Decay of $^{121}Sn^m$

J. M. R. Hutchinson, F. J. Schima, and B. M. Coursey
Radioactivity Section, National Bureau of Standards, Washington, D.C. 20234
(Received 23 March 1978)

$^{121}Sn^m$ has been found to decay by a highly converted isomeric transition to $^{121}Sn^g$ with a branching fraction of 0.776 ± 0.020. The K-shell internal-conversion coefficient α_K for the 37.2-keV transition in Sb, has been found to be 9.45 ± 0.33.

[RADIOACTIVITY $^{121}Sn^m$; $\beta\gamma$ coin, I_γ, ICC in Sb; deduced isomeric branching fraction.]

INTRODUCTION

In recent measurements at the National Bureau of Standards (NBS), $^{121}Sn^m$ has been found to decay directly to $^{121}Sn^g$ (see Fig. 1, [1]), as well as by the previously observed β decay to the 37.2-keV level in ^{121}Sb. β-low-energy photon coincidence counting of a sample containing $^{121}Sn^m$ yielded an activity which was approximately 4 times the activity obtained from Ge and Si(Li) 37.2-keV Sb γ-ray counting and a published[2] value for the internal-conversion coefficient. This discrepancy in the deduced activity values is explained here by the postulated existence of isomeric decay. It has previously been assumed[2] that $^{121}Sn^m$ decays 100% to the 37.2-keV level in ^{121}Sb, although in 1968, Synder and Beard[3] noted that their investigations had not ruled out the presence of the isomeric transition. Fogelberg *et al.*[4] investigated the ^{121}In decay to levels in ^{121}Sn and placed the ^{121}Sn isomer at 6.29 ± 0.01 keV above the ^{121}Sn ground state, which is consistent with the earlier value of 8 ± 5 keV of Synder and Beard.[3] Low-lying isomeric levels, which depopulate by $M4$ transitions (with $\frac{11}{2}^- \rightarrow \frac{3}{2}^+$), occur in nearby Sn and Te nuclei, and the isomeric-transition probablities of these levels can be used to ascertain if the isomeric decay mode in $^{121}Sn^m$ can compete with the β decay. The transition probability, $T(M4)$, for an $M4$ transition can be written as follows[5]:

$$T(M4) = A(M4) B_R E^9 , \tag{1}$$

where E is the energy of the isomeric transition, B_R is the reduced transition probability[5] and is dependent on factors relating to the nuclear structure, and $A(M4)$ is a constant for a given transition multipole. Table I summarizes data on five such transitions for which we have calculated $A(M4)B_R$ values. Also, the partial half-life for isomeric decay, $\tau_{1/2}^P$ is given by

$$\tau_{1/2}^P = \ln 2 / [T(M4)(1+\alpha_T)] , \tag{2}$$

where α_T is the total conversion coefficient. Using Eq. (1) to obtain the estimate of $T(M4)$ for $^{121}Sn^m$ from the average $A(M4)B_R$ value of Table I and an $\alpha_T = 9.36 \times 10^{10}$, from Ref. 7, then $\tau_{1/2}^P = 76$ years, which is comparable to the measured value of 50 years for $^{121}Sn^m$. This indicates that the isomeric decay should compete with the β decay and it seems reasonable to ascribe the above noted discrepancy in deduced activity values to this cause.

The existence of the isomeric transition has been confirmed, as will be described, by observing a

