Internal-Conversion Studies with Sn^{119m} and Sn^{117m}†

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Measurements were made with a magnetic spectrometer of the internal conversion-electron lines from the $M4$ and $M1(E2)$ transitions which occur in cascade in the decay of Sn^{117m} and Sn^{119m}. In the case of Sn^{117m}, the transition energies are 156.0±0.1 keV ($M4$) and 158.6±0.1 keV [$M1(E2)$]; $\alpha_K(158.6)$ was found to be 0.1373±0.0030. Sn^{117m} is shown to be a convenient reference standard for conversion-coefficient measurements, and working values of the "coefficient" for several kinds of measurement are given. In the case of Sn^{119m}, the energies are 65.66±0.01 keV ($M4$) and 23.875±0.010 keV [$M1(E2)$]. Comparisons are made of measured subshell intensities with theoretical values, including those of Hager and Seltzer for the M subshells; and upper limits are set for the amounts of $E2$ admixture in the two $M1$ transitions. For the 23.875-keV $M1$ transition, the $L_1:M_1:N_1$ conversion ratios observed agree with the ratios of the electron densities near the nucleus of the bound $2s$, $3s$, and $4s$ electrons, in accord with the results of Drell and of Church and Monahan.

I. INTRODUCTION

BOTH Sn^{119m} and Sn^{117m} decay by emission of $M4$ and mixed $M1$-$E2$ radiations in cascade. Decay schemes of these two isomers as given in a recent compilation[1] are shown in Fig. 1. Internal-conversion studies with these nuclides were undertaken for several reasons: Predictions of $M1$ and $E2$ transition probabilities are available from the work of Sorensen,[2] and it is especially worthwhile to compare them with experimental values for single closed shell nuclei, such as the isotopes of Sn. Independent of this, an accurate total conversion coefficient for the 24-keV Mössbauer transition in Sn^{119} is useful in the determination of recoilless fractions.[3] Furthermore, the simplicity of the decay schemes of these isomers facilitates the determination of intensities of lines produced in the M and higher shells; these lines are of interest in connection with effects of screening on the wave functions of these electrons and hence on their internal conversion. Comparison of observed intensities with line intensities calculated from screened wave functions is important for the interpretation of chemical effects on the internal conversion of outer-shell electrons.[4] Finally, the fact that the two transitions in Sn^{117m} decay have approximately the same energy makes it possible to measure accurately the absolute K conversion coefficient of the $\frac{3}{2}+ \rightarrow \frac{1}{2}+$ transition; Sn^{117m} may thus find some application as an internal-conversion-coefficient standard.

II. EXPERIMENTAL METHODS

Sn^{117m} was produced by irradiation of natural Cd metal with α particles from the Brookhaven cyclotron; the α-particle energy was degraded to 15 MeV in order to favor the α, n reaction. After short-lived activities had decayed, the target was dissolved, and the Sn was chemically separated from Cd, Sb, and other activities. In this separation, the most important steps were repeated distillations of $SnBr_4$ from acid solutions containing hold-back carriers. The purified Sn activity, in which there were minor components of Sn^{113} and Sn^{119m}, was coprecipitated with a few milligrams of $Fe(OH)_3$; and the precipitate was transferred to a boat-shaped heater made of rhenium. At about 1200°C in vacuum the Sn activity sublimed. It was condensed onto a Ti metal foil held just over the heater opening. The final deposit was thin and fairly uniform and of the size required for measurement in the β-ray spectrometer, about 1 mm×15 mm.

Sources of Sn^{119m} were prepared in the Brookhaven electromagnetic isotope separator from a sample of Sn metal, highly enriched[5] in Sn^{118}, which had been heavily irradiated with neutrons.[6] The combination of high

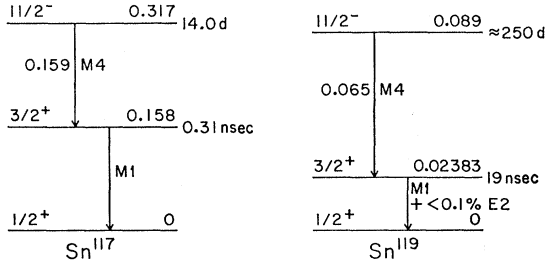

FIG. 1. Decay schemes of Sn^{119m} and Sn^{117m}, taken from Ref. 1; energies are given in MeV. Note that the transition energies measured in this work differ from the values given in this figure.

