

Half-life of the 6.3-keV isomer in  $^{121}\text{Sn}$ G. A. Rech,<sup>1</sup> E. Browne,<sup>1</sup> I. D. Goldman,<sup>2</sup> F. J. Schima,<sup>3</sup> and E. B. Norman<sup>1</sup><sup>1</sup>*Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720*<sup>2</sup>*Instituto de Fisica, Universidade de Sao Paulo, Sao Paulo, Brazil*<sup>3</sup>*National Institute of Standards and Technology, Gaithersburg, Maryland 20899*

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We present a measurement of the half-life of  $^{121}\text{Sn}^m$ , obtaining a value of  $43.9 \pm 0.5$  yr. In the same experiment, we measured a half-life of  $21.8 \pm 0.3$  yr for  $^{210}\text{Pb}$  to verify the reliable performance of our data acquisition system and analysis methods. Our experimental  $^{121}\text{Sn}^m$  half-life is consistent with 49 yr, a value predicted on the basis of the systematic trend of the M4 isomeric  $\gamma$ -ray transition in  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$ .

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The ground and first two excited states in  $^{117}\text{Sn}$ ,  $^{119}\text{Sn}$ , and  $^{121}\text{Sn}$  originate from the  $s_{1/2}$ ,  $d_{3/2}$ , and  $h_{11/2}$  shell-model configurations. Consequently, these nuclides each have a long-lived isomer that predominately decays by an M4 isomeric  $\gamma$ -ray transition. The  $h_{11/2}$  configuration assignment may be tested with this transition provided that reliable values of the isomer half-life were available for all three isotopes of tin. However, this is not the case for  $^{121}\text{Sn}^m$ , where the following discrepant values were reported in the literature:  $76.3 \pm 6.6$  [1],  $55 \pm 5$  [2],  $40 \pm 10$  [3], and 25 yr [4]. The first was determined by counting a thick sample with a gas-flow proportional counter. The next two are private communications quoted in an evaluation paper by Tamura [5]. The fourth value, which is significantly lower than the other two, is a preliminary result [4] measured with a NaI  $\gamma$ -ray detector. We therefore decided to measure the half-life of  $^{121}\text{Sn}^m$  and have obtained a value of  $43.9 \pm 0.5$  yr. In the same experiment, we measured a half-life of  $21.8 \pm 0.3$  yr for  $^{210}\text{Pb}$ . This latter result agrees well with the recommended value of  $22.3 \pm 0.2$  yr [6] and thus confirms the good performance of our equipment and adequate analysis of our data. Our result of  $43.9 \pm 0.5$  yr for the half-life of  $^{121}\text{Sn}^m$  may be compared with 49 yr, an estimate that we have deduced from the reduced transition probabilities of the M4 isomeric transition in  $^{117}\text{Sn}^m$  and  $^{119}\text{Sn}^m$ . This agreement supports the  $11/2^-$  spin and parity assignments to  $^{121}\text{Sn}^m$  shown in the partial decay scheme of Fig. 1 [7].

We used the following radioactive sources.

A sealed  $^{121}\text{Sn}^m$  point source with an activity of about 1.5  $\mu\text{Ci}$  dried as stannous chloride onto a polystyrene tape. This source was produced by thermal neutron irradiation of an isotopically pure  $^{120}\text{Sn}$  target, and was supplied to us by the National Institute of Standards and Technology (NIST). Its preparation is given in Ref. [7].

A sealed  $^{241}\text{Am}$  source with an activity of 1  $\mu\text{Ci}$ , purchased from Amersham Corporation.

A sealed  $^{210}\text{Pb}$  source with an activity of 1  $\mu\text{Ci}$ , purchased from Isotope Product Laboratories, which contains the  $^{210}\text{Bi}$  (5.01d) and  $^{210}\text{Po}$  (138.4d) descendant nuclei in equilibrium.

All three sources were sealed between thin plastic layers to reduce the absorption of emitted low-energy ( $<60$  keV)  $\gamma$  rays.

