

gest the possible existence of beta-beta-coincidences. A search was made for them, but after carefully eliminating all scattering effects arising from the 2.22-Mev beta-rays, none were found. This is not surprising, because the conversion electrons and associated soft nuclear beta-rays are very few in number as compared to the 2.22-Mev nuclear beta-rays.

Note added in proof: The "Table of Isotopes" compiled by G. T. Seaborg and I. Perlman [Rev. Mod. Phys. **20**, 585 (1948)], has appeared since this paper was presented for publication. In the table, reference is made to a report having only a restricted distribution by W. C. Peacock, J. W.

Jones, and R. T. Overman [Plutonium Project Report Mon N-432, p. 56 (Dec. 1947)]. Their value of 1.65 Mev is quoted in the isotopic table as the energy of the hard gamma-ray of Pr^{142} . This is in agreement with the deductions of the present paper.

ACKNOWLEDGMENTS

The writer wishes to express his particular appreciation to Henrietta Fricke of Bartol for having made the drawings for this paper and for the entire preceding series of papers dealing with pile-induced activities. He also acknowledges the continued interest of Dr. W. F. G. Swann, director of this laboratory.

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Radioactive Isotopes of Platinum and Gold

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Using the 60-inch Crocker Laboratory cyclotron, bombardments have been made of iridium and platinum with 19-Mev deuterons, of iridium with 38-Mev α -particles, and of platinum and gold with fast and thermal neutrons. Radioactive isotopes of iridium, platinum, and gold were chemically separated and the radiation characteristics studied.

THE results of a survey of the radioactive isotopes of platinum and gold have been reported previously.¹ Table I summarizes present knowledge of these isotopes. The present paper describes in more detail the experimental techniques used and the radiation characteristics of the radioactive isotopes observed.

I. EXPERIMENTAL

A. Bombardment

Using the 60-inch cyclotron at the Crocker Laboratory, bombardments were made of iridium with 38-Mev helium ions and 18-Mev deuterons, of platinum with 18-Mev deuterons, fast and thermal neutrons, and of gold with fast neutrons.

In all bombardments, reagent quality metals were used. Platinum and gold were bombarded as 2- and 5-mil foils. Iridium was bombarded as the powdered metal, in order to facilitate subsequent solution; in short bombardments where no chemical separations were made, iridium foil was used. The metal powder was tamped into the trough of a water-cooled copper "interceptor" target, which received about one-third of the total beam from the cyclotron. The

iridium was protected by wrapping the target with 0.2-mil tantalum foil.

After bombardment, platinum and gold targets were dissolved in boiling aqua regia. Iridium was dissolved by the procedure of Wichers *et al.*² The metal powder (100 mg) was transferred to a thick-walled glass tube, and an equal weight of sodium chlorate was added. While the tube was cooled in liquid air to prevent chemical reaction, 5 ml of fuming hydrochloric acid were added. After sealing, the tube was allowed to warm and then heated at 300°C for about three hours.

B. Chemical Separations

The chemical methods used in both the initial separations and in the separation of daughter activities were variants on the procedures outlined below. The steps were carried out in the given order, the usual radiochemical techniques being employed with milligram amounts of carriers present. "Holdback" and "scavenger" carriers were used at appropriate points, and separations were repeated until acceptable radiochemical purity was obtained.

¹ Geoffrey Wilkinson, Phys. Rev. **73**, 252 (1948).

² E. Wichers, W. G. Schlect, and C. L. Gordon, J. Nat. Bur. Stand. Research **33**, 363 (1944).

TABLE I. Summary of present knowledge on gold and platinum isotopes.

Isotope	Type of radiation	Half-life	Energy of radiation in Mev		Produced by
			Particles	γ -rays	
Pt ¹⁹¹	<i>K</i> , e^- , γ	3.00±0.02 days	0.5 e^-	<i>L</i> , <i>K</i> x-rays 0.57, 1.5	Ir- <i>d</i> -2 <i>n</i> Au ¹⁹¹ decay Pt- <i>n</i> -2 <i>n</i>
Pt ¹⁹³	<i>K</i> , e^- , γ	4.33±0.03 days	0.11 e^-	<i>L</i> , <i>K</i> x-rays 1.5, 0.18	Ir- <i>d</i> -2 <i>n</i> Pt- <i>n</i> - γ Pt- <i>n</i> -2 <i>n</i> Au ¹⁹³ decay Pt- <i>d</i> - <i>p</i>
Au ¹⁹¹	<i>K</i>	~1 day			Pt- <i>d</i> -3 <i>n</i> Ir- α -4 <i>n</i>
Au ¹⁹²	<i>K</i> , e^- , $\gamma\beta^+$	4.7±0.1 hr.	~0.4, ~15 e^- ~1.9 β^+	<i>K</i> x-rays, 2-3	Pt- <i>d</i> -2 <i>n</i> Ir- α -3 <i>n</i>
Au ¹⁹³	<i>K</i> , e^-	15.8±0.3 hr.	<0.2 e^-	<i>L</i> , <i>K</i> x-rays	Pt- <i>d</i> - <i>n</i> , 3 <i>n</i> Ir- α -2 <i>n</i>
Au ¹⁹⁴	<i>K</i> , e^- , γ β^+ (~3%)	39.5±0.5 hr.	0.3 e^- 1.8 β^+	<i>L</i> , <i>K</i> x-rays ~0.4 ~1.8	Ir- α -3 <i>n</i> , Pt- <i>d</i> -2 <i>n</i> , 3 <i>n</i>
Au ¹⁹⁶	<i>K</i> , e^- , γ	185±3 days	0.1 e^-	<i>L</i> , <i>K</i> x-rays ~1.6, ~0.19	Ir- α -2 <i>n</i> Pt- <i>d</i> - <i>n</i> , 2 <i>n</i> , 3 <i>n</i>
Au ¹⁹⁶	<i>K</i> , $\gamma\beta^-$ (~20%)	5.55±0.05 days	0.34 β^-	<i>L</i> , <i>K</i> x-rays 0.41, ~1.6	Pt- <i>d</i> - <i>n</i> , 2 <i>n</i> Au- <i>n</i> , 2 <i>n</i>
Au ¹⁹⁶	<i>K</i> or I.T.	14.0±0.3 hr.		x-rays	Au- <i>n</i> -2 <i>n</i>

1. Osmium was separated by distillation of the volatile tetroxide from nitric acid solutions, followed by reduction of the collected distillate to the metal by magnesium. This step was performed only in deutron bombardments of iridium. Experiments showed that no osmium produced was from impurities in platinum bombardments.

The absence of any ruthenium activities resulting from impurities in iridium and platinum was shown by distillation of the tetroxide from boiling perchloric acid solutions after removal of osmium. The collected distillate was reduced to the oxide by alcohol.

2. Gold was extracted by ethyl acetate from 2-6*N* hydrochloric acid solutions containing nitric acid. After removal of the solvent by evaporation, gold was precipitated as the metal by saturation of a hot 2*N* nitric acid solution with sulfur dioxide.

3. After removal of gold contamination by repeated solvent extractions, excess stannous chloride was added to the cold hydrochloric acid solution, and the orange-red platinum complex, presumably H₂PtCl₄, which is immediately formed, was extracted with ethyl acetate.³ The solvent was evaporated and platinum reduced to the metal by magnesium. Iridium and gold do not extract or interfere in this procedure even when present in large quantities. Palladium, however, behaves like platinum and is extracted, while rhodium reacts very slowly and extracts in small quantities. The absence of palladium contaminations was shown by the specific precipitation by dimethylglyoxime in 4*N* acid solution.

