metal arc plus hot copper oxide to

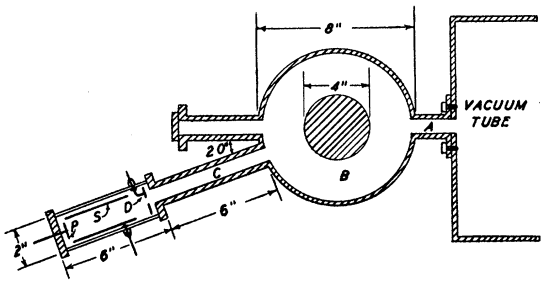

FIG. 4. Cross section of apparatus for measuring secondary electrons.

remove the hydrogen. Evidently, not all the active components were removed since the arc filaments became completely worthless after about five hours' use. On removal the filaments were found coated with a hard greenish compound, and no recoating restored the emitting properties. This deterioration of the filament also made for unsteadiness in the He^+ currents, which reduced the reliability of these measurements. The magnetic field was not strong enough at the highest energies used to make possible an adequate mass-spectrographic analysis of the He^+ beam. Protons and H_2^+ ions were present in small amounts. These ion groups decreased with time of use of the arc and must in part have been due to hydrogen left on the walls of the arc system. Since the magnetic field was insufficient to bend the helium particles through 20° for all energies, the measurements with helium were generally taken with the target directly in line with the tube, which has the disadvantage of including all ions of different mass. However to check the yields obtained with the unanalyzed beam, measurements were made at the low energies, 107 kv and below, with the helium beam bent through 20°. These readings checked within 10 percent of the readings made without magnetic analysis indicating that the effect of ion impurities in the helium was small.

Results

The yield of secondaries per primary for Mo, Cu, Al and Pb bombarded by protons, hydrogen molecular ions and He^+ ions over the range from 43 to 426 kv is summarized in Table I. For molybdenum, which was studied the most thoroughly, the results are plotted in Fig. 5.

TABLE I. *Yield of secondary electrons per primary particle from several targets at various energies.*

Metal	Energy (kv) 43	78	107	142	213	284	355	426
				Protons				
Mo		4.10	3.77	3.27	2.76	2.35	2.18	2.01
Cu		3.88	3.61	3.41	2.90	2.52	2.44	2.21
Al		4.19	3.82	3.51	3.01	2.68	2.19	2.17
Pb		4.23	3.87	3.56	3.12	2.89	2.70	2.50
				H_2^+ Ions				
Mo		6.31	6.59	6.40	6.19	5.56	5.45	
Cu		6.68	6.64	6.45	6.26	6.30	5.71	5.60
Al		6.08	6.30	6.23	6.43	6.29	5.98	5.46
Pb		8.24	7.74	7.58	7.46	7.34	6.99	6.61
				He^+ Ions				
Mo	11.1	12.8	13.3	13.9	14.1	14.3	14.3	14.1

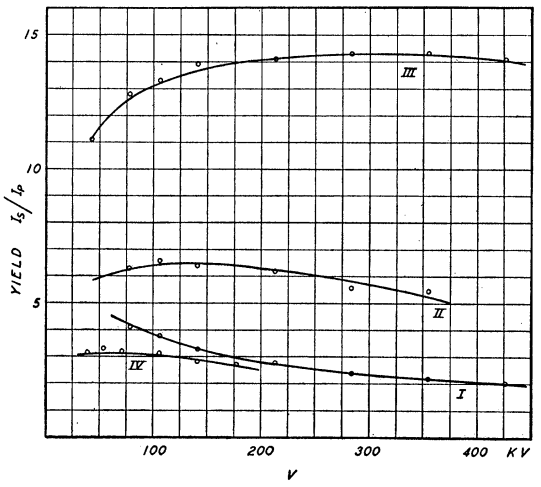

FIG. 5. Yield of secondaries per primary for molybdenum. I. protons; II. H_2^+; III. He^+; IV. yield/2 *vs.* $V/2$ for H_2^+.

