

The Nuclear Spectrum of Ge⁷⁷*

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The disintegration of Ge⁷⁷ (12 hour) has been studied with the help of a magnetic lens spectrometer, coincidence counters, and scintillation counters. The beta-ray spectrum consists of three groups of whose end-point energies are 2.196 Mev, 1.379 Mev, and 0.710 Mev. In addition there are 13 gamma-rays appearing in the product nucleus As⁷⁷, some of which are internally converted. By coincidence counting techniques it has been shown that the beta-ray of energy 2.196 Mev does not go to the ground state but to an excited state 0.264 Mev above the ground state. A disintegration scheme is suggested.

I. INTRODUCTION

EARLY work on germanium activities produced by cyclotron bombardment gave rise to somewhat conflicting results. Among several activities produced, one of approximately 12 hours half-life was assigned^{1,2} to Ge⁷⁷. Work on fission products by Steinberg and Engelkemeir³ established that Ge⁷⁷ has a half-life of 12 hours and is the parent of a 40-hr As⁷⁷. They obtained a beta-ray end point by absorption of 2.0 Mev. Arnold and Sugarman⁴ showed that in addition to the 12-hr period, there is an isomeric state of Ge⁷⁷ having a half-life of 59 sec. They showed that this state decays, in part, directly to the ground state of As⁷⁷ by the emission of a beta-ray whose end-point energy they determined by absorption to be 2.8 Mev. Finally, Mandeville, Woo, Scherb, Keighton, and Shapiro⁵ studied the radiations from the 12-hr Ge⁷⁷ by means of absorption and coincidence counting techniques. The end point of the beta-ray spectrum, which they obtained by absorption in aluminum, was 1.74 Mev. Coincidences were found

between beta-rays and a gamma-ray, estimated to have an energy of approximately 0.5 Mev.

A detailed nuclear spectroscopic examination of the radiations from Ge⁷⁷ was undertaken in order to determine its disintegration scheme. The determination of this scheme completes a group at mass number 77 consisting of As⁷⁷, Se^{77m}, and Br⁷⁷ recently under investigation in this laboratory.

II. METHOD OF PREPARATION OF SOURCES

Sources of the 12-hr Ge⁷⁷ were prepared by bombarding germanium metal with deuterons in the Indiana University cyclotron. The germanium metal was dissolved in aqua regia, and arsenic carrier was added to the solution. The germanium was separated by distilling GeCl₄ from the solution in the presence of chlorine gas. The resultant germanium ion was precipitated as sulfide.

Germanium prepared in this manner contains, in addition to Ge⁷⁷, Ge⁷¹ (11 days), Ge⁷⁵ (82 minutes), and some As⁷⁷ which grows from the Ge⁷⁷. Since none of these isotopes except Ge⁷⁷ emit gamma-rays, the material can be used directly to measure the gamma-ray spectrum. In order to investigate the beta-ray spectrum, the source was not separated until 12 hours after bombardment in order to let the Ge⁷⁵ die out. The germanium was then separated from the arsenic and the beta-ray spectrum was measured over short periods of time so that the amount of As⁷⁷ growing from the Ge⁷⁷ would be negligible. Several sources were used in the measurement of the spectrum and a composite of the results prepared.

III. MEASUREMENT OF THE GAMMA-RAYS

The energies of the gamma-rays were determined by measuring the energies of secondary electrons ejected from lead or uranium radiators in a magnetic lens spectrometer. Figure 1 shows the results of a series of measurements taken in this way. The main curve of Fig. 1 was obtained using a lead radiator of surface density 16 mg/cm² and a counter window whose cutoff was about 16 kev. *K*-lines and, in most cases, the accompanying *L*-lines for five gamma-rays are shown on this curve. In addition there is evidence for an *L*-line (*L*₂) for a gamma-ray of low energy.

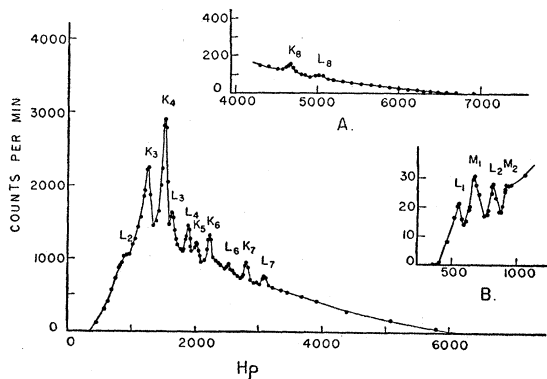


FIG. 1. Photoelectrons ejected by γ -rays of Ge⁷⁷.