The  $^{121}\text{Sn}^m$  and  $^{241}\text{Am}$  sources were rigidly mounted in a Lucite holder with a 0.25 mm thick foil of zirconium placed between them to attenuate low-energy x rays from the  $\alpha$  decay of  $^{241}\text{Am}$ , which would have interfered with the measurement. The  $^{210}\text{Pb}$  source was mounted into a separate Lucite holder. Either source assembly could then be snugly fitted into slots cut in a larger Lucite holder, and placed at fixed distances from the beryllium end cap of a dedicated  $36 \times 13$  mm thick planar high-purity Ge detector. We collected  $\gamma$ -ray spectra of 4096 channels from 0 to 170 keV in four-day intervals and recorded the data to magnetic disk using an ORTEC ACE data acquisition system on a PC. The detector had a full width at half maximum energy resolution of 0.57 keV at 59.5 keV.

We began the experiment using the  $^{121}\text{Sn}^m/^{241}\text{Am}$  assembly placed 10 cm away from the Ge detector cap. Figure 2(a) shows the relevant portion of the  $\gamma$ -ray spectrum of our first ( $^{121}\text{Sn}^m$  and  $^{241}\text{Am}$ ) run, which we used to determine the half-life of  $^{121}\text{Sn}^m$ . After about six months we placed the  $^{210}\text{Pb}$  source assembly in front of the  $^{121}\text{Sn}^m/^{241}\text{Am}$  source 7.5 cm away from the Ge detector to simultaneously measure the spectrum of both source assemblies ( $^{210}\text{Pb}$ ,  $^{121}\text{Sn}^m/^{241}\text{Am}$ ) for a month. Figure 2(b) shows a spectrum measured in four days for the same energy region displayed in Fig. 2(a). We then removed the  $^{210}\text{Pb}$  source assembly and again measured the  $\gamma$ -ray spectrum of just the  $^{121}\text{Sn}^m/^{241}\text{Am}$  source for a month. We repeated the latter two steps four times and collected spectral data of  $^{121}\text{Sn}^m$  and  $^{210}\text{Pb}$  for 1.2 and 0.7 yr, respectively.

We deduced the half-life of  $^{121}\text{Sn}^m$  by measuring the decrease in the 37.1 keV  $\gamma$ -ray count rate over a period of 1.2 yr. Since such a change is expected to be about 1.5%, good statistics (more than  $\approx 1 \times 10^6$  events in the peak) and a precise reckoning of the spectral areas are required as well as a stable response of the data acquisition system. In each four-day spectrum the 37.1 keV  $\gamma$ -ray peak contained about  $5 \times 10^6$  counts, and we estimated its background by averaging the areas (for the same number of channels) above and below this peak. Because of its long and well known half-life of  $432.2 \pm 0.7$  yr [8] we used the 59.5 keV peak from  $^{241}\text{Am}$   $\alpha$  decay to test the stability of the electronics and to correct the data for systematic errors that could have originated from changes in the Ge detector response. Thus, we fitted an ex-

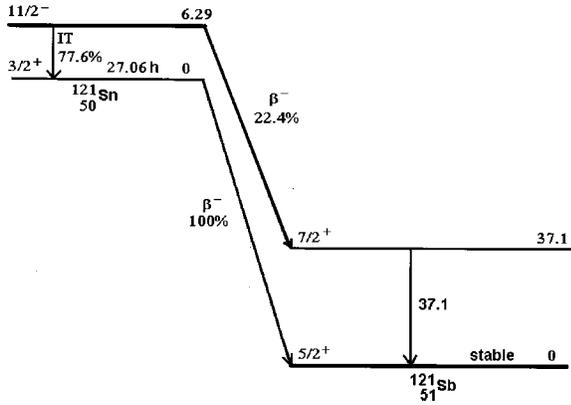


FIG. 1. Partial decay scheme of  $^{121}\text{Sn}^m$  [7]. Energies are in keV.