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[2] R. A. Sorensen, Phys. Rev. **132**, 2270 (1963); **133**, B281 (1964).

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[5] This material 97% Sn^{118} and 0.64% Sn^{119} was supplied by the Y-12 Plant, Union Carbide and Carbon Corp., through the Isotopes Division, U. S. Atomic Energy Commission, Oak Ridge, Tenn.

[6] The neutron irradiated Sn was purchased from New England Nuclear Corp., Boston, Mass.

TABLE I. Measured intensity ratios for the internal-conversion lines of Sn^{117m}.

Intensity ratio	Measured value
$L156.0/K156.0$	0.414 ± 0.014[a]
$MNO156.0/K156.0$	0.118 ± 0.006
$K158.6/K156.0$	0.1851 ± 0.0028
$L158.6/K158.6$	0.13 ± 0.03

[a] Values for this ratio obtained by interpolation in tabulations of Rose (see Ref. 9) and of Sliv and Band (see Ref. 10) happen to be the same (0.408); that from Hager and Seltzer (see Ref. 11) is 0.403.

specific activity and great depletion of stable Sn^{119} was required to obtain, even after isotope separation, a source thickness adequately small for the resolution of the low-energy electron lines. An ion retardation system at the collector of the separator served to reduce the energy of the Sn^{119} ions to 1.5 keV. At this energy their range is estimated to be $\sim 2\ \mu g/cm^2$. Ion focussing also results from the configuration of the retardation field, and the final deposits were approximately rectangular 0.5 mm wide by 20 mm long. As described in Ref. 4, sources were prepared both in metallic form and in the form of SnO_2. These sources were used for the studies reported in Ref. 4 and for the work reported here.

Measurements of the internal-conversion-electron spectra were made with a 50-cm radius, double-focussing, iron, β-ray spectrometer at momentum resolutions of 0.10–0.16%. The electron detector employed was a side-window proportional counter; a window thickness of $\sim 130\ \mu g/cm^2$ was used with the Sn^{117m} sources, but for Sn^{119m} a thinner window, $\sim 50\ \mu g/cm^2$, was needed. In both cases absorption corrections were thus negligible.

III. RESULTS: Sn^{117}

A. Transition Energies

Energies of the conversion lines of the two transitions in Sn^{117m} decay were measured by comparison with the L conversion lines of a 130.53 ± 0.02-keV transition occurring in Yb^{169} decay.[7] The two energies were found to be

$$E(\tfrac{11}{2}- \rightarrow \tfrac{3}{2}+) = 156.0 \pm 0.1 \text{ keV},$$
$$E(\tfrac{3}{2}+ \rightarrow \tfrac{1}{2}+) = 158.6 \pm 0.1 \text{ keV}.$$

TABLE II. Theoretical values of $\alpha_K(158.6)$ calculated for a pure $M1$ transition and for 1% $E2$ admixture. These α_K values may be compared with the experimental result $\alpha_K(158.6) = 0.1373 \pm 0.0030$.

$\delta^2 = E2/M1$	α_K (Sliv and Band)[a]	α_K (Rose)[b]	α_K (Hager and Seltzer)[c]
0%	0.135	0.139	0.136
1%	0.136	0.140	0.137

[a] Reference 10. [b] Reference 9. [c] Reference 11.

[7] E. N. Hatch, F. Boehm, P. Marmier, and J. W. M. Du Mond, Phys. Rev. **104**, 745 (1956); Z. Grabowski, J. E. Thun, and B. Lindström, Z. Physik **169**, 303 (1962); P. Van Assche, M. Neve de Mevergnies, and J. Vervier, J. Phys. **24**, 850 (1963).

