The Second Transition in the Decay of Sn