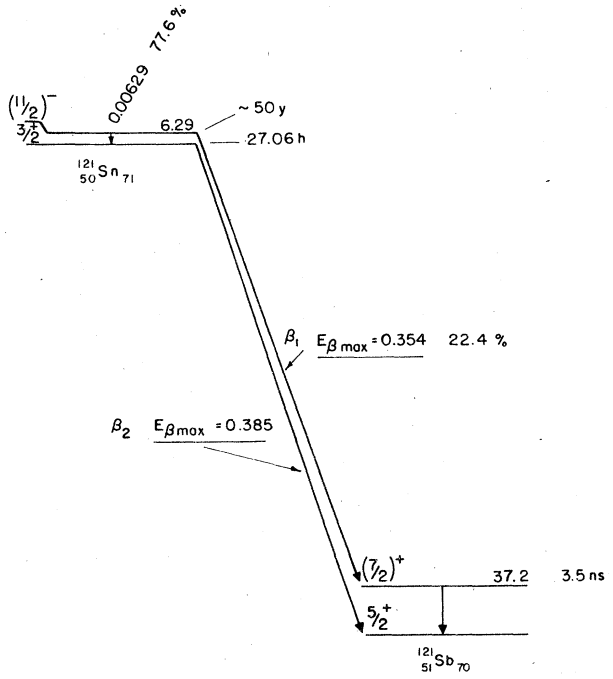

FIG. 1. Proposed $^{121}Sn^m$ decay scheme. Spin and parity assignments, half-lives, β_1 β-end-point energy and the Sb first excited state level energy are taken from Ref. 2. The energy of the isomeric level in ^{121}Sn is from Ref. 4. The β_2 β-end-point energy was obtained from an energy balance.

TABLE I. Summary of $M4$ transition data in several Sn and Te nuclei. All decay data, unless indicated otherwise, are taken from Ref. 6.

Nuclide	Neutron number	E (MeV)	α_T [a]	Branching fraction	Partial half-life (yr)	$A(M4)B_R \times 10^{-7}$ (MeV^{-9} yr^{-1})
^{117}Sn	67	0.156	47.8	1.0	1.87	0.678
^{119}Sn	69	0.065	5.2×10^3	1.0	3560.0	0.928
^{121}Te	69	0.08178	1.79×10^3	0.9	838.5	0.505
^{123}Te	71	0.08846	1.16×10^3	1.0	3702.0	0.564
^{125}Te	73	0.1094	3.63×10^3	1.0	57.8	0.534
Average value						0.642

[a]Internal-conversion coefficients are from Ref. 7.

low-energy conversion electron spectrum, as measured with a liquid-scintillation system, superimposed on the β-particle spectrum. A further confirmation, which will be described, is obtained from the observation of Sn L x rays in addition to those of Sb in a Si(Li) detector spectrum of $^{121}Sn^m$. Sn L x rays would result from L-shell internal conversion in the isomeric decay.

EXPERIMENTAL

A. β-γ coincidence counting

$^{121}Sn^m$ was produced[8] by thermal neutron irradiation of an isotopically pure ^{120}Sn target. Somewhat more than two years after the irradiation, examination of the photon and β-particle spectra indicated no other significant activity was present. Quantitative aliquots of between 3 and 60 mg of dilute HCl containing $^{121}Sn^m$ were deposited onto 10-mg cm^{-2} polyester tape and dried. Another layer of tape was then placed over each source to form a sandwich. The activities of six of these sources were determined by the method of β-photon coincidence counting using the apparatus and technique described previously by Mann and Hutchinson[9] for ^{129}I. It can be shown that the decay through the $^{121}Sn^g$ will not significantly change the coincidence equations given in Ref. 9 so long as the β-spectra from the $^{121}Sn^m$ and $^{121}Sn^g$ decay are sufficiently similar. It is estimated that a systematic error in the value for the activity per gram from this cause is less than 0.1%. The β-channel discriminator setting was varied over a range corresponding to energies from 40 to 350 keV and no systematic variation in the measured activity values for a given source was observed. Neither was there a systematic variation of measured activity per gram of $^{121}Sn^m$ solution with weight of active material. The result for 21 measurements with 6 sources was 1.147×10^6 s^{-1} g^{-1}. Table II summarizes all these measurements which are ordered by the mass of the deposited solution. These data yield a random error with a limit at the 99% confidence level of 0.63%, which is $2.845S_m$, where S_m is the standard error computed from the 21 measurements. An overall uncertainty of 0.73% is assigned to this determination. The validity of the coincidence measurements was checked also by varying the source position with respect to the two detectors, and at another time, by introducing absorbing material between first one then the other detector. In no case was there a significant change in the measured values for the activity per gram of active material.

TABLE II. Summary of the β-γ coincidence data for the $^{121}Sn^m$ activity determination.