The nature of the uncertainties attached to these values is such that the energy difference is accurately established as 2.59 ± 0.02 keV. Some confusion about the energies of these transitions exists in the literature, and it should be emphasized that from results reported here on line intensities, as well as from results already published,[8] the $M4$ transition has the lower energy.

B. K-Conversion Coefficient of the 158.6-keV Transition

Determination of the K-conversion coefficient of the 158.6-keV transition is based principally on the intensity of the electron line $K158.6$ relative to the sum of the intensities of the lines from the almost entirely converted 156.0-keV $M4$ transition. Because the lines compared are very close in energy, the usual difficulties arising from slight energy dependence of spectrometer transmission and line shape are at a minimum. Furthermore, the value obtained for $\alpha_K(158.6)$ is very insensitive to uncertainty in the value assumed for $\alpha_K(156.0)$. The measured intensity ratios are given in Table I.

Values for the ratio $L156.0/K156.0$ obtained from the tabulations of Rose,[9] and of Sliv and Band,[10] and from the new tabulation of Hager and Seltzer,[11] are in agreement with the measured result.

With the value assumed[12] for $\alpha_K(156.0)$, 31.5 ± 3.1, and the first two ratios from Table I one obtains

$$(K156.0+L156.0+MNO156.0+\gamma 156.0)/K156.0 = T156.0/K156.0 = T158.6/K156.0 = 1.564 \pm 0.015, \quad (1)$$

where T is the total transition intensity. It is, of course, assumed that the two transition intensities are equal. The K-conversion coefficient of the 158.6-keV transition may be written

$$\alpha_K(158.6) = K158.6/\gamma 158.6 = K158.6/[T158.6 - (K158.6+L158.6+MNO158.6)]. \quad (2)$$

All quantities on the right side of Eq. (2) may be expressed in terms of $K156.0$ by use of the result of Eq. (1), the ratios in Table I, and the assumption $MNO158.6/L158.6 = 0.3 \pm 0.1$ [13]; one thus obtains

$$\alpha_K(158.6) = 0.1373 \pm 0.0030.$$

[8] J. W. Mihelich and R. D. Hill, Phys. Rev. **79**, 781 (1950); J. M. Cork, A. E. Stoddard, C. E. Branyan, W. J. Childs, D. W. Martin, and J. M. LeBlanc, *ibid.* **84**, 596 (1951).

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[10] L. A. Sliv and I. M. Band, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Co., Amsterdam, 1965), p. 1639.

[11] R. S. Hager and E. C. Seltzer, California Institute of Technology, Report No. CALT-63-60, 1967 (unpublished).

[12] This value is the mean of 31.6 (Ref. 9), 31.1 (Ref. 10), and 31.7 (Ref. 11); a generous but arbitrary 10% uncertainty is attached.

[13] This assumption is based on results which may be found in the literature for transitions of a range of energies and of several multipole orders. Examples may be found in *Internal Conversion Processes*, edited by J. H. Hamilton (Academic Press Inc., New York, 1966).

TABLE III. Observed relative intensities of the internal-conversion lines of Sn^{119m}, and theoretical values for the conversion coefficients.

Atomic shell	Relative intensity	Theoretical coefficient[a] Rose		Sliv and Band		Hager and Seltzer	
		23.875-keV transition					
		$M1$	$(E2)$	$M1$	$(E2)$	$M1$	$(E2)$
L_1	3.75[b]	3.66	2.14	3.68	2.23	3.75	2.34
L_2	0.304±0.007	0.267	129	0.289	119	0.306	119
L_3	0.082±0.002	0.062	171	0.078	185	0.080	179
M_1	0.73 ±0.01					0.728	0.58
M_2	0.072±0.002					0.062	23.8
M_3	0.013±0.001					0.016	36.3
N_1	0.156±0.003						
		65.66-keV transition					
		$M4$		$M4$		$M4$	
L_1	814 ±16	769		768		784	
L_2	163 ± 7	167		171		173	
L_3	1742[b]	1673		1685		1742	
M_1	187 ± 7					179	
M_2	55 ± 6					39	
M_3	410 ± 8					401	
$M_{4,5}$	15.0± 1.5					18.2	
$\sum N$	119 ± 5						

[a] Theoretical values were derived from Refs. 9–11 by interpolation.
[b] Intensity normalized to theoretical value from Ref. 11 for a pure multipole order.

