Decay of ${}_{50}$ Sn¹¹³ (112 days) and ${}_{49}$ In¹¹³^m (1.73 hr)*

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Samples of ${}_{50}$ Sn¹¹⁸(112 days) were produced by neutron irradiation of tin enriched in Sn¹¹². The gamma rays were studied by scintillation methods using the Argonne 256-channel analyzer; the internal-conversion electrons were investigated by means of a 180° β -ray spectrometer. In addition to the well-established 393-kev gamma ray, a gamma ray of 255 kev was found. Studies of all the tin radioisotopes showed the latter to be in the Sn¹¹³ activity. Energy levels in In¹¹³ at 393 and 648 kev, the latter having spin $\frac{1}{2}$ or $\frac{3}{2}$, are indicated.

INTRODUCTION

THE nuclide 50 Sn¹¹³ (112 days) decays to 49 In¹¹³ m by orbital-electron capture. The isomeric state of In¹¹³ lies at 393 kev and decays to the ground state by means of a gamma-ray or internal-conversion transition.

For some time we have observed a gamma ray of 255 kev in our tin samples. This radiation has also been observed by others and reported in the literature.¹ A number of independent investigations have been carried out concurrently. In particular, the work of Bhatki *et al.*² which appeared a few weeks prior to the preliminary report of this work³ and that of Girgis and Lieshout⁴ are both in excellent agreement with the experimental results reported here. While some latitude of interpretation still exists, the substantial agreement of all the recent work leaves little in doubt concerning the experimentally observable properties of this decay.

APPARATUS

The gamma-ray spectra were studied by means of scintillation methods. The detectors were $2\frac{1}{4}$ -inch cubic NaI(Tl) crystals coupled to Dumont Type 6292 photomultipliers. Pulse-height analysis was accomplished with the Argonne 256-channel analyzer. Coincidence measurements were carried out by adding a single-channel analyzer and a conventional "fast-slow" coincidence circuit to the 256-channel analyzer. The resolving time of this system is 0.04 microsecond.

The analyses of gamma-ray spectra are greatly facilitated by the use of a collimator which is interposed between the source and crystal. This collimator consists of a cylinder of lead, 3 inches in diameter and 4 inches long, through which a tapered hole is bored. The hole flares from $\frac{1}{2}$ -inch diameter at the source end to 1.0inch diameter at the crystal. The bore of the collimator is lined with a laminated filter to selectively suppress secondary x-rays. The liner comprises layers of tantalum (nearest the lead), tin, and steel. This detail improves the performance of the collimator in the energy region below 100 kev where the lead x-rays are normally troublesome. The collimator confines the incident beam to the central region of the crystal, reducing fringe effects as well as preventing scattered radiation from reaching the crystal; both effects result in a reduction of the height of the Compton distribution relative to the photopeak. The clear resolution of the 255-kev photopeak in Fig. 1(a) illustrates the quality of the results.

Measurements of the relative intensities of the internal-conversion-electron lines were made using a conventional 180° flat-field beta-ray spectrometer. The spectrometer was operated with a momentum spread of 1.5%.

EXPERIMENTS

Assignment of 255-kev Gamma Ray to Sn¹¹³

Several experiments were carried out in an effort to establish with certainty whether the 255-kev radiation was correctly assigned to the Sn¹¹³ decay. Samples of all the enriched stable isotopes of tin were irradiated in the Argonne reactor CP-5. After the shorter periods had died out, the intensity of the 255-kev gamma ray was compared to that of the 393-kev radiation (the latter being known to represent the Sn¹¹³ decay) for each of the samples. The 393-kev gamma ray was readily observable in all of the isotopes because of the relatively high capture cross section of Sn¹¹² and incomplete isotopic separation. The relative intensities were determined by measuring the areas under the photopeaks of the two gamma rays and correcting for the variable efficiency of the counter. The principal difficulty lies in the fact that the photopeak associated with the 255-kev radiation is extremely small compared to that of the 393-kev gamma ray and lies on the rapidly changing portion of the Compton distribution of the latter [Fig. 1(a)]. This fact makes it very difficult to ascertain what spectral background to subtract from the curve in order to determine the shape of the 255-kev photopeak. This difficulty was largely obviated by the following method.