4. After removal of other activities, iridium was finally precipitated as the dioxide by the bromate oxidation method.⁴ Rhodium reacts similarly, but contaminating rhodium activities were shown to be absent by solution of the oxide in hydrochloric acid and reduction of rhodium to the metal by titanous chloride.

In iridium bombardments, where large amounts of the element were present, separation was made by the sparingly soluble ammonium iridium chloride; the solution, after removal

of platinum, was re-oxidized with nitric acid and saturated with ammonium chloride.

C. Measurements

The final precipitates were either washed with acetone and mounted as the metal or were dissolved in acid and aliquots of the solution evaporated for counting. Samples were mounted on microscope cover slides or on very thin mica.

"End on" type counters 7.5 cm high, 3 cm diameter, with 2- to 3-mg/cm² mica windows, filled with 10 cm argon and 0.5 cm alcohol were used; a Lauritsen type quartz fiber electroscopie was used to measure some very active samples. Measured counting rates were corrected for coincidences and for variations in counter efficiency by use of a uranium reference. The geometry of the shelves of the counter were calibrated using a UX₂ standard.

Radiation characteristics were studied by absorption methods using aluminum, beryllium, and lead foils. For the reported ranges of electrons, the total absorber thickness for removal was computed, i.e., added absorbers plus air gap and counter window. Electrons and soft electromagnetic radiation were distinguished by their differential absorption in aluminum and beryllium. The average energy of *L* x-radiation in this region is of the order of 10 kev with an absorption half-thickness in beryllium of ~800 mg/cm². Unless the electrons are very energetic and require several *L* x-ray half-thicknesses of beryllium for their removal, electrons can be removed by a beryllium absorber and the absorption of residual soft quantum radiation then

³ H. Wöbling, *Berichte* 67, 773 (1934).

⁴ R. Gilchrist and E. Wichers, *J. Am. Chem. Soc.* 57, 2565 (1935).

measured in aluminum. The absorption curve of electrons in aluminum is obtained by subtraction from the measured aluminum curve of the electromagnetic radiation background. This, in turn, is obtained from the measured aluminum absorption curve of this radiation; in appropriate correction for reduction in the measured intensity of L x-rays by the beryllium filter is made and the true absorption curve constructed. If the electrons have a range much less than the half-thickness of the L x-rays in beryllium, the former are most easily determined from a direct absorption measurement with beryllium screens. When the electrons and L x-rays have comparable absorption in beryllium, the problem of resolution is more complicated, requiring complete absorption measurements in both aluminum and beryllium and a knowledge of the expected energy of L radiation.

Harder electromagnetic radiation was determined by lead absorbers. The active sample was placed between beryllium absorbers sufficient to remove electrons, and a third beryllium foil, of adequate thickness to remove all secondary electrons emitted from the lead absorbers, was placed immediately below the counter window. The lead absorbers were sandwiched between the beryllium foils. In addition, the whole counter set-up was removed from the usual lead housing in order to

minimize scattering effects of γ -rays and secondary electrons from the walls.

For comparison with measured values, x-ray absorption half-thicknesses in beryllium, aluminum, and lead absorbers were calculated from the energies and relative intensities of the various L and K x-ray components, together with the absorption coefficients given by Compton and Allison.⁵ Because the critical absorption edges of iridium, platinum, and gold lie in the region of the heavy rare earths, hafnium and tantalum, measurements to distinguish between x-rays of these elements were not made, inasmuch as absorber materials were not available.

In the determination of ratios of electrons and electromagnetic radiation, corrections were made for absorption of electrons in the air gap and counter window and for counting efficiencies of quantum radiations. For radiations of energies between 30 keV and 0.5 MeV a counting efficiency of 0.5 percent was taken, with an increase of one percent per additional MeV energy. For softer radiations the efficiency was calculated from the counters used from the absorption coefficients in argon; the efficiencies at 7, 10, and 15 keV were respectively 12 percent, 2 percent, and 1 percent. No allowance was made for fluorescence yields of

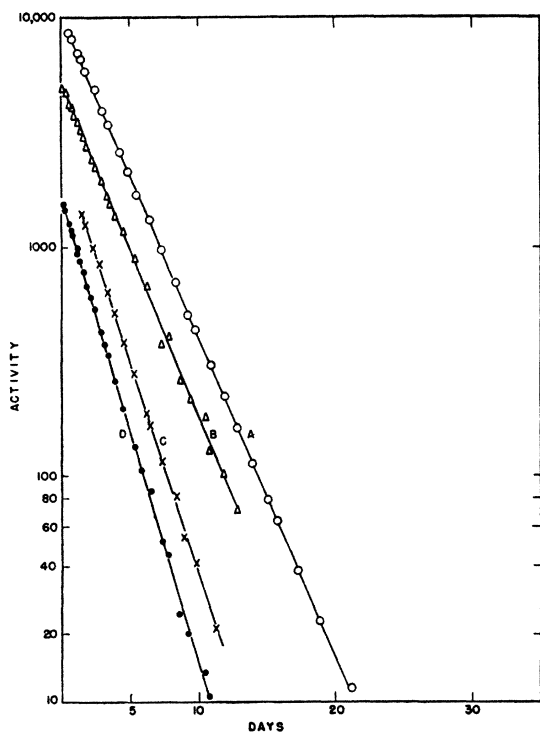


FIG. 1. Decays of 4.33-day Pt^{193} and 3.0-day Pt^{191} . (A) Gross decay of 4.33-day Pt^{193} from $\text{Pt}+d$ bombardment. (B) Decay of soft electron radiation from $\text{Ir}+d$, half-life 4.33 days. (C) 3.0-day Pt^{191} from $\text{Ir}+d$, γ -decay through 5 g/cm^2 lead. (D) Decay of hard electron radiation from $\text{Ir}+d$, half-life 3.0 days.

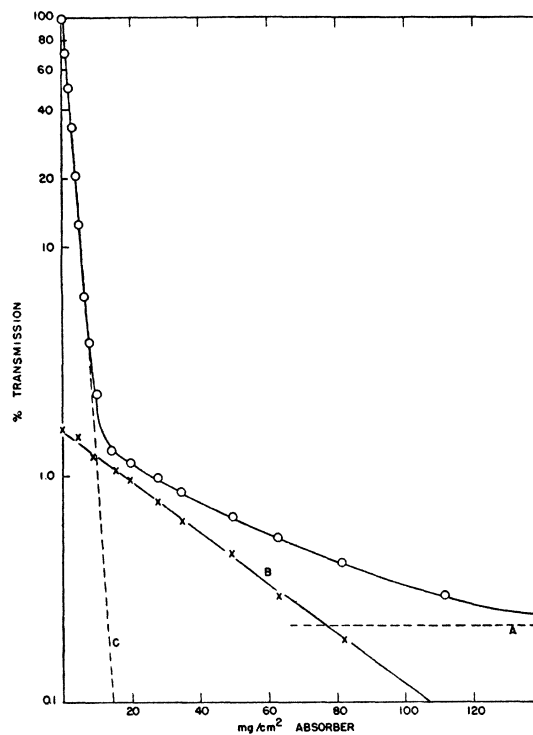


FIG. 2. Aluminum absorption of 4.33-day Pt^{193} from $\text{Pt}+d$. (A) K x-ray background. (B) L x-rays 28 mg/cm^2 Al half-thickness. (C) Electrons, total range 17 mg/cm^2 .

