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# The Capture of Orbital Electrons by Nuclei

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**Abstract**

The simple theory of electron capture is outlined and three general methods for its detection are suggested. The first experimental evidence for the process (in activated titanium) is described. A rigorous experimental proof of the hypothesis ie given for the case of $Ga^{67}$. A summary of several isotopes whose properties are best explained on this hypothesis is appended. The properties of $Ga^{67}$ are described in considerable detail, and include the first evidence for internal conversion in artificially radioactive atoms.

## Introduction

The suggestion that positron emitters might decay by the alternate process of electron capture was first advanced by Yukawa[1] from considerations based on the Fermi theory of beta-ray emission. In this theory, the electrons and positrons are pictured as being created at the moment they are ejected, during neutron-proton transitions. The continuous beta-ray spectrum and the conservation of spin are explained by the simultaneous emission of a neutrino and electron. One may represent the transition involved in electron and positron decay by the following equations:

$$N \rightarrow P + e^- + \nu \tag{1}$$

$$P \rightarrow N + e^+ + \nu. \tag{2}$$

[1]Yukawa and Sakata, Proc. Phys. Math. Soc. Japan **17,** 467 (1935); **18,** 128 (1936).

On the basis of Dirac's theory, however, the positron is merely the "hole" left in the continuum of negative energy electrons when one of these electrons is given a positive energy by the addition of at least $2mc^2$. The proton in (2) does not transform into a neutron and positron, but rather captures a negative energy electron, and turns into a neutron, leaving the hole in the negative energy sea, or positron. Eq. (2) may then be written

$$e^- + P \rightarrow N + \nu. \tag{3}$$

The experimental observation that positrons may be annihilated (a positive energy electron falling into the hole), shows that there is no essential difference between electrons in the two energy states. Therefore, there is no a *priori* reason why Eq. (3) demands the use of a negative energy electron. In fact, when the energy difference between parent and daughter nucleus is less than $2mc^2$, it would be impossible for the relation to be satisfied unless a proton could capture an ordinary electron. Since there are many cases of negative beta-ray decay with an energy release of less than this value, it is natural to suppose that there would be excited nuclei whose desire to emit positrons could not be allowed on energetic grounds. Yukawa suggested that in these cases, the decay would proceed by the capture of an orbital electron. In addition, he calculated, and others[2], [3], [4], [5] extended the calculations, that even when there was enough energy to create a pair, a certain fraction of the excited nuclei would decay by electron capture. The branching ratio of the two processes was found to depend on the energy available, the spin change involved, and the nuclear charge (density of electrons at the nucleus). Electron capture should become more probable as the energy decreases, and as the spin change, atomic number and half-life increase.

## Experimental Methods

Alpha-decay and the two well-established methods of beta-decay are easy to observe, since ionizing radiations are emitted in these processes. Eq. (3) shows that only the undetectable neutrino is given off by the nucleus in this new type of transition, so that more refined experimental methods must be employed if the effect is to be demonstrated.

[2]Mercier, Nature **139,** 797 (1937); Comptes rendus **204,** 1117 (1937).
[3]Hoyle, Nature **140,** 235 (1937); Proc. Camb. Phil. Soc. **33,** 286 (1937).
[4]Moller, Physik. Zeits. d. Sowjetunion **11,** 9 (1937); Phys. Rev. **51,** 84 (1937).
[5]Uhlenbeck and Kuiper, Physica **4,** 601 (1937).

(1) It is possible in theory at least to count the number of positron-radioactive nuclei formed in a given reaction, and to compare this with the number of positrons emitted in the subsequent decay. Unfortunately, most reactions leading to $e^+$ emitters are formed in $(\alpha, n)$, $(d, n)$, or $(p, n)$ reactions, which makes the accurate counting of the disintegrations very difficult. In the one case where the number of neutrons has been compared with the number of positrons-in the reaction $C^{12} + d \rightarrow N^{13} + n$; $N^{13} \rightarrow C^{13} + e^+$ – a discrepancy was found[6], which might be interpreted as evidence for electron capture[7]. But the data are not sufficiently precise to make that conclusion necessary. In addition, from theoretical considerations, it is unlikely that $N^{13}$, which is energetic, light, and involves no spin change, should have a capture branching-ratio large enough to observe by this methoda[7]

(2) If it were possible to count the number of atoms formed when a positron-active substance decayed, this number could be compared with the total number of positrons given off, to give a measure of the branching ratio. This might be applicable if there were any known cases of successive positron activities, i.e., radioactive series, where one could compare the activities of the parent and daughter substances. Since no such cases have as yet been discovered, the alternate, but much more difficult, method of counting the number of stable product nuclei-is worth investigation. A radioactive sample with an initial strength of 1 millicurie and a half-life of $t$ days contains $1.6 \times 10^{12} t$ active atoms, which will give rise to $2.6 \times 10^{-12} tA$ gram of decay product. ($A$ = atomic weight.) For anything but a noble gas, this is beyond the limit of chemical or spectroscopic detection. It is very fortunate that $Na^{22}$[8], the longest lived positron-active substance, decays into Ne, the most easily detectable rare gas[9]. Calculation shows that a one day bombardment of Mg in the cyclotron will yield enough $Na^{22}$ to allow the easy measurement of the quantity of Ne produced each month, if no electrons are captured. Theory suggests that 30 times as much Ne will be formed[10] by electron capture as is expected from the positron emission. The small bulk of Na containing the activity can be freed of all gases by prolonged heating in a vacuum, and then the Ne can be allowed to grow in the cold, an ideal case for measurement of small quantities of noble gases by the Paneth method[11].

[6] Alvarez, Phys. Rev. **53,** 326 (1937).

[7] Roberts and Heydenburg, Phys. Rev. **53,** 374 (1937).

[7] Note added in proof. Crane and Halpern [Phys. Rev. **54,** 306 (1938)] have recently shown that electron capture plays a negligible role in the decay of $N^{13}$

[8] Laslett, Phys. Rev. **52,** 529 (1937).