^{*} Work performed under the auspices of the U. S. Atomic Energy Commission.

¹ Cork, Stoddard, Branyan, Childs, Martin, and LeBlanc, Phys. Rev. 84, 596 (1951); Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953); Y. Deshamps and P. Avignon, Compt. rend. 236, 478 (1953); P. Avignon, Ann. phys. 1, 10 (1956); G. Gardner and J. I. Hopkins, Phys. Rev. 101, 999 (1956); Achor, Phillips, Hopkins, and Haynes, Bull. Am. Phys. Soc. 2, 259 (1957).

² Bhatki, Gupta, Jha, and Madan, Nuovo cimento 6, 1461 (1957).

³ Grench, Burson, and Schmid, Bull. Am. Phys. Soc. 3, 207 (1958).

⁴ R. K. Girgis and R. Van Lieshout, Physica 24, 672 (1958).



After the gamma-ray spectrum of the Sn¹¹³ source had been accumulated by the 256-channel analyzer and the data were recorded, the tin source was removed from the counting apparatus and replaced by a source of Au¹⁹⁸. The high voltage applied to the photomultiplier was lowered slightly so that the photopeak associated with the 411-kev gamma ray of the gold coincided exactly with the channel position in which the 393-kev peak of the tin had been observed. After this matching was accomplished the gold spectrum was accumulated and recorded. The 411-kev gold peak was then normalized to match the 393-kev tin peak and the resulting gold spectrum was subtracted, in the region of interest, from that of the tin [Fig. 1(a)]. It was thus possible to subtract an empirically determined spectral background from beneath the 255-kev gamma-ray peak. The only deviation from exactness lies in the approximation that the spectral shape of the 411-kev gamma ray is essentially the same as that for the 393-kev gamma ray. This procedure was repeated for each of the active tin isotopes. In every case, the area of the 255-kev peak was found to be approximately 3.6% of that of the 393-kev photopeak. After corrections, this indicates that the intensity of the 255-kev gamma ray is $2.7 \pm 0.2\%$ of that of the 393-kev gamma ray. The enrichment process would have drastically upset this relationship if the 255kev radiation were associated with any isotope other than Sn¹¹³.

Detailed studies of the Sn¹¹³ activity were carried out using sources prepared by neutron irradiation of metallic tin enriched to 58.9% Sn¹¹². These samples were subjected to chemical purification⁵ to remove slight traces of radioactive antimony present in the sources after the irradiation. After the relative-intensity measurements described above were made, the samples were again subjected to the same chemical procedure. The chemistry was highly specific for tin and in any case would have greatly upset the relative abundances of any impurities. After the second purification, counting was commenced approximately one hour after the separation and the 255-kev peak was seen to have an initial intensity comparable to that of the 393-kev peak. The latter was then observed to grow in with the 1.7-hour period characteristic of the In^{113 m} activity. After equilibrium had been reached, the intensities of the two radiations were found to be in the same ratio as before the chemical separation.

Decay of the 255-kev peak was followed for about 7 months and found to exhibit a half-life of about 107 days (equal to that of the Sn^{113} within experimental error). These experiments lead to two clear-cut conclusions: (1) The 255-kev gamma ray is associated with the Sn^{113} isotope, and (2) it is associated with the parent Sn^{113} activity rather than with the 1.7-hour In^{113} isomer.

Coincidence Experiments

Coincidence experiments were conducted in an effort to establish the position of the 255-kev radiation in the decay scheme. The 256-channel analyzer was adjusted to observe the spectrum from zero to about 600 kev.

⁵ Courtesy of E. P. Steinberg and K. F. Flynn, Argonne National Laboratory, Chemistry Division, Lemont, Illinois.



FIG. 2. Internal-conversion-electron spectrum of Sn¹¹³.