⁵ A. H. Compton and S. K. Allison, *X-rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York), second edition 1935.

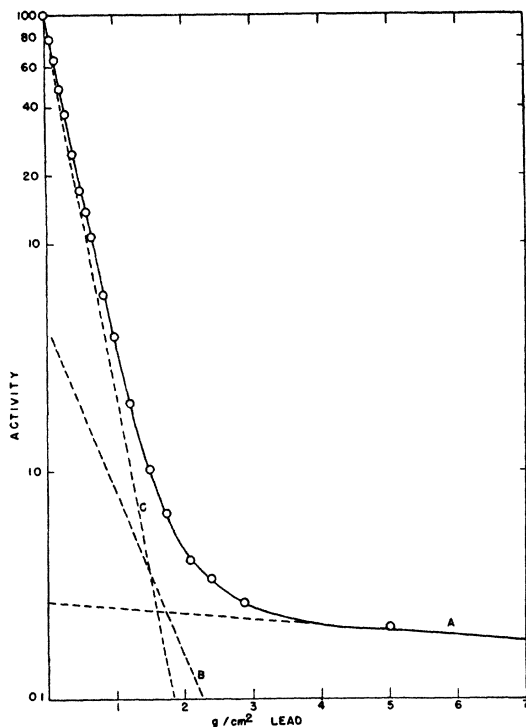


FIG. 3. Lead absorption of 4.33-day Pt^{198} from $Pt+d$. γ -ray half-thicknesses 15 g/cm^2 (A), 530 g/cm^2 (B), 185 mg/cm^2 (C) lead.

x-rays, which were assumed to be unity in the region studied. While it is realized that ratios so obtained may be in considerable error in view of the various assumptions made regarding counting efficiencies, etc., it seems justifiable to attempt to obtain an approximate idea of the radiations arising from one disintegration in order to allow calculation of reaction yields. Beam intensities were taken from the cyclotron instruments.

II. GENERAL SURVEY OF THE LONGER LIVED ACTIVITIES

A. Deuteron Bombardment of Iridium

Two platinum isotopes, with half-lives of 3.0 days and 4.33 days only are observed after chemical separation. The iridium fractions contain only the well-known Ir^{192} (60-day) and Ir^{194} (19-hr.)⁶ activities from d,p reactions on iridium.

B. α -Particle Bombardment of Iridium

Osmium and iridium fractions from bombardments of iridium with 38-Mev α -particles were almost completely inactive. The platinum fraction contained the 4.33-day activity with a small proportion of the 3.0-day isotope. The chemically separated gold fraction which contained most of the activity gave a complex decay (Fig. 6), which was resolvable into activities of 4.0 hours, 15.8 hours,

⁶ Glenn T. Seaborg, Rev. Mod. Phys. 16, 1 (1944).

39.5 hours, and 185 days. Chemical separations of platinum from the bulk of the gold fraction showed that the 4.33-day activity grows from a gold parent of half-life 16 ± 1 hours. Very low activities of the 3.0-day platinum were also found to grow in the gold sample and appeared to have a parent with a half-life of approximately 1 day.

C. Deuteron Bombardment of Platinum

Gold, platinum, and iridium fractions were chemically separated from platinum foils bombarded with 19-Mev deuterons for various times from a few seconds to several hours. The platinum

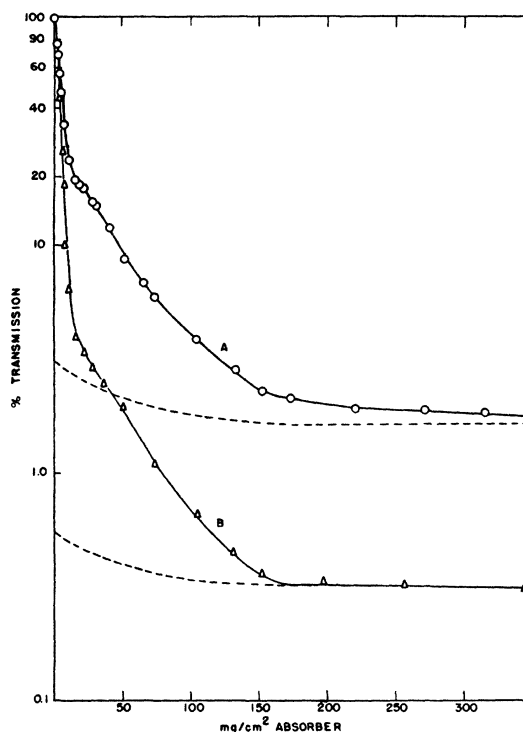


FIG. 4. Aluminum absorption of 4.33-day Pt^{198} + 3.0-day Pt^{199} from $Ir+d$. Measured at 0.5 day (A) and at 16 days (B) after bombardment.

fraction contained the β -active isotopes⁶ Pt^{199} (31-min.), and Pt^{197} (18-hour) produced by the d,p reaction. Chemical separation of the platinum fraction was repeated after decay of the shorter-lived activities. The platinum then contained the 4.33-day activity together with a small amount of the 3.0-day activity. No longer-lived isotopes have been observed. Both the well-known isotopes Ir^{192} and Ir^{194} are observed in the iridium fraction and probably arise from the reactions $Pt^{194}(d,\alpha)Ir^{192}$, $Pt^{196}(d,\alpha)Ir^{194}$. The saturation activities are approximately equal, in agreement with the abundances of Pt^{194} and Pt^{196} ; further, the activities are too large to be due to iridium impurity in the platinum used,

The resolution of the decay and absorption curves of the gold fraction from deuteron bombardment of platinum is complicated by the presence of β -emitting Au^{198} and Au^{199} . Short bombardments, however, produce the shorter-lived activities in a relatively greater yield and allow comparatively simple resolution. In long bombardments, the decay of hard γ -radiation shows the 39.5-hour activity; after decay of this isotope and also Au^{198} and Au^{199} , the gold fraction contains only 5.5-day and 185-day activities. Separation of daughter activities from the gold fraction confirms the data in α -bombardment of iridium.

D. Neutron Bombardments of Platinum and Gold

The only gold activities observed from $n,2n$ reaction were the previously reported⁸ 5.5-day isotope, together with a low yield of an isotope with a 14-hour half-life which is presumably an isomer.

Fast and thermal neutrons on platinum form the 4.33-day isotope but only in the fast bombardment was any evidence of the 3.0-day isotope seen.