[9] Günther and Paneth, Zeits. f. physik. Chemie **173,** 401 (1935).

[10] Lamb, Phys. Rev. **50,** 388 (1936).

[11] Professor H.E. White and Mr. H. Weltin are working on this problem at present, in

(3) In the two previous methods suggested for detecting the capture of electrons, no advantage was taken of the fact that the electron was originally part of the stable electronic system of the parent atom, and that this system is disturbed by the loss of one of its component parts. It is well known that $x$-rays are given off by an atom which has lost one of its inner electrons by photo-ionization, and the same will be true of one which has lost an inner electron to the nucleus.. The vacant place in the inner shell will be immediately filled, and a quantum of $x$-radiation (or an Auger electron) will be emitted in the process. It is this phenomenon which is the basis of the third method of detection. One has again a technical difficulty that the $x$-rays from the light elements (and no positron emitters are known among the heavy ones[12]) are soft, and difficult to observe in the presence of positrons and gamma-rays. Jacobsen[13] was the first to try this method, in the case of $Sc^{43}$, but he found no trace of the expected $x$-rays in a cloud chamber.

## Characteristic $x$-rays from Active Titanium[14]

Walke[15] has shown that a strong positron activity of 16 days half-life is induced in titanium when it is bombarded with high energy deuterons. The energy of the positrons shows that the transition is not ,an allowed one (second Sargent curve), and this fact, together with the long life and relatively high atomic number for a positron emitter, suggested that it would be an ideal starting point in a search for the $x$-rays following electron capture. The isotope responsible for the activity has been identified chemically by Waike as vanadium, so any $x$-rays would have the wave-length characteristic of the daughter element, titanium. The fact that this wave-length, 2.7 A, is just below the "vacuum region" had much to do with the choice. From the arguments given above, $Na^{22}$ would have been a far better choice, but Ne $K\alpha$ has a wavelength of about 14A.

The detection of the soft x-ray quanta presented several difficulties. It was, of course, necessary to eliminate the positrons, and this was easily accomplished with the aid of an electromagnet. Activated Ti has an abnormally high ratio of gamma-rays to positrons, so the problem resolved itself into the detection of a weak component of very soft radiation superposed

collaboration with the author.

[12]Alvarez, Phys. Rev. **53,** 213 (1938).

[13]Jacobsen, Nature **139,** 879 (1937).

[14] Alvarez, Phys. Rev. **52,** 134 (1937).

[15]Walke, Phys. Rev. **5l,** 1011 (1937).

on a strong, hard one. A counter has an obvious advantage over an electroscope here, but the problem of getting the soft quanta into the counting volume is a serious one. This was solved by constructing the envelope of thin Cellophane, and filling it with argon at atmospheric pressure. This last feature greatly increases its relative sensitivity to 2.7A $x$-rays, as almost all the quanta are stopped in the gas, while very few gamma-rays are absorbed in the thin walls or gas.

The experimental arrangement is shown in Fig. 1. The Ti sample was placed in a five-sided aluminum box between the poles of an electromagnet, in a field of 2000 oersteds, so that all positrons were kept from reaching the counter. The sample was aged for two weeks, so that all short periods were of negligible intensity. The tube between source and counter was. filled with He at atmospheric pressure and capped with Cellophane ends. This reduced the solid angle (counting rate) without discriminating against the soft $x$-rays. The counter cathode was. made of copper foil in the shape of a C, so that $x$-rays could enter the active volume without passing through too much absorber. The copper foil was only 0.00025 cm thick, to prevent the absorption and counting of gamma-rays. Lead blocks screened the glass tubes which supported the Cellophane wall and copper cathode, for. the latter reason also. The anode wire was of 0.0025 cm tungsten wire to keep the counter voltage low–the working potential was about 2000 volts. Alcohol in a side tube made the counter more reliable, and to keep the sensitivity constant, it was found necessary to counteract leaks in the Cellophane system by flowing argon through the counter whenever it was in operation. The background was high, but the counting rate was so great that this caused no inconvenience. Absorbers of thin aluminum foil were arranged on sliding frames so that it Was a simple matter to take absorption data.

The experimental procedure consisted in taking 2000 counts at each of seven values of absorber thickness, and in repeating each setting eight times. Each point then had a probable error of less than one percent. The background count and the linearity of the counting circuit were checked after each series. The counting rate dropped from 21.3 to 15.4 per second as the absorbers were introduced into the beam. The rate remained at the latter value from 0.0025 cm to 0.0075 cm of Al, indicating that the decrease was due to a soft component in addition to the hard gamma-rays.

Figure 2 shows the actual absorption curve, and Fig. 3 is the absorption curve of the soft component. The three lines are drawn through the upper, most accurately known point, with slopes calculated from the measured absorption coefficients in Al of $K$-radiation from Sc, Ti and V. The agreement of the data with the Ti line is very striking, and indicates that the radia-

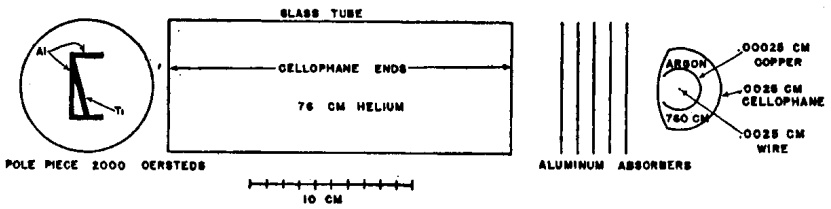


Figure 1: Experimental arrangement.

tion is homogeneous, and of the expected wavelength, giving support to the electron capture hypothesis.

