

## A Mass Spectrometric Investigation of Branching in $\text{Cu}^{64}$ , $\text{Br}^{80}$ , $\text{Br}^{82}$ and $\text{I}^{128}$

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An investigation of branching in certain neutron induced activities has been carried out by a mass spectrometric technique. Gram quantities of the elements in question were subjected to prolonged irradiation in a graphite moderated pile and were allowed to cool. The occurrence of negatron emission in the induced activities was identified with the presence in the irradiated sample of decay products of atomic number  $Z+1$ ; positron emission and orbital electron capture, with the presence of decay products of atomic number  $Z-1$ . The amounts of these decay products were determined by adding to a solution of the irradiated sample, as tracers, weighed milligram amounts of the daughter elements in a form isotopically *different* from the radiogenic material. When isotopic mixing had occurred, the daughter elements were isolated for mass spectrometric analysis. In such a procedure, quantitative recovery is unnecessary: the branching ratio can be determined from the change in the isotopic constitution of the tracers. The following values of the branching ratio  $\lambda_{\pm}$  (defined as the ratio of  $Z \rightarrow Z-1$  transitions to  $Z \rightarrow Z+1$  transitions) were determined:

$$\text{Cu}^{64} 1.62 \pm 0.11; \quad \text{Br}^{80} 0.090 \pm 0.002; \quad \text{Br}^{82} < 0.00027; \quad \text{I}^{128} 0.053 \pm 0.002.$$

In addition, the following neutron absorption cross section ratios were evaluated. For moderated pile neutrons:

$$\frac{\sigma_a(\text{Cu}^{63})}{\sigma_a(\text{Cu}^{66})} = 2.32 \pm 0.11; \quad \frac{\sigma_a(\text{Br}^{79})}{\sigma_a(\text{Br}^{81})} = 3.86 \pm 0.05.$$

### I. INTRODUCTION

AMONG the stable nuclear species now known, there are 62 even-even isobaric pairs which differ in atomic number by two and which are separated by an unstable odd-odd nucleus of intermediate atomic number. According to present theories of beta-decay, these intermediate nuclei are unstable with respect to both the isobaric transitions  $Z \rightarrow Z-1$  and  $Z \rightarrow Z+1$ . Hence branching may be expected to take place in the decay schemes of these nuclei to an extent which may or may not be experimentally observable, depending chiefly on the energies available for the alternate modes of decay. Experimentally, branching has been reported as definitely occurring in only 12 of these 62 cases. This fact alone is good evidence that the decay schemes of such nuclei deserve more careful study.

The purpose of the work described in this paper was to examine the branching properties of a group of these nuclei which can be formed by the  $(n, \gamma)$  reaction. All such nuclei are known to emit negatrons so that the efforts of this investigation are necessarily focused on the question of whether or not the processes of positron emission or orbital electron capture are concurrent. The mass spectrometric method of investigation, first employed for artificial activities by Hayden, Reynolds, and Inghram<sup>1</sup> in a study of neutron irradiated europium, takes advantage of the fact that, whereas negatron emission leads to product nuclei of atomic number  $Z+1$ , positron emission or orbital electron capture lead to product nuclei of atomic number  $(Z-1)$ ,  $Z$  being the atomic number of the irradiated element. The amount of a decay product in an irradiated sample, even though

present only in parts per million, can be measured with precision by use of the mass spectrometer and an isotope dilution technique.

Results for four nuclei are presented in this paper:  $\text{Cu}^{64}$ ,  $\text{Br}^{80}$ ,  $\text{Br}^{82}$ , and  $\text{I}^{128}$ . Of these,  $\text{Cu}^{64}$  has long been known to decay by all three possible modes and the triple branching scheme has been studied with counting techniques by many investigators.<sup>2-6</sup>  $\text{Br}^{80}$ ,  $\text{Br}^{82}$ , and  $\text{I}^{128}$  have been examined for positron emission by Barber<sup>7</sup> with a trochoidal apparatus. He was able to detect positrons only in the case of  $\text{Br}^{80}$ . No reliable evidence has existed heretofore on the question of orbital electron capture by these three nuclei.