ponentially decreasing function of time to the ratio between the peak areas of the 37.1 keV ( $^{121}\text{Sn}^m$ ) and 59.5 keV ( $^{241}\text{Am}$ )  $\gamma$  rays [shown in Fig. 3(a)]. The mathematical procedure involved taking the logarithms of the ratios between the peak areas of the 37.1 and 59.5 keV  $\gamma$  rays and then performing a least-squares fit to a straight line (same procedure and computer program as those used in Ref. [9] to measure the half-life of  $^{44}\text{Ti}$ ). We obtained an effective decay constant  $\lambda_{\text{eff}}=0.014\ 19\ (19)\ \text{yr}^{-1}$ , and a  $\chi^2/\nu$  value of 1.04. Using  $\lambda(^{241}\text{Am})=0.001\ 603\ 8\ (26)\ \text{yr}^{-1}$ , the decay constant of  $^{121}\text{Sn}^m$  is given by

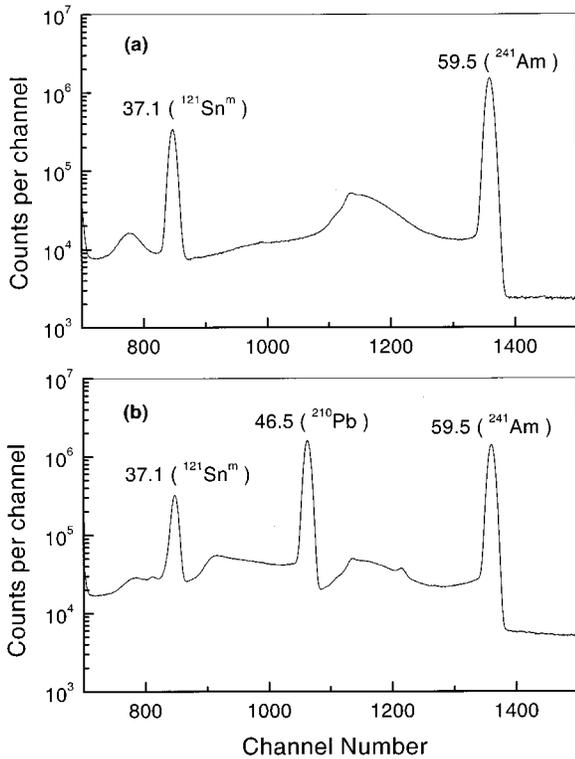


FIG. 2. Relevant portions of  $\gamma$ -ray spectra observed in four days from (a)  $^{121}\text{Sn}^m/^{241}\text{Am}$  source, and (b)  $^{210}\text{Pb}$  and  $^{121}\text{Sn}^m/^{241}\text{Am}$  sources. Energies are in keV.

