Radioactive Isotopes of Cu, Zn, Ga and Ge

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A study has been made of the radioactive isotopes produced in Cu, Zn, Ga and Ge by bombardment with both fast and slow neutrons. Some of the isotopes were also produced by deuteron bombardment. The results may be summarized in the following table of decay periods.

Element	*Mass No.*	*Sign*	*Half-life*	*β-Ray Energy*	*γ-Ray*
Nickel	63	e^-	2.5 ∓ 0.3 hr.	medium	—
Copper	62	e^+	10 ∓ 0.5 min.	hard	γ
Copper	64	$e^\mp$	12.8 ∓ 0.3 hr.	soft	γ
Copper	66	e^-	5 ∓ 1 min.	medium	—
Zinc	63	e^+	35 ∓ 3 min.	hard	γ
Zinc	65	e^+	210 ∓ 30 min.	very soft	γ
Zinc	69	e^-	57 ∓ 3 min.	hard	—
Gallium	68	e^+	66 ∓ 2 min.	hard	γ
Gallium	70	e^-	20 ∓ 1 min.	hard	—
Gallium	72	e^-	14.1 ∓ 0.2 hr.	hard	γ
(Gallium	74?	e^-	6 ∓ 1 day	—	—)
Germanium	69	e^+	30 ∓ 3 min.	hard	—
Germanium	71	e^+	26 ∓ 3 hr.	medium	γ
Germanium	75	e^-	81 ∓ 3 min	medium	—
Germanium	77	e^-	8 ∓ 1 hr.	—	—
Arsenic	71	e^+	50 ∓ 3 hr.	soft	γ
Arsenic	73	e^+	88 ∓ 2 min.	medium	γ
Arsenic	74	$e^\mp$	16 ∓ 2 days	medium	γ
Arsenic	77	e^-	55 ∓ 5 days	very soft	γ

The writer failed to find the Cu^{61} (e^- $T=3.5h$) suggested by the Michigan group in fast neutron bombardments of copper. No trace of the 23-hr. period reported by Fermi and his collaborators was found in neutron bombardments of gallium.

INTRODUCTION

IN CONTINUATION of work[1] at the Cavendish Laboratory on nuclear reactions involving a loss of a neutron from a nucleus, the writer started a more detailed study of the "neutron loss" periods. He soon noticed that in the many investigations[2] of the radioactivity induced in Cu, Zn and Ga but few of the isotopes produced had been identified. There still remain many discrepancies in the experimental data. As for germanium very little has been reported so far.

It is the purpose of the present paper to report the results of the study[3] of the radioactivity induced in these elements by fast and slow neutrons and by deuteron bombardments which have been carried out in the Radiation Laboratory of the University of California.

APPARATUS

The cyclotron in the Radiation Laboratory was used as a source of deuterons and neutrons. At the time of the bombardments it was adjusted to furnish currents of 20 to 150 μa of deuterons of energy from 5.5 Mev to 7.6 Mev.

Ordinary copper plates which are usually of sufficient purity were used as copper targets. The

* Now at the Tokyo Imperial University.
[1] Chang, Goldhaber and Sagane, Nature **139**, 962 (1937).
[2] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti and Segrè, Proc. Roy. Soc. **149**, 522 (1935). Livingood, Phys. Rev. **50**, 425 (1935). McLennan, Grimmet and Read, Nature **135**, 505 (1935). Van Voorhis, Phys. Rev. **50**, 895 (1936). Heyn, Nature **138**, 723 (1936); Physica **4**, 160 (1936). Pool, Cork and Thornton, Phys. Rev. **52**, 212 (1938).
[3] Most of the results except those of Ge+D were reported at the Stanford meeting held on Dec. 17, 1937. Sagane, Phys. Rev. **53**, 212 (1938).

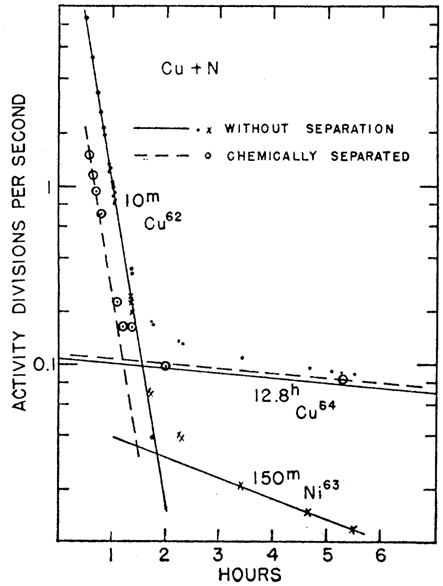

FIG. 1A. Decay curves for copper when bombarded by fast neutrons. The curve taken without chemistry can be analyzed in three periods, while the curve taken after chemical separation consists from two periods showing that the 150-min. period is not caused by a copper isotope.

zinc, gallium and germanium targets were also in metallic form and were of high purity, claimed to be 99.9 percent.

