$M3$ Isomeric Transition in Tin-113†

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The isomeric state of Sn^{113} has been produced by thermal neutron irradiation of tin enriched in Sn^{112}. Its half-life is found to be (20±1) min. The scintillation spectrum shows K x rays and a single gamma ray of 79±3 kev. The K-conversion coefficient ($\epsilon_K=95\pm15$) and the half-life show that this isomeric transition is $M3$, and not $M4$, as has been reported. A (9±2)% electron capture branch from the isomeric state was found, and an upper limit of 10^{-4} was placed on the ratio of positron emission to electron capture. The latter value limits the decay energy to the ground state of In^{113} to $\leq$1.15 Mev. Spins and parities of 7/2+ for Sn^{113m} (20 min) and 1/2+ for Sn^{113g} (118 day) are assigned. The isomeric ratio $\sigma(Sn^{113m})/\sigma(Sn^{113g})$ was found to be (0.41±0.06), and a value of (0.35±0.08) barn was obtained for the production cross section of the isomer.

INTRODUCTION

An isomer in Sn^{113} with a half-life of (27±3) min has been reported by Selinov and Chikhladze.[1] Conversion lines were observed in a beta-ray spectrometer following positron decay of Sb^{113}; a chemical separation confirmed the assignment to tin. The energy of the isomeric transition was found to be 79.3 kev and its K/L ratio 1.75.

Previous to the work of Selinov and Chikhladze, Naidu[2] had reported an (18±2)-min activity in tin exposed to slow neutrons; a Geiger counter with tin walls was used as source and detector. Nelson *et al.*[3] had reported a (30±5)-min activity produced by the exposure of tin enriched in Sn^{112} to slow neutrons. Also, Petroff[4] had reported a (19±1)-min activity emitting gamma rays of 78 kev produced by a Cd(α,n) or Cd(α,2n) reaction and had assigned it to a tin isotope of mass number greater than 112.

By analogy with other isomers in this region, and from their internal conversion results, Selinov and Chikhladze concluded that the multipolarity of the isomeric transition was $M4$.[4a] However, the half-life does not fit the well-established systematics of $M4$ transitions (theoretical single-particle $M4$ half-life, 5×10^7 sec; theoretical single-particle $M3$ half-life, 40 sec). The reported K/L ratio also fits better an $M3$ rather than the $M4$ assignment, as the theoretical ratios are 0.95 for $M4$ and 2.2 for $M3$.[5] Because of this inconsistency in the multipolarity assignment, and because of recent interest in the tin isotopes as a testing ground for the pairing-correlation nuclear model,[6] we have reinvestigated this isomer.

The ground state of Sn^{113} has a half-life of 118 days. It decays by electron capture, with about 98% of its decays leading to the 1/2− isomeric state in In^{113}, which has a half-life of 1.7 hr and an energy of 393 kev above the 9/2+ ground state of In^{113}. It has been established by the work of Bhatki *et al.*,[7] Girgis and Van Lieshout,[8] Burson *et al.*,[9] and Phillips and Hopkins,[10] that about 2% of the decays of Sn^{113g} take place to a state in In^{113} at 648 kev. The only de-excitation of this state observed by the first three groups was a 255-kev gamma ray; the absence of a crossover led these three groups to assign the spin of the 648-kev state as 1/2 or 3/2. The logft value for the transition from Sn^{113g} to the 648-kev state in In^{113}, when computed for any reasonable decay energy, fitted best a nonunique first-forbidden transition, and since the ground state of Sn^{113}, like all the low-spin near-ground states of the odd tin isotopes, most probably has even parity, the 648-kev state in In^{113} probably has odd parity. Phillips and Hopkins, in contradiction to the other three groups, seemed to observe a 648-kev crossover transition whose intensity was (6.0±0.7)% of that of the 255-kev transition; they concluded that the 648-kev state was 5/2−. In the recent work of Greenwood and Brannen,[11] however, an upper limit of 0.1% of the 255-kev transition is put on the intensity of the possible 648-kev crossover, confirming the conclusion of the first three groups.[7–9] Whereas Bhatki *et al.*[7] had previously reported a decay energy of 36 kev for the decay of Sn^{113g} to the 648-kev state in In^{113}, Greenwood and Brannen found a lower limit of 150 kev for this energy.