For protons these results show a relatively rapid decrease in yield with increasing energy. This characteristic might be explained in a qualitative way by reference to the theory of stopping power of matter for high speed ions. As is well known the rate of energy loss increases with decreasing energy, hence more energy is lost near the surface by the low speed ions, and hence more electrons presumably can escape from the metal. However, this energy variation is not nearly so pronounced when H_2^+ and He^+ ions are used for the bombarding particles; and therefore it must be due to some property of the target or the bombarding protons. For both protons and H_2^+ molecules the yields show very little dependence on the type of metal used, which may indicate that the presence of foreign atoms on the surface is the controlling factor in our measurements.

The yield of secondary electrons when H_2^+ molecules are used as the bombarding particles shows much less energy dependence than the proton curves show. The yield is more nearly constant over the energy range and the average value is roughly twice that for protons. From a naive point of view one might expect the H_2^+ yield to be equal to that of two protons each of one-half the energy of the molecule, V, plus one

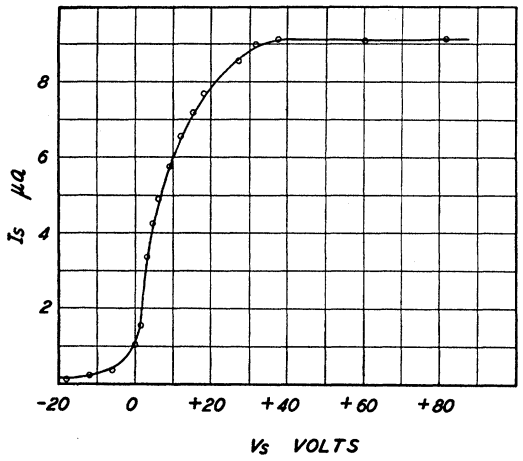

FIG. 6. Secondary current as a function of collector potential.

electron of energy $V/3680$, since the electron binding energy is small compared to the kinetic energy of the molecule. If we plot one-half the yield against one-half the energy (curve IV of Fig. 5) we should expect from this point of view that the difference between this curve and the proton curve would then be half the electronic yield at energy $V/1840$, where V is now the proton energy. This difference is seen to be negative, and this point of view of rather questionable validity.

The high yields obtained with helium are very interesting, and for the three particles used, indicate an average yield almost proportional to the mass of the bombarding particle. Here again it is instructive to plot $\frac{1}{4}$ the yield against $\frac{1}{4}$ the energy to compare with the corresponding H_2^+ curve. It is hoped to continue these experiments with heavier ions, possibly argon and nitrogen in the near future to investigate how much further such a relation holds. In this connection the work of Linford using Hg ions of between 0.7 and 2.3 Mev energy is very interesting. Linford found yields from various targets varying from 7 to 20, depending on the condition of the target. The yield curve for He^+ indicates a maximum in the neighborhood of 300 kv. For H_2^+ particles on Mo and Al there is some indication of a maximum around 100 kv, but no maxima were found for protons. Since a maximum must exist between zero and 78 kv (the lowest voltage for which protons could be focused in our apparatus), we see that the voltage at the maximum rises with the mass of the bombarding particle.

It is interesting here to note the results of Healea and Chaffee using H_2^+ ions. At 1600 volts they obtained a yield of 0.22 electron per primary ion for a heated nickel target, and 0.80 electron per primary for a cold target. Similarly, Oliphant and Moon, using He^+ ions at 1000 volts obtained a yield of 0.7 for heated and 1.2 for cold molybdenum targets. Below these voltages the yields of secondary electrons appear to increase linearly with the energy of the bombarding ion.

In order to get some idea of the energy of the secondaries the current to the collector (for a fixed positive ion voltage) was measured as a function of the voltage on the collector. The results of these measurements are shown in Fig. 6 for 213-kv protons on Mo. This indicates that the majority of secondaries have energies below 30 volts. Similar curves for other proton energies are identical with the one shown in the figure, and when H_2^+ molecules are used the figure is changed very little. No such curve was taken using He^+ particles as the arc was somewhat unsteady with helium and precluded any long measurements where both the voltage and positive ion current must be kept constant. However, the maximum energy of the secondaries was not far different from that observed for the protons and hydrogen molecules.