For bombardments with fast neutrons, both lithium and beryllium targets were bombarded and the samples were placed inside a small box made of cadmium sheets one to two millimeters in thickness, filled with boric acid. The box was generally put in the forward direction of the beam. For bombardments with slow neutrons, the samples were placed inside a paraffin block which was generally put at right angles to the beam.

All the measurements of activity were carried out with Lauritsen-type quartz fiber electroscopes[4] with aluminum windows of 1/10,000″ thickness. Absorption measurements were made by placing aluminum or lead sheets between the source and the window.

[4] I am indebted to Dr. A. H. Snell for the use of one of his electroscopes.

Copper

This element has been reported by many authors[2] to give the well-known 5-min. and 12.8-hr. periods with slow neutrons. With fast neutrons from Li+D, Heyn reported first that six-min. and 10.5-min. periods were produced.

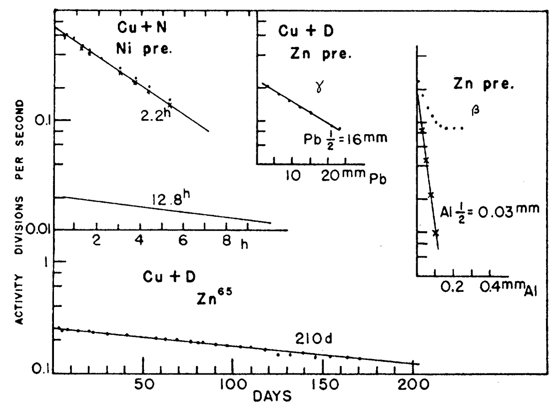

FIG. 1B. A decay curve for a nickel precipitate from a copper sample bombarded by neutrons, and a decay curve for a long lived zinc isotope obtained when copper was bombarded by deuterons. Absorption curves for the zinc isotope are also shown.

On the other hand, Pool, Cork and Thornton[5] suggested the formation of Cu^{61} (3.5 hr.) when bombarded with fast neutrons.

Bombardment with slow neutrons on Cu resulted in the formation of two periods, 5 min. and 12.8 hr. as was predicted. With fast neutrons, three periods 10 min., 2.5 hr. and 12.8 hr. were noticed as shown in Fig. 1A. Chemical separations showed that the 10-min. and the 12.8-hr. periods were caused by Cu isotopes and the one of 2.5 hr. was caused by a nickel isotope. With a

[5] Pool, Cork and Thornton, Phys. Rev. **52**, 41 (1937).

TABLE I. *Summary of results obtained in the bombardment of copper.*

Isotopes	Cu^{66}	Cu^{62}	Ni^{63}	Cu^{64}	Co	Co	Zn^{65}
Bombardment							
Slow neutrons	5 min. (n, γ)		2.5 hr. (n, p)	12.8 hr. (n, γ)			
Fast neutrons Li		10 min. $(n, 2n)$	2.5 hr. (n, p)	12.8 hr. (n, γ) $(n, 2n)$			
Fast neutrons Be			2.5 hr. (n, p)	12.8 hr. (n, γ)			
Deuterons	5 min. (d, p)			12.8 hr. (d, p)	(30 days?)*	(215 days?)*	210 days (d, γ)
γ-rays		10 min. (γ, n)					

* Segrê and others.

simple magnet close to the electroscope[6] the signs of the β-rays were determined and it was found that the 10-min. period was caused by a positron emitter and the others were caused by negative electron emitters. When copper samples were bombarded with deuterons, the 12.8-hr. period was very strongly activated and a rather weak 5-min. period was also obtained.

In addition to these two, some very long periods have been reported by Segrè and his collaborators[7] who find a zinc isotope with a half-life of 245 days and cobalt isotopes with half-lives of about a month and 215 days.

This long lived zinc isotope was studied carefully by separating zinc isotope from the copper filings scraped from the copper deflecting plate of the cyclotron. The decay curve and the absorption curve which are shown in Fig. 1B shows that this isotope has a half-life of about 210 days. It was found that this isotope emits low energy positrons[8] and hard γ-rays of about 2.5 Mev in energy. The results are summarized in Table I.