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[1] I. P. Selinov and U. L. Chikhladze, J. Exptl. Theoret. Phys. (U.S.S.R.) **38**, 1012 (1960) [translation: Soviet Phys.—JETP **11**, 728 (1960)].

[2] R. Naidu, Nature **137**, 578 (1936).

[3] C. M. Nelson, B. H. Ketelle, and G. E. Boyd, Oak Ridge National Laboratory Report ORNL-828, 1950 (unpublished).

[4] M. Petroff, University of California Radiation Laboratory Report UCRL-3538, 1956 (unpublished).

[4a] *Note added in proof.* In a more recent report on their work [I. P. Selinov *et al.*, Izvest. Akad. Nauk S.S.S.R., Ser. Fiz. **25**, 848 (1961)], these authors suggest an ($M3+E4$) or ($E3+M4$) assignment for the multipolarity, rather than the original $M4$ assignment.

[5] The theoretical internal conversion coefficients quoted in the paper are taken from M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

[6] L. S. Kisslinger and R. A. Sorensen, Kgl. Danske, Videnskab. Selskab, Mat.-fys. Medd. **32**, No. 9 (1960); R. A. Sorensen Nuclear Phys. **25**, 674 (1961), and private communication.

[7] K. S. Bhatki, R. K. Gupta, S. Jha, and B. K. Madan, Nuovo cimento **6**, 1461 (1957).

[8] R. K. Girgis and R. Van Lieshout, Physica **24**, 672 (1958).

[9] S. B. Burson, H. A. Grench, and L. C. Schmid, Phys. Rev. **115**, 188 (1959).

[10] W. E. Phillips and J. I. Hopkins, Phys. Rev. **119**, 1315 (1960).

[11] R. C. Greenwood and E. Brannen, Phys. Rev. **122**, 1849 (1961).

EXPERIMENTAL WORK AND CONCLUSIONS

Samples of tin enriched to 72% in Sn^{112} were irradiated in the Brookhaven reactor for periods of several minutes. The sources were investigated with a sodium iodide crystal spectrometer coupled to a 256-channel analyzer. Two prominent peaks were observed, corresponding to energies of (79±3) kev and (26±1) kev, both decaying with a (20±1)-min half-life. (The K x-ray energy in tin is 25.8 kev.) As this activity was much less prominent after irradiation of tin enriched in other isotopes,[12] the assignment of the 20-min activity to an isomer in Sn^{113} is confirmed. A sample spectrum is shown in Fig. 1.

Assuming that all the K x rays observed are due to internal conversion, an absolute K-conversion coefficient of about 100 follows from the x/γ intensity ratio, which fits $M3$ rather than $M4$ (theoretical K-conversion coefficients; $\beta_3=94$, $\beta_4=720$). If there is any K-electron capture branch from the isomeric state, the K-conversion coefficient will be reduced and therefore be still further from the $M4$ conversion coefficient.

To check the possibility of K capture from Sn^{113m} to In^{113}, two experiments were performed.

(1) A Sn^{113m} source in fine powder form was placed between two thin plastic scintillators, so that the conversion electrons were counted in a 4π geometry (see Fig. 2). The gamma rays and K x rays were detected in a thin sodium iodide crystal protected by a beryllium absorber. The photon spectrum in anticoincidence with the electrons was displayed on a 256-channel analyzer. For calibration a Sn^{117m} source was used. In Sn^{117m} all the x rays should be in coincidence with conversion electrons; since the number of

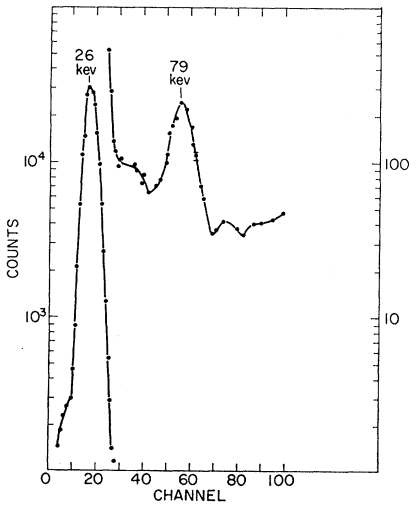

FIG. 1. NaI scintillation pulse-height spectrum of Sn^{113m}, showing K x rays and the 79-kev gamma ray. Both peaks showed a pure 20-min decay for many half-lives. The scale on the left refers to the 26-kev peak, and the scale on the right to the 79-kev peak.