### II. EXPERIMENTAL METHOD

Gram quantities of reagent grade chemicals, checked spectroscopically for impurities, were used for the irradiations. Special attention was paid to impurities of atomic number  $Z+1$  and  $Z-1$  which would decrease the sensitivity of the technique. The elements for study were irradiated in a chemical form which met the following requirements:

- The form must be a convenient one from the standpoint of the chemical operations which are to be carried out after the irradiation,
- the form must be chemically stable with respect to physical conditions prevailing during the irradiation,
- the elements in chemical combination with the element of interest must not be strong neutron absorbers,

<sup>2</sup> Bradt, Gugelot, Huber, Medicus, Preiswerk, Scherrer, and Steffen, *Helv. Phys. Acta* **19**, 219 (1946).

<sup>3</sup> C. S. Cook and L. M. Langer, *Phys. Rev.* **73**, 601 (1948).

<sup>4</sup> C. S. Cook and L. M. Langer, *Phys. Rev.* **74**, 1241 (1948).

<sup>5</sup> R. Bouchez and G. Kayas, *J. de phys. et rad. Ser. 8*, **10**, 110 (1949).

<sup>6</sup> Huber, Ruetschi, and Scherrer, *Helv. Phys. Acta* **22**, 375 (1949).

<sup>7</sup> W. C. Barber, *Phys. Rev.* **72**, 1157 (1947).

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<sup>1</sup> Hayden, Reynolds, and Inghram, *Phys. Rev.* **75**, 1500 (1949).

(d) the elements in chemical combination with the element of interest must not yield activities which will constitute a health hazard following the irradiation.

The samples were packed for irradiation in such a way that they could be unpacked by remote control. The samples in which rare gas decay products were sought were de-gassed before the irradiation and were packed in sealed quartz ampules to prevent any of the radiogenic gas from escaping.

After irradiation by moderated neutrons in a graphite moderated pile, the samples were allowed to cool for periods long in comparison with the half-lives in question. The samples were then unpacked by remote control and dissolved. The samples for rare gas analysis were outgassed in vacuum as will be described later. To the solutions there was added immediately a tracer solution containing weighed amounts of electromagnetically enriched isotopes of the elements  $Z+1$  and  $Z-1$ . Electromagnetically enriched isotopes were used as tracers, rather than normal materials, so as to be able to correct for possible chemical contamination; ( $Z+1$ ) or ( $Z-1$ )-impurities in the original material or in chemical reagents used subsequently always have normal isotopic constitution, and could be observed during the final isotopic analyses at a mass number different from that of the electromagnetically enriched tracer and that of the isobaric transition under investigation. When possible, the two tracer elements were weighed into the same container and dissolved together. Thereafter the *ratio* of the amounts of the two tracer elements present in the tracer solution was fixed (was unaltered by spilling for example) and it is this quantity which enters, finally, into calculations of the branching ratio.

When isotopic mixing had taken place in the joint solution of irradiated material and tracers, quantitative chemistry was no longer required. It remained only to determine the isotopic constitution of the elements  $Z+1$  and  $Z-1$  as present in the solution. This was done by isolating the elements, converting them to a chemical form suitable for mass analysis, and carrying out the analysis with a single focusing, 6 in. radius,  $60^\circ$  magnetic sector type mass spectrometer of standard design.<sup>8</sup> Ion currents were measured with a  $5 \times 10^{10}$  ohm resistor and vibrating reed electrometer; they were automatically recorded on a moving chart. The method of magnetic scanning of ion peaks was used throughout.

Since only a fraction of a milligram of the element was available for analysis, these measurements had to be made with great care. The chief source of error in measurements of small samples is the possible contribution of impurities to the mass spectrum. Thus extraordinary care had to be taken to keep the apparatus clean. The entire spectro meter was disassembled and re-built, prior to the series of measurements, with cleanliness as the prime object. New ion sources were

installed thereafter only after parts had been vacuum and hydrogen fired at  $1000^\circ\text{C}$ . Frequent baking of the entire spectrometer tube, while under vacuum, was carried out in order to cut background to a minimum. As a further safeguard against impurity errors, for each element other than nickel, which was ionized thermally, the mass spectrum was observed in several different positions; e.g., singly charged metal ions, doubly charged metal ions, singly charged oxide ions, etc. Results were accepted as valid only when measurements at the several positions gave identical results.