There is no doubt that the 5-min. and the 12.8-hr. periods are caused by Cu^{66} and Cu^{64}, respectively. The 10-min. period is found when copper samples are bombarded by neutrons of more than 12 Mev in energy[9] and is also obtained by photodisintegration.[1, 10] Moreover, it emits positrons. These three facts rather definitely determine the assignment of this isotope to be Cu^{62}.

The 2.5-hr. period must be either Ni^{63} or Ni^{65}. If the relative abundance of Cu^{63} and Cu^{65} and that of Ni^{62} and Ni^{64} is taken into account it is most likely to be Ni^{63}. The three-hour period found in nickel with slow neutron bombardments[11] may be caused by the same isotope.[12]

As for the long life zinc isotope, it should be Zn^{65} which must be formed as the result of a rare

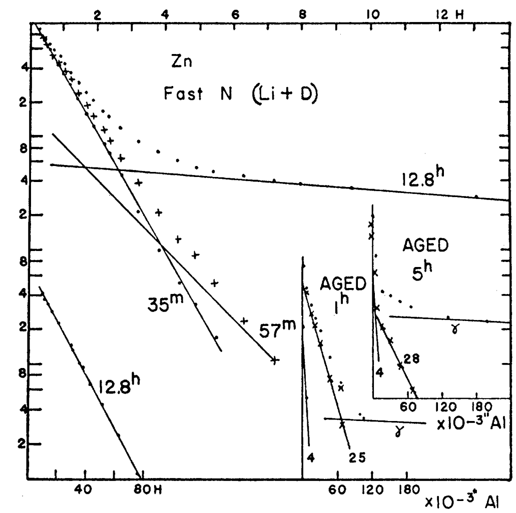

FIG. 2B. A decay curve for zinc when bombarded by fast neutrons. This is analyzed in three periods, 35 min., 57 min. and 12.8 hr. Note that the 57-min. period appears very weak compared with Fig. 2A and no trace of the 210-day period is found.

process of deuteron capture. Discussions for the assignment are given in the next section. Nothing has been done for the investigation of Co isotopes reported by Segrè.

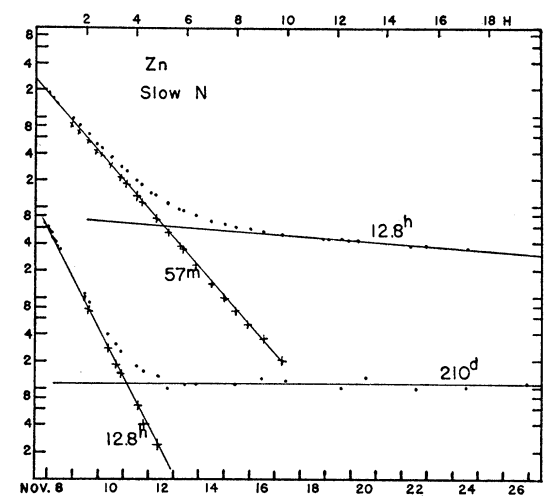

FIG. 2A. A decay curve for zinc when bombarded by slow neutrons. This is analyzed in three periods, 57-min. 12.8-hr. and 210 day.

ZINC

With slow neutrons, a 57-min. period was produced very strongly. In addition to this, a

[6] I am indebted to Dr. A. H. Snell for the use of his device.

[7] Perrier, Santangelo and Segrè, Phys. Rev. **53**, 104 (1938).

[8] A study of β-rays with a cloud chamber has been done in Tokyo and the results will be published soon.

[9] Sagane, Phys. Rev. **53**, 492 (1938).

[10] Bothe and Gentner, Nature, pp. 25, 90, 126, 191, 284 (1937).

[11] Naidu, Nature **137**, 578 (1936). Rotbalt, Nature **136**, 515 (1935). Oeser and Tuck, Nature **139**, 1110 (1937).

[12] Simultaneously with my publication at the Stanford meeting, Heyn reported (Physica **4**, 1224 (1937)) his recent results which are in good agreement with mine except a slight discrepancy of a numerical value for half-value periods.