On the remote chance that this radiation could have been excited by the positrons in passing through the Ti sample, the sample was covered with a thin layer of chromium (actually a 0.00025 cm Ni foil plated on each side with 0.0012 cm of Cr). This foil was thick enough to absorb all the observed Ti $K\alpha$ and if these $x$-rays were of secondary origin, the positrons should have excited about equal amounts of Cr $K\alpha$ in the Cr foil. Absorption data showed that no soft component was present under these circumstances, so it seemed safe to assume that the $x$-rays arose from Ti atoms which had been formed by electron capture from the 16-day vanadium period. The assumption as to the period was based on Walke's finding that all but a negligible fraction of the activity in a two-week-old sample of Ti bombarded with deuterons was due to this period. This fact, plus the theory, which suggested that $K$-radiation should be found in this period made it seem unnecessary to repeat the expensive (because of the argon waste) experiment two weeks later just to check the period.

However, Waike carried this Ti sample to Liverpool shortly after, and Williams and Pickup[16] have followed its activity in a cloud chamber since then. They improved Jacobsen's technique by bending the positrons out of the chamber by a magnetic field, and were thus able to see $x$-ray photoelectrons much closer to the source, where they are more plentiful. A count of these short tracks against distance verified that they had approximately the correct absorption coefficient to be Ti $K\alpha$. The surprising observation was made that there were about 500 photoelectrons per positron, and that while the positrons decayed with the 16-day period, the number of photoelectrons remained constant for several months, within the statistical error. This indicated that some of the $x$-rays observed in this laboratory were due

[16]Williams and Pickup, Nature **141,** 199 (1938).

to a new, long period in Ti, which had escaped Walke's attention because of its soft radiation. To find what percentage, if any, of the $x$-rays were due to the 16-day period, the author prepared and sent a fresh sample of Ti to Liverpool, where Williams and Pickup showed that no appreciable fraction was emitted by the 16-day isotope. While this changes the identification of the responsible isotope, the original interpretation that electron capture was probably the cause of the radiation, was still the most plausible guess.

But this interpretation is not the only one which will fit the facts, and if one is to establish the existence of a new phenomenon, there must be no alternative explanations. It is well known that $x$-rays appear when gamma-rays are internally converted. Here the shell is vacated by an expelled electron, instead of one lost to the nucleus. It is entirely possible that the long-

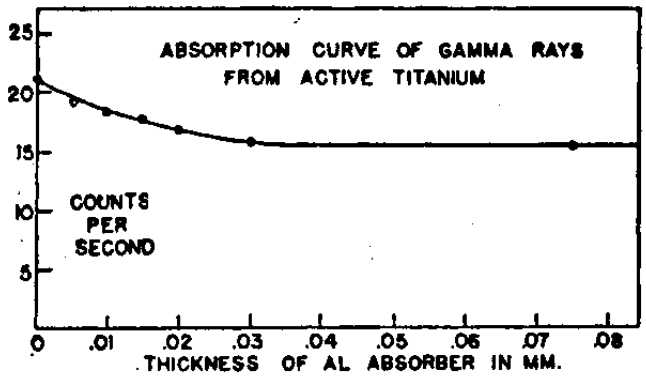


Figure 2:

lived $x$-ray emitting isotope is a metastable state of a stable or radioactive Ti or V isotope. Low lying states of this kind are usually assumed to be responsible for the existence of nuclear isomers, of which this long period would then be an example. It is assumed that the higher level may radiate a gamma-ray to the ground state, and if these states are close, as they must be for a long life to be possible, the soft gamma-rays could be largely internally converted. The soft conversion electrons would not be seen in Williams and Pickup's chamber. While this is not suggested as the probable explanation, it fits all the observed data as well as the electron capture hypothesis. It serves best to emphasize that a more decisive test must be made if electron capture is to be accepted as an established fact.

## Proof of the Electron Capture Hypothesis[17]

A glance at the table of isotopes showed that several blank spaces existed on the positron side of the stability band, in the region between Mn and Ge, although this section of the table has been very carefully studied by several workers. It seemed quite probable that some of these missing isotopes might capture electrons, and that they had not been discovered because their soft $x$-rays had been masked by. the strong beta- and gamma-rays of the other periods.

An exploratory investigation was therefore made of the soft quantum radiations emitted by several of these elements after bombardment with deuterons. Of the four elements tried, Fe, Ni, Cu, and Zn, all were found to give off various amounts of soft $x$-rays. Since the wave-lengths are shorter in this region, it was possible to use an air-filled Lauritsen type electroscope as the detector; the magnet was still necessary to suppress the beta-rays. The absorption curve on the iron radiation showed that about half of gamma-ray ionization was due to a component with an absorption coefficient in Al about equal to that of Cr $K\alpha$. There are several long periods induced in iron, so it did not lend itself easily to investigation. Ni showed about 8 percent $K$-radiation following the well-known 3.5 hr. period of $Cu^{61}$. Copper showed about 40 percent $x$-ray ionization following the 12 hr. $Cu^{64}$. This is interesting, as it is what one would predict on theory[5]. The activity with the shorter life has a smaller capture branching ratio, and in fact the numbers quoted above fit very well on the theoretical curves. The ratio of these two percentages is a number which does not depend on geometry or the relative sensitivity of the electroscope to various types of radiation, or on atomic number or spin change; so it is well adapted to a comparison with theory. It is encouraging that it fits so well, but it cannot be taken too seriously at present.

Activated zinc was the most interesting of the four elements bombarded, and the rest of this section will be devoted to it. Four of the activities induced in zinc by fast deuterons were found to be accompanied by $x$-rays with the correct absorption coefficient in Al to be Zn $K\alpha$. Electron periods as well as the positron periods were among these four, so it did not seem possible to explain them all as due to electron capture. And if there was an alternate explanation, why could it riot explain all the cases?

Most of the work was done on the 83-hour period. This activity has had

[17] Alvarez, Phys. Rev. **53,** 606 (1938).