The single-channel analyzer was set on the intense 24kev x-ray peak. In the coincidence spectrum so obtained [Fig. 1(b)], the 255-kev photopeak is seen to stand out strongly; the 393-kev peak is unobservable.

With the apparatus adjusted as described in the previous paragraph, a series of critical absorbers were interposed between the source and the crystal detecting the x-ray. It was possible to conclude unambiguously from these experiments that the radiations in coincidence with the 255-kev gamma ray are the characteristic Kx-rays of indium. It is thus concluded that there is a weak K-electron-capture branch from Sn^{113} to In^{113} which is followed promptly by a gamma ray of 255 kev. The mean life of the state in indium must be less than 4×10^{-8} second, the resolving time of the coincidence circuit.

Clearly, an excited state, other than the one at 393 kev, must exist in In¹¹³ and lie either 255 or 648 kev above the ground state. A search was made for possible 648-kev radiation, but none could be detected. An upper limit for the intensity of such possible radiation can be estimated to be 10^{-3} of that of the 393-kev gamma ray. Coincidences between the 255- and the 393-kev gamma rays were sought. None were observed but, if the 255kev radiation originated from a state of 648 kev, no such coincidences would be expected because of the long lifetime of the intermediate state.

Postulating a state at 255 kev above the ground state, the possibility of a 138-kev transition between the two excited states was examined. No such gamma ray could be directly observed, but no attempt is made to place an upper limit on its possible intensity since the source was still exhibiting residual Sn¹¹⁷ activity, which is characterized by 159-kev radiation. Therefore, none of these experiments provides any positive evidence with regard to the placement of the 255-kev transition in the level scheme of In¹¹³.

The relative intensities of the internal-conversionelectron groups (Fig. 2) were measured with the 180° beta-ray spectrometer. The K/(L+M) ratio was found to be 4.3 ± 0.1 for the 393-kev gamma ray. This value is in agreement with previously reported measurements. The relative intensity of the internal-conversion-elec-



FIG. 3. The decay 50Sn113(112

trons from the 255-kev transition is extremely low; that of the K-conversion group being 0.0024 ± 0.0002 that of the K-line of the 393-kev transition. A value of 8_{-2}^{+3} is determined for the K/(L+M) ratio for the 255-kev transition. From the scintillation measurements, the intensity of the 255-kev gamma ray was found to be $2.7 \pm 0.2\%$ that of the 393-kev radiation. Assuming the 393-kev transition to be an M4 type and using a theoretical K-conversion coefficient⁶ of 0.44 for this transition, the intensity of the branching into the 255-kev transition is calculated to be $1.8 \pm 0.2\%$ and the Kconversion coefficient of the 255-kev transition to be 0.039 ± 0.003 . (The K/(L+M) ratio and α_K for the 255-kev transition are not considered to be in conflict with the less precise values reported earlier.³) When the value of $\alpha_{\mathcal{K}}$ is considered together with the nature of the electron-capture branch to the 648-kev level (to be discussed below), one is led to the conclusion that the 255-kev transition is most likely M1 or (M1+E2).

INTERPRETATION

The proposed decay scheme (Fig. 3) is deduced as follows: The ground state of In¹¹³ has been measured to have a spin of $9/2^7$ and, according to single-particle shell theory, this would be $g_{9/2}$. Childs and Goodman have measured the spin of the 393-kev isomeric state to be $\frac{1}{2}$.⁸ This is expected to be a $p_{\frac{1}{2}}$ state. It is clear that the state from which the 255-kev transition takes place cannot lie below 393 key when the spins of the ground state and the 393-kev level are considered together with the transition probabilities. However, a state at 648 kev with negative parity and spin of $\frac{1}{2}$ or $\frac{3}{2}$ complies with all the conditions which the experimentally observed phenomena indicate to be necessary. Transitions from such a state to the ground state would be highly forbidden, whereas decay to the 393-kev level would be probable. From systematics of nuclei, one would expect the ground state of the parent Sn¹¹³ to be an $s_{\frac{1}{2}}$ state. A total decay energy lying between 0.7 and 1.0 Mev may be inferred