III. PLATINUM ISOTOPES

A. 4.33-Day Pt^{193}

This isotope is formed both in deuteron and α -particle bombardments of iridium, in the deuteron

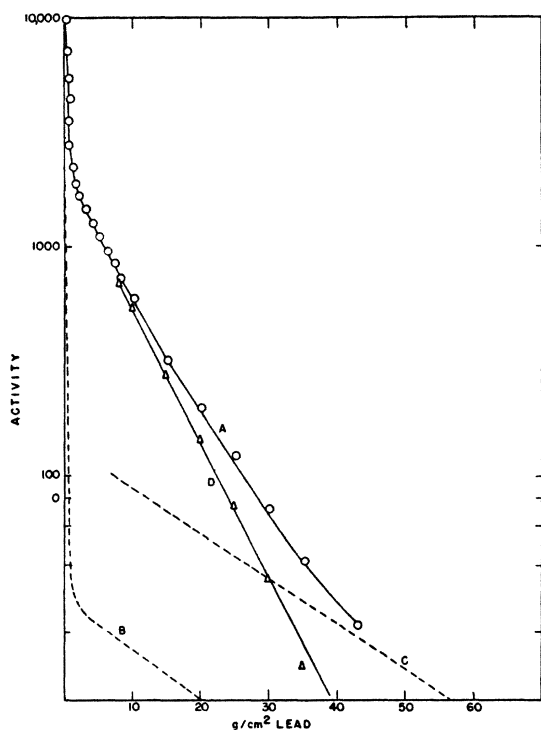


FIG. 5. Lead absorption of 4.33-day Pt^{193} +3.0-day Pt^{191} from Ir+d. (A) Measured absorption curve. (B) Calculated 4.33-day Pt^{193} contribution. Estimated hard γ -ray (C) and γ -ray of half-thickness 5.4 g/cm^2 lead (D) of 3.0-day Pt^{191} .

bombardment of platinum, and in both fast and thermal neutron bombardments of platinum. It has been also found to grow from a gold parent isotope with a 16-hour half-life. The radiations, all of which decay (Fig. 1) with the same half-life, are as follows:

1. Electrons of range $16.5 \pm 1 \text{ mg}/\text{cm}^2$, which corresponds to an energy of 110 kev. The absorption curve of these electrons together with the soft electromagnetic radiation background is shown in Fig. 2.

2. The electromagnetic radiation consists of four components. Aluminum absorption shows soft radiation of half-thickness $29 \pm 1 \text{ mg}/\text{cm}^2$, which corresponds well with the energy of 10.3 kev which would be expected for L x-rays of iridium or platinum. In the lead absorption of the harder components (Fig. 3), there is a small percentage of a very hard γ -ray of half-thickness $14 \pm 0.5 \text{ g}/\text{cm}^2$ lead, corresponding to an energy of 1.5 Mev. After subtraction of this component, the residue was still complex, having a noticeable tail. If it is assumed that the "tail" is attributable to a γ -ray of energy $\sim 180 \text{ kev}$ ($350 \text{ mg}/\text{cm}^2 \text{ Pb}$), a half thickness of $185 \pm 5 \text{ mg}/\text{cm}^2$ is obtained for the softer component. This agrees with that expected for iridium K radiation, which has an energy of 67 kev.

The ratios of electrons and electromagnetic radiation were determined from an "infinitely thin" sample mounted on mica and from lead absorption measurements. After making the corrections discussed above, the following ratios were obtained:

Electrons: L x-rays: K x-rays: 170 kev γ : 1.5 Mev γ
 $= 0.3:2:1:1 \times 10^{-2}:5 \times 10^{-3}$.

The measured energies of electrons and soft quantum radiation agree well with values reported by Krishnan and Nahum⁷ for a "2.9-day" activity produced by deuteron bombardment of platinum. A similar isotope has also been reported⁸ as having a half-life of 3.3 days. These activities were unquestionably due to the same isotope as is here observed. The gross decay was, however, measured through fourteen half-lives, the gross electromagnetic radiation through 8.5 half-lives, and the hard γ -radiation through 6.5 half-lives. A half-life of 4.33 ± 0.03 days was deduced from the various measurements. Similar values of the half-life were obtained for the isotope produced in neutron bombardments of platinum and in the deuteron bombardment of iridium.

In order to calculate the cross sections for formation in the various reactions, it has been assumed that the isotope decays by orbital electron capture and that one K x-rays quantum represents one disintegration. However, the observed ratio of L to K x-radiation of 2:1 is abnormally high and may indicate decay by L electron capture in addition to K capture, or γ -ray conversion in the L shell.

The isotope has been allocated to mass 193.

⁷ R. S. Krishnan and E. A. Nahum, Proc. Camb. Phil. Soc. **37**, 422 (1941).

⁸ E. M. McMillan, M. Kamen, and S. Ruben, Phys. Rev. **52**, 375 (1937).

It is produced in a yield comparable with that of the 3.0-day activity (see below) by deuteron bombardment of iridium. Assuming one K x-ray per disintegration and a counting efficiency of 0.5 per cent for this x-ray, the cross section for $\text{Ir}^{193}\text{-}d\text{-}2n$ reaction is ~ 0.07 barn; with the same assumptions, the cross section for formation of the 3.0-day activity by $\text{Ir}^{191}\text{-}d\text{-}2n$ reaction is ~ 0.09 barn.

In the deuteron bombardment of platinum, the cross section for the production of Pt^{193} by d,p reaction on Pt^{192} is ~ 0.05 barn. This value compares reasonably, in view of the unavoidable assumptions, with values of 0.08 to 0.1 barn for d,p reactions in iridium and platinum, where β^- -active isotopes Ir^{192} , Ir^{194} , and Pt^{197} are formed.

No iridium daughter of half-life greater than a few minutes has been detected, which is in agreement with the assignment to Pt^{193} .

B. 3.00-Day Pt^{191}

In the platinum fraction from deuteron bombardment of iridium, the isotopes Pt^{191} and Pt^{193} are to be expected in high yield from $d,2n$ reaction. The aluminum absorption curves (Fig. 4), after subtraction of electromagnetic radiation backgrounds, show electrons of ranges ~ 16 mg/cm², 145 ± 5 mg/cm², and ~ 400 mg/cm², corresponding to energies of 0.1, 0.5 Mev, and ~ 1 Mev, respectively. The decay of the soft electron was obtained by sub-

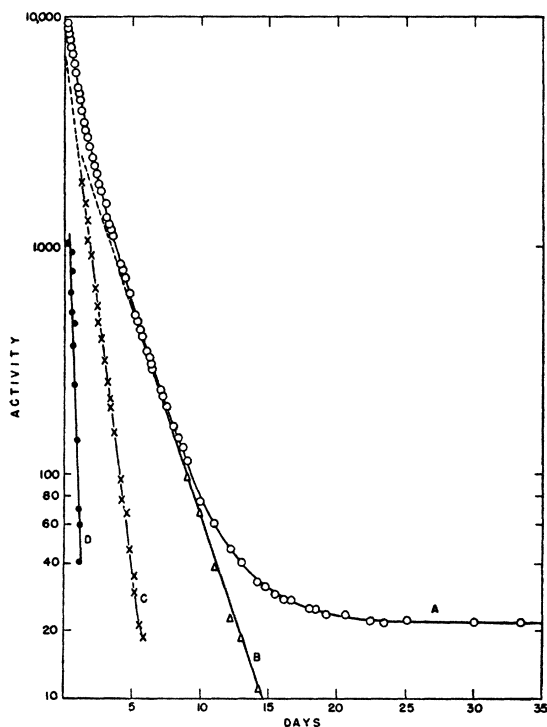


FIG. 6. Gross decay of gold fraction from $\text{Ir}+\alpha$ bombardment. A, B, C, and D are, respectively, the resolved decays of half-lives 185 days, 39.5 days, 15.8 hours, and 4.0 hours.