TABLE II. *Summary of results obtained in the bombardment of zinc.*

ISOTOPES	Cu^{66}	Zn^{63}	Zn^{69}	Ni^{63}	Cu^{64}	Zn^{65}
Bombardment						
Slow neutrons			57 min. (n, γ)		12.8 hr. (n, p)	210 days (n, γ)
Fast neutrons Li	5 min. (n, p)	35 min. $(n, 2n)$	w57 min. (n, γ)		12.8 hr. (n, p)	
Fast neutrons	(6 min.)	(40 min.)		(2.5 hr.) (n, γ)	(12.5 hr.)*	
Fast neutrons Be	5 min. (n, p)	35 min. $(n, 2n)$	w57 min. (n, γ)		12.8 hr. (n, p)	
Deuterons			(1 hr.)		12 hr., 25 hr., 97 hr.†	
γ-rays	38 min.‡					

* Heyn.
† Livingood.
‡ Bothe, Gentner, Chang, Goldhaber, Sagane.

5-min. and a 12.8-hr. period were also activated, as had been reported by Fermi and others.[2] There appeared also a very long period of about 200 days.

With fast neutrons, Heyn[2] reported 6-min. and 60-min. periods, while Pool, Cork and Thornton[13] reported 6-min., 40-min. and 12.8-hr. periods. According to the present experiment a 35-min. period is produced as well as the 5-min. and 12.8-hr. periods. The 57-min. period was also obtained with a very weak activity, but no trace of the 210-day period was found. Examples of the decay curves obtained are shown in Figs. 2, A and B.

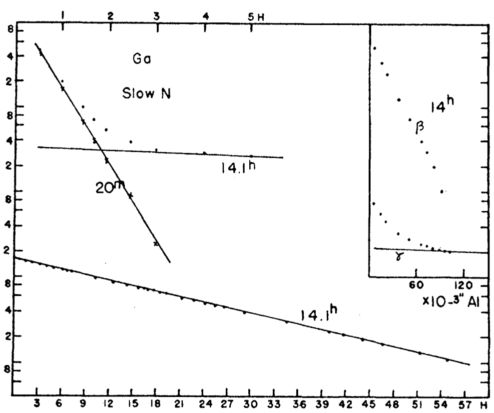

FIG. 3A. Decay curves obtained when gallium samples were bombarded by slow neutrons. No trace of the 23-hr. period was found.

Chemical separations showed that the 35-min., 57-min. and 210-day periods were all zinc isotopes and the 12.8-hr. period was a copper isotope.

No trial was made for deuteron bombardment, as Dr. Livingood has already bombarded zinc samples with deuterons, and intends to repeat it to check his previous results. According to him, four periods, 1 hr., 12 hr., 25 hr. and 97 hr. were reported.

By hard γ-ray bombardments,[1, 10] it has been reported that a 38-min. period was obtained in zinc. The results are summarized in Table II.

The 35-min. period was only activated appreciably with fast neutrons of energy higher than 9 Mev[3] and it emits positrons. These facts suggest that this period is produced by the neutron loss process, i.e., the $(n, 2n)$ reaction. So this must be Zn^{63}. The 38-min. period reported by γ-ray bombardments and the 34-min. period reported by Ridenour and Henderson[14] on the α-particle bombardments of nickel, the 40-min. period reported by Pool, Cork and Thornton[13] must be the same.

The 57-min.[8] period was very water-sensitive, and it appeared rather weak in activity when bombarded with fast neutrons. This isotope emits negative electrons. If the relative abundance of Zn^{68} (17.8 percent) and Zn^{70} (0.4 percent) are taken into account, it is most likely that this isotope is Zn^{69}, in agreement with the new results of Heyn[12] and contrary to the assignment of Thornton.[15]

The one-hour period reported by Livingood[2] may be the same isotope.

The long life period of order of about 210 days was obtained only by slow neutrons. It emits positrons of rather low energy accompanied by Compton electrons produced by its hard γ-rays. An absorption measurement of γ-rays showed that this isotope emits very hard γ-rays ($Pb\frac{1}{2}$ $\sim$15 mm) which is in good agreement with the results obtained on the zinc isotope produced by Cu+D. There remains only one possibility for a positron emitting zinc isotope which can be

[13] Pool, Cork and Thornton, Phys. Rev. **52**, 239 (1937).

[14] Ridenour and Henderson, Phys. Rev. **52**, 889 (1937).

[15] Thornton, Phys. Rev. **53**, 326 (1938).

produced by Cu+D and Zn+n, so this isotope must be Zn^{65}.