[12] E. der Mateosian and M. Sehgal (private communication).

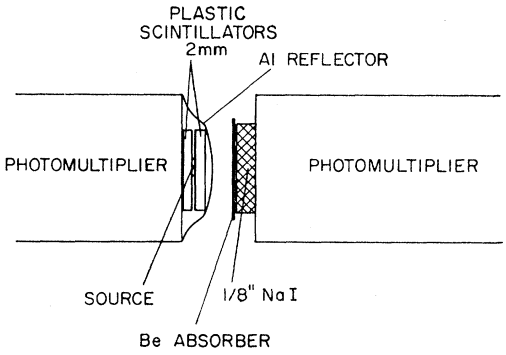

FIG. 2. Anticoincidence arrangement used in searching for electron capture from Sn^{113m} (20 min).

K x rays in anticoincidence with electrons was 5% of the singles, presumably 5% of the electrons were not counted, either due to absorption in the source or due to the electronic bias necessary to eliminate noise. In Sn^{113m} (25±1%) of the K x rays were in anticoincidence with electrons. As the energy of the conversion electrons is on the average 60 kev in Sn^{113m} as against 140 kev in Sn^{117m}, and since the same counting arrangement was used and the source thicknesses were similar, more electron losses are expected in Sn^{113m}. Therefore, we arrive at an upper limit of 20% for the relative number of K x rays not in coincidence with K-conversion electrons, giving, after correction for higher-shell conversion of the isomeric transition, an upper limit of 14% for K-electron capture.

(2) The ratio of In to Sn K x rays, and thus the ratio of K capture to K internal conversion, was then determined by a critical absorption measurement. Absorber foils of Rh and Pd were calibrated by means of sources emitting pure In K x rays [Sn^{113} (118 day)] and pure Sn K x rays [Sn^{117m} (14 day)]. It was found that (12±3)% of the Sn^{113m} K x rays were In K x rays and the remaining 88% were Sn K x rays.

As no 255-kev gamma ray is seen immediately after irradiation (see Fig. 3), we conclude that the electron capture occurs predominantly to the 9/2+ ground state of In^{113}. Since the decay energy to that state is larger than 0.88 Mev, the L/K capture ratio is approximately 0.1;[13] from this we conclude, after correcting for conversion of the isomeric transition in the higher shells, that the total electron capture branch is (8.9 ±2.3)%.

As mentioned above, no other gamma ray of 20-min half-life was detected in the spectrum of Sn^{113m}. A transition of less than 30 kev might have escaped detection in the singles spectrum due to high internal conversion or by being masked by the x-ray peak or by noise. However, in the coincidence experiments the 79-kev gamma ray was found to be not in coincidence

[13] Orbital electron capture ratios are taken from *Nuclear Spectroscopy Tables*, edited by A. H. Wapstra *et al.* (North-Holland Publishing Company, Amsterdam, 1959).

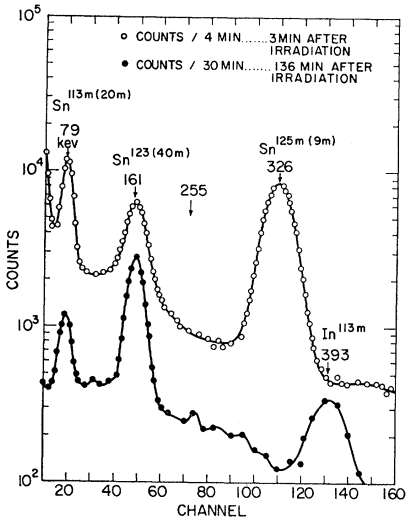

FIG. 3. Scintillation spectra of tin enriched in Sn^{112} irradiated with thermal neutrons. From these and other similar spectra the activation cross section of Sn^{113m} relative to that for other tin activities was deduced, and an upper limit was placed on the intensity of a 255-kev gamma ray in the decay of Sn^{113m}.

