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Decay of As^{72}†

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The gamma rays following the decay of As^{72} have been studied with scintillation spectrometers both single and in coincidence. Sixteen gamma rays have been resolved and have necessitated the addition of levels at 2.91 and 3.74 Mev to the levels of Ge^{72} known from previous studies of the radiations of Ga^{72}. From delayed coincidence measurements gamma rays of 1.75, 2.24, and 3.0 Mev, in addition to annihilation radiation, have been observed to populate the isomeric (0^+) state in Ge^{72} at 0.69 Mev.

I. INTRODUCTION

The decay of Ga^{72} has been recently studied in this laboratory[1] and by Johns, Chidley, and Williams.[2] The two investigations led to very similar conclusions concerning the rather complicated level structure of Ge^{72}. Because of the anomalous state at 0.69 Mev, that has spin zero and even parity, it was felt important, however, to obtain additional and confirmatory information concerning the levels of Ge^{72} by studying the decay of As^{72}.

The decay of As^{72} (26-hour) has been studied by several investigators.[3–6] Mei *et al.*[5] found five positron groups at 3.339 Mev (19.3%), 2.498 Mev (61.6%), 1.844 Mev (12.1%), 0.669 Mev (5.0%) and 0.271 Mev (2.0%), a gamma ray of 0.835 Mev, and a conversion electron line at 0.69 Mev (intensity equal to 1.2% of the positrons present). The presence of higher energy gamma rays was noted but their weak intensity prevented any quantitative measurements. Stoker and Ong Ping Hok[6] while studying As^{71} have also observed the As^{72} conversion electron line at 0.69 Mev (intensity equal to 0.9% of the positrons present).

II. PREPARATION OF THE SOURCES

Sources of As^{72} were obtained by bombardment of Ga_2O_3 by alpha particles for 14 microampere-hours with the 60-inch cyclotron of the Crocker Radiation Laboratory of the University of California. Two irradiations were carried out, the first at 17 Mev and the second at 20 Mev. The chemical separations of the radioactive arsenic were carried out by means of a distillation of $AsCl_3$. The arsenic was then precipitated as As_2S_3. In the case of the first source 100 mg of As carrier were added, and 10 mg for the second source.

III. THE GAMMA-RAY SPECTRUM

The gamma-ray spectrum was determined using the single NaI crystal, collimated-beam, scintillation spectrometer that has been described previously.[1] The source was placed in a Lucite cup with a $\frac{1}{2}$-inch thick bottom to absorb the positrons. In Fig. 1, curve (a), is shown one of the spectra obtained. The intensities of the various gamma rays were determined by measuring the areas of the photopeaks and applying corrections for the efficiency of NaI and ratios of photopeak area to total area under the spectrum.[7] The method of successive subtractions of gamma-ray distributions from a complex spectrum yielded good agreement with beta-ray spectrometer measurements of gamma-ray intensities in the case of Ga^{72} (see references 1 and 2). Nevertheless the method may lead to systematic errors in the case of the lower energy gamma rays of small intensity (see Tables I and II).

The presence of other As activities in our sources required a background subtraction. In addition to As^{72} there were present As^{71} (60-hour), As^{73} (76-day), and As^{74} (17-day). In order to determine the proper background to be subtracted, the singles spectra were carefully followed as a function of time. As^{71} was found to be the major contributor to the background. In addition to the known[6,8,9] line at 0.175 Mev and annihilation radiation, other lines have been resolved in this study and are reported in Appendix A. As^{73} decays[10] solely by K-capture and emission of gamma rays of 0.054 and 0.013 Mev and hence contributed nothing to the energy region of interest in As^{72}. In an investigation of As^{74} currently being conducted at this laboratory there has been resolved a weak gamma ray of 1.20 Mev, in addition to the known[11] lines at 0.51 (annihilation quanta), 0.59, and 0.63 Mev.

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† This work was supported in part by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

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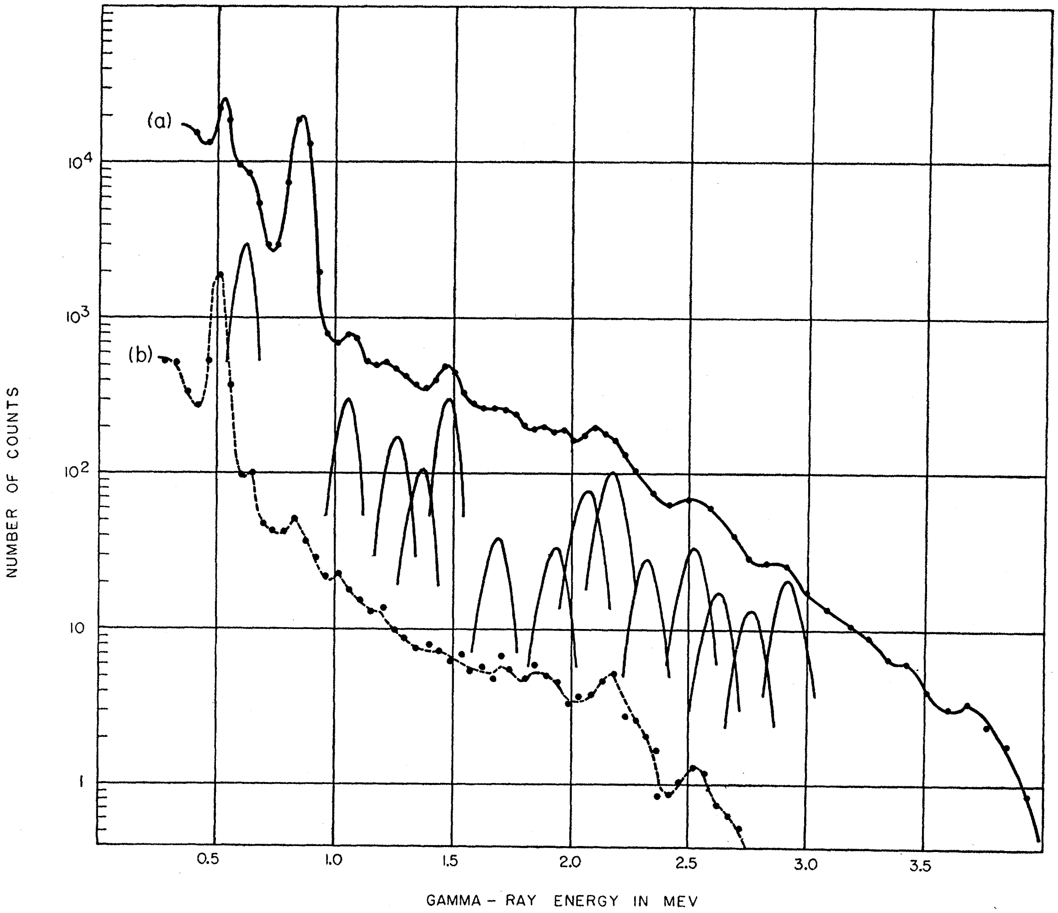

FIG. 1. Curve (a): Gamma-ray spectrum obtained with single NaI(Tl) crystal scintillation spectrometer. Curve (b): Gamma-gamma coincidence spectrum with gamma-ray discriminator set at 0.84 Mev.

In Table I are listed the energies and corrected intensities of the gamma rays that have been resolved. The values given represent averages from several runs at different gains. The presence of weak gamma rays below 0.63 Mev, found in the Ga^{72} work, cannot be excluded. Certainly the outstanding difference between the gamma-ray spectra of As^{72} and Ga^{72} is the relatively low intensity of the high-energy lines in As^{72}.

In order to determine the ratio of the total number of positrons to the number of 0.835-Mev gamma rays, a source was mounted between two $\frac{3}{16}$-inch flat pieces of copper. The gamma-ray spectrum below 0.9 Mev was measured with the source 14 cm from the counter and without any lead collimator. A Na^{22} source was then mounted in a position identical with the As^{72} source, and its gamma-ray spectrum was determined. After making the usual corrections, an analysis of the gamma-ray spectra showed that the intensity ratio of the positrons to the 0.835-Mev gamma ray in As^{72} was 0.96 ± 0.10.

IV. GAMMA-GAMMA COINCIDENCE MEASUREMENTS

For these measurements the source was placed between two flat pieces of copper, $\frac{1}{16}$ inch thick, and two flat pieces of lead also $\frac{1}{16}$ inch thick. This sandwich was put between two NaI scintillation counters. The coincidence circuitry, which was described previously,[1] was set to register prompt gamma rays in coincidence with gamma-ray pulses of 0.84 Mev. The coincident gamma-ray spectrum was displayed on a twenty-channel pulse-height analyzer. In order to subtract coincidences due to Compton electrons from higher-energy gamma rays under the 0.84-Mev photopeak, the gamma-ray spectrum coincident with 0.95-Mev pulses was determined next. The difference in the two spectra was analyzed in the usual fashion.[1] In Table II are listed the gamma-ray energies and intensities that resulted from that analysis.

V. DELAYED COINCIDENCE MEASUREMENTS

In order to determine what gamma rays populated the 0.3μsec isomeric state in Ge^{72}, the gamma-ray spectrum in coincidence with delayed conversion electrons of 0.69 Mev was measured. The electron counter consisted of a $\frac{1}{4}$-inch-thick piece of anthracene mounted on an RCA 5819 photomultiplier tube. A small amount of source was placed on cellophane tape between the beta- and gamma-ray counters. The gamma-ray counter was shielded by $\frac{1}{16}$ inch of copper.

TABLE I. Analysis of gamma-ray spectra.

Gamma-ray energy (Mev)	Relative intensity[a]	Percent per disintegration[b]
0.63 ±0.01	10.3	7.9 ±0.8
0.835[c]	100	76.6
1.05 ±0.01	2.7	2.1 ±0.5
1.25 ±0.02	2.2	1.7 ±0.3
1.37 ±0.02	1.4	1.1 ±0.5
1.46 ±0.01	4.0	3.1 ±0.5
1.68 ±0.02	1.1	0.9 ±0.9
1.92 ±0.03	1.0	0.8 ±0.4
2.08 ±0.02	2.6	2.0 ±0.4
2.20 ±0.02	3.7	2.8 ±0.5
2.32 ±0.03	1.2	0.9 ±0.5
2.51 ±0.02	1.7	1.3 ±0.3
(2.63)	(0.9)	(0.7)
(2.76)	(0.7)	(0.5)
2.91 ±0.03	1.5	1.2 ±0.4
3.74 ±0.05	0.3	0.25±0.1

[a] The intensities have been normalized by setting the intensity of the 0.835-Mev gamma ray equal to 100.
[b] See text for normalization procedure.
[c] This gamma ray has been used for the energy calibration in addition to the well-known gamma rays from Na^{22}, Ce^{144}, ThC″, and Po − Be sources.

The delayed (0.5μsec) coincidence gamma-ray spectrum thus determined was corrected for the chance background. An analysis of the resulting spectrum indicated the presence of four gamma rays of energy 3.0, 2.24, 1.75, and 0.51 Mev with relative intensities of 25, 100, 49, and 17, respectively.[12] The weak line at 0.51 Mev presumably represents quanta from the annihilation of positrons that feed either directly or indirectly the 0.69-Mev isomeric state. After correction for the fact that not all of the positrons were annihilated in the vicinity of the source, the intensity of positrons relative to 2.24-Mev gamma rays was calculated to be 18 to 100. The low-energy delayed gamma ray (0.115 Mev) found in the Ga^{72} decay[1] is also expected to be present in As^{72}, but at a lower relative intensity. The large chance background in the low-energy region in our As^{72} sources prevented its being detected however.

TABLE II. Energies and relative intensities from 0.84-Mev gamma-gamma coincidence spectrum.

Gamma-ray energy in Mev	Relative intensity[a]	Percent per disintegration[b]
0.63±0.01	100	7.9
0.73±0.02	40	3.2
0.90±0.02	18	~1.4
1.02±0.03	43	3.4
1.22±0.03	38	3.0
1.40±0.05	20	~1.6
1.90±0.03	13	~1.0
2.17±0.02[c]	66	5.0
2.30±0.05	19	~1.5
2.54±0.02	26	2.0

[a] These intensities have been normalized setting the 0.63-Mev gamma ray equal to 100.
[b] See text for normalization procedure. The errors on these values are approximately ±50% except in those cases where an approximation sign indicates a possible additional uncertainty.
[c] The shape of this line indicates that it is complex, consisting most likely of gamma rays of 2.08 and 2.20 Mev.

VI. THE DECAY SCHEME

The knowledge of the level structure of Ge^{72} from the study[1] of Ga^{72} is of considerable aid in interpreting the data from As^{72}. The gamma rays of 0.63, 0.84, 1.05, 1.25, 1.46, 1.68, 1.92 (1.88), 2.20, and 2.51 Mev (see Table I) were also found in the decay of Ga^{72}, and thus to a first approximation, can be immediately assigned. The lines in Table I at 1.37, 2.08, 2.32, 2.91, and 3.74 Mev can be accommodated by the addition of levels in Ge^{72} at 2.91 and 3.74 Mev. These levels, as well as the eleven known[1] levels of Ge^{72} are shown in Fig. 2. By the use of this decay scheme, the intensities of the gamma rays listed in Table I have been renormalized, in a manner to be described, to percent per disintegration, and are thus shown in Fig. 2.

The 1.68-Mev gamma ray can be accommodated in the 2.51–0.84 Mev transition, as well as in the 3.74–2.06 Mev transition. The intensity of this gamma ray has been shared in Fig. 2 between these two transitions using the experimental intensity ratio of the 1.05- to 1.68-Mev gamma rays found from the Ga^{72} work.

It is interesting to note that the ratio of intensities of the 1.92- (1.88-) Mev and the 2.51-Mev line in Table I (1.0 to 1.7) is approximately that of the 1.88-Mev to the 2.49-Mev lines (1.0 to 1.4) from the Ga^{72} work. This presumably indicates that the level at 3.32 Mev is populated in the decay of As^{72} to a much greater extent than the level at 3.34 Mev. This is in contrast to the Ga^{72} decay where the 3.34-Mev state is populated to a greater degree.

The gamma ray at 1.25 Mev can represent three different transitions. A fraction of its intensity is assigned to the 3.32–2.06 Mev transition on the basis of intensity ratios from Ga^{72}. The rest of the intensity is assigned to the 3.74–2.51 Mev transition, since one would expect the other alternative (2.06–0.84 Mev) transition to be weak.

The line at 1.46 Mev is considerably too intense to represent entirely the crossover transition from the 1.46-Mev state to ground state. Using again the intensity ratio from Ga^{72} (1.46- to 0.63-Mev lines) the remainder has been assigned to the 2.91–1.46 Mev transition. This assignment is in agreement with the coincidence measurements (Table II) where the 1.46-Mev gamma ray was observed fairly strongly in coincidence with the 0.84-Mev gamma ray.

Another ambiguity arises in the assignment of the 2.91-Mev gamma ray as it could represent the 2.91–0.00 Mev or the 3.74–0.84 Mev transitions. Because it was not observed in coincidence with the 0.84-Mev gamma ray, it has been assigned entirely to the former transition. The line at 2.17 Mev in Table II is presumably a composite of the lines at 2.08 and 2.20 Mev from intensity considerations and from the fact that it was

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[12] The errors on the relative intensities are approximately ±50%. The presence of a gamma ray of relative intensity less than 30 between 0.5 and 1.7 Mev is not excluded by our work. An additional 1.35-Mev gamma ray has been found [H. W. Kendall (private communication)].

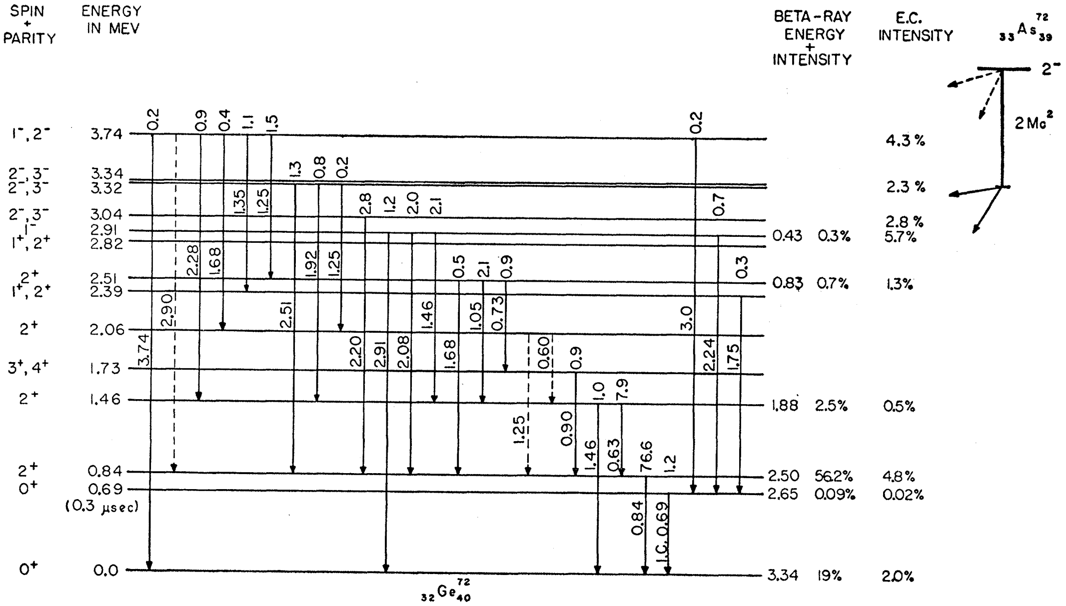

FIG. 2. Proposed level scheme of Ge^{72}. The intensities of the gamma rays are subject to the errors shown in Tables I and II. Assignments of gamma rays shown in dotted lines are uncertain. Branching ratios in the As^{72} decay have been calculated from the gamma-ray intensity balance alone and are subject to the errors indicated in Table III.

resolved into these two components in an earlier 0.84-Mev gamma-gamma coincidence measurement.

The weak and uncertain lines at 2.63 and 2.76 Mev in Table I remain unassigned. The lines at 0.73 and 0.90 Mev, observed in coincidence with the 0.84-Mev gamma ray, can be assigned to the 2.51–1.73 Mev, and 1.73–0.84 Mev transitions. It is evident that a gamma ray of 0.60 Mev should be present in the decay of As^{72} but has not been resolved in the present work because of its low intensity.

The three gamma rays at 1.75, 2.24, and 3.0 Mev, that were found in the delayed-coincidence spectrum are readily assigned as transitions from the 2.39-, 2.91-, and 3.74-Mev levels, respectively, to the isomeric state at 0.69 Mev. The sum of intensities of these three lines and the positrons in coincidence with the delayed electrons has been normalized (see Fig. 2) to equal 1.2 percent per disintegration. The value of 1.2 was obtained by taking an average of the determined[5,6] ratios of 0.69-Mev conversion electrons to total positrons and correcting this average for electron capture.

VII. DISCUSSION

In order to normalize the gamma-ray intensities to percent per disintegration, it was necessary to assume the ratio of the intensities of the positrons populating the ground and second excited states of Ge^{72} from the data of Mei *et al.*[5] Further, the ratio of electron capture to positron emission must be known for the two transitions. It has been shown by Mei *et al.*,[5] and will be discussed later, that both transitions can be classified as first forbidden. The K-capture-to-positron ratio for the transition to the 0.84-Mev level ($\Delta J=0$, first forbidden) has been estimated to be 1.6 times the allowed value of 0.052 (see Table III). This estimate is based on recent experimental results[13–15] and earlier theoretical ones.[16] The K-capture-to-positron ratio for the ground-state transition ($\Delta J=\pm 2$, unique first forbidden) has been similarly estimated to be 5 times the allowed value of 0.022. In order to include L-capture, the K-capture ratios for both transitions have been increased by 10%.[17]

If the 0.845-Mev gamma ray is represented by 100 units of intensity, the ground-state (electron-capture-plus-positron) transition as calculated by the above procedure is represented by 26.0 units of intensity. In addition, the 0.69-Mev conversion electrons would have

TABLE III. Branching ratios in As^{72} decay.

Level energy in Mev	Total E.C.+β^+ intensity[a]	Allowed K/β^+ ratio[b]	Calculated β^+ intensity[a]	log f^+t	log f_1t[c]	log $(f^++f^K)t$[b]
0.0	20±3	0.022	18	8.4	8.8	8.3
0.69	0.11±0.07[d]	0.048	~0.09	~10	~10	10
0.835	61±3	0.052	56	7.3		7.2
1.46	~3	0.138	~2.5	~8		8
1.73	<1	0.214	···			>8
2.06	<1	0.436	···			>7
2.39	<1	1.15	···			>7
2.51	~2	1.82	~0.7			~7
2.82	~0	9.55	···			··
2.91	6±2	18.2	0.3			~6
3.04	3±1	66	~0			~6
3.32	~2	∞	0			~6
3.34	~0	∞	0			··
3.74	4±2	∞	0			~5

[a] The intensities are given in percent of total disintegrations. The estimated errors are based on the decay scheme and the errors of the relative intensities of the gamma rays.
[b] Values of f^K/f^+ and f^++f^K have been taken from the graphs of E. Feenberg and G. Trigg [Revs. Modern Phys. **22**, 399 (1950)].
[c] See reference 18.
[d] See text for normalization procedure for this branch.

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1.4 units of intensity, and the other gamma rays feeding the ground state would have 2.1 units of intensity. This yields a total normalization factor of 1.305, which has been applied to the gamma-ray intensities listed in Table I, column 2. The result is shown in Table I, column 3, and the intensities are also shown in the decay scheme in Fig. 2. The normalization factor is quite insensitive to the K-capture-to-positron ratios which were assumed.

Table III, column 2, shows the feeding to the various levels of Ge^{72} by electron capture and positrons, as required by the measured gamma-ray intensities. In Table III, column 3, are shown the allowed K-capture-to-positron ratios. Column 4 gives the positron intensities calculated using the allowed electron-capture ratios for all transitions except those going to the ground, 0.69-, 0.84-, and 1.46-Mev levels, for which the electron-capture-positron ratios have been corrected as mentioned above, because these transitions are presumably first-forbidden. The sum of the positron intensities is $77.5 \pm 10\%$, which is in good agreement with the value $73.5 \pm 8\%$ derived from measurements of the ratio of intensities of annihilation radiation to 0.835-Mev gamma rays (see Sec. III). This assures the correctness of the main features of the As^{72} decay scheme as shown by Fig. 2.

In Table III, column 5, are listed the $\log f^+ t$ values for the positron branches whose intensities are shown in column 4. The nomographs of Moszkowski[18] have been used in obtaining the values listed. A unique first-forbidden shape factor has been observed[5] for the ground-state positron spectrum. This is quite consistent with the fact that the $\log f_1^+ t$ value (8.8) is close to those of other unique first-forbidden transitions, as summarized by Davidson.[19] A spin and parity of 2^- is thus required for As^{72}. Such as assignment can be accounted for by a shell-model configuration of $f_{5/2}$ for the (33rd) proton and $g_{9/2}$ for the (39th) neutron and by application of Nordheim's[20] "strong" rule. As^{74} and As^{76} have also a spin and parity of 2^- and apparently the same nucleon configuration.

The transition to the 0.69-Mev (0^+) level should also be unique first forbidden. The $\log f_1^+ t$ value (~ 10), however, is quite high for this type of transition. A possible explanation of this could be that the ground and first excited states of Ge^{72} differ by the excitation of a pair of equivalent neutrons or protons.[21] Thus if the ground-state positron transition is essentially a one-particle transition, the first excited state positron transition cannot be.

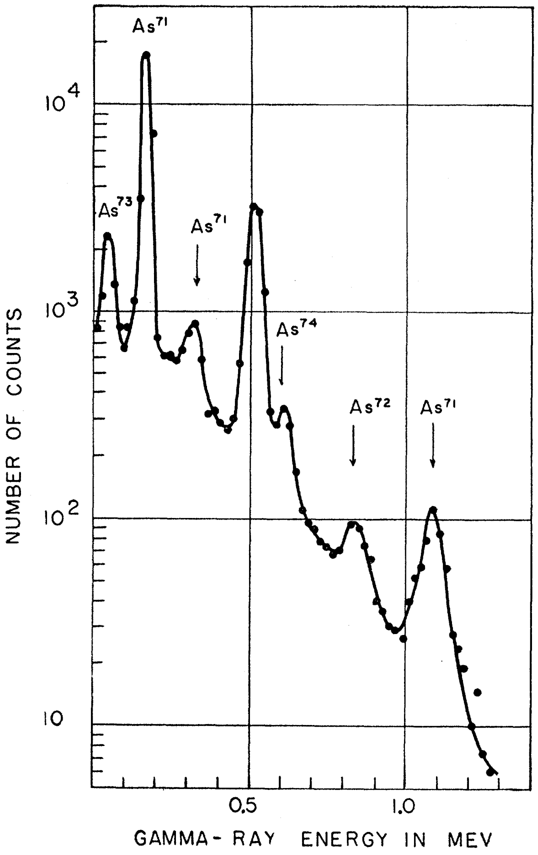

FIG. 3. Gamma-ray spectrum obtained two weeks after irradiation of the second source.

The positron transitions to the 0.835-, 1.46-, and 2.51-Mev levels have $\log ft$ values that permit the transitions to be classified most readily as first forbidden ($\Delta J = 0, \pm 1$). This is in agreement with the 2^+ assignments (see Fig. 2) from the study of Ga^{72}. The transitions to the levels at 1.73, 2.06, 2.39, and 2.82 Mev should also be first-forbidden in accordance with the spins and parities assigned in the Ga^{72} decay[1] and no contrary information is obtained from the As^{72} decay. These transitions should be weak, and the fact that they were not observed at all is probably not significant in view of the experimental uncertainties.

The four levels at 2.91, 3.04, 3.32, and 3.74 Mev are fed essentially by electron capture. The $\log[(f^+ + f^k)t]$ values listed in Table III, column 7, indicate that these transitions are most likely allowed. This is in agreement with the 2^- or 3^- assignment made for the 3.04- and 3.32-Mev levels from the Ga^{72} data. The new level at 2.91 Mev has been given a 1^- assignment in view of the allowed electron-capture transition and the fact that transitions associated with the level were not observed in the Ga^{72} decay. Similar reasoning can be applied to the other new level at 3.74 Mev, except that the small

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amount of energy available makes it appear unlikely that this level would be populated from Ga^{72} in any case. Either a 1^- or 2^- assignment seems appropriate for this level. The intensities of the gamma-ray transitions from the 2.91- and 3.74-Mev levels to the first excited state relative to the ground-state transitions are in good agreement with the relative intensities expected for single-particle transitions,[22] although presumably the transitions are not of this simple nature.

Our results seem to indicate only a weak feeding of the 3.34-Mev level in Ge^{72} from As^{72}. If this would imply a spin 4^- for this level, one would find disagreement with the observed 3.34-Mev ground-state transition in Ge^{72}.[1]

VIII. ACKNOWLEDGMENTS

We wish to thank Me. L. Piette for performing the chemical separations and Mr. H. Enderton for help in the analysis of the data.

APPENDIX A

In Fig. 3 is shown the collimated gamma-ray spectrum of our second source two weeks after the time of irradiation. The following known lines are in evidence: 0.053 Mev (As^{73}), 0.175 Mev (As^{71}), 0.59 and 0.63 Mev (As^{74}), and 0.835 Mev (As^{72}). In addition to these, there are lines at 0.310 and 1.10 Mev which have been assigned to As^{71} since they decay with the 60-hour half-life characteristic of As^{71}. The intensities of these two lines relative to the 0.175-Mev gamma ray are 5 and 9%, respectively.

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Spontaneous Fission Kinetics of Cf^{252}†

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A double ionization chamber is used to determine the spontaneous fission modes of Cf^{252} [$\tau_{\frac{1}{2}}$(fission) = 66 years]. The most probable energies of the light and heavy fragments are measured to be 107 and 79 Mev, respectively. The asymmetry of the process is found to agree with chemical mass-yield determinations. The results of the experiment are compared with the kinetics of other fission processes.

INTRODUCTION

THE kinetics of particle-induced fission have been studied in detail.[1–5] It has been theoretically predicted[6,7] and experimentally verified[5,8] that heavy, naturally occurring isotopes spontaneously fission with long half-lives. The low probability of the process makes experimental measurements exceedingly difficult. However, the availability of transuranic elements with short fission lifetimes provides an excellent opportunity for the study of the spontaneous process. These nuclei, existing in relatively high states of excitation, should exhibit the same fission characteristics as stable nuclei excited by relatively energetic particles. This is partially born out by the experimental mass-yield measurements and kinetic energy determinations[5,9,10] in Pu^{240} and Cm^{242}.

Among the transuranic elements, $_{98}Cf^{252}$ is ideally suited to an investigation of spontaneous fission. It has a fission half-life[11] of 66 years and a relatively long-lived alpha activity (2.2 years), making good measurements possible in a short period of time. A careful study of the alpha decay[12] has shown this isotope to follow the systematics of even-even alpha emitters. Glendenin and Steinberg[13] have determined chemically the spontaneous fission mass yields. They obtain a most probable mass ratio of ~1.34 along with evidence of typical mass-yield fine structure. The most probable number of neutrons released in the fission of Cf^{252} is 3.54–4.06.[14]

In the present experiment the total fragment energy in the spontaneous fission of Cf^{252} is measured. The relative probability of the various modes of fission is obtained and a comparison made with other fission processes.

SOURCE

The californium was taken from a sample that had been made by neutron irradiation of Pu^{239} in the materials testing reactor.[11] It was a radiochemically pure sample of californium consisting, by mass, of 50% Cf^{252}, 10% Cf^{251}, and 40% Cf^{250} so that essentially

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† This work performed under the auspices of the U. S. Atomic Energy Commission.

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