Gallium

With slow neutrons, two periods, 20 min. and 23 hr. were reported first by Fermi and his collaborators.[2] When bombarded with fast neutrons. Pool, Cork and Thornton[9] reported that they observed 20 min. (e^-), 55 min. (e^+), 1.7 hr. (e^+) and 22 hr. (e^-).

As shown in Fig. 3A the decay curves obtained with slow neutron bombardments gave only two periods; the well-known 20-min. period and a new 14-hr. period. No trace of the 23-hr. period reported by Fermi and others or by the Michigan group was found. With fast neutrons only three periods, 20 min., 66 min. and 14 hr. were found. Not even a trace of the 1.7-hr. period or the 23-hr. period was found. Chemical separations showed that all three periods were due to gallium isotopes.

As this sample has a very low melting point, no attempt has as yet been made at deuteron bombardment. The results are summarized in Table III.

Ga^{70} and Ga^{72} are the only two possible isotopes for a radioactive gallium which emits negative electrons. If the fact is taken into account that the 20-min. period is activated not only by slow neutrons but also by fast neutrons and γ-rays, it is quite certain that this 20-min.[16] period must be caused by Ga^{70}. The 18.2-min. period reported by DuBridge and his collaborators[17] in $Zn+p$ may be caused by the same isotope. On the other hand, the other 14-hr. period which is a negative electron emitter is not produced by γ-ray bombardments. These two points support very well the conclusion that this 14-hr. period should be caused by Ga^{72}. The 66-min.[8] period can only be obtained when the energy of the bombarding neutrons is higher than 6 Mev.[3] This isotope emits positrons and is produced also by hard γ-ray bombardments. These facts afford good evidence that this isotope is Ga^{68}, which is most probably a positron-emitting a Ga isotope.

The author must add a line about a negative result in attempting to check the reaction Ga^{69} (n, α) Cu^{66} which we have reported in *Nature*.[1] Although several very short exposures to fast neutrons, less than one minute, were made, no trace of the five-min. period was found.

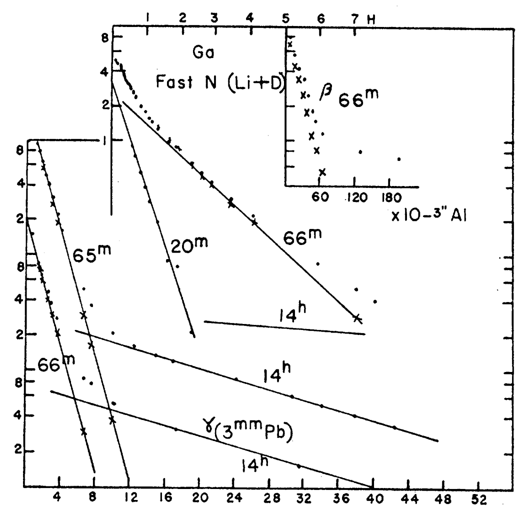

FIG. 3B. Decay curves obtained when gallium samples were bombarded by fast neutrons.

Germanium

Germanium with slow neutrons gives four periods, 81 min., 8 hr., 26 hr. and six days. With fast neutrons 29-min., 81-min., very weak 8-hr., 26-hr. and six-day periods were produced. As for

[16] Mann, Phys. Rev. 52, 405 (1937).
[17] DuBridge, Barnes and Buck, Phys. Rev. 51, 995 (1937).

TABLE III. *Summary of results obtained in the bombardment of gallium.*

Isotope	Ga^{70}	Ga^{68}	Ga^{72}
Bombardment			
Slow neutrons	20 min. (n, γ)		14 hr. (n, γ)
Fast neutrons Li	20 min. (n, γ) $(n, 2n)$	66 min. $(n, 2n)$	14 hr. (n, γ)
Fast neutrons Be	20 min. (n, γ) $(n, 2n)$	66 min. $(n, 2n)$	14 hr. (n, γ)
γ-rays	20 min. (γ, n)	66 min. (γ, n)*	

* Bothe, Gentner, Chang, Goldhaber, Sagane.