An upper limit of 0.4% is given for the amount of $E2$ admixture in the predominantly $M1$ 158.6-keV transition by angular-correlation results[14]; and, as may be seen from Table II, this limit is consistent with the experimental value for α_K.

Since the 158.6-keV transition is an l-forbidden magnetic dipole transition, one might anticipate that penetration terms[15] in the conversion might be visible. If the conversion coefficient is expressed in the approximate form

$$\alpha_K(\lambda)=[1-C\lambda]^2\alpha_K(\lambda=0),$$

then the value of C can be estimated from the recent tabulation of Church,[16] and the value of λ can be taken as the ratio of the shell-model estimate for the penetration matrix element and the experimental value (derived from the measured half-life[17]) of the γ-ray matrix element. One finds $C=0.006$ and $\lambda^2\approx 1$. Thus, the penetration effects probably do not alter the coefficient by more than about 1% and can be neither confirmed nor denied in the present case.

C. Sn^{117m} as a "Standard Source" for Conversion-Coefficient Measurements

If the measurement is such that the two γ rays are unresolved and the K electron lines are resolved from L lines but not from each other, then the effective conversion coefficient is

$$\epsilon_A=(K156.0+K158.6)/(\gamma 156.0+\gamma 158.6)=0.859\pm 0.027.$$

If the γ rays are unresolved, but the K electron lines are resolved, a convenient coefficient is

$$\epsilon_B=K156.0/(\gamma 156.0+\gamma 158.6)=0.725\pm 0.009.$$

If both γ rays and electron lines are resolved, the most easily used coefficient is

$$\epsilon_C=K156.0/\gamma 158.6=0.742\pm 0.010.$$

IV. RESULTS: Sn^{119}

A. Transition Energies

Calibration of the β-ray spectrometer for measurements of Sn^{119} lines was determined by the Siegbahn method[18]; the K and L_3 lines of the $M4$ transition were used. The required binding energies were taken from Hagström, Nordling, and Siegbahn.[19] Figure 2 shows the observed L multiplet of the $M1$ transition. Energies of the two transitions were found to be 65.66±0.01 and 23.875±0.010 keV.

B. Subshell Ratios

In Table III there are given the conversion-line intensity values measured, as well as theoretical values for the internal-conversion coefficients. There appears to be a small discrepancy between measurement and

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[19] S. Hagström, C. Nordling, and K. Siegbahn, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Co., Amsterdam, 1965), p. 845.