The question of the effect of surface impurities and their elimination is a very important one. In this type of apparatus there is necessarily present about 10^{-4} mm pressure of the gas of the bombarding particles. An ideal method of eliminating this gas, and any absorbed impurities, would be to enclose the target in its own vacuum chamber where it could be thoroughly cleaned up. This would require using a window between the two parts of the apparatus which would be rugged and still not slow down the particles to any great extent. For an apparatus limited to 450 kv this would necessitate a window of thickness equivalent to 1 mm of air for protons assuming a 50-kv loss. Since this is not feasible perhaps the best method would be to use heated targets with differential pumping as has been done by Oliphant and by Healea and Chaffee. An objection to this could be raised

in that there might exist a temperature effect to obscure the effect of impurities. This is unlikely since the temperature effect for electronically produced secondaries is small. While the quantitative interpretation of results for cold targets is difficult, nevertheless, the conditions under which such measurements are made make the results immediately applicable in the understanding of some high energy phenomena.

In the region below 300 kv energy, our measurements of the yield for copper are roughly 1.5 to 2 times higher than those obtained by Allen for heated targets. For cold targets Allen found the yield increased by about 50 percent. It is probable then that at least at low energies the yield is greatly influenced by gas molecules absorbed on the surface. As the energy is raised our yields begin to approach the yields found by Allen, although his maximum energy was less than ours, and the results are not strictly comparable. This decrease in yield with increasing energy may be due to the increased heating effect at the higher energies. In this connection we have already pointed out that our results were reproducible only after a bombardment of several hours at high energy, which evidently serves to clean up the surface until a stable point is reached.

The relative independence of the yield on the type of surface bombarded is difficult to interpret. It lends support to the point of view that for cold targets the surface impurities are the main factors in determining the yield, and that the base metal is of secondary importance. However for heated targets of C, Cu, N and Pt bombarded by protons Allen found this same independence of material, the yields being close to 2 in all cases over his energy range of 72 to 212 kv. Beryllium, it is true, showed a yield of over 7, but beryllium is the metal showing an abnormally high yield for electronically produced secondaries. In this case an oxide film is known to be the cause of the high yield as has been shown by Schneider,[13] and it is possible that this oxide film is causing the high yield found by Allen, even though the surfaces were heated during the bombardment.

In conclusion, the authors wish to express their thanks to Professor R. J. Van de Graaff and Professor E. S. Lamar for much helpful advice throughout the course of this work. We also wish to gratefully acknowledge a grant of the Research Corporation for aid in construction of the high voltage apparatus.

[13] E. G. Schneider, Phys. Rev. **54**, 185 (1938).

MARCH 1, 1939 PHYSICAL REVIEW VOLUME 55

A New Infra-red Absorption Band of Liquid Water at 2.52μ

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Cornell University, Ithaca, New York
(Received January 17, 1939)

The absorption of liquid water in the spectral region from 2.00 to 2.65μ was measured by means of a thermopile and glass prism spectrometer with a slit width of 100 angstroms in a search for a weak absorption band predicted by Ellis as a combination band occurring in liquid water but not in water vapor. A band was found with a peak at 2.52μ and with a maximum absorption coefficient of about 34. This is the fifth absorption band to be found in the spectrum of liquid water that has no counterpart in the spectrum of water vapor.

THE description of the liquid state of water by Bernal and Fowler[1] as a quasi-crystalline state and the elaboration of this idea by Cross, Burnham and Leighton[2] lends weight to the suggestion of Ellis[3] that certain absorption bands

[1] J. D. Bernal and R. H. Fowler, J. Chem. Phys. **1**, 515 (1933).

[2] P. C. Cross, J. Burnham and P. A. Leighton, J. Am. Chem. Soc. **59**, 1134 (1937).

[3] J. W. Ellis, Phys. Rev. **38**, 693 (1931).

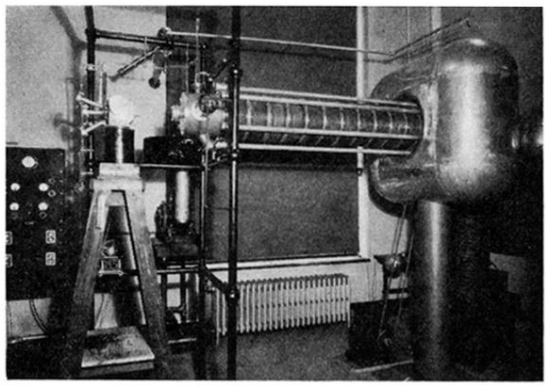

FIG. 1. Assembly.