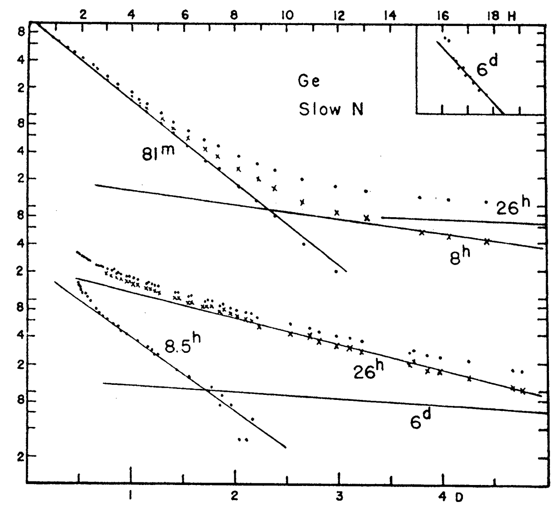

FIG. 4A. A decay curve obtained when germanium was bombarded by slow neutrons from Be+D surrounded by paraffin. This curve may be analyzed in four periods, 81 min., 8 hr., 26 hr. and 6 day.

deuteron bombardments, not much has been done yet and some of the results are still inaccurate. The results so far obtained show seven periods 80 min., 88 min., 12 hr., 50 hr., 16 days and 55 days. Examples of the decay curves obtained are reproduced in Figs. 4-A, B, C, D, E. The results are summarized in Table IV.

Since the positron-active 29-min. period was produced in appreciable intensity when the energy of the bombarding neutrons is higher than 5.5 Mev,[3] it is most likely caused by Ge^{69}. The 81-min.[8] period was formed in strong intensity in each bombardment. This isotope emits negative electrons and is sensitive to slow neutrons. Because of the relative abundance of Ge^{74} (37 percent) and Ge^{76} (6.5 percent), the isotope in question is probably Ge^{75}.

The 88-min. period obtained in deuteron bombardments is positron active and decay curves were followed by measuring only the positron activity. Although no chemical separation has as yet been carried out this period may be caused by As^{73}.

The 8-hr. period was obtained appreciably only in slow neutron bombardments. With fast neutrons only a trace of this period was noticed, indicating very clearly that this period is very sensitive to slow neutrons. There remains only one

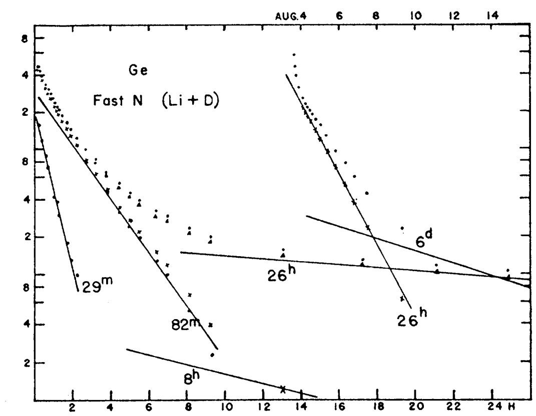

FIG. 4B. A decay curve obtained when germanium was bombarded by fast neutrons from Li+D. This curve may be analyzed in five periods, 29 min., 82 min., 8 hr., 26 hr. and 6 day. Note that the 81-min. and 8-hr. periods appear rather weak compared with Fig. 4A.

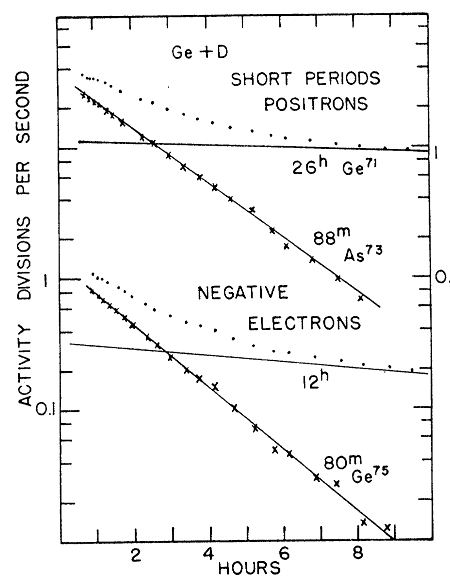

FIG. 4C. Parts of decay curves showing short periods obtained when germanium was bombarded by deuterons. These curves were taken by measuring only positrons or negative electrons with a simple magnet close to the electroscope.

possibility for this kind of negative electron active period, that is Ge^{77}.

The 26-hr. period is positron active and can be obtained with both fast and slow neutrons. These points are good enough to assign this period to Ge^{71}.

The 50-hr. period found in deuteron bombardments is expected to be caused by an arsenic isotope, probably As^{71}, because it emits positrons. The positrons emitted are of very low energy and are accompanied by strong γ-rays.