Source mass (mg)	Activity per mg	Residual
3.668	1.1450	−0.0020
	1.1213	−0.0257
	1.1379	−0.0091
	1.1376	−0.0094
	1.1487	0.0017
	1.1520	0.0050
	1.1673	0.0203
	1.1381	−0.0089
	1.1410	−0.0060
18.05	1.1350	−0.0120
22.83	1.1436	−0.0034
25.65	1.1430	−0.0040
	1.1338	−0.0132
36.38	1.1570	0.0100
60.16	1.1529	0.0059
	1.1459	−0.0011
	1.1457	−0.0013
	1.1519	0.0049
	1.1653	0.0183
	1.1622	0.0152
	1.1618	0.0148
Average value	1.1470	

B. γ-ray-emission-rate measurements

The 37.2-keV γ-ray-emission rate per gram of source material for four of these sources was determined using Ge and Si(Li) spectrometers which were calibrated with NBS low-energy photon-emission-rate standards of ^{109}Cd, ^{125}I, and ^{139}Ce. A problem arose due to the fluorescence of the K shell of the Sn carrier in the more massive sources by the β particles and γ rays. A corrected value for the emission rate per gram of source material was obtained from an extrapolation to zero mass of the integrated peak counts per second, recorded by the Ge system. The corrected emission rate per gram of source material was 2.14×10^4 s^{-1} g^{-1}. An uncertainty of 4.9% is assigned to this value, which consists of the linear sum of 0.9%, which is the limit of the random error at the 99% confidence level ($4.032S_m$, where S_m is the standard error computed from six sets of measurements), and 4.0%, which is the upper limit of conceivable systematic errors. Additionally, the K-shell internal-conversion coefficient for the 37.2-keV transition was found to be 9.45, based on the Sb $K\alpha$ and $K\beta$ x-ray emission rates relative to that of the 37.2-keV γ ray and agrees well with the theoretical value of 9.53 (Ref. 2). A value[10] of 0.867 was used for the Sb K-shell fluorescence yield. The overall uncertainty of 3.5%, assigned to the α_K determination, was lower than the emission-rate error because of cancellation of some systematic errors.