(less than 5%) with any electrons. Therefore, an upper limit of about 10 kev can be placed on the energy of any gamma ray in cascade with the 79-kev $M3$ transition, unless its half-life is longer than 10^{-7} sec.

A search for annihilation radiation coincidences led to an upper limit of 10^{-5} for positron emission per isomeric transition. Hence we obtain an upper limit for the ratio $\beta^+/\text{E.C.} \leq 10^{-4}$. This leads to an upper limit for the total decay energy, $Q(\text{Sn}^{113m} \rightarrow \text{In}^{113}) \leq 1.15$ Mev. The lower limit of Greenwood and Brannen[11] for the decay energy from Sn^{113g} to In^{113} (648-kev state) implies a lower limit of 0.88 Mev for the decay energy from Sn^{113m} to In^{113g}. The logft value for electron capture of 1.15 Mev is 4.6 ± 0.1; for electron capture of 0.88 Mev it is 4.4 ± 0.1.

After correcting for the measured electron capture branch and also for the Auger effect, we obtain from the x/γ ratio a K-conversion coefficient $\epsilon_K = 95 \pm 15$ for the isomeric transition. This is to be compared to the theoretical value $\beta_3 = 94$.

From the measured electron-capture branch and from the intensity ratio of the 79-kev and 393-kev gamma rays we can estimate the ratio of the cross sections for the production of Sn^{113m} and Sn^{113g}. We find

$$\sigma_m = (0.41 \pm 0.06)\sigma_g.$$

The cross section for the activation of Sn^{113g} has been reported as (1.3 ± 0.3) b.[3,14] We conclude that this value represents

$$\sigma_g + 0.91\sigma_m,$$

and then we find

$$\sigma_g = 0.9 \pm 0.3 \text{ b}, \quad \sigma_m = 0.39 \pm 0.10 \text{ b}.$$

An independent check on the isomeric cross section has been made by comparing it with the activation of the 160-kev gamma ray in the decay of 40-min Sn^{123m}. We find

$$\sigma_m = \sigma_{113m} = (2.0 \pm 0.2)\sigma_{123m}.$$

Since the tabulated value of σ_{123m} is (0.16 ± 0.04) b,[14] we find

$$\sigma_m = 0.32 \pm 0.08 \text{ b},$$

in agreement with our other measurements. Combining the two values, we arrive at a cross section of (0.35 ± 0.08) b. The results are in agreement with the isomeric ratio rule,[15] which states that the ratio of cross sections for the production of two isomeric states by slow neutron capture is such that the isomeric state with spin closest to that of the compound nucleus is favored.

The partial half-life of the isomeric transition, after correction for the electron-capture branch, is 22 min, and the radiative part alone has a partial half-life of 2.0×10^5 sec. This implies a hindrance of 33 compared to the Weisskopf single-particle estimate with a nuclear radius constant of 1.2×10^{-13} cm. The transition is l forbidden by one unit. Little is known about the speed of l-forbidden $M3$ transitions. The transition in Sn^{113} may, however, be compared with the $M3$ transition in In^{112}, which is probably best interpreted as the transition

$$[(g_{9/2}^{-1})_p(s_{1/2})_n]_{4+} \rightarrow [(g_{9/2}^{-1})_p(g_{7/2})_n]_{1+},$$

and thus corresponds to the same odd-neutron transi-

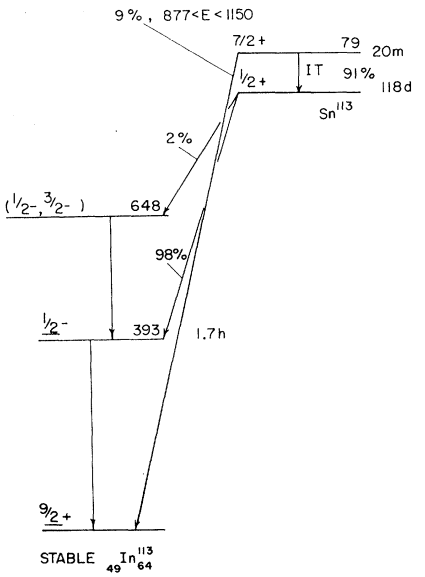

FIG. 4. Proposed decay scheme of Sn^{113m}.