The 6-hr. period is rather too weak in activity to check the sign of the β-particles emitted. Thus not much can be said about this period, but I suspect that it may be caused by a gallium isotope, probably Ga^{74}.

The 12-hr. period found in Ge+D may be identical to Ge^{77} (8 hr.) or might be Na^{24} contamination.

The 16-day period was obtained by deuteron bombardments only. This isotope emits both positive and negative electrons and was proved to be an arsenic isotope by chemical separations. These points allow one to ascribe this period to As^{74}, as has been reported recently.[18]

In addition to these periods a very soft negative β-ray emitter was noticed, the half-life of which is about 55 days. This was also proved to be an arsenic isotope by chemical test and is most likely As^{77}.

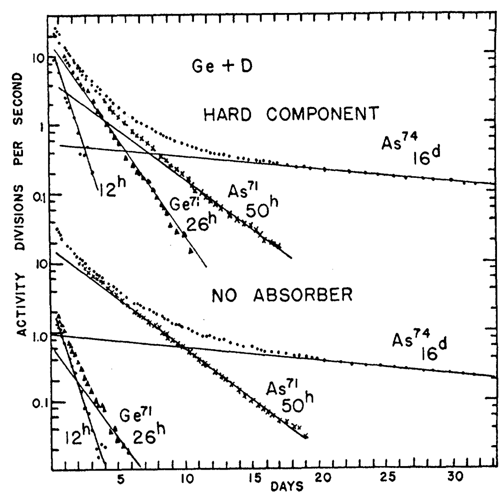

FIG. 4D. Parts of decay curves showing 26-hr. and 50-hr. periods obtained when germanium was bombarded by deuterons. A decay curve obtained by measuring the activity with an aluminum absorber 0.024″ thick is mentioned as hard component.

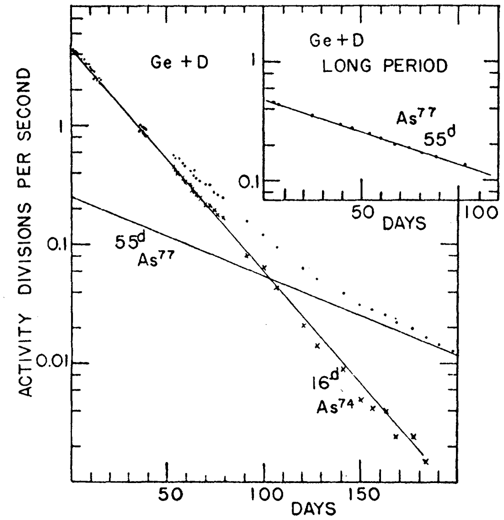

FIG. 4E. Parts of a decay curve showing long periods obtained when germanium was bombarded by deuterons.

Summary of Reactions

Figure 5 shows a portion of the periodic table in the neighborhood of Cu, Zn, Ga and Ge. The radioactive isotopes are shown in circles and the arrows indicate the reactions studied in this article. In some of these samples, existence of K radiation was found. But to get definite conclusions, further work must be done, because internal conversion of γ-rays must be taken into account.

The values of half-value thickness or maximum range of β-rays are not given intentionally, because they depend too much on the geometry of the measuring apparatus; on the other hand, the study of the β-rays of those isotopes discussed has been started in Tokyo and some of the results are expected to be published soon.

Acknowledgment

The author is very greatly indebted to Professor E. O. Lawrence for the opportunity of using

[18] Sagane, Kojima and Ikawa, Phys. Rev. **54**, 149 (1938).

TABLE IV. *Summary of results obtained in the bombardment of germanium.*

Isotopes	Ge^{69}	Ge^{75}	As^{73}	Ge^{77}	Ge^{77}	As^{61}	Ga?	As^{74}	As^{77}
Bombardment									
Slow neutrons		81 min. (n,γ)		8 hr. (n,γ)	26 hr. (n,γ)		6 days (n,p)		
Fast neutrons Li	29 min. $(n,2n)$	81 min. (n,γ)		w 8 hr. (n,γ)	26 hr. (n,γ)		6 days (n,p)		
Fast neutrons Be	29 min. $(n,2n)$	81 min. (n,γ)		w 8 hr. (n,γ)	26 hr. (n,γ)		6 days (n,p)		
Deuterons		80 min. (d,p)	88 min. (d,n)	12 hr.?	26 hr. (d,p)	50 hr. (d,n)		16 days (d,n)	55 days (d,n)