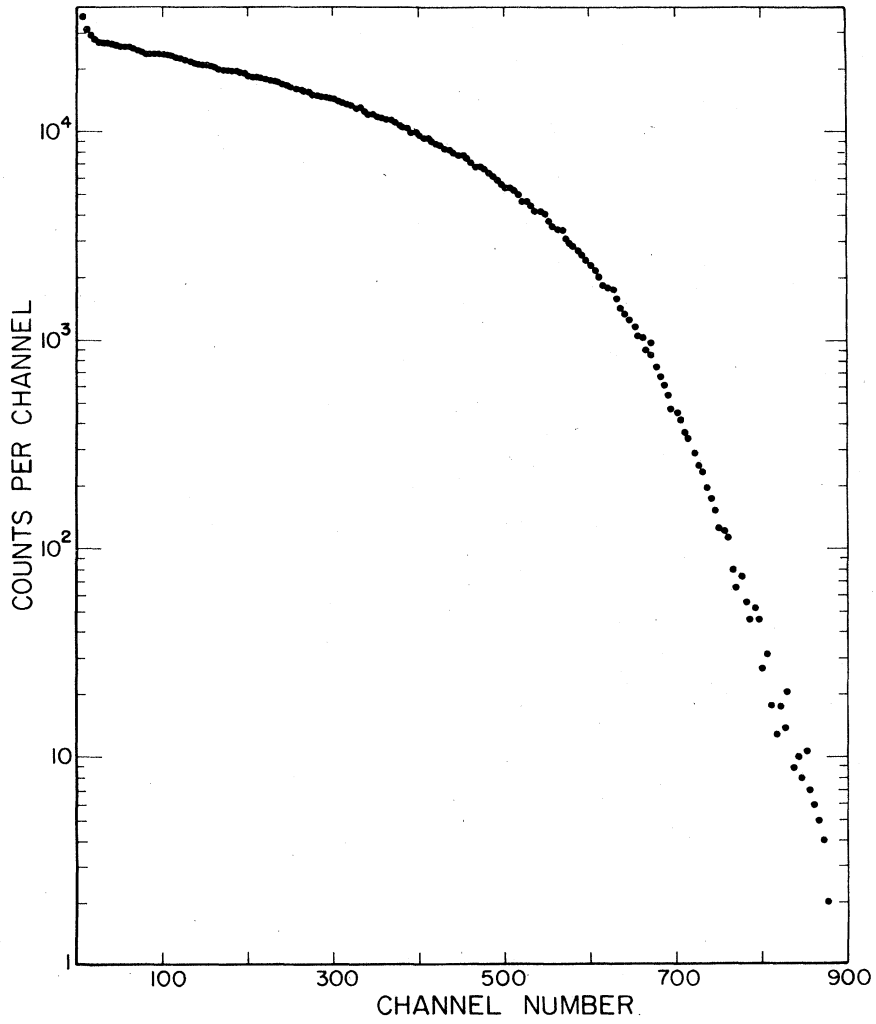

FIG. 2. Liquid-scintillation spectrum at low gain. The logarithm of the data is plotted against channel number.

C. Liquid-scintillation counting

Fifty milligrams of an aqueous solution containing $^{121}Sn^m Cl_2$ was deposited in a glass bottle and evaporated to dryness. 100 mg of a chelating agent, diethylhexylphosphoric acid (HDEHP), was added followed by 50 ml of toluene scintillator. The solution was allowed to mix thoroughly and then was purged with N_2 gas. 2.3 ml of the mixture was transferred to a cylindrical spectrophotometer cell, 2.2 cm in diameter and 1.2 cm long. The scintillations from this cell were observed with two RCA 8850 phototubes (ultra-low noise with GaP first dynodes) optically coupled to the two flat faces of the cell. The output pulses from the phototubes were amplified and summed, and the resulting spectrum was displayed on a multichannel analyzer (MCA), with the results shown in Fig. 2 for low gain and Fig. 3 for high gain. Figure 2 shows the β-particle spectrum with an indication of the existence of some pulses of low amplitude. With the amplifier gain increased by a factor of 32, the low-energy end of the spectrum is seen, in Fig. 3, to exhibit a large number of pulses, which could not be in coincidence with the β particles and which, therefore, cannot originate from the assumed β-decay mode.

A number of authors have developed procedures for determining the detection efficiencies of liquid-scintillation systems to low-energy emissions.[11,12] In this work, the procedure adopted is that described by Houtermans[13] who assumes, as also do other authors,[14] that for a given energy deposited in the scintillator, the probabilities of producing n photoelectrons from the photocathodes are distributed, for a monoenergetic source, according to the Poisson distribution P_n as

$$P_n(M) = M^n \exp(-M)/n! \,, \tag{3}$$

where M is the mean value of n. The pulse-height spectrum, $y_1(x)$ where x is the channel number in the MCA for single photoelectrons emitted from the photocathode, was obtained by exposing the photocathodes to weak incandescent light. The