traction of electromagnetic and hard electron radiation from the gross activity, and a value of 4.35 ± 0.05 days measured through eight half-lives was obtained. The decay and electron energy of this radiation corresponds closely with the values obtained for the 4.33-day Pt^{193} described above and formed in the deuteron bombardment of platinum. The decay of the 0.5-Mev electron, obtained by subtraction of electromagnetic radiation background from the gross activities measured through a 20 mg/cm² beryllium absorber was 3.05 ± 0.05 days through seven half-lives. Since no 1-Mev electron was observed in the radiation of the 4.33-day activity, the small fraction of this electron present

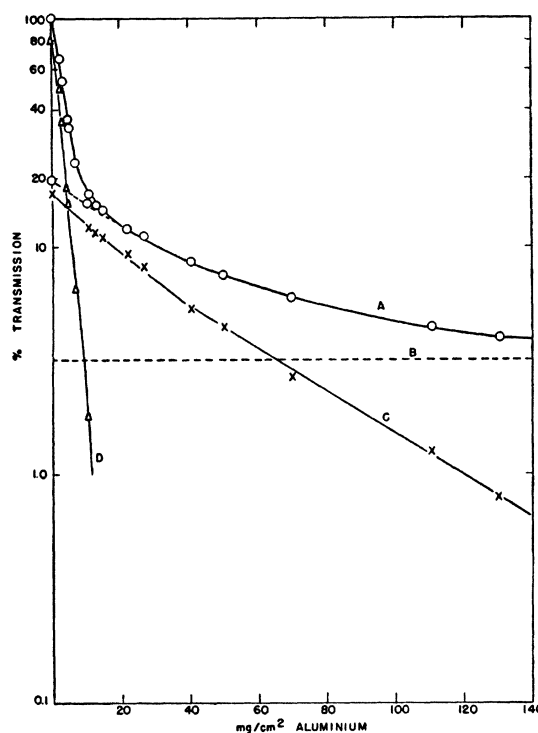


FIG. 7. Aluminum absorption of 185-day Au^{196} from $\text{Pt}+d$. (A) Measured curve. (B) K x-ray and γ -ray background. (C) L x-rays. (D) Electrons.

is assumed to arise from the 3.0-day activity. The various decay curves are shown in Fig. 1.

The hard γ -radiation had a gross decay of 3.00 ± 0.05 days through eight half-lives. This was consistent with the small contribution to the total quantum radiation to be expected from the electron activities of the 4.33-day isotope present. This contribution was computed and the absorption of the electromagnetic radiation of the 3-day activity obtained by subtraction. The aluminum absorption of the 3.0-day activity showed L x-rays while the lead absorption curve (Fig. 5) showed components of half-thicknesses ~ 190 mg/cm², 5.4 ± 0.2 g/cm², and 15 g/cm². With the heavier absorbers there was

a marked divergence from linearity of the 5.4-g/cm² line. The assumption of a small background of a γ -ray of ~ 15 g/cm² half-thickness corrected this, and it would seem that the 3-day isotope also has a hard γ -ray similar to that of the 4.3-day isotope. This assumption is supported by the fact that the contribution to the hard γ -radiation computed for the 4.33-day isotope is less than 0.2 of the activity which has to be subtracted to give a linear absorption for the 5.4-g/cm² ray. The latter, which corresponds to 0.57-Mev energy, is undoubtedly the unconverted γ -rays corresponding to the 0.5-Mev electrons.

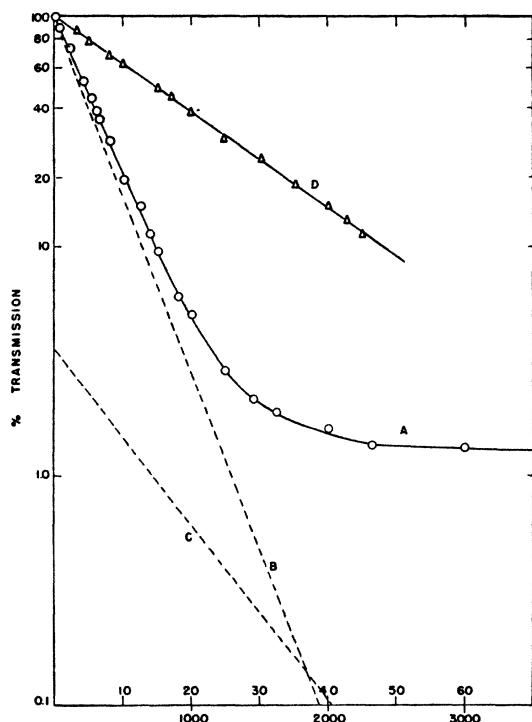


FIG. 8. Lead absorption of 185-day Au^{196} from $\text{Pt}+d$. *A*, *B*, *C* are components of half-thicknesses of 15 g/cm², ~ 400 g/cm², and 185 g/cm² lead. *D* is the measured absorption of hard γ -ray.

The ratio of electrons to quantum radiation for the 3.0-day activity has been computed from measurements on an "infinitely thin" sample mounted on very thin mica and from the lead absorption. The following ratios were obtained after all corrections had been made:

$$\begin{aligned} 0.1\text{-Mev electron} : 0.5\text{-Mev electron} : L \text{ x-rays} : \\ K \text{ x-rays} : 0.57 \text{ Mev } \gamma : 1.7 \text{ Mev } \gamma \\ = 2 \times 10^{-3} : 0.2 : \sim 1 : 1 : 0.2 : 1 \times 10^{-2}. \end{aligned}$$

The 3.0-day platinum activity is allocated to mass 191. It is produced with a cross section of ~ 0.09 barn assuming $\text{Ir}^{191}\text{-}d\text{-}2n$ reaction with 19-Mev deuterons; no iridium daughter of half-life greater than a few minutes has been detected. It is

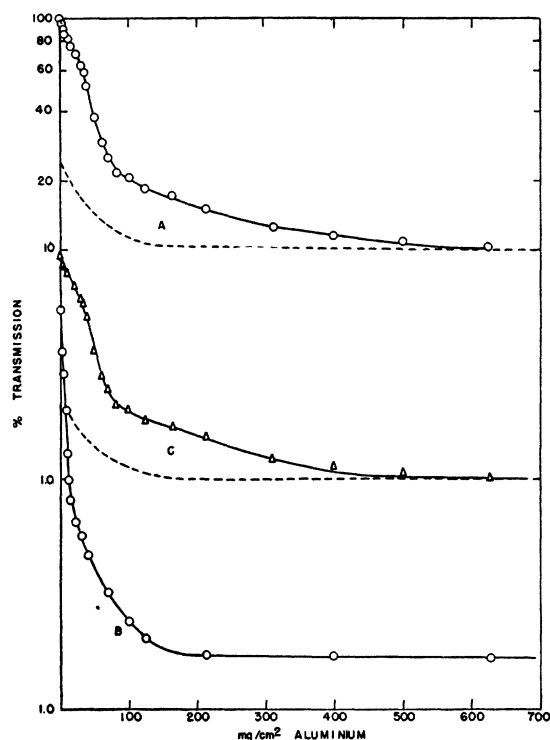


FIG. 9. Aluminum absorption of 39.5-hour Au^{194} and 185-day Au^{196} from $\text{Ir}+\alpha$. (*A*) Measured absorption of mixture at 8.5 days after bombardment. (*B*) Contribution of 185-day activity at 8.5 days after bombardment obtained from subsequent measurements. (*C*) Resolved absorption of 39.5-hour Au^{194} .

not observed in the thermal neutron bombardment of platinum. In the bombardment of platinum with fast neutrons from a $\text{Be}+d$ source, the platinum fraction, after decay of the shorter-lived 31-min. and 19-hour activities, showed a complex decay caused by a mixture of the 4.33-day activity with a small yield of the 3.0-day activity.