After a satisfactory measurement of the "branching sample" was completed, the tracer for that element was subjected to mass analysis using the same technique. The tracer or "blank," of which milligram quantities were available, was always examined second in case there should be spectrometer "memory" of the first sample.

### III. EXPERIMENTAL RESULTS

#### A. Copper

A quantity of neutron-irradiated copper metal was dissolved in nitric acid. To this solution there was added an aliquot of a tracer solution which had been prepared by dissolving weighed amounts of  $\text{Ni}^{58}\text{O}$  and  $\text{Zn}^{68}\text{O}$  in nitric acid. The combined solution was evaporated to eliminate excess nitric, diluted, and electrolyzed with platinum electrodes (rotating anode) until no trace of the blue cupric ion remained. The solution was made strongly acid with  $\text{H}_2\text{SO}_4$  and treated with  $\text{H}_2\text{S}$  to precipitate final traces of copper. The nickel in the solution was then isolated by the standard dimethylglyoxime precipitation. The zinc was isolated by precipitating  $\text{ZnS}$  from a weakly acid solution.

In order to examine the isotopic constitution of the nickel isolated in this way, the organic precipitate was digested in nitric acid and a portion of the resulting solution was pipetted onto a wolfram ribbon which served, when heated in the mass spectrometer, as a source of  $\text{Ni}^+$  ions. The ion peak at mass 64, negligible in the tracer nickel, was now prominent and could be compared with the principal peak at mass 58. The fact that the  $\text{Ni}^{60+}$  ions were no more abundant in this sample than in the tracer placed an upper limit of 0.2 percent on the amount of normal or contamination  $\text{Ni}^{64}$  present, and proved that the excess  $\text{Ni}^{64}$  was radiogenic in origin.

The zinc precipitate was converted to  $\text{ZnBr}_2$  by the action of hydrobromic acid. A portion, in aqueous solution, was pipetted into a small quartz crucible which rested in a wolfram heater directly above the ion box of an electron impact ion source. By raising the heating current, the  $\text{ZnBr}_2$  could be volatilized into the cross electron beam in a controlled way. Measurements were made on  $\text{Zn}^+$ ,  $\text{ZnBr}^+$ , and  $\text{ZnBr}_2^+$  ions. The amounts of  $\text{Zn}^{64}$  and  $\text{Zn}^{66}$  were greatly increased over those present in the tracer zinc; the constancy of  $\text{Zn}^{67}$

<sup>8</sup> A. O. C. Nier, Rev. Sci. Inst. 11, 212 (1940).

placed an upper limit on the amount of normal zinc present and proved that the excess  $Zn^{64}$  and  $Zn^{66}$  was due to negatron decay from  $Cu^{64}$  and  $Cu^{66}$  respectively.

From the mass spectrometer measurements and from the weights recorded in the preparation of the tracer solution, the branching ratio  $\lambda_{\pm}$ , which can be defined as the ratio of  $Z \rightarrow Z-1$  transitions to  $Z \rightarrow Z+1$  transitions, was calculated. The result was

$$\lambda_{\pm}(Cu^{64}) = \text{radiogenic Ni}^{64} / \text{radiogenic Zn}^{64} = 1.62 \pm 0.11.$$

The quoted probable error of 6.8 percent is the result of an analysis of all possible sources of error. The largest uncertainty in this particular case arises from an uncertainty in the abundance of  $Zn^{67}$  in the "branching" sample of zinc; this introduces, in turn, an uncertainty in the amount of  $Zn^{64}$  present due to contamination. The cause of the uncertainty was the presence of small amounts of impurities which contributed a non-negligible ion background at mass 67.

It was also possible to calculate the ratio of the neutron absorption cross sections of  $Cu^{63}$  and  $Cu^{65}$ . For pile neutrons:

$$[\sigma_a(Cu^{63})] / [\sigma_a(Cu^{65})] = 2.32 \pm 0.11.$$

This is in poor agreement with Seren's<sup>9</sup> value of  $1.5 \pm 0.4$  obtained by activation. The probable explanation for this discrepancy is that Seren's counting efficiency for the  $K$  captures in  $Cu^{64}$ , which constitute about 43 percent of the total disintegrations, was low.