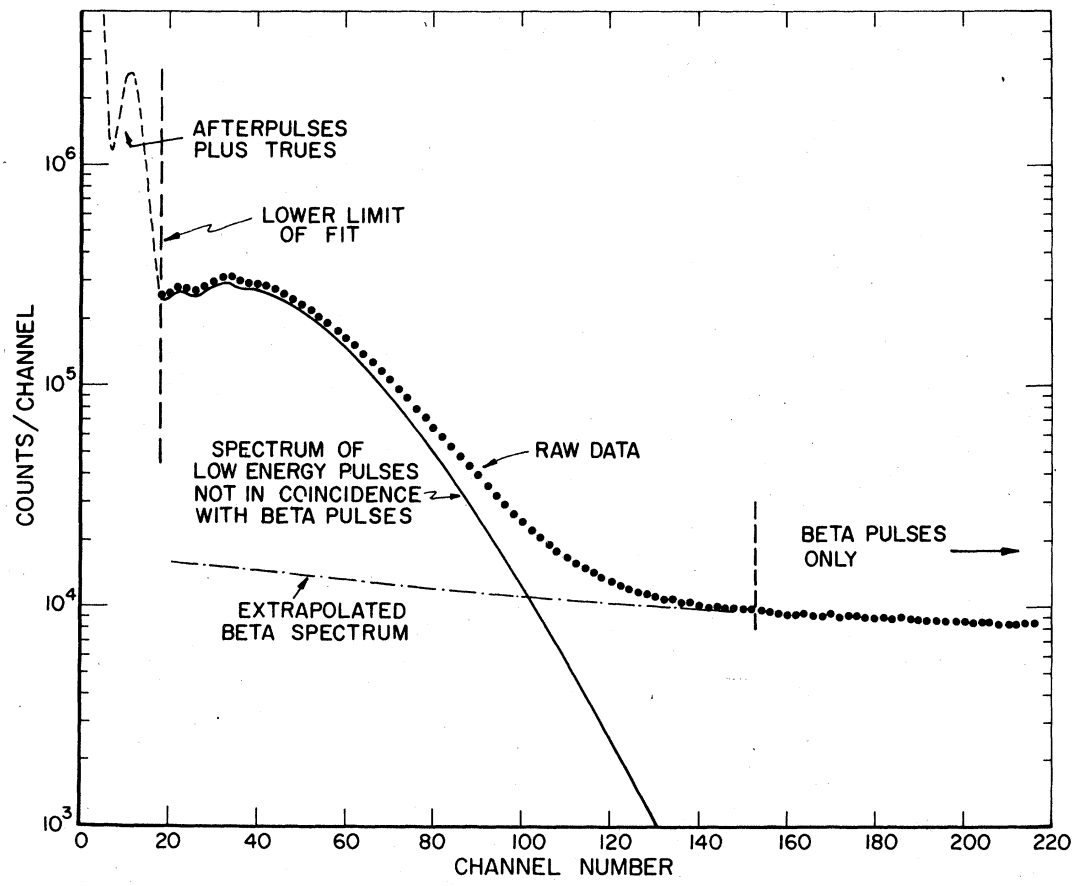

FIG. 3. Liquid-scintillation spectrum, with gain increased by a factor of 32 over the low-gain spectrum. Data points are shown as dots, while lines indicate the extrapolated β spectrum and the difference spectrum, which is assigned to the low-energy isomeric transition in the $^{121}Sn^m$ decay.

pulse-height spectrum corresponding to the production of n photoelectrons, $y_n(x)$, can be constructed from $y_1(x)$ as described by Houtermans.[13] The pulse-height spectrum $S_M(x)$ for a monoenergetic source such as the 6.29-keV transition in $^{121}Sn^m$ is given by

$$S_M(x)=\sum_{n=1}^{\infty} P_n(M)y_n(x). \quad (4)$$

The low-energy spectrum after subtracting an extrapolated β-particle background, as seen in Fig. 3, was fitted from just above the single-photoelectron peak for three values of M, the best value being $M=3.6$ as shown in Fig. 4. This value corresponds to a transition energy of approximately 6.2 keV which is in good agreement with the expected value of 6.29 keV. This extrapolation procedure was verified using β-particle pulse-height distributions of ^{3}H, ^{60}Co, ^{89}Sr, and ^{90}Sr. The single-photoelectron peak was not included in the fit because it contained spurious pulses due to afterpulsing. However, knowing $M=3.6$ from the fit, the total rate of isomeric decay events was calculated from the integrated rate for $n>1$ and using Eq. (3) to obtain corrections for $n=0$ and $n=1$. Also, the total β-particle count rate (which is equivalent to the activity of the $^{121}Sn^m$) was obtained from the data in Fig. 2, including counts from the extrapolation at low energies shown in Fig. 3. Thus, the probability of isomeric decay, P_I, defined as the ratio of the isomeric decay events to activity is estimated to be 0.814.