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¹ Seaborg, Livingood, and Friedlander, *Phys. Rev.* **59**, 320 (1941).

² Sagane, Miyamoto, and Ikawa, *Phys. Rev.* **59**, 904 (1941).

³ E. P. Steinberg and D. W. Engelkemeir, *National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Radiochemical Studies; The Fission Products, Paper No. 54* (McGraw-Hill Book Company, Inc., New York, 1951), p. 566.

⁴ J. R. Arnold and N. Sugarman, *J. Chem. Phys.* **15**, 703 (1947).

⁵ Mandeville, Woo, Scherb, Keighton, and Shapiro, *Phys. Rev.* **75**, 1528 (1949).

In order to investigate the part of the spectrum of low energy a thinner window, supported by a grid, was fitted to the counter. This window transmitted electrons of energy 5 kev or greater. The results of this investigation are shown in Curve B of Fig. 1, where it will be seen that *L* and *M* lines were found for gamma-rays having energies of 42 kev (*L*₁, *M*₁) and 73 kev (*L*₂, *M*₂).

In order to check the energies obtained using the lead radiator and to bring up lines of high energy and low intensity, an experiment was made using a uranium radiator of surface density 90 mg/cm². Curve A of Fig. 1 shows the high energy portion of this curve. The *K*- and *L*-lines designated as *K*₈, *L*₈ correspond to a gamma-ray of energy 1.105 Mev. In addition, it will be seen that there are Compton electrons for a weak gamma-ray of higher energy extending beyond the photolines for the gamma-ray at 1.105 Mev. The energy of this line, estimated from the energy of the Compton electrons, is 1.75 Mev.

The energies of these gamma-rays, together with

TABLE I. Energies of the gamma-rays from Ge⁷⁷.

Line No.	Energy ^a (Mev)	Line No.	Energy ^a (Mev)
(a) Lines obtained from measurement of photoelectrons.			
1	0.042	6	0.418
2	0.073	7	0.564
3	0.213	8	1.105
4	0.264	13 from	1.75
5	0.368	Compton	...
		end point	
(b) Lines obtained from measurements of internal conversion electrons.			
3	0.209	5	0.366
4	0.268	11	0.408
9	0.300	6	0.425
10	0.327	12	0.466

^a Energies in this table are averages of all determinations from Pb and U radiations and internal conversion lines.

some additional gamma-rays appearing as internal conversion lines in the electron spectrum, are shown in Table I. It is difficult to make an estimate of the relative intensities of the lines in a spectrum as complicated as this. Nevertheless, an attempt has been made to do this, using the photoelectric cross sections for lead obtained from Gray's curves. The results serve as a guide in constructing the decay scheme. The strongest line in the spectrum is that at 0.264 Mev. Those at 0.213 and 0.564 Mev are of about equal intensity, and each has an intensity about 0.6 that of the line at 0.264 Mev.

IV. THE BETA-RAY SPECTRUM

The measurement of the beta-ray spectrum posed a difficult problem. In the method of preparing the source, as mentioned earlier, there is always some Ge⁷⁵ (82 min) produced. In order to get rid of the Ge⁷⁵ activity, the source was allowed to decay for 12 hours after bombardment before making a chemical separation. This, of course, decreased the initial activity of the Ge⁷⁷ to

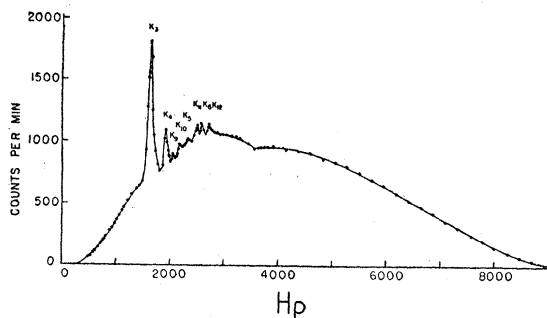


FIG. 2. Beta-spectra of Ge⁷⁷.

one-half. The second difficulty concerns the growth of As⁷⁷ from Ge⁷⁷. As⁷⁷ is a pure beta-ray emitter having an end-point energy⁶ of 0.700 Mev. Measurements on the Ge⁷⁷ beta-spectrum at energies between 0.700 Mev and the end point (2.196 Mev) could therefore be made without interference from the beta-rays of As⁷⁷. At the lower energies, experiments were made in such a way that only a short time elapsed during the counting period so that the As⁷⁷ did not have time to grow in appreciably. The sources consisted of a slurry of GeS₂ deposited on a thin zapon film.

The results are shown in Figs. 2 and 3. Figure 2 is a plot of the beta-ray spectrum together with the internal conversion lines. The most intense internal conversion line corresponds to the gamma-ray of energy 0.213 Mev and the next intense to that of 0.264 Mev. The energies computed from the internal conversion lines are 0.209 and 0.268 Mev, in good agreement with the values obtained from the photoelectron spectrum. Of the remaining lines, those corresponding to gamma-ray energies of 0.366 and 0.425 Mev correspond to the gamma-rays obtained from the photoelectron spectrum with energies of 0.368 and 0.418 Mev. The other lines at 0.300, 0.327, 0.408, and 0.466 Mev were not seen in the photoelectron spectrum, possibly because of overlapping.

The results of a Fermi analysis of the data are shown in Fig. 3. It will be seen that there are three groups of beta-rays whose end-point energies, relative abundances, and log*f**t* values are shown in Table II.

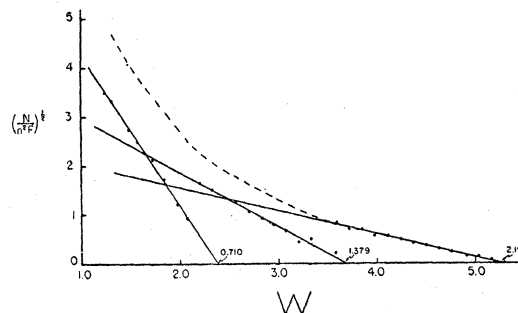


FIG. 3. Fermi analysis of beta-groups from Ge⁷⁷.

⁶ R. Canada and A. C. G. Mitchell, Phys. Rev. **81**, 485 (1951).

TABLE II. Characteristics of the beta-ray spectrum.

Group	End-point energy (MeV)	Relative abundance (percent)	$\log_{10} ft$
1	2.196	42	7.6
2	1.379	35	6.6
3	0.710	23	5.7

V. COINCIDENCE EXPERIMENTS

From the large number of gamma-rays observed it is apparent that the disintegration scheme of Ge^{77} is quite complicated. In order to get a starting point for constructing the scheme, it is necessary to know whether or not the most energetic beta-ray leads to the ground state of As^{77} . In order to determine this, the coincidence experiments of Mandeville *et al.*⁵ were repeated under somewhat better controlled conditions. The apparatus and procedures used in this experiment were the conventional ones which have been in use in this laboratory for a number of years.^{7,8} A cylindrical lead cathode counter was used for counting gamma-rays and a counter with an end window of mica for counting beta-rays. Since some Ge^{71} was always present in the sources which were used, care had to be taken not to confuse the x-rays from Ge^{71} with the electrons from Ge^{77} . Since it was found⁹ in an earlier experiment on Ge^{69} that 0.10-cm aluminum reduced the x-ray count from Ge^{71} to a negligible value, coincidence experiments were made at absorber thicknesses greater than this. A curve showing the number of beta-gamma coincidences ($N_{\beta\gamma}/N_{\beta}$) against absorber thickness in cm aluminum, from 0.25- to 0.29-cm of aluminum, is shown in Fig. 4. This region corresponds to energies greater than 1.38 Mev, the end point of the middle group, out toward the end point of the highest energy group. It will be seen that beta-gamma coincidences are present and that the number per disintegration is independent of the energy over this region. This shows that the beta-ray group of highest energy does not go the ground state.

VI. DISCUSSION

From the data given in the preceding sections it is possible to construct a decay scheme which gives a good energy fit with the gamma-ray and beta-ray data and is consistent with the relative intensities of the gamma-rays as far as they can be determined. The scheme which results from these considerations is extremely complicated and for this reason must be considered as tentative. It is shown in Fig. 5.

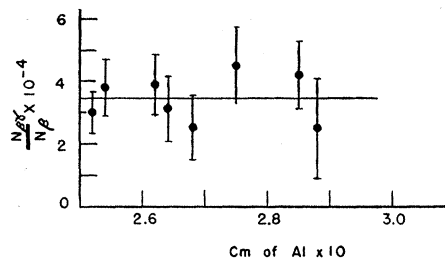
Leaving aside that part of the scheme having to do with the isomeric state of Ge^{77} of half-life 59 sec, which will be discussed later, it has been shown that the beta-ray transition of energy 2.196 Mev does not lead to the

ground state. Since the most intense gamma-ray is that whose energy is 0.264 Mev, it probably lies at the bottom of the scheme as shown. This gamma-ray and the highest energy beta-ray fix the energy of the ground state of Ge^{77} at 2.460 Mev. The remaining two beta-rays determine levels in As^{77} at 1.750 and 1.10 Mev from which there are gamma-rays to the ground state. The remaining gamma-rays are placed in the scheme in such a way as to give the best fit energy-wise. The fit is good to about 2 percent in the worst case.

It is surprising that two such low energy gamma-rays as those of energies 0.042 and 0.073 Mev occur so high up in the scheme and in addition are not appreciably internally converted. A test of this scheme would be to show that the 0.073-Mev gamma-ray is in coincidence with those of 0.564-Mev or of 0.213-Mev and 0.264-Mev energy. Such an experiment has been performed for us by Mr. M. M. Miller of this laboratory with two crystal counters in coincidence, and indeed shows that the 0.073-Mev line is in coincidence with those at 0.213 and 0.264 Mev. This experiment is described in the Appendix.

The energy level diagram also shows the energy relation between the 59-sec isomeric state of Ge^{77} and the ground state. Experiments by Arnold and Sugarman⁴ have shown that the 59-sec state of Ge^{77} decays to the ground state of As^{77} with the emission of a 2.8-Mev beta-ray. Mitchell and Smith¹⁰ have measured the energy of the gamma-ray transition between 59-sec and 12-hr states of Ge^{77} by means of a scintillation spectrograph and have found a value of 0.380 ± 0.020 Mev. Since the value of $\log ft$ for the 59-sec beta-transition is 4.8, the transition is characterized by no change in parity and $\Delta j = 0, \pm 1$. Since the ground state of As^{77} has the configuration $p_{3/2}$, the 59-sec metastable state has the configuration $p_{1/2}$.

From the results of the present work considered in the light of the shell theory, it is possible to give the configurations of the two lowest states as As^{77} and that of the ground state of Ge^{77} . The ground state of As^{77} is known to have a configuration $p_{3/2}$. From shell model predictions the first excited state of As^{77} would be expected to have the configuration $f_{5/2}$. Ge^{77} has 45 neutrons or 7 neutrons beyond neutron number $N=38$. One would expect the configuration of one of the

FIG. 4. Beta-gamma coincidences in Ge^{77} .

⁷ A. C. G. Mitchell, *Revs. Modern Phys.* **20**, 296 (1948).

⁸ J. L. Meem, Jr., and F. Maienschein, *Phys. Rev.* **76**, 328 (1949).

⁹ C. M. Huddleston and A. B. Smith, *Phys. Rev.* **84**, 289 (1951).

¹⁰ A. C. G. Mitchell and A. B. Smith, *Phys. Rev.* **85**, 152 (1952).

isomeric^{10,11} states, the lower one in this case, to be $G_{7/2}$ and the other $p_{1/2}$. The present experiments are consistent with this assumption. The beta-ray transition leading from the ground state of Ge⁷⁷ to the first excited state of As⁷⁷ shows a $\log ft = 7.6$ and has an allowed shape, which is consistent with a transition of the type $\Delta j = \pm 1$, yes. If, on the other hand, the ground state of Ge⁷⁷ had the configuration $g_{9/2}$, the beta-ray spectrum would have a forbidden shape ($\Delta j = \pm 2$, yes) and a much larger value of $\log ft$. The present experiments are therefore consistent with the configurations predicted by the shell model and shown in Fig. 5 for the two states of Ge⁷⁷ and the ground and first excited states of As⁷⁷. It is impossible at the present time to predict the configurations of the many other excited states of As⁷⁷.

The author wishes to express his thanks to Dr. A. C. G. Mitchell, who suggested this problem, for help and

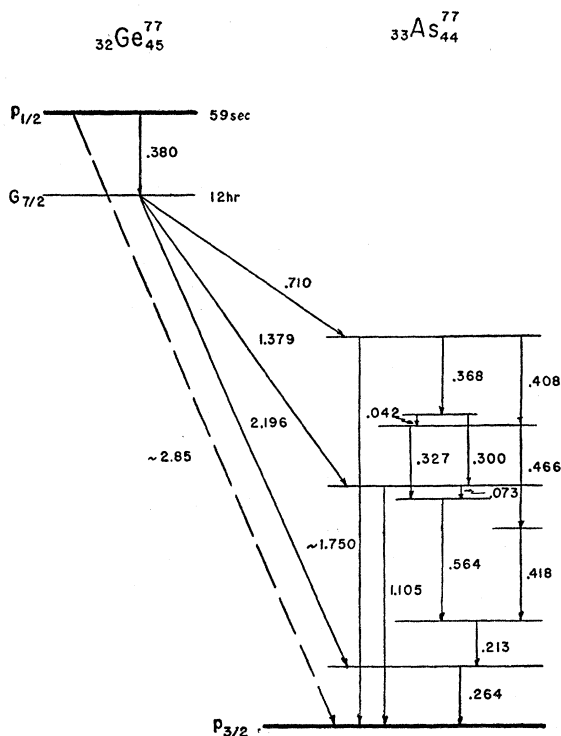


FIG. 5. Tentative decay scheme of Ge⁷⁷.

¹¹ M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

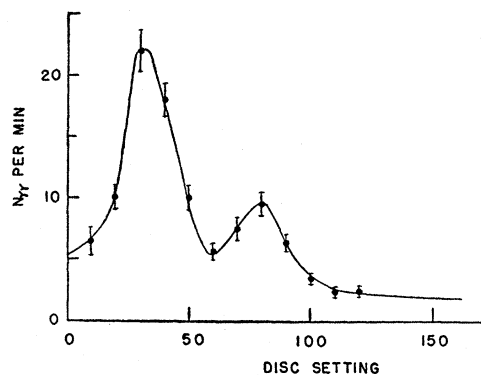


FIG. 6. Gamma-gamma coincidences in Ge⁷⁷ obtained with scintillation counter.

suggestions during the carrying out of the experiments, to Mr. C. M. Huddleston for taking some of the readings, and to Mr. A. E. Lessor for making the chemical separations. He also wishes to thank Dr. M. B. Sampson and the cyclotron crew for making the bombardments. He also wishes to acknowledge that two sources of Ge⁷⁷ irradiated by neutrons in a pile were received, one from the Oak Ridge National Laboratory and one from the Argonne National Laboratory. Unfortunately neither of these sources was strong enough to measure.

APPENDIX

The proposed disintegration scheme of Ge⁷⁷ assumes that a gamma-ray of energy 0.073 Mev lies high in the disintegration scheme and is in cascade with lines at 0.263 and 0.213 Mev. A test of this part of the scheme would be to show that the 0.073-Mev line is in coincidence with the lines at 0.263 Mev or 0.213 Mev or both.

The apparatus consisted of two scintillation counters with NaI(Tl) crystals and 5819 photomultiplier tubes.¹² The output of each tube was fed through a linear amplifier to a differential pulse-height discriminator and then to either a scalar or a fast coincidence circuit. After a preliminary investigation of the peaks obtained in the singles channels, channel 1 was set on the high side of the peak arising from the 0.263-Mev and 0.213-Mev lines, and the coincidences were measured when channel 2 was run over the spectrum. The result is shown in Fig. 6, in which the number of coincidences per minute is plotted against the dial setting in channel 2. It will be seen that two well-defined peaks occur. The peak at dial setting 30 corresponds to a gamma-ray of approximately 0.070 Mev, while that at dial setting 80 corresponds to a gamma-ray of energy about 0.21 Mev. This shows that the lines at 0.073 Mev and 0.213 Mev are in coincidence with that at 0.263 Mev, as is required by the decay scheme.

¹² For a description of the method see Miller, Pruett, and Wilkinson, Phys. Rev. 84, 849 (1951).