In order to check on the over-all accuracy of the isotopic dilution method, a standard solution containing normal nickel and normal zinc in known proportions was prepared by weighing and dissolving gram quantities of these metals. A small fraction of this solution was then combined with a second aliquot of the  $Ni^{58}-Zn^{68}$  tracer solution used in the branching study. The nickel and zinc in the combined solution were isolated and subjected to mass analysis as before. The constants of the tracer solution and the mass spectrometer results were then used to calculate the ratio of  $Ni^{60}$  to  $Zn^{66}$  in the standard solution. The result checked the true value within 2.5 percent.

## B. Bromine

For this investigation, a sample of neutron irradiated sodium bromide was processed in two weighed parts. The larger fraction was dissolved in water and combined with a weighed quantity of electromagnetically enriched  $Se^{78}$  in nitric acid. Excess nitric acid oxidized any metallic selenium present in the irradiated salt so that isotope mixing occurred on an atomic scale. The solution was then evaporated to dryness over the water bath in order to destroy nitric acid and was redissolved in 7.5 percent hydrochloric acid. Metallic selenium was precipitated by treating the solution with  $SO_2$ . The precipitate was filtered, washed, and dissolved in nitric

acid. The selenium was reprecipitated from 18 percent HCl prior to mass analysis in order to increase the purity.

Mass analysis was carried out by depositing a nitric acid solution of the selenium in a small quartz crucible and following the same procedure that was used with zinc. Measurements were made on  $Se^+$ ,  $SeO^+$ , and  $SeO_2^+$  ions. A comparison of the mass spectrum of this sample with that of the  $Se^{78}$  tracer, analyzed immediately thereafter, showed a pronounced enrichment at mass 80 due to positron emission and/or orbital electron capture by  $Br^{80}$ . There was no enrichment at mass 82, showing that  $Br^{82}$  decays almost entirely by negatron emission. The abundance of  $Se^{76}$  was unchanged over that in the tracer, proving that less than 0.3 percent of the  $Se^{80}$  present was due to contamination by normal selenium.

The second smaller fraction of NaBr was processed in the apparatus shown in Fig. 1. This is a greaseless vacuum system designed specifically for the manipulation of rare gases and patterned after an arrangement used by Epstein and co-workers.<sup>10</sup> Symbols V1 through V5 indicate mercury valves which, normally open, may be closed by raising the level of the appropriate mercury column. C1 and C2 are activated charcoal traps for gas handling; C3 is an activated charcoal trap in which the final gas sample is isolated and sealed off. The special McLeod gauge at the right hand end of the system has been provided with an enlarged precision capillary so that amounts of gas up to  $0.4 \text{ cm}^3$  can be measured accurately. With the apparatus arranged as in Fig. 1, the system was degassed by pumping while baking C1, C2, and C3 at  $450^\circ\text{C}$ . After raising V1, V3, and V5 so as to isolate the system from the pumps, a small amount of normal krypton, which served as the tracer, was introduced into the system from the gas pipette, after which the gas pipette was sealed off, completely isolating the system from stopcock grease and possible extraneous krypton. By cooling C2 with liquid air for several minutes, raising V4, and then heating C2 to  $100^\circ\text{C}$ , most of the tracer krypton was transferred to the bulb of the special McLeod gauge where it was isolated by compressing the gas in the bulb. The remainder of the system was then degassed by lowering V1, V3, and V5 to resume pumping and by baking C1, C2, and C3 at  $450^\circ\text{C}$  for 12 hours. The amount of tracer krypton stored in the McLeod gauge was measured to within 0.5 percent, meanwhile, by making repeated readings of the McLeod mercury levels.

The isotopic mixing was carried out by isolating the system from the pumps, transferring the tracer krypton to the left hand side of the system by cooling C1 and raising V4, breaking open the sealed ampule containing the irradiated salt, and allowing the gases to mix. To

<sup>9</sup> Seren, Friedlander, and Turkel, *Phys. Rev.* **72**, 888 (1947).