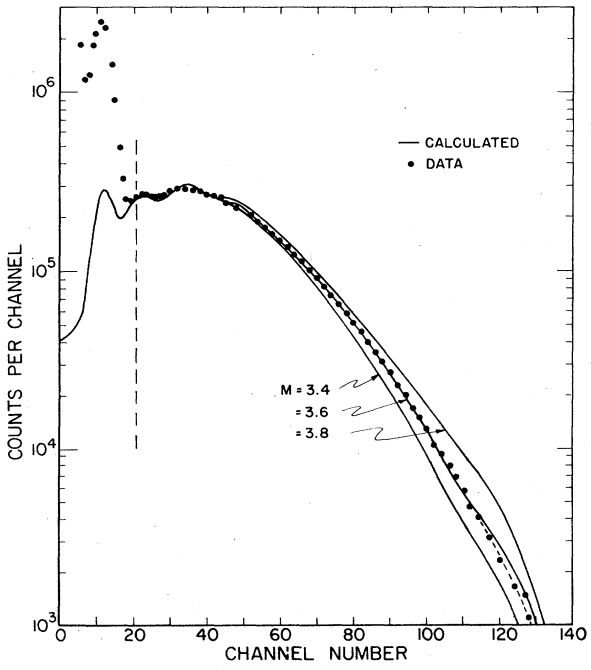

FIG. 4. The corrected low-energy liquid-scintillation spectrum shows the data as dots. The solid lines are fits to the data showing agreement with a Poisson distribution with a mean number of photoelectrons emitted per decay, M, equal to 3.6 as the best value.

D. L-x-ray analysis

Ten milligrams of diluted source solution together with a wetting agent were deposited onto a Mylar film and dried. The Si(Li) L-x-ray spectrum of this source is shown in Fig. 5, which displays principal peaks at 3.44 and 3.90 keV, which correspond to the Sn $L\alpha_1$-$L\alpha_2$ doublet and the Sn $L\beta_2$ (using the Siegbahn notation). These are the L x rays of greatest intensity resulting from Sn L_3 subshell vacancies. The theoretical[7] $M4$ internal conversion coefficient for the isomeric 6.29-keV transition predicts that 98.9% of the total number of L-shell conversions would occur in the L_3 subshell. Salem *et al.*[15] give the relative intensities for the Sn $L\alpha_1$, $L\alpha_2$, and $L\beta_2$ x-ray transitions as 100, 11, and 16, respectively, for vacancies in the Sn L_3 subshell, which is in rough agreement with the data in Fig. 5. As can be seen in Fig. 6, no Sn K x ray is observed, so the L_3 subshell vacancies cannot result from K-shell fluorescence. Internal conversion of the 37.2-keV $M1$ transition would produce the Sb $L\beta_1$ and Sb $L\alpha_1$-$L\alpha_2$ doublet at 3.84 and 3.60 keV, respectively, in addition to

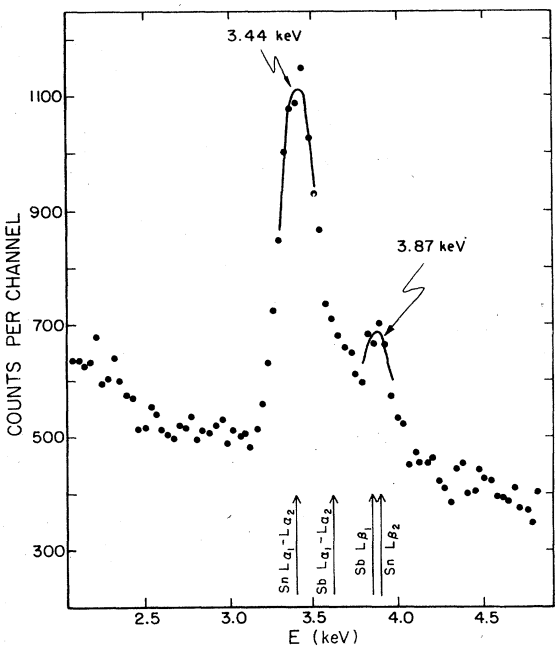

FIG. 5. A Si(Li) spectrum is displayed for the 10-mg source. The vertical bars along the energy axis indicate the location of the indicated L-x-ray emissions.

the Sb K x rays shown in Fig. 6 The Sb L-x-ray peaks must be present from the conversion of the 37.2-keV transition, but do not predominate in Fig. 5. We conclude that the observed L-x-ray spectrum supports the existence of the $M4$ isomeric transition.

CONCLUSION

We conclude that $^{121}Sn^m$ decays, not only by the emission of β particles but also by isomeric decay to the ^{121}Sn ground state. From the coincidence and γ-ray counting data, P_I is deduced to be 0.776 ± 0.020 as follows:

$$P_I = (1.0 - P_\beta), \tag{5}$$

where P_β is the probability for β decay to the 37.2-keV level in ^{121}Sb and

$$P_\beta = I^\gamma_{37.2}(1.0 + \alpha_T)/N_0, \tag{6}$$

where $I^\gamma_{37.2}$ is the γ-ray-emission rate per gram of source material, N_0 is the activity per gram of source material, and $\alpha_T = 11.0$ was obtained from our measurement of the α_K and the theoretical α_T/α_K ratio obtained from Hager and Seltzer.[7] The error for P_β was obtained from a linear sum of fractional errors of the quantities appearing in Eq. (6), multiplied by P_β. The above value of P_I is supported by a liquid-scintillation measurement, which gives $P_I = 0.814 \pm 0.067$. The low-energy liquid-scintillation pulse-height distribution can be fitted, after β-particle distribution subtraction, by a calculation which assumes a monoenergetic source as is the case for atoms undergoing isomeric decay. A further confirmation of the existence of the isomeric decay is obtained from the observation of Sn $L\alpha_1$-$L\alpha_2$ x rays in the photon spectrum from a thin $^{121}Sn^m$ source using a Si(Li) detector system.

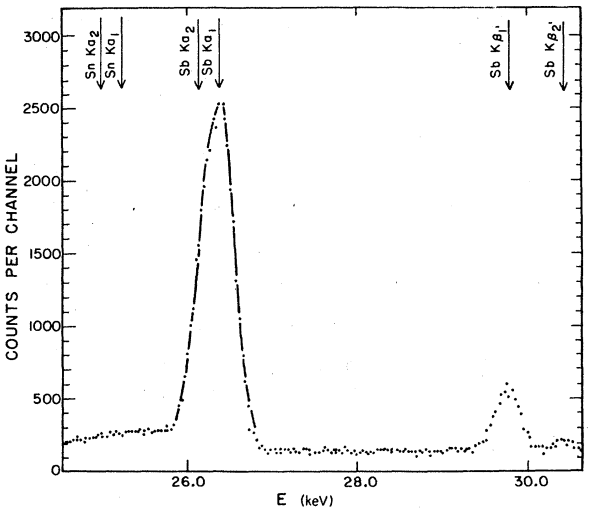

FIG. 6. A Si(Li) spectrum of the 10 mg source displays the K-x-ray region. The vertical bars along the energy axis indicate the expected location of the indicated K-x-ray emissions. Note, no Sn K x ray is discernable.

The authors would like to thank P. A. Mullen for the assistance given in the accumulation of the coincidence data, Dr. J. R. Noyce for preparing some of the sources, and J. K. Whittaker for many useful discussions about the low-noise phototubes.

[1]J. M. R. Hutchinson, F. J. Schima, and B. M. Coursey, Bull. Am. Phys. Soc. 23, 90 (1978).
[2]D. J. Horen, $A = 121$, Nucl. Data B6, 75 (1971).
[3]R. E. Synder and G. B. Beard, Nucl. Phys. A113, 581 (1968).
[4]B. Fogelberg, L.-E. De Geer, K. Fransson, and M. af Ugglas, Z. Phys. A276, 381 (1976).
[5]S. A. Moszkowski, *Alpha-, Beta-,* and *Gamma-Ray Spectroscopy*, edited by K. Seigbahn (North-Holland, Amsterdam, 1965), Vol. 2, p. 883.
[6]C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes*, 6th ed. (Wiley, New York, 1968), p. 260.
[7]R. S. Hager and E. C. Seltzer, Nucl. Data A4, 1 (1968).
[8]Obtained from the New England Nuclear Corporation (private communication).
[9]W. B. Mann and J. M. R. Hutchinson, Int. J. Appl. Radiat. Isot. 27, 187 (1976).
[10]W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. 44, 716 (1972).
[11]R. Vaninbroukx and A. Spernol, Int. J. Appl. Radiat. Isot. 16, 289 (1965).
[12]K. F. Flynn, L. E. Glendenin, and V. Prodi, in *Organic Scintillators and Liquid Scintillation Counting*, edited by D. L. Horrocks and C.-T. Peng (Academic, New York, 1971), p. 687.
[13]H. Houtermans, Nucl. Instrum. Methods 112, 121 (1973).
[14]J. A. B. Gibson, in *Liquid Scintillation Science and Technology*, edited by A. A. Noujain, C. Ediss, and L. I. Weibe, (Academic, New York, 1976), p. 153.
[15]S. I. Salem, S. L. Panossian, and R. A. Krause, At. Data Nucl. Data Tables 14, 91 (1974).