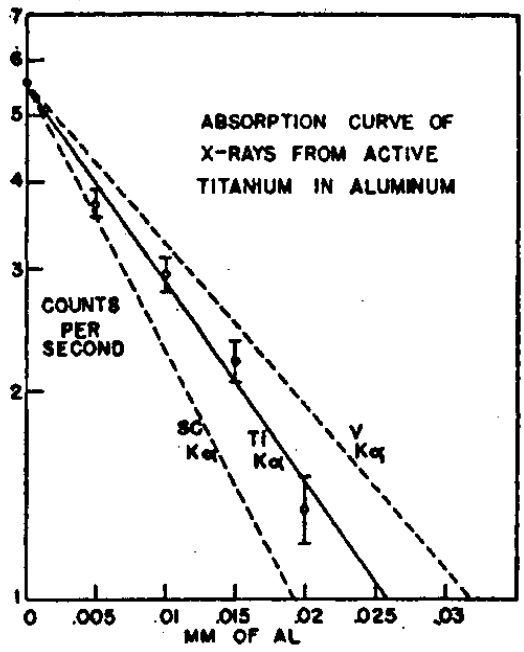


Figure 3:

the following interesting history: It was first found by Livingood[18], who gave its half-life, without chemistry, as 97 hours. Later, Thornton[19] identified it as a zinc isotope with the same period, and assigned it to $Zn^{71}$. Next, Du Bridge, Barnes, Buck, and Strain[20] quoted its period as 82 hours, but said it was surprising that although it was prepared from Zn with protons, it was not detected in either copper, zinc, or gallium, precipitates. The author gave the half-life as 83 hours, and showed that it was chemically similar to Ga. Strain and Buck[21] have since confirmed this, and Mann[22], who reported a Ga isotope with a period of about 55 hours, has also shown that his activity is identical with the 83-hour Ga. The isotope has thus been prepared from zinc by deuteron, alpha-particle, and proton bombardment, which pins it down at $Ga^{67}$.

The $Ga^{67}$ could be separated from the rest of the activities induced in

[18]Livingood, Phys. Rev. **50,** 425 (1936).
[19]Thornton, Phys. Rev. **53,** 326 (1938).
[20]Du Bridge, Barnes, Buck, and Strain, Phys. Rev. **53,** 447 (1938).
[21]Strain and Buck, Phys. Rev. **53,** 943 (1938).
[22]Mann, Phys. Rev. **53,** 212 (1938).

Zn by means of the ether extraction process[23]. This procedure depends upon the fact that $GaCl_3$ is soluble in ether, while the chlorides of the neighboring elements are not. If one then shakes an HCl solution of these metals with ether in a separatory funnel, most of the $GaCl_3$ will be found in the ether layer. The ether may be then evaporated on a metal sheet, leaving an invisible layer of $GaCl_3$ which contains almost all of the original activity. This is probably the simplest way of preparing a pure radioactive sample–no carrier is needed. In the case under discussion, there were no doubt small traces of Ga impurities in the activated Zn, but very recently, Grahame and Seaborg[24] have activated specially prepared, Ga-free Zn and found that the ratio of activity in the ether layer to activity in the aqueous layer is the same for pure radioactive $GaCl_3$ as it is for macroscopic amounts of stable $GaCl_3$.

When a weak, separated sample of $Ga^{67}$ was measured without a magnetic field on a thin walled electroscope, a very soft component was found, which was electronic in nature as shown by the sign of the magnetic deflection. Rough range measurements showed that the energy of these electrons was about 100 kev. This was quite surprising for a period as short as 83 hours. The Sargent relations show that for any given half-life, there is a minimum beta-ray energy, corresponding to an allowed transition of spin change zero. For greater spin changes, the energy released is considerably higher. There are no known beta-ray energies less than that called for by the first Sargent curve, and there is no theoretical reason for expecting them[25]. So it seemed probable that the electrons .observed here were conversion electrons instead of primary disintegration particles. No previous example of internal conversion in the artificially radioactive elements has' been reported, and it has generally been assumed that it should be very unlikely.

To check this possibility, the absorption curve of these electrons was carefully investigated with a separated sample of $Ga^{67}$. The data are shown in Fig. 4. It is at once evident that the curve is not exponential, nor a combination of exponentials as one finds for beta-rays, since it is concave toward the origin. Its shape is strong evidence that the electrons responsible have a line structure. The intensity available in the sample was enough to allow its measurement on a high resolution mass spectrograph. Mr. D.C. Kalbfell photographed the electron spectrum in an instrument of his design, and showed that there was indeed a line structure. His plates showed an

[23]Noyes and Bray, *Qualitative Analysis for the Rare Elements*(Macmillan Company).
[24]Grahame and Seaborg, Phys. Rev. **54,** 240 (1938).
[25]See later section on $Hg^{197}$.

intense line at 90 kev, and a fainter one at about 99 kev. This is precisely what one would expect if the electrons were due to the internal conversion of a gamma-ray of 100 kev. The $K$ and $L$ absorption edges of zinc are at 1.2 and 12A, respectively, corresponding to energies of 10 and 1 kev. The strong line is then due to $K$-conversion of the gamma-ray, and the weak one to $L$-conversion. Dr. E.M. Lyman examined the specimen in his high resolution beta-ray spectrograph equipped with a Geiger counter, and

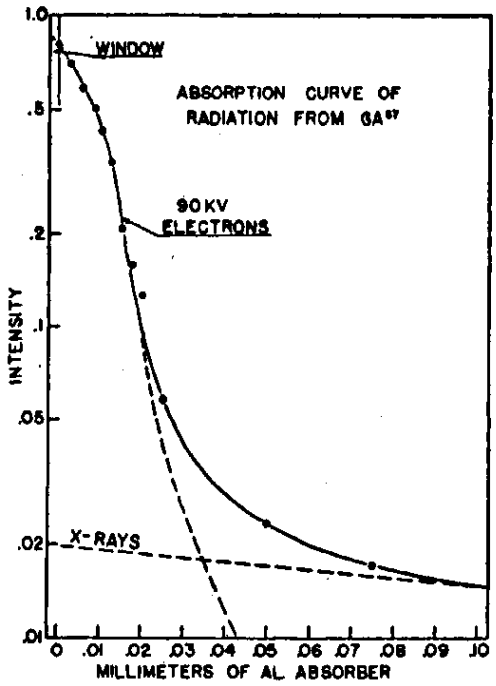


Figure 4:

obtained the spectrum shown in Fig. 5[26]. This electron group is interesting in that it is the purest radioactive line source known.