<sup>10</sup> S. Epstein, *Proc. Conf. on Nuclear Chem.*, McMaster University, pp. 108-116 (May 1947).

make sure that all of the radiogenic gas mixed with the tracer gas, the temperature of the salt was raised until it distilled out of the ampule and refluxed vigorously down the walls of the outer quartz tube. The final gas mixture was purified by volatilizing calcium in the calcium furnace: calcium forms solid products with all but the rare gases. Purification was considered complete when the gas showed no reduction in volume, as measured in the McLeod, after successive treatments to calcium vapor. The final sample of purified krypton was isolated in the sample tube by cooling C3 and sealing off.

A mass analysis of the krypton so obtained was carried out by the standard technique of passing the gas through a small leak into the source region of the mass spectrometer. A greaseless spectrometer sample handling system was used so that there could be no possibility of contaminating the gas sample with other gases absorbed in the grease. The mass spectrum of the krypton showed heavy enrichment at masses 80 and 82 and measurements of the abundance of these isotopes were made by comparison with  $\text{Kr}^{83}$  and  $\text{Kr}^{84}$ .

With these data, the branching ratios for  $\text{Br}^{80}$  and  $\text{Br}^{82}$  were calculated:

$$\lambda_{\pm}(\text{Br}^{80}) = \text{radiogenic Se}^{80} / \text{radiogenic Kr}^{80} = 0.090 \pm 0.002;$$

$$\lambda_{\pm}(\text{Br}^{82}) = \text{radiogenic Se}^{82} / \text{radiogenic Kr}^{82} < 0.00027.$$

The data also yield the ratio of the neutron absorption cross sections of  $\text{Br}^{79}$  and  $\text{Br}^{81}$ . For pile neutrons:

$$[\sigma_a(\text{Br}^{79})] / [\sigma_a(\text{Br}^{81})] = 3.86 \pm 0.05.$$

This is in fair agreement with Seren's<sup>9</sup> value of  $4.8 \pm 1.4$  obtained by activation.

### C. Iodine

The procedure with irradiated NaI was very similar to that with NaBr. Again the sample was processed in two parts.

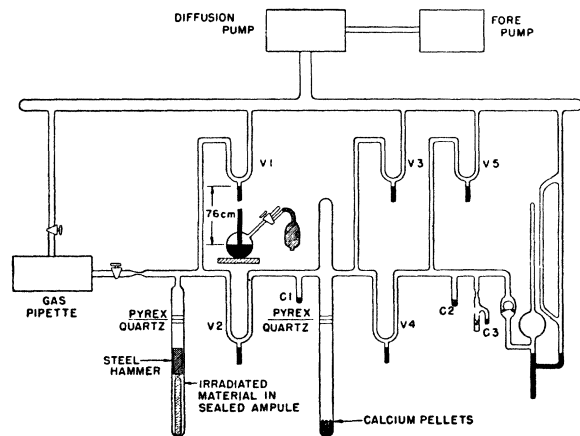


FIG. 1. Apparatus for the extraction and purification of radiogenic noble gases from neutron irradiated salts.

A tracer solution of  $\text{Te}^{130}$  in HCl was added to the solution of the irradiated salt, and, although tellurium appears to be reduced by the iodide ion, it remains in solution under these conditions so that isotope mixing takes place. Metallic tellurium was precipitated, after stirring, by neutralizing with  $\text{NH}_4\text{OH}$  and treating with  $\text{SO}_2$ . The mass analysis of the tellurium was carried out in the same way as with selenium. A reproduction of the mass spectrometer recorder tracings for both the "branching sample" and the tracer is shown in Fig. 2. The curves have been normalized at mass 130 for ease of comparison. As is evident on inspection, there has been considerable enrichment at mass 128. The constancy of the peak at mass 126 (not evident in Fig. 2) rules out the presence of normal tellurium and proves that the excess  $\text{Te}^{128}$  is radiogenic in origin.

The xenon procedure, utilizing the apparatus shown in Fig. 1, was identical with that for krypton. A small amount of normal xenon was used as a tracer. A mass spectrometer recorder tracing of the xenon obtained is shown in Fig. 3 along with the mass spectrum of normal xenon. The huge enrichment at mass 128 is evident upon inspection.