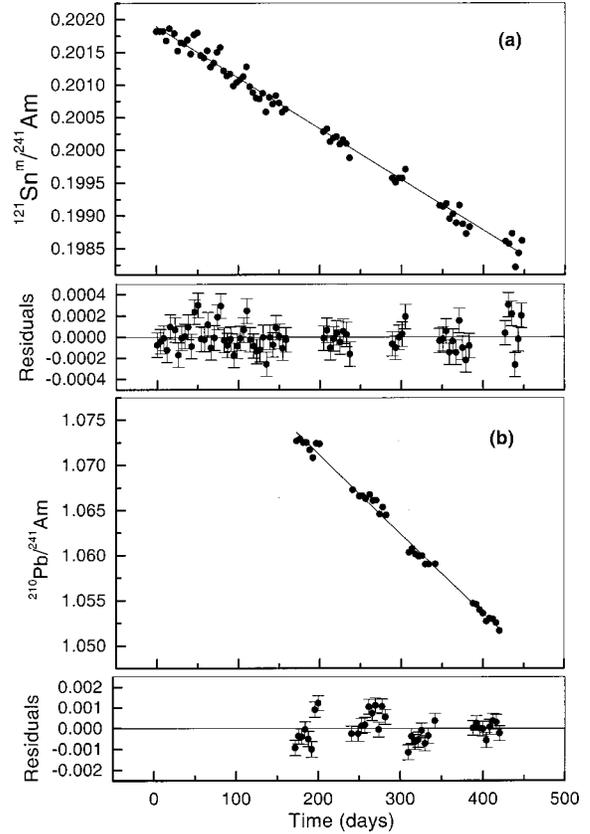


FIG. 3. Measured decay curve for (a)  $^{121}\text{Sn}^m/^{241}\text{Am}$ , and (b)  $^{210}\text{Pb}/^{241}\text{Am}$ . The differences between measured and fitted values are shown as residuals. The half-life values determined from these fits are (a)  $t_{1/2}(^{121}\text{Sn}^m)=43.9\pm 0.5\ \text{yr}$ , and (b)  $t_{1/2}(^{210}\text{Pb})=21.8\pm 0.3\ \text{yr}$ .

$$\begin{aligned} \lambda(^{121}\text{Sn}^m) &= \lambda_{\text{eff}} + \lambda(^{241}\text{Am}) \\ &= 0.014\ 19\ (19)\ \text{yr}^{-1} + 0.001\ 603\ 8\ (26)\ \text{yr}^{-1} \\ &= 0.015\ 79\ (19)\ \text{yr}^{-1}, \end{aligned}$$

and thus its corresponding half-life is 43.9 (5) yr. The number within parentheses represents the  $1\sigma$  uncertainty in the least significant digit. It is apparent that the statistical uncertainty of  $\approx 0.06\%$  in the spectral area ratios was quite adequate to detect the decrease in the  $^{121}\text{Sn}^m$  activity with time. We similarly determined the half-life of  $^{210}\text{Pb}$ . Figure 3(b) shows the decrease of the ratio between the spectral areas of the 46.5 keV  $\gamma$  ray, from  $^{210}\text{Pb}$   $\beta^-$  decay, and the 59.5 keV  $\gamma$  ray from  $^{241}\text{Am}$   $\alpha$  decay. The effective fitted decay constant is  $\lambda_{\text{eff}}=0.030\ 14\ (26)\ \text{yr}^{-1}$  (with  $\chi^2/\nu=2.97$ ), or 0.030 14 (45) for  $\chi^2/\nu=1.0$ . The decay constant of  $^{210}\text{Pb}$  is given by

$$\begin{aligned} \lambda(^{210}\text{Pb}) &= 0.030\ 14\ (45)\ \text{yr}^{-1} + 0.001\ 603\ 8\ (26)\ \text{yr}^{-1} \\ &= 0.031\ 74\ (45)\ \text{yr}^{-1}, \end{aligned}$$

and then its corresponding half-life is 21.8 (3) yr. The significantly larger value of 2.97 (compared to 1.04 in the  $^{121}\text{Sn}^m/^{241}\text{Am}$  case) for  $\chi^2/\nu$  may be due to repositioning the

TABLE I. Parameters used in Eq. (1) for  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$  and the respective values obtained for  $B_r$ .

Nuclide	$E_\gamma$ (MeV)	$\alpha_T$ (M4)	$B$ (IT)	$t_{1/2}(d)$	$B_r$ ( $\text{MeV}^{-9}\text{yr}^{-1}$ )
$^{117}\text{Sn}$ [11]	0.156 02 (3)	47.8 (24)	1.0	13.60 (4)	509 (25)
$^{119}\text{Sn}$ [12]	0.065 66 (1)	$5.16(26)\times 10^3$	1.0	293.1 (7)	521 (25)

$^{210}\text{Pb}$  source between the  $^{121}\text{Sn}^m/^{241}\text{Am}$  source and the Ge detector for each separate measurement. The geometry of the  $^{121}\text{Sn}^m/^{241}\text{Am}$  assembly, however, remained unchanged during the experiment, which explains the better value of 1.04 for  $\chi^2/\nu$ . Our experimental value for the half-life of  $^{210}\text{Pb}$  agrees very well with the recommended value of  $22.3 \pm 0.2$  yr [6], and thus confirms the quality and reliability of our result for the half-life of  $^{121}\text{Sn}^m$ .