Attention was next directed to the $x$-rays. Although it would have been possible to determine the atomic number of the element whose characteristic radiation was emitted from active Ti, by careful absorption measurements in Al, this is not true of the radiation from Ga. The absorption coefficient of the $K$-radiation from Sc, Ti, and V is a very sensitive function of $Z$, as

[26]I wish to thank Dr. Lyman and Mr. Kalbfell for examining the active Zn in their spectrographe.

shown in Fig. 3, but it is a slowly varying function in the neighborhood of Zn. One must therefore resort to a more refined method to identify the wave-length of the Ga $x$-rays. There is enough intensity available to diffract the rays in a bent crystal spectrograph[27], and this method was the first tried. If this were the only possibility, it is certain that results could have been obtained without too much difficulty. But the attempt was abandoned in favor of the simpler method described below.

It is well known that if one plots absorption coefficient in a given element against wavelength, sharp discontinuities appear at certain wave-lengths which are known as "critical absorption limits," and which correspond to the binding energies of the $K$, $L$, etc. shells of the absorbing atom. If one now plots the same type of curves for the two neighboring elements, he finds that the curves are very similar, except that the discontinuities have shifted slightly to either side.

If one could find two neighboring elements which absorbed the Ga $x$-rays very differently, it would show that the wave-length of this radiation lay between the two critical limits. Experiments showed that Ni and Cu foils exhibited this property. The data are plotted in Fig. 6. The Ni and Cu absorption limits are at 1.48 and 1.38A, respectively, so most of the radiation is between these limits. Zn $K\alpha$ is the only strong line satisfying these conditions, so one may conclude that it is responsible for most of the effect. Zn $K\beta$ should accompany it, and since it has a shorter wave-length than either of the two absorption edges, it is strongly absorbed in both Ni and Cu. It is seen that the copper curve resolves into two components corresponding to $K\alpha$ and $K\beta$ of zinc, and the ratio of the intensities of the two lines is approximately correctly given by the intercepts of the resolved components on the vertical axis. That the absorption in Ni is not as great as one would expect from the tabulated absorption coefficients is due to the imperfection of the geometrical conditions. Ni $K\alpha$ is excited as fluorescence radiation when Zn $K\alpha$ is absorbed in Ni foil, and some of these x-rays are detected in the chamber, to give too high a reading. When the geometry was made still poorer, the apparent absorption coefficient fell further, showing that this explanation of the discrepancy was correct. The evidence is therefore conclusive that the soft component of the undeflectable radiation is composed of zinc characteristic $x$-rays, and that therefore, the daughter substance is an isotope of zinc.

The gamma-ray spectrum was next investigated by two methods. Ab-

[27]Du Mond and Kirkpatrick, Rev. Sci. Inst. **1,** 88 (1930); Cauchois, J. de Physique **3,** 320 (1932).

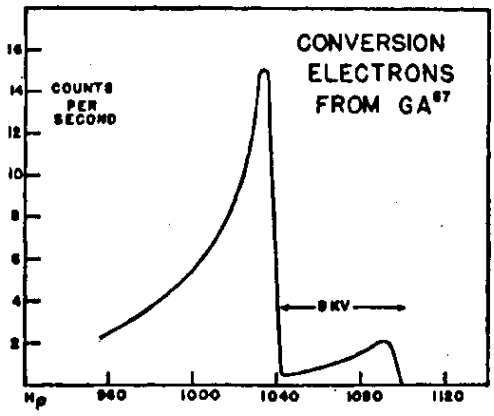


Figure 5:

sorption data in copper and lead showed that there was a component in the neighborhood of 250 kev. It was particularly important to establish whether or not any annihilation radiation accompanied the decay. The absorption curve in lead was compared with one taken on $Cu^{64}$ with identical geometry. The latter emits only annihilation radiation[28]. From the two curves (Fig. 7) one can see that if any of this type of gamma-rays is given off from $Ga^{67}$, it is a very small fraction indeed. Confirmatory evidence was obtained in a cloud chamber, where no positrons could be observed. No trace of the unconverted fraction of the 100 kev line was detectable in the absorption data, so a more sensitive method based on transition effects was used. The gamma-ray effects in an electroscope are due to Compton and photoelectrons ejected in the material between the source and the sensitive region of the chamber. As one piles lead over the source, the gamma-ray ionization at first increases, and then decreases. For lighter absorbers, the maximum will not be as high, and the rate of decrease will, of course, be smaller. These initial rises are due to the production of electrons in the absorber (in heavier elements the equilibrium ratio of electrons to gamma-rays is higher). If one places enough lead over the source to bring the ionization current up to near its maximum value, and then absorbs out the electrons from the lead with aluminum sheets, the ionization current will fall to the low value for equilibrium in Al. The amount of Al necessary to accomplish this lowering is equal to the range of the electrons, and therefore is a measure of the energy

[28]Van Voorhis, Phys. Rev. **50,** 895 (1936).