From the data the branching ratio of  $\text{I}^{128}$  was calculated:

$$\lambda_{\pm}(\text{I}^{128}) = \text{radiogenic Te}^{128} / \text{radiogenic Xe}^{128} = 0.053 \pm 0.002.$$

### IV. DISCUSSION

The outstanding advantage of branching investigations of this type is the great sensitivity of the mass spectrometer method for detection of orbital electron capture. In a conventional study, using ion detectors, orbital electron capture is an elusive phenomenon, detectable only by secondary events such as nuclear  $\gamma$  rays, x-rays, or Auger electrons. In the present study, the sensitivity of detection for capture is equal to that for electron emission since the primary event—nuclear transformation—is the one observed.

Another advantage of the method is the precision obtainable. The branching ratios quoted here have an average probable error of 4.3 percent; the ultimate accuracy of the method is probably about 1.5 percent. Since relative measurements can be made precisely with ion detectors, it is probable that results obtained by the mass spectrometer method will be useful as standards. The mass spectrometric method is free of the common errors due to absorption and scattering of radiation, counter insensitivities and end effects, background, and the presence of impurities in the activities under investigation.

The obvious disadvantage of the method is its lack of discrimination between, (a) orbital electron capture and positron emission, and (b) isomeric activities with different decay periods (unless one is very long). Hence the studies must, for completeness, be supplemented by ion detector observations. The activities investigated

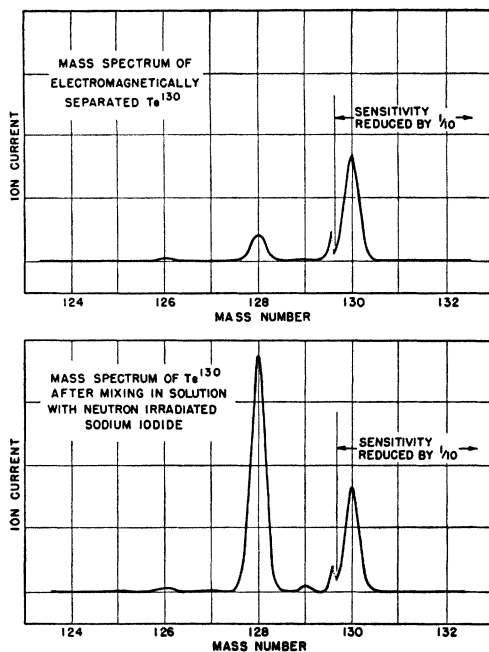


FIG. 2. Mass spectrum of electromagnetically enriched  $\text{Te}^{130}$  before and after mixing in solution with neutron irradiated NaI. The excess  $\text{Te}^{128}$  is due to branching in the decay of  $\text{I}^{128}$ . The peak at mass 129 is due to an impurity.

in this paper have all been studied before by conventional methods and there appears to be no ambiguity with regard to the periods with which the decays took place.

There follows a discussion of the results in light of the decay schemes of the particular nuclei investigated.

### 12.8 Hour $\text{Cu}^{64}$

This is an activity which has been much studied. It decays by negatron emission, by positron emission, and by orbital electron capture. The decay scheme was believed to be everywhere simple until a weak nuclear gamma-ray was detected by Bradt *et al.*<sup>2</sup> and confirmed by Deutsch<sup>11</sup> in a subsequent study. Deutsch placed the  $\gamma$ -energy at 1.35 Mev with a frequency of 1  $\gamma$ -ray per  $40 \pm 5$  positrons. This complicates the scheme only slightly and for most purposes this complexity may be overlooked. The negatrons and positrons have been studied with great care<sup>3, 12, 13</sup> and both serve as excellent examples of a simple spectrum with the allowed Fermi distribution.

Two investigators have determined the overall  $\beta^-/\beta^+$  ratio by integration of the spectra. Cook and Langer<sup>3</sup> report a value of 2.0 negatrons per positron; Bradt *et al.*,<sup>2</sup> a value of 2.1. The value  $2.05 \pm 0.10$  will be adopted for this quantity in the calculations to follow. This value, together with the ratio  $1.62 \pm 0.11$  of  $Z-1$  transitions to  $Z+1$  transitions measured mass spectro-