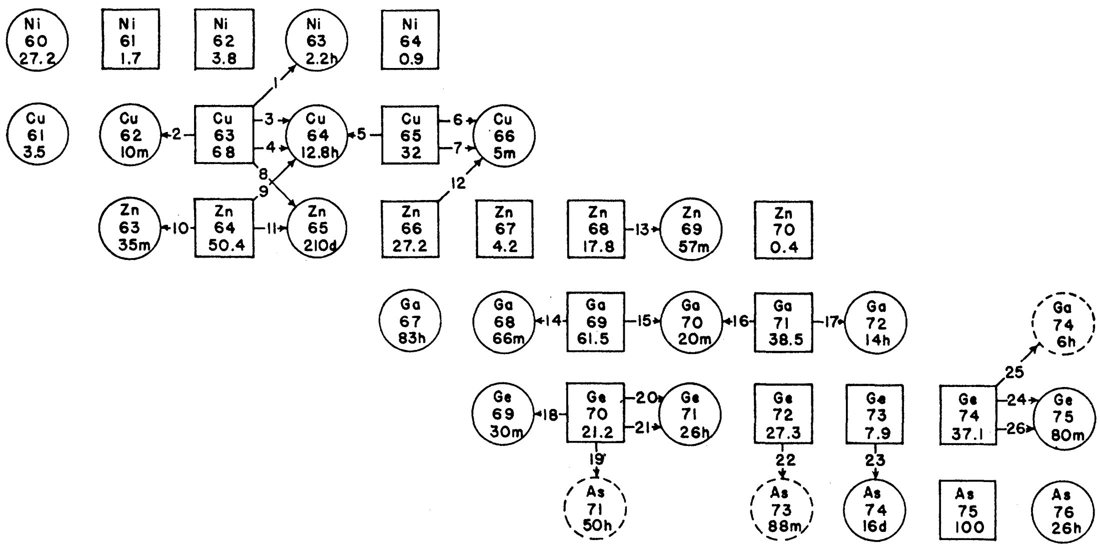

FIG. 5. This diagram gives a summary of all the transmutations described in this paper. The stable isotopes (squares) are given with their percentage abundance in small figures. Radioactive isotopes are given in circles, which are dotted when there is still some ambiguity as to their exact identification. The numbered arrows indicate transmutations which have been observed, and the reactions describing them are as follows:

‡1	Cu^{63} (n, p)	Ni^{63}	9	Zn^{64} (n, p)	Cu^{64}	16	Ga^{71} $(n, 2n)$	Ga^{70}	*23	Ge^{73} (d, n)	As^{74}
2	Cu^{63} $(n, 2n)$	Cu^{62}	10	Zn^{64} $(n, 2n)$	Zn^{63}	*17	Ga^{71} (n, γ)	Ga^{72}	*24	Ge^{74} (n, γ)	Ge^{75}
3	Cu^{63} (n, γ)	Cu^{64}	*11	Zn^{64} (n, γ)	Zn^{65}	*18	Ge^{70} $(n, 2n)$	Ge^{69}	†*25	Ge^{74} (n, p)	Ga^{64}
4	Cu^{63} (d, p)	Cu^{64}	12	Zn^{66} (n, p)	Cu^{66}	†*19	Ge^{70} (d, n)	As^{71}	*26	Ge^{74} (d, p)	Ge^{75}
5	Cu^{65} $(n, 2n)$	Cu^{64}	‡13	Zn^{68} (n, γ)	Zn^{69}	*20	Ge^{70} (n, γ)	Ge^{71}	*27	Ge^{76} (d, n)	As^{77}
6	Cu^{65} (n, γ)	Cu^{66}	14	Ga^{69} $(n, 2n)$	Ga^{68}	*21	Ge^{70} (d, p)	Ge^{71}	*28	Ge^{76} (n, γ)	Ge^{77}
7	Cu^{65} (d, p)	Cu^{66}	15	Ga^{69} (n, γ)	Ga^{70}	†*22	Ge^{72} (d, n)	As^{73}	*29	Ge^{76} (d, p)	Ge^{77}
‡8	Cu^{63} (d, γ)	Zn^{65}									

* Reactions which are newly reported in this paper.
† Reactions which were observed but not yet checked carefully enough.
‡ Reactions which have not been established well, but are proved in this paper to exist.

the cyclotron and for his helpful discussion and encouragement. He is also very greatful to the staff of the Radiation Laboratory for their valuable suggestions and discussions of this work. This work has been aided by grants from the Research Corporation.