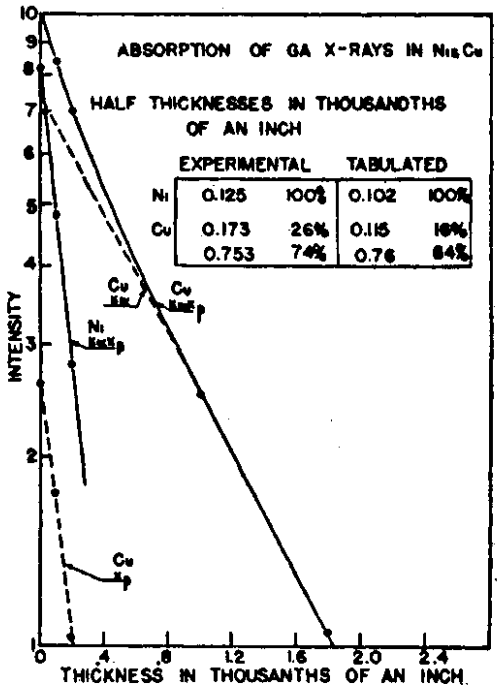


Figure 6:

of the gamma-ray which projected them from the lead.

This method was applied to the annihilation radiation from $Cu^{64}$, and gave a value near 500 kev. When $Ga^{67}$ was substituted, the transition curve showed a large group of electrons from a gamma-ray at about 250 kev, in agreement with the absorption data. A careful search with very thin absorbers showed another group due to the unconverted 100 kev line. (See Fig. 8.) Thus the line is not totally internally converted, as had at first been suspected. An examination of the transition curve near the 500 kev portion showed no trace of a drop, so it is quite certain that positrons play no important part in this activity.

Summing up the evidence, we see that a radioactive Ga isotope has changed into a zinc isotope, and during the process, no positrons have been emitted. (The gamma-ray evidence eliminates the possibility that the positrons might have been too soft to be detected.) To investigate the small chance that protons were emitted during the disintegration, a sample

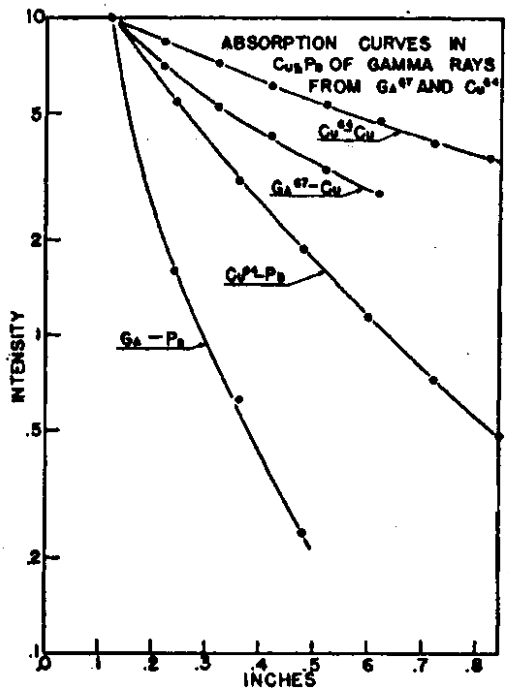


Figure 7:

was examined in a linear amplifier, and also placed inside the electroscope chamber. No evidence was found for protons, and there are in addition good theoretical reasons for believing that extremely slow protons could not emerge in such numbers with a half-life as short as 83 hours. It must then be concluded that the transition from Ga to Zn has not been accompanied by the emission of any known particle of single positive charge. (The possibility that a heavy electron is responsible for the transition is ruled out on energetic grounds. There is not enough energy available for the transition to $Zn^{67}$ and, if it went to $Zn^{66}$, the latter would be left too highly excited.)

We are therefore forced to the conclusion that the $Ga^{67}$ nucleus has captured a negative electron from its orbital system, and been transformed into stable $Zn^{67}$.

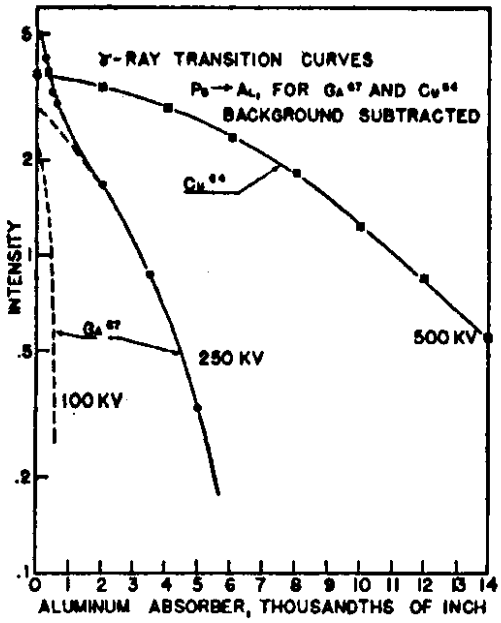


Figure 8:

## Other Data Bearing on Electron Capture

**Ta**$^{180}$

The second artificially radioactive body shown to emit characteristic $x$-rays was Ta$^{180}$, reported by Oldenberg[29]. He irradiated tantalum with fast neutrons from the Li + D reaction, and separated an active form of tantalum by chemical methods. Since this period could not be induced by slower neutrons, it was assumed to be formed by neutron loss, and therefore assigned to the isotope with atomic weight one less than the only known stable isotope of Ta, Ta$^{181}$. The beta-rays from this isotope were all negative, as determined by a trochoid analyzer. The gamma-rays had an absorption coefficient in Al equal to that of Hf $K$-radiation. No magnetic field was necessary, as the first Al absorber cut out all the electrons. The curve could then be followed for several half-thicknesses of Al, with the aid of a Geiger counter. Oldenberg interpreted the $x$-rays as due to electron capture, giving Hf$^{180}$. Since W$^{180}$ was not known, he suggested that the beta-rays

[29]Oldenberg, Phys. Rev. **53,** 35 (1938).

were conversion electrons from an internally converted gamma-ray from an excited level of $Hf^{180}$ to ground.