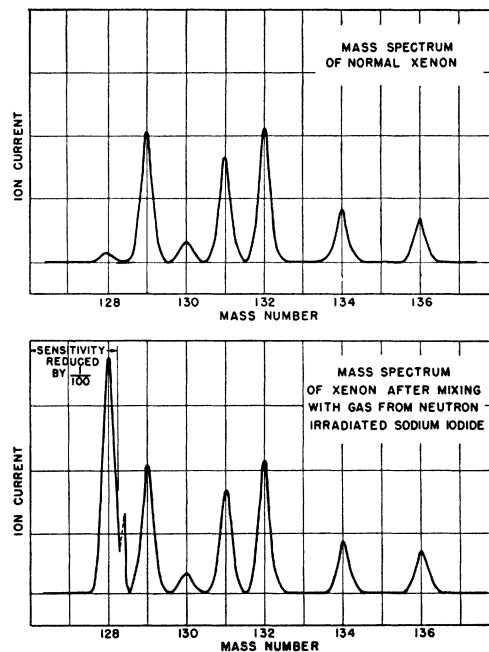


FIG. 3. Mass spectrum of normal xenon before and after mixing with the gas dissolved in neutron irradiated NaI. The excess  $\text{Xe}^{128}$  is due to beta-decay of  $\text{I}^{128}$ .

metrically, determines the branching scheme: (a) by negatron emission,  $38.2 \pm 1.6$  percent; (b) by positron emission,  $18.6 \pm 1.2$  percent; (c) by orbital electron capture,  $43.2 \pm 2.0$  percent (with  $0.46 \pm 0.06$  percent decaying by electron capture to the 1.35-Mev state of  $\text{Ni}^{64}$  according to Deutsch<sup>11</sup>).

The  $K/\beta^+$  ratio is of particular interest since it is independent of nuclear matrix elements and can be calculated from the Fermi theory. Huber, Ruetschi, and Scherrer<sup>6</sup> and Bouchez and Kayas<sup>5</sup> have each calculated this quantity using a formula due to Moeller<sup>14</sup> and the latest value of 0.657 Mev for  $E_{\text{max}}$  for  $\text{Cu}^{64}$  positrons. They find, respectively:

$$(K/\beta^+)_{\text{Theor.}}^{\text{Cu}^{64}} = 2.08 \text{ and } 2.35.$$

The new experimental value is:

$$(K/\beta^+)_{\text{Exp.}}^{\text{Cu}^{64}} = 2.32 \pm 0.28$$

and is in agreement with both the calculated values. This quantity has been measured before, but by very different methods and not with the present accuracy. Table I compares the past and present experimental values.

It should be mentioned that for the "allowed"  $\text{Cu}^{64}$  positrons, all variants of the Fermi theory predict about the same  $K/\beta^+$  branching ratio.

### 18 Minute $\text{Br}^{80}$

Unlike the 4.4 hour metastable state of  $\text{Br}^{80}$ , which has been carefully investigated,<sup>15</sup> the 18-minute  $\text{Br}^{80}$

<sup>11</sup> M. Deutsch, Phys. Rev. **72**, 729 (1947).

<sup>12</sup> L. Cranberg and J. Halpern, Phys. Rev. **73**, 259 (1948).

<sup>13</sup> C. S. Wu and R. D. Albert, Phys. Rev. **75**, 1107 (1949).

<sup>14</sup> C. Moeller, Physik Zeits. Sowjetunion **11**, 9 (1937).

<sup>15</sup> E. Segrè and A. C. Helmholtz, Rev. Mod. Phys. **21**, 2 (1949).

TABLE I. Summary of experimental determinations of the quantity ( $K/\beta^+$ ) for  $\text{Cu}^{64}$ .

Investigator	Value of ( $K/\beta^+$ ) reported	$K$ -capture evaluated by	$\beta^+$ emission evaluated by
Cook and Langer <sup>a</sup>	$3.5 \pm 1.0$	Counted Auger electrons from $\text{Ni}^{64}$ . Apparatus: beta-ray spectrometer with 4-kev accelerator.	Counted positrons. Apparatus: beta-ray spectrometer.
Bouchez and Kayas <sup>b</sup>	$2.65 \pm 0.4$	Counted x-rays from $\text{Ni}^{64}$ . Apparatus: thin window counter. Calibration: calculated.	Counted annihilation quanta. Apparatus: ionization chamber. Calibration: unspecified.
Huber, Ruetschi, and Scherrer <sup>c</sup>	$1.75 \pm 0.2$	Counted x-rays from $\text{Ni}^{64}$ . Apparatus: thin window counter. Calibration: tandem counting with identical counters.	Counted annihilation quanta. Apparatus: thick-walled counter. Calibration: coincidence counting of annihilation quanta.
Present study	$2.32 \pm 0.28$	Mass spectrometric determination (by difference).	Counted positrons. <sup>d,e</sup> Apparatus: beta-ray spectrometers.