[14] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed.

[15] E. der Mateosian and M. Goldhaber, Phys. Rev. **108**, 766 (1957).

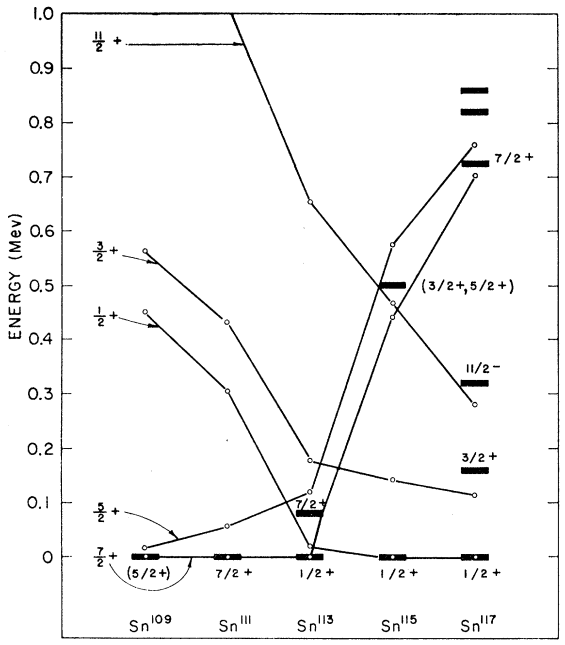

FIG. 5. Experimental levels in odd-mass tin isotopes near Sn^{113} compared with results of pairing-correlation model calculations by Kisslinger and Sorensen and by Sorensen.[6] The experimental levels are shown as heavy bars and the theoretical level positions are shown as open circles, with a solid line connecting levels of the same spin. The theoretical energies include terms due to coupling to electric quadrupole vibrational modes. The figure was drawn from numbers supplied in a private communication from R. A. Sorensen. It is seen that in Sn^{113} the relative positions of the 7/2+ and 1/2+ levels are found to be reversed from the order predicted by the calculation. However, the discrepancy is only 0.1 Mev, which is about the accuracy expected from the calculation.

tion. The In^{112} transition is hindered 5.3 ± 0.5 times more than the Sn^{113} transition.

LEVEL SCHEME

Although the ground-state spin of Sn^{113} has not been definitely measured, the 118-day decay is consistent only with a spin of 1/2 or 3/2. The low-lying (shell model, or one-quasi-particle) states in the odd tin isotopes are 1/2+, 3/2+, 5/2+, 7/2+, and 11/2−. Our results, indicating an $M3$ transition directly to the ground state, are consistent only with a 7/2+ isomeric level, decaying partly by isomeric transition to what then must be assigned as the 1/2+ ground state of Sn^{113}, and partly by electron capture to the 9/2+ ground state of In^{113} (see Fig. 4).

From the absence of the 255-kev gamma ray in our experiment a lower limit of one year is obtained for the partial half-life for electron capture from Sn^{113m} to the 648-kev level in In^{113}. This corresponds to a lower limit of 7.3 for the logft value if the decay energy is 0.50 Mev and a lower limit of 6.6 if the decay energy is 0.23 Mev. These limits are consistent with the assignments (1/2−, 3/2−, 5/2−) for the 648-kev level. However, the fact that the internal conversion coefficient of the 255-kev transition has been measured by Burson *et al.*,[9] and is consistent only with an $M1$ or mixed $M1+E2$ multipolarity, seems to limit the allowed spins for the 648-kev level to 1/2 and 3/2.

Recent work by Kisslinger and Sorensen[6] uses the pair-correlation model to predict level positions and wave functions for the tin (and other single-closed-shell) nuclei. The model has a number of successes and, with the parameters the authors select, seems to predict the positions of the low-lying levels in the odd-mass tin isotopes to within about 0.1 Mev. For Sn^{113} it predicts a ground state of 7/2+ and a first excited state at 0.02 Mev of 1/2+. We find that the ground state is 1/2+ and that the 7/2+ state is 0.08 Mev above it. Figure 5 compares the predictions of the model with experimentally known levels in odd-mass tin nuclei in the neighborhood of Sn^{113}.

ACKNOWLEDGMENTS

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