For 19-Mev deuterons, the $d,3n$ reaction on Ir^{191} might be expected to form Pt^{190} , since the $d,3n$ reaction with platinum and other elements in this region has a cross section ~ 0.1 that of the $d,2n$ reaction. However, no activity other than the 3.0- and 4.33-day activities were observed in the platinum fraction from deuteron bombardments of iridium, nor was the 10.7-day Ir^{190} ,⁹ which would be the daughter of Pt^{190} , found in the iridium fraction. From general considerations of nuclear stability, Pt^{190} is expected to be stable or very long-lived; from the present data, the half-life is greater than five hundred years.

IV. GOLD ISOTOPES

A. 5.55-Day and 14-Hour Au^{196}

The 5.55-day gold activity has been previously reported⁸ and its radiations have been studied by

⁹ L. J. Goodman and M. L. Pool, Phys. Rev. **71**, 288 (1947).

the β -ray spectrometer.¹⁰ The radiation characteristics of this isotope produced here in the deuteron bombardment of platinum and in the fast neutron bombardment of gold, and measured in the same geometry in both cases, agree well with previous data. McMillan, Kamen, and Ruben⁸ also reported that a 14-hour activity was produced in the fast neutron bombardment of gold. Measurement of the electromagnetic radiation from the present samples with sufficient beryllium to remove electrons showed a period of 14.0 ± 0.3 hours. The activity was not observed through a 2000-mg/cm^2 lead absorber, and presumably the 14-hour activity emits x-rays or soft γ -radiation. It was impossible to resolve the aluminum absorption and lead absorption curves for the 14-hour radiations because the 5.5-day Au^{196} and the 3.7-day Au^{198} formed simultaneously.

B. 185-Day Au^{195}

Cork and Halpern¹¹ reported a 164-day gold activity having a 0.45-Mev β -ray and 0.11-Mev γ -ray which they allocated to Au^{199} . Krishnan and Nahum⁷ have also reported a long period activity, but this was in too small an amount to characterize. The gold fractions from deuteron bombardments of platinum and α -particle bombardments of iridium measured in the same geometry, show, after decay

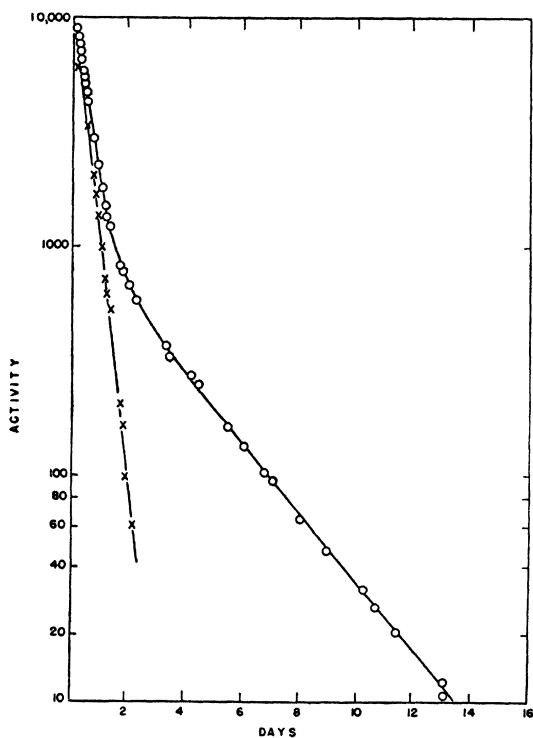


FIG. 10. γ -ray decay of 4.0-hour Au^{199} and 39.5-hour Au^{194} from $\text{Ir} + \alpha$ bombardment. Measurements made through 10 g/cm^2 lead absorber.

¹⁰ J. L. Lawson, and J. M. Cork, Phys. Rev. **58**, 580 (1940).

¹¹ J. M. Cork and O. Halpern, Phys. Rev. **58**, 201A (1940).

of the shorter periods, a long-lived activity whose radiation characteristics are identical in the two cases. A very active sample of the gold activity was obtained from a platinum target which had received a total of 490 microampere hours of 19-Mev deuterons and which had decayed for five months prior to separation. A sample of about 0.4 millicurie was obtained on four milligrams of gold carrier.

The radiations of the isotope consist of soft electrons of range $14 \pm 1\text{ mg/cm}^2$ aluminum, corresponding to 0.1 Mev, and electromagnetic radiation of half-thicknesses $29 \pm 1\text{ mg/cm}^2$ aluminum, $196 \pm 3\text{ mg/cm}^2$ lead, $\sim 400\text{ mg/cm}^2$ lead, and $14 \pm 0.5\text{ g/cm}^2$ lead, corresponding, respectively, to 9.3 kev, 69 kev, 190 kev, and 1.6 Mev. The two soft quantum radiations agree well with values for the L and K x-radiation of gold or platinum.

The gross decays of samples from both $\text{Pt} + d$ and $\text{Ir} + \alpha$ -bombardments, and x-ray and hard γ -ray decays of the very active gold samples have been followed for two years. All the radiations decay with the same half-life, the best value for which at present is 185 ± 3 days.

The isotope has been further examined on a crude β -ray spectrometer; no positrons, but only a peak of conversion electrons ~ 100 kev in energy were observed.

The ratios of the various radiations were obtained from aluminum absorption measurements (Fig. 7) on an "infinitely thin" gold sample, and from lead absorption measurements (Fig. 8). The following ratios corrected for counting efficiency, etc., were obtained.

$$\text{Electrons} : L \text{ x-rays} : K \text{ x-rays} : 190 \text{ kev } \gamma : 1.6 \text{ Mev } \gamma \\ = 1.5 : 3 : 1 : 3 \times 10^{-2} : 1 \times 10^{-2}.$$

As was observed with the platinum isotopes, the ratio of electrons and L x-radiation to K x-radiation is very high; this, and also the high yield of electrons, might arise from L electron capture or conversion of a γ -ray in the L shell. Since the decay scheme is somewhat indeterminate, bombardment yields were estimated assuming that one K x-ray represents one disintegration.

C. 39.5-Hour Au^{194}

After decay of the shorter-lived gold activities, the decay curve (Fig. 7) of the gold fraction from the α -particle bombardment of iridium shows only two components of half-lives 39.5 hours and 185 days. No 5.5-day Au^{196} , which could be formed by α, n reaction, was observed; the yield of an α, n reaction in this region at 38 Mev would be expected to be very small.

The 39.5-hour isotope has been further observed in high yield consistent with $d, 2n$ reactions in the gold fraction from deuteron bombardment of platinum; in this case, however, resolution of the

radiation characteristics is complicated by presence of the 5.5-day Au^{196} and 2.7-day Au^{198} .