<sup>a</sup> Reference 4.<sup>b</sup> Reference 5.<sup>c</sup> Reference 6.<sup>d</sup> Reference 2.<sup>e</sup> Reference 3.

activity needs considerably more study by modern methods. The presence of a 2.0 to 2.2-Mev group of negatrons is well established. A penetrating  $\gamma$ -ray has been reported,<sup>16</sup> but its energy is unknown. Barber<sup>7</sup> was the first to observe branching in this nucleus. He observed 18-minute positrons with a trochoidal apparatus and gave the ratio of negatrons to positrons as about 35. By absorption measurements, he found  $E_{\text{max}}$  for the positrons to be  $0.73 \pm 0.1$  Mev. The positron activity has recently been confirmed in beta-ray spectrometer measurements by Dzhelepov, Anton'eva, and Shestopalova.<sup>17</sup> However, they report the ratio of negatrons to positrons as 98 with a maximum positron energy of one Mev.

The present study reveals a total branching of  $0.090 \pm 0.002$ , or a ratio of negatrons to captures plus positrons of 11.1. It is evident that there is considerable orbital electron capture associated with this activity; i.e., the  $K/\beta^+$  ratio is 2 or 8 depending upon whether one takes the result of Barber or of Dzhelepov *et al.* It is to be hoped that the positron-negatron ratio will be measured with greater precision. In order to compare the  $K/\beta^+$  ratio with theory, it is also essential that a precise determination of the maximum positron energy be made, since the theoretical branching ratio is sensitive to this quantity.

### 34-Hour $\text{Br}^{82}$

Recent measurements on 34-hour  $\text{Br}^{82}$  by Siegbahn, Hedgran, and Deutsch<sup>18</sup> with beta-ray spectrometers of high resolution, indicate that the spectrum is complex: there are seven  $\gamma$ -rays present of different intensities. The mass spectrometric investigation shows that the number of  $Z \rightarrow Z-1$  transitions per negatron is less than 1 in 3700 or 0.00027. Barber<sup>7</sup> has previously found the ratio of positrons to negatrons to be less than 0.004.

It appears that much less energy is available for

the transition  ${}_{35}\text{Br}^{82} \rightarrow {}_{34}\text{Se}^{82}$  than for the transition  ${}_{35}\text{Br}^{82} \rightarrow {}_{36}\text{Kr}^{82}$ . This is in agreement with Kohman's conception of the stability line<sup>19</sup> at mass 82 ( $Z_A = 35.7$  for  $A = 82$ ).

### 25 Minute $\text{I}^{128}$

The present study shows that the transition  $\text{I}^{128} \rightarrow \text{Te}^{128}$  takes place in  $5.0 \pm 0.2$  percent of the disintegrations. This transition has not been reported heretofore. Barber<sup>7</sup> has found less than 0.002 positrons per negatron. It is probable that positron emission is energetically forbidden ( $Z_A = 53.3$  for  $A = 128$ ).

Siegbahn and Hole<sup>20</sup> have examined the beta-spectrum of  $\text{I}^{128}$ . They showed that there are prominent conversion lines in the spectrum due to a  $\gamma$ -ray of energy 0.429 Mev which is present to about seven percent. Accordingly they propose a decay scheme with two groups of negatrons separated in energy by 0.429 Mev, although the two groups are by no means clearly indicated by the Fermi plot.

It is possible that  $\gamma$ -rays may be associated with the 5.0 percent orbital electron capture. Certainly the decay scheme should be investigated more carefully with coincidence techniques and critical absorbers.

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<sup>18</sup> Siegbahn, Hedgran, and Deutsch, *Phys. Rev.* **76**, 1263 (1949).

<sup>19</sup> T. P. Kohman, *Phys. Rev.* **73**, 16 (1948).

<sup>20</sup> K. Siegbahn and N. Hole, *Phys. Rev.* **70**, 133 (1946).