From the four previously available half-life values for  $^{121}\text{Sn}^m$ , only that of Flynn, Glendening, and Steinberg [1] ( $76.3 \pm 6.6$  yr, average of two values) is accompanied by a description of the measurement, although certainly very limited. Their activity was chemically separated and a thick (20  $\text{mg}/\text{cm}^2$ ) source was  $\beta$  counted for seven years using a gas-flow  $\beta$  proportional counter. Since with this detector it was not feasible to select the radiations from  $^{121}\text{Sn}^m$ , the result of their measurement relies entirely on the chemical and isotopic purity of their source. Moreover, although the stability of their data acquisition system for seven years was tested by counting the  $\beta$  decay of a  $^{210}\text{Pb}$  (22.3 yr) sample, their result for the half-life of this radionuclide is not given in their paper, and thus it is difficult to judge the attained level of stability. Finally, it is somewhat surprising that in a measurement that lasted seven years they obtained a precision of only  $\approx 10\%$  in the half-life value. Such a result suggests that very few measurements were done during the 7 yr period, and it casts additional doubts on their verifying of equipment stability and ultimately, on their result for the half-life of  $^{121}\text{Sn}^m$ .

For the other three half-life values, there is no information given on the methods that were used in these measurements, rendering a useful comparison virtually impossible. On the other hand, we can test the systematics of the M4 isomeric transition in  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$  to produce a half-life estimate for  $^{121}\text{Sn}^m$ , and then compare this estimate with our measured value. Using a reduced transition probability

for the isomeric transition deduced from  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$ , as well as their energies, conversion coefficients, and branching ratios we have estimated a half-life for  $^{121}\text{Sn}^m$  as follows.

The reduced  $\gamma$ -ray transition probability for the M4 isomeric transition is given by [10]

$$B_r = B(\text{IT}) \times \ln 2 / \{E_\gamma^9 [1 + \alpha(\text{M4})] A^2 t_{1/2}\}, \quad (1)$$

where the parameters for the isomeric transition are the following  $B(\text{IT})$  is the fraction of the isomer that decays by the isomeric transition (IT).  $E_\gamma$  is the  $\gamma$ -ray energy,  $\alpha(\text{M4})$  is the total conversion coefficient,  $t_{1/2}$  is the isomer half-life, and  $A$  is the atomic mass number. Table I shows the values of these parameters for  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$ , as well as their reduced transition probabilities ( $B_r$ ) calculated with Eq. (1).

Now, using  $B_r(^{121}\text{Sn}^m) = 515$  (average from  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$ ), together with  $B(\text{IT}) = 0.776$  (20) [7],  $E_\gamma = 0.00629$  (8) MeV [5],  $\alpha(\text{M4}) = 9.4 \times 10^{10}$  [5], and  $A = 121$  in Eq. (1), we have obtained  $t_{1/2}(^{121}\text{Sn}^m) = 49$  yr. This result is within 11% of our measured value of  $43.9 \pm 0.5$  yr. However, because of the very low energy of the isomeric transition [0.00629 (8) MeV], even the small uncertainty in its energy produces a systematic error of about 50% in its theoretical conversion coefficient, and thus about the same fractional uncertainty in the estimated half-life of  $^{121}\text{Sn}^m$ . Our measured  $^{121}\text{Sn}^m$  half-life is consistent with that calculated on the basis of the systematics of M4 isomeric transitions in  $^{117}\text{Sn}$  and  $^{119}\text{Sn}$ , which supports the  $h_{11/2}$  assignment to the 6.3 keV isomer in  $^{121}\text{Sn}$ .

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