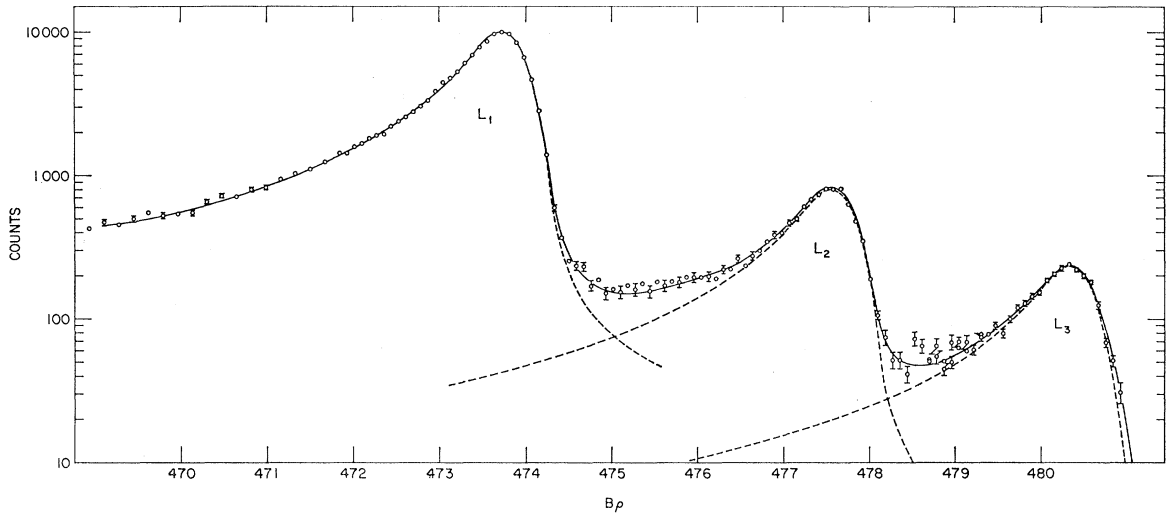

FIG. 2. L subshell internal-conversion lines of the 23.875-keV transition of Sn^{119m}. The $B\rho$ scale gives values of the line momenta measured at the extrapolated upper edges of lines on a linear plot of count rate. For this figure, background has been subtracted.

theory in the L_2:L_3 ratio for the 65.66-keV transition; the values derived from the three tabulations agree within about 2%, but the experimental value is about 6% lower. The discrepancy cannot be accounted for by $E5$ admixture. The measured M_2:M_3 ratio, on the other hand, comes out higher than the value derived from Hager and Seltzer.

In the case of the 23.875-keV transition, the differences for the L subshells between the theoretical $M1$ values and those measured can be ascribed to an $E2$ admixture; the experimental subshell ratios and the mixing ratios deduced from them are shown in Table IV. The coefficients derived from Rose[9] do not lead to a consistent mixing ratio. Those derived from Sliv and Band[10] and from Hager and Seltzer[11] are both consistent with no $E2$ admixture. Because the magnetic dipole L_3:L_1 ratios of Sliv and Band do not show a smooth energy-dependence for $Z=50$, we adopt, for the present, the mixing ratio derived from the coefficients of Hager and Seltzer, $\delta^2 \leq 0.33\times10^{-4}$. This limit is based on our arbitrary estimate of 3% for the accuracy of the relative coefficients for L_3 and L_1 conversion for the magnetic dipole part of the conversion. As in the case of the 158.6-keV transition of Sn^{117}, penetration effects in the magnetic dipole conversion should alter the L_1 and L_2 coefficients by only about 1% and thus do not appreciably affect the limit on the $E2$:$M1$ ratio.

The emission of $E2$ radiation in transitions between states having the same number of quasiparticles can be hindered by pair correlations.[20] The evidence for this effect in the tin isotopes comes only from transitions between two-quasiparticle states in the even-mass nuclei.[21] The present failure to detect any $E2$ component in the 23.875-keV transition is consistent with the systematics, but the upper limit which can be placed on the $E2$ transition probability, in single-particle units, $B(E2)\leq 0.6\ B(E2, \text{s.p.})$, is still much higher than the systematics or the predictions of Sorenson[2]: $B(E2) \approx 0.0006\ B(E2, \text{s.p.})$.

C. Shell Ratios

In magnetic dipole conversion the shell ratios for conversion in s states (e.g., L_1:K, M_1:L_1, etc.) are generally found to be nearly independent of energy; it is therefore plausible that they should be nearly equal to the bound-state electron density ratios at the nucleus, as implied by the early calculations of Drell and of Church and Monahan.[22] The ratio of M_1 to L_1 electron densities at the nucleus derived from the Hartree-Fock-Slater wave functions of Herman and Skillman[23] is 0.194, in excellent agreement with the ratio of coefficients derived from the tables of Hager and Seltzer (0.194). The measured ratio (0.196±0.003) also agrees well.

In the case of the N_1:M_1 ratio the measured value is (0.212±0.005), while the Herman-Skillman ratio of electron densities is 0.203, lending further support to the assumptions of the latter part of Ref. 4. It is interesting to note that the O:N_1 ratio found experimentally[4] for white tin metal (0.108±0.004) is in agreement with the free-atom electron density ratio (for equal numbers of $5s$ and $4s$ electrons) of Herman and Skillman (0.110).