Sizoo[30] has shown on simple theoretical considerations that if the middle one of three neighboring isobars is radioactive, and emits negative electrons, it is almost certain to transform some of the time by electron capture. (If it could not do this, i.e., if the isotope with smallest $Z$ had greater mass, the latter would transform by negative electron emission to the middle one, and would therefore not be observed as stable.) On the chance that the negative electrons observed here were disintegration betas–that this case were an example of Sizoo branching–the author asked Professor A.J. Dempster to reexamine his mass spectra of W to see if there was any evidence for $W^{180}$. The old plates showed faint lines, and new plates confirmed the existence of this isotope[31]. Thus Oldenberg's activity is probably the first example of electron emission-electron capture branching.

**$K^{40}$**

Weizsäcker[32] has made the suggestion that the natural radioactivity of potassium may be a branching reaction of the Sizoo type. In addition to the well-known electron decay to $Ca^{40}$, he postulates electron capture to $A^{40}$. This would explain several anomalies in the abundance of argon. The position of argon in the periodic table is not given by its sequence in the list of atomic weights. This situation would be corrected if the percentage abundance of $A^{40}$ were smaller. The abundance of argon in the atmosphere is about one percent, which is vastly greater than any of the other rare gases. This might be termed circumstantial evidence, but it does fit well with theoretical ideas about electron capture.

**$Zn^{65}$**

Barnes and Valley[33], and Livingood and Seaborg[34], have reported an 8-month zinc period which emits positrons, and which has an abnormally high ratio of gamma-rays to positrons. This is no doubt the longest of the four periods in zinc mentioned above, which emit $x$-rays. Absorption measurements on the $x$-rays in Ni and Cu were made at that time, which showed that the radiation was characteristic of copper[35]. This is what one

[30]Sizoo, Physica **4,** 467 (1937).

[31]Dempster, Phys. Rev. **52,** 1074 (1937).

[32]Weizsäcker, Physik. Zeits. **38,** 623 (1937).

[33]Barnes and Valley, Phys. Rev. **53,** 946 (1938).

[34]Livingood and Seaborg, Phys. Rev. **54,** 239 (1938).

[35]The shortest of these four periods in unseparated, activated zinc was a new electron emitting isotope with a half-life of 15 minutes. The $x$-rays are definitely Zn $K$-radiation, and since this period is unknown, it might be due to $Ga^{65}$ capturing electrons.

would expect from Zn" if it captured electrons. It seems relatively safe to assume that the two zinc activities are identical and to assume, as Barnes and Valley, and Livingood and Seaborg did, that this is the first case of positron emission-electron capture branching.

**$Hg^{197}$**

Heyn[36] has reported a period of about 43 minutes induced in mercury by very fast neutrons. No details of the radiations emitted were noted by him, but he made the reasonable suggestion that the reaction was of the $n - 2n$ type. McMillan, Kamen and Ruben[37], in their survey of the neutron induced activities in the heavy metals, investigated this activity in some detail. They-showed that the period was due to mercury, and that the electrons were all negative. They noted that the beta-ray energy was too low for even the first Sargent curve, and in private conversations, they suggested that the electrons might be conversion electrons from a gamma-ray after electron capture. An absorption curve of the gamma-rays showed a "possible complex structure with energies in the range 70-250 kev." On this basis, the assignment would have been to $Hg^{197}$. But these authors preferred the idea that the electrons were disintegration betas, and so assigned the activity to $Hg^{203}$.

An obvious difficulty with this assignment is the failure to observe the period with slow neutrons. Slow neutron capture is always energetically possible, and could not have escaped detection with such a short period. This would lead one to assign the period to $Hg^{197}$, since $Hg^{196}$ is present only to 0.1 percent. The electrons then would not. be primaries, which would eliminate the only point on a "negative Sargent curve." Since no positrons were observed, and since the gamma-ray spectrum does not extend to 500 kev, this is probably a case of electron capture.

**$Ag^{106}$**

Pool, Cork and Thornton[38] discovered an interesting case of, isomerism in $Ag^{106}$. The 24-minute period emits positrons, while the one of 8-days halt-life is electron active. Pool[39] has investigated the radiations from the isotope, and concluded that the 8-day period also captures electrons. If this explanation were correct, one would expect to find about three quanta of Pd $K$ $x$-rays, and one Auger electron for every four gamma-rays. Pool's gamma-ray spectrum shows no electrons in this energy, range,, but they would have been a little difficult to measure. To check this point, a search

[36]Heyn, Nature **139,** 842 (1937).
[37]McMillan, Kamen and Ruben, Phys. Rev. **52,** 375 (1937).
[38]Pool, Cork and Thornton, Phys. Rev. **52,** 380 (1937). "
[39]Pool, Phys. Rev. **53,** 116 (1938).

for Pd $K\alpha$ was made in this laboratory. The source of radio-silver was covered with paraffin to absorb the beta-rays, and was thin enough to have negligible self-absorption for the $x$-rays. The gamma-ray ionization was measured in an electroscope filled with $CH_3Br$[40]. This type of instrument is much more sensitive to Pd $x$-rays than to gamma-rays, so almost all of the observed ionization should have been due to the former, if electron capture were taking place. Aluminum absorbers had very little effect on the ionization current, so it must be concluded that electron capture does not play an important part in the decay.

**$Cd^{107}$** or **$Cd^{109}$**

Ridenour, Delsasso, White, and Sherr[41] have found a 6.7-hour proton-induced activity in silver, which they ascribe either to $Cd^{107}$ or $Cd^{109}$. The radiations emitted by this isotope are a soft electron group, a strong $x$-ray with the correct Al absorption coefficient, and a very weak gamma-ray. They conclude that the whole situation in this radioelement is similar to the case of $Ga^{67}$.