The radiation characteristics of the isotope were determined by resolution of decay and aluminum absorption curves of the 39.5-hour and 185-day mixture from $\text{Ir} + \alpha$ -bombardments, and also of the gold fraction from short deuteron bombardments of platinum. The aluminum absorption curve (Fig. 9c) shows electrons of ranges $75 \pm 5 \text{ mg/cm}^2$ (0.3 Mev) and $\sim 800 \text{ mg/cm}^2$ (~ 1.8 Mev) aluminum. Using a crude beta-ray spectrometer, the harder electron has been shown to be a positron with a maximum energy of 1.8 Mev.

The softer electron has negative sign and may be a beta-particle or a conversion electron, or a mixture of both. The electromagnetic radiation of half-thicknesses $\sim 28 \text{ mg/cm}^2$ aluminum, and $15 \pm 0.5 \text{ g/cm}^2$ lead; complex radiations of intermediate energy can be resolved by assuming them to be a mixture of platinum K x-rays (195 mg/cm^2 lead) and γ -rays of energy 0.4–0.5 Mev. From the measurements the following ratios for the various radiations were obtained:

$$\begin{aligned} 0.3 \text{ Mev } e^- : 1.8 \text{ Mev } \beta^+ : L \text{ x-rays} : K \text{ x-rays} : \\ \sim 0.5 \text{ Mev } \gamma : \sim 1.8 \text{ Mev } \gamma \\ = 0.2 : 0.03 : \sim 1.5 : 1 : \sim 0.1 : 1.5. \end{aligned}$$

All the various radiations decay with the same half-life; the decay of electrons and positrons was followed through eight half-lives and electromagnetic radiation through six half-lives to give a value of 3.9 ± 0.5 hours.

Assuming that one K x-ray represents one disintegration by orbital electron capture, about 3 percent of the disintegrations of the 39.5-hour activity occurs by positron emission. The isotope is the heaviest positron emitter yet characterized.

D. 4.0 ± 0.2 -Hour Au^{192}

In the gold fraction from long bombardments of iridium with 38-Mev α -particles, an activity of 4–6 hours was observed (Fig. 7). The isotope has been produced in short α -particle bombardments of iridium foil and was studied without chemical separation. Essentially similar results for the half-life and radiation characteristics were obtained for the separated gold fraction from short deuteron bombardments of platinum. The aluminum absorption curves are somewhat difficult to resolve, even in short bombardments, as a result of the presence of other gold activities. Electrons of range $\sim 100 \text{ mg/cm}^2$ and with the 4.0-hour half-life. The electromagnetic radiation consists of K x-rays, and some soft γ -rays, but comprise mainly a very hard γ -ray of half-thickness $16\text{--}17 \text{ g/cm}^2$, corresponding to 2–3 Mev energy. The gross decay was followed through six half-lives and the decay of the very hard γ -radiation (Fig. 10) through seven half-lives

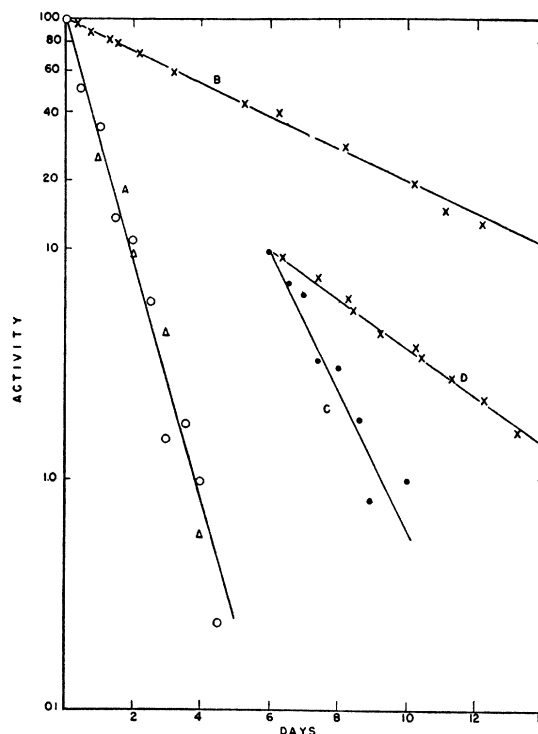


FIG. 11. Growth of Pt^{191} and Pt^{193} from Au^{191} and Au^{193} . (A) Decay of 15.8-hour Au^{193} as determined from "milked" activities of 4.33-day Pt^{193} . (B) Decay of "milked" 4.33-day Pt^{193} . (C) and (D) are similar curves for Au^{191} and Pt^{191} . Δ : data from $\text{Ir} + \alpha$ bombardments; \circ : data from $\text{Pt} + d$ bombardments.

to give a value of 4.0 ± 0.2 hours. The activity produced in short bombardments of iridium with 38 Mev α -particles was studied on a crude beta-ray spectrograph. The radiations were found to consist of positrons of maximum energy ~ 1.9 Mev. The negative electrons consisted of two distinct groups of maximum energies ~ 0.4 and ~ 1.5 Mev. The ratios of positron and negative electrons were obtained from the crude beta-ray spectrometer; comparing these values with those obtained from aluminum absorption measurements, and assuming that the gross electromagnetic radiation has a counting efficiency of 1 percent, the following ratios were obtained:

$$\begin{aligned} \sim 0.4 \text{ Mev } e^- : \sim 1.5 \text{ Mev } e^- : \sim 1.9 \text{ Mev } \beta^+ : \gamma\text{-radiation} \\ = \sim 0.07 : \sim 0.01 : \sim 0.01 : 1 \end{aligned}$$

Assuming that one quantum of the gross electromagnetic radiation represents one disintegration by orbital electron capture, then about 1 percent of the disintegrations occur by positron emission.

E. 15.8-Hour Au^{193} and ~ 1 -Day $\text{Au}^{191(?)}$

In both the deuteron bombardment of platinum and the α -particle bombardment of iridium, there should be produced Au^{193} which should be the parent of the 4.33-day Pt^{193} . Accordingly, in both

cases the bulk of the separated gold fractions were examined for growth of platinum daughter activities, and it was found that such growth did occur. The aluminum and beryllium absorption curves of the "milked" platinum activity, however, resembled that of the mixture of the 4.33-day Pt^{198} with a small percentage of the 3.0-day Pt^{191} activities. It was necessary, therefore, to follow separately the decays of the soft and hard electrons in the separated platinum fractions by use of appropriate beryllium absorbers. The energies of the soft and hard electrons in the aluminum absorption curve agreed with the values for the 4.33-day and 3.0-day activities, and the resolved decay curves for the two electrons confirmed this.

Chemical separations of platinum from the gold stock solution were made at equal time intervals under standard conditions.

The activities of the soft electrons, range 16 mg/cm^2 characteristic of the 4.33-day activity and the electron range 150 mg/cm^2 characteristic of the 3.0-day activity were determined from the absorption curve measured in standard geometry.

Corrections to the observed activities were made for chemical yield, i.e., the fraction of added platinum carrier counted, for decay of the activity between the time of separation and time of measurement and for self-weakening of the electrons in the various samples. In Fig. 11 activities of the platinum daughter activities, corrected to the time of extraction from the gold, are plotted as a function of time. The decay of these initial activities shows that the 4.33-day Pt^{198} grows from a parent with a half-life of 16 ± 1 hours. Separations were made over a period of seven days from the gold fraction produced in an 80-microampere hour bombardment of platinum with deuterons. The yields of the 3.0-day platinum and its parent, gold, were lower than those of the 4.33-day activity and its parent by a factor of ~ 15 ; measurements were consequently less decisive and the half-life of the gold parent can be placed only as about one day.

The 16-hour Au^{191} has been directly observed in the gross decay of the gold fraction from the 38-Mev α -particle bombardment of iridium (Fig. 8). The platinum daughter of this activity would be unobservable in the measured sample. Since the 5.5-day Au^{196} was not observed in the α -bombardment of iridium, it is then unlikely that the 15.8-hour activity observed was actually the 14-hour isomer of Au^{196} , formed by an α, n reaction. It has proved very difficult to determine the radiation characteristics of the 15.8-hour activity from absorption curves resulting from the presence of the other gold activities of not too dissimilar half-lives. The activity, has, however, x-radiation and soft electrons of range less than 50 mg/cm^2 ; no hard γ -radiation was observed to decay with a 15.8-hour half-life.

V. DISCUSSION

The gold activities have been allocated as in the accompanying Table I. The 5.5-day and 14-hour activities can unequivocally be allocated to mass 196 on the basis of their formation in the $n, 2n$ reaction on gold; the absence of the 5.5 day activity in the gold fraction from the 38-Mev α -particle bombardment of iridium is in agreement with this allocation since the α, n cross section at this energy is expected to be very low, i.e., less than $\sim 10^{-4}$ barn.

Since the 4.33-day and 3.0-day platinum activities have been allocated to masses 193 and 191, respectively, the respective 15.8-hour and ~ 1 -day gold parents are allocated similarly.

Assuming that one quantum of the 2- to 3-Mev γ -ray of the 4.0-hour activity represents one disintegration and that the counting efficiency of such a ray is two percent, the cross section for formation of the isotope in the deuteron bombardment of platinum by $\text{Pt}^{192-d-2n}$ reaction is ~ 0.1 barn, which agrees reasonably with order $d, 2n$ yields estimated; with the same assumptions, the yield for the $\text{Ir}^{191-\alpha-3n}$ reaction is 1.5 barns, again a reasonable value. Allocation to mass 192 agrees with the yields.

The 39.5-hour activity is allocated to mass 194 on the basis of reaction yields and absence of platinum daughter activity; in the α -particle bombardment of iridium, the yield, assuming $\text{Ir}^{193-\alpha-3n}$ reaction and that one K x-ray represents one disintegration, is ~ 1 barn; in the deuteron bombardment of platinum, Au^{194} could be formed both by $d, 2n$ and $d, 3n$ reactions, and the yields agree with the allocation.

The remaining unallocated isotope, the 185-day gold, is allocated to mass 195; the yield in the α -bombardment of iridium, for $\text{Ir}^{193-\alpha-2n}$ reaction, and assuming one K x-ray represents one disintegration, is 0.1 barn, which agrees reasonably with values expected for $\alpha, 2n$ reactions in this region. The present allocations agree reasonably with empirical rules for the stability of isotopes of odd atomic number and from comparison with half-lives of radioactive isotopes in similar positions of other heavy elements. The short half-life of 15.8 hours for Au^{198} , however, seems anomalous; no longer-lived isomer has, however, been detected, and no activities of the 4.33-day platinum could be separated from the 0.4-millicurie source of the long-lived gold from deuteron-bombarded platinum.

Since the work on gold isotopes was terminated and a preliminary note published,¹ Steffen *et al.*¹² have reported that gold activities of half-lives 39 ± 1 hours and 180 ± 10 days are produced in the bombardment of platinum with 7-Mev protons. The mass allocation and radiation characteristics re-

¹² R. Steffen, O. Huber, F. Humbel, and W. Zünti, *Helv. Phys. Acta* **21**, 195 (1948).

ported here were confirmed, but the latter, obtained from β -ray spectrometer and coincidence measurements, are given in greater detail than in the present paper.

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Boundary Conditions and Range of Force for S State of Two Protons

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The paper is divided into five sections the first of which is an introduction. In the second the possibilities of describing phase shifts by means of a boundary condition at a distance small compared with $e^2/mc^2 = 2.8 \times 10^{-13}$ cm is discussed. It is brought out that the irregular solution of the wave equation in a Coulomb field has a logarithmic infinity which masks the features of the wave function which have to do with phase shifts and therefore with observation. In Table I approximate values of essential quantities are listed. In Section III boundary conditions at moderate distances are studied. It is found that one can replace the "potential energy curve" description by the requirement that the logarithmic derivative of the wave function have an energy independent value at a distance of $\sim 0.47 e^2/mc^2$. Similarly the 1S proton-neutron interaction can be approximately described by requiring the logarithmic derivative to have an energy independent value at $\sim 0.49 e^2/mc^2$. In the convention of dealing with distance times radial function the values of the logarithmic derivatives are $\sim 0.08, 0.06$ for the proton and neutron cases, respectively.

I. INTRODUCTION

THE work reported on in this paper has two objects: (1) To investigate on an empirical basis the possibility of replacing the potential energy point of view for proton-proton scattering by boundary condition requirements; (2) to systematize the treatment of theoretically expected proton-proton scattering by bringing out the way in which different compact potential energy curves can give similar results and to make available convenient ways of adjusting the nuclear potential well parameters to experimental data.

The first of the two objects is related to the general desire of describing the collision process with a minimum of detailed hypothesis concerning the mechanism of the interaction. It has been brought out¹ that a description entirely by means of phase shifts is a possible one and that the isotropy of space implies certain restricting conditions on the

It is also possible to require a linear variation of energy for the logarithmic derivative within limits and to retain agreement with experiment. It is pointed out in the introduction that theoretical arguments for considering a failure of the potential energy viewpoint exist and that the agreement of the boundary conditions of Section III with observation may be more than an accident. In Section IV the adjustment of the range of force is treated and evidence for a somewhat smaller value than 2.8×10^{-13} cm, perhaps 2.6×10^{-13} cm is discussed. Use is made of simple relationships between effective depth variation with energy and range. In Section V the function f of BCP is expanded in powers of energy E , the relations for potential energy curves of different shapes are taken up regarding equivalence of range, the deviations from linearity of f with E are discussed from the viewpoint of equivalent error in scattering, and a rapid procedure for finding the equivalent square well range by means of successive approximations is given.

possible set of phase shifts. This point of view is closely related to Heisenberg's S matrix.² While it is possible to describe the collision process in such a generalized manner, it is difficult to make such a theory quantitatively specific and to establish relations between different phenomena such as those of proton-proton scattering and of the meson field. For this reason it appears advisable not to neglect an approach of intermediate generality with the hope that the investigation might eventually be of help in the formation of a theory of nuclear forces. The question of representing the proton-neutron interaction by means of a suitable boundary condition at zero distance between nuclear particles has been thought about by Wigner about 15 years ago³ but subsequent calculations on binding energies and other nuclear phenomena have pointed to the potential energy curve of finite width as the more promising temporary expedient. The possibility that the interaction between protons might be confined to distances of negligible amount on the

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¹ G. Breit, nuclear physics volume of the *University of Pennsylvania Bicentennial Conference*, N-15-15 (University of Pennsylvania Press, Philadelphia, 1941).

² W. Heisenberg, *Zeits. f. Physik*, **120**, 513, 673 (1943).

³ E. P. Wigner, private communication.