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TABLE IV. $E2/M1$ mixing ratio for the 23.875-keV transition from L subshell ratios.

Subshell ratio	Measured ratio	$\delta^2=E2/M1$[a] Rose	Sliv and Band	Hager and Seltzer
L_1/L_2	12.35±0.3	(2.05±0.72)×10^{-4}	(≤1.75)×10^{-4}	≤1.19×10^{-4}
L_1/L_3	45.7 ±1.5	(0.82±0.13)×10^{-4}	(≤0.36)×10^{-4}	≤0.33×10^{-4}

[a] In the calculation of these values, account was taken of the uncertainties in the experimental intensity ratios and of an assumed uncertainty in the theoretical ratios for order $M1$. The latter was taken to be 3%, except in the case of L_1:L_3 from Sliv and Band (Ref. 10), where 5% was used, for a reason given in Sec. IV B.

Relativistic corrections for these nonrelativistic L_1, M_1, N_1, and O wave functions are all very nearly the same.

D. Total Conversion Coefficient of the 23.875-keV Transition

Because of the difference in line shape between $K65.66$ and the other lines, it was not possible accurately to sum line intensities of the 65.66-keV transition and compare this sum with the intensity sum from the 23.875-keV transition in order to obtain α_{total} (23.875). One can, however, obtain the α_{total} value from the sum of the observed 23.875-keV intensities (Table III) by assuming the theoretical value for any one line. Thus, if one takes L_1 23.875 to be 3.75±0.07, α_{total} (23.875) is found to be 5.12±0.14. In this value the O intensity is included; M_4 and M_5 make negligible contributions. One may compare this value for α_{total} with that of Benczer-Koller[3] 5.2±0.3.

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Spin Dependence of the U^{235} Low-Energy Neutron Cross Section*

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The interaction of polarized monochromatic neutrons with polarized U^{235} nuclei has been used to study the spin dependence of the total cross section of U^{235} at a number of neutron energies between 0.075 and 2.04 eV. Measurements were made primarily with a $^{235}U_{0.2}La_{0.8}Cl_3$ target in which appreciable nuclear polarization was produced. The results show that the 0.275-eV resonance is in the opposite spin state to the 1.14-eV resonance and to the major part of the thermal cross section. The 2.08-eV resonance is probably in the same spin state as the 0.275-eV resonance. If we assume that U^{235} has a negative magnetic moment, then $J=I-\frac{1}{2}=3$ at 0.275 eV. None of the existing cross-section analyses are completely consistent with the observations for $E\leq 1.14$ eV. In U^{235} metal a small "brute-force" nuclear polarization was detected, but there was no evidence of any hyperfine interaction. A somewhat larger nuclear polarization was observed in $^{235}UFe_2$, due to a negative hyperfine field of undetermined magnitude.

I. INTRODUCTION

ALTHOUGH the low-energy neutron cross section of U^{235} has been the subject of intensive study, no completely satisfactory analysis of it in terms of resonance theory has been possible. The difficulty originates in two basic facts: first, fission is a few channel process; second, the fission widths of the levels are comparable to their spacings. Thus there is appreciable interference between resonances in the same spin state and as a consequence the simple Breit-Wigner single-level formula is inadequate for the resonance analysis.[1] This is a situation in which a more general approach such as the Wigner-Eisenbud formalism[2] must be used. In principle, the resulting multilevel analysis should provide the basis for a complete description of the cross section; however, in practice, there is no systematic way to apply it to U^{235}. The problem is that the levels which interfere have to be specified in order to get a unique set of parameters from the analysis. Unfortunately, there is no objective procedure, based on the cross-section data alone, for grouping the resonances according to spin.

The large thermal cross section of U^{235} is also difficult to explain. It is usually attributed to one or more compound states lying just below the neutron binding energy, i.e., "negative energy" resonances. However, if only

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