**$Be^7$**

Roberts, Heydenburg, and Locher[42] have very recently observed an interesting example of electron capture in $Be^7$. This new radioactive isotope may be formed in either of the following two reactions:

$$Li^6 + D \rightarrow Be^7 + n,$$

or

$$B^{10} + D \rightarrow Be^7 + He^4.$$

This isotope is the only one known which emits nothing but gamma-rays; their energy is about 425 kev. The neutron yield in the reaction leading to $Be^7$ from Li is ten times the radioactive gamma-ray yield, which is a large enough discrepancy to mean something. The interpretation given by these authors is that in 90 percent of the disintegrations, the neutrino carries away all the available energy, leaving the Li7 nucleus in the ground state. In 10 percent of the cases, which are those observed, the $Li^7$ is left in the well-known level at 440 kev, from which it radiates to the ground state.

**$Mn^{54}$**

Livingood and Seaborg[43] have found a long lived manganese isotope which may be prepared in three different reactions, and thus assigned to

[40] I am indebted to Mr. Philip Abelson for the use of this instrument.
[41] Ridenour, Delsasso, White, and Sherr, Phys. Rev. **53,** 770 (1938).
[42] Roberts, Heydenburg, and Locher, Phys. Rev. **53,** 1016 (1938).
[43] Livingood and Seaborg, Phys. Rev. **54,** 391 (1938).

$Mn^{64}$. The radiations from this isotope are a strong $x$-ray absorbed in Al as Cr $K\alpha$, a gamma-ray of about 1 Mev, and a very weak negative electron group which is probably of secondary origin. These data are best interpreted as evidence for electron capture. The gamma-rays are internally converted to a very small extent, if at all.

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# Appendix

**Additional data on $Ga^{67}$**

Since this paper is primarily concerned with electron capture, only those data about $Ga^{67}$ which bore directly on the problem were included in the section above. This appendix will complete the discussion of $Ga^{67}$, merely as an interesting case of artificial radioactivity. It was established earlier that the following radiations were emitted by this isotope: a line spectrum of electrons, the characteristic $x$-rays of zinc, a gamma-ray of about 250 kev and one at about 100 kev. In order to obtain a more complete picture of the processes involved, it is necessary to have some idea of the relative intensities of the various radiations.

An absorption curve in aluminum, of the total radiation from a thin, separated sample of $Ga^{67}$, showed that the ionization current due to the electrons was about 40 times that due to the $x$-rays. The chamber had a depth of 6 cm; from the absorption coefficient, one can calculate that 6 percent of the $x$-rays were absorbed in the active volume of the electroscope. The electrons have a range of about 10 cm in air, so about half of their energy was expended in the chamber. Each electron has ten times as much energy as an $x$-ray quantum. The relative intensity of $x$-rays and electrons is then

$$10/(40 \times 0.06 \times 2) \cong 2.$$

The fluorescent yield for Zn is 0.5, so the ratio of electron excitations to conversion electrons is about 4.

The intensity of the gamma-rays may be estimated in the following manner: since the gamma-ray ionization was obtained with aluminum absorbers over the sample, it is necessary to calculate the equilibrium ratio of electrons to quanta in aluminum. The mass absorption coefficient of the gamma-rays is 0.11 $cm^2/g$, and the range of the electrons is 0.045 $g/cm^2$, so 0.005 of the gamma-ray energy will be converted into electronic form in the effective upper layer of the aluminum. The range of the electrons in air is $0.045/0.001 = 45$ cm, but only 6 cm of this range is effective in the chamber. The ratio of $x$-ray ionization to gamma-ray ionization

wag observed to be 3.5, so the relative intensity of these two types of radiation is then

$$\frac{3.5 \times 0.005 \times 6 \times 250}{0.06 \times 45 \times 9} \cong 1.$$

The relative intensities of $K$-excitations, gamma-rays, and electrons are therefore approximately $4 : 2 : 1$. These values suggest that there are two excited states in the $Zn^{67}$ nucleus–one at 350 kev, and the other either at 100, or 250 kev above ground. Every case of electron capture leaves the $Zn^{67}$ in the upper state, from which it cascades to ground in two steps. One of the transitions gives rise to the 250 kev gamma-ray, and the other is responsible for the internally converted radiation at 100 kev. The internal conversion coefficient is fairly high, since the transition curves show the 100 kev line to be weaker than the one at higher energy. The relative intensities of electrons and $x$-rays confirm this. If the line were totally internally converted, the $K$-excitation would be twice as great as the electron emission, since the $K$ shell would be emptied by capture and conversion in each transition. The observed ratio is 4, which indicates that the conversion coefficient is about 40 percent. All the intensities are consistent with the scheme outlined above.

The question then arises:–how is it possible to have such a large internal conversion factor in an element ag light as zinc? It has been quite generally thought by theoretical physicists that internal conversion would play a negligible part in artificial radioactivity, since the equations predict a $Z^3$ dependence. (Total conversion in a 0-0 transition would be possible, however.) When it first be came apparent that $Zn^{67}$ had an anomalously high internal conversion coefficient, Dancoff and Morrison[44] reexamined the theory, and found that in the energy range near 100 kev for zinc, the factor could be quite high. The 40 percent observed (internal conversion coeff. $\sim 0.7$) would mean the gamma-rays were due to a quadripole transition. No detailed calculations of this nature had been made previously for light nuclei, as it had seemed certain from an inspection of the formulae that there were no terms entering which could make the ratio appreciable. All details of the picture, both experimental and theoretical, are now self-consistent.

It might be interesting to look for coincidences between the two $x$-rays ejected almost simultaneously in 1/8 of the disintegrations. (40 percent $\times$ (fluorescent yield)$^2$ = 1/8.) An apparatus for this experiment has been constructed, but has not as yet been used. The $x$-ray spectrum might yield interesting information about the relative lifetimes of $x$-ray and gamma-ray states. If the $K$ shell were doubly excited, the wave-length of the characteristic radiation would be slightly changed, and this could be detected on a bent crystal spectrograph. No such case of a doubly excited $K$ shell is known.

[44] Dancoff and Morrison, Phys. Rev. **54,** 149 (1938).