⁶ M. E. Rose, Internal Conversion Coefficients (North Holland Publishing Company, Amsterdam, 1958). 7 R. F. Bacher and D. H. Tomboulian, Phys. Rev. 52, 836

^{(1937).}

⁸ W. J. Childs and L. S. Goodman, Bull. Am. Phys. Soc. Ser. II, 1, 342 (1956).

from the tabulated data for nuclei in this region.⁹ Within the upper and lower limits imposed by this assumption, the log ft value for the capture branch to the 393-kev level would lie between 6.5 and 7.1. Thus this branch is

⁹ K. Way and M. Wood, Phys. Rev. 94, 119 (1953).

PHYSICAL REVIEW

VOLUME 115, NUMBER 1

JULY 1, 1959

Isomeric State of Platinum-199*

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An isomeric state of platinum-199 has been produced by thermal-neutron irradiation of normal and enriched platinum samples. The isomer decays with a half-life of 14.1 ± 0.3 seconds by the emission of γ rays of 32 ± 2 and 393 ± 2 kev energy. The thermal-neutron activation cross section of Pt¹⁹⁸ for the formation of the isomer is 0.028 ± 0.003 barn. Tentative level assignments are made, consistent with systematics and shell theory.

I. INTRODUCTION

'HE existence of an \sim 1-minute isomeric transition in Pt¹⁹⁹ of \sim 500-kev energy was predicted by Åström¹ from the systematics of the known transitions of the odd-mass-number isotopes of platinum, mercury, and lead. He was not able to detect the activity by the (d, p) reaction on platinum because of the masking effect of the annihilation radiation of the 72-second O¹⁴ obtained from nitrogen present in the metal. We have been able to produce the isomer by thermal-neutron irradiation of natural and of enriched platinum samples.

II. EXPERIMENTAL

Short irradiations of 2- to 10-second duration were performed, using the pneumatic tube facilities² of the Ford Nuclear Reactor at the University of Michigan, at a thermal neutron flux of $1.4 \times 10^{12} n \text{ cm}^{-2} \text{ sec}^{-1}$. The activity produced was studied on a special 100-channel pulse-height analyzer² designed for work with shortlived radioisotopes. This analyzer utilizes a rapid printer and a dual memory which permits a continuous spectral record to be made. A 3 in. \times 3 in. NaI(Tl) γ crystal in a 40 in. \times 40 in. \times 40 in. lead-shielded cave, along with a $1\frac{1}{2}$ in. $\times 1$ in. NaI(Tl) γ crystal and a hollow β -scintillation crystal³ mounted at the pneumatic tube terminal, were used as detectors for the spectrometer.

III. RESULTS

probably first-forbidden. The log ft value for the 1.8%

branch to the 648-kev level would lie between 6.5 and

8.2 for the same energy limits. Therefore, the transition

to the 648-kev state is probably also first-forbidden, as is consistent with the indicated character of this level.

Figure 1 shows a typical series of γ spectra taken at 24-second intervals after a 6-second irradiation of enriched platinum. Measurement was started 20 seconds after the end of irradiation. The short-lived component, assigned to Pt^{199m}, can be resolved from the data and is shown in Fig. 2. The data below 50 kev have been normalized to the γ peak at ~70 kev and were obtained from spectra taken with the $1\frac{1}{2}$ in. $\times 1\frac{3}{4}$ in. NaI(Tl) crystal mounted at the pneumatic tube terminal by scanning the low-energy spectra at higher gain.



FIG. 1. Gamma-ray spectra showing decay of 14-second Pt^{199m} and background due to 31-minute Pt¹⁹⁹. Spectra taken at intervals of 24 seconds.

^{*} This work was supported in part by the Michigan Memorial Phoenix Project and the U. S. Atomic Energy Commission. ¹ B. Aström, quoted by I. Bergström and G. Andersson, Arkiv Fysik 12, 415 (1958). ² W. W. Meinke, Nucleonics (to be published).

³ D. G. Gardner and W. W. Meinke, Intern. J. Appl. Radiation and Isotopes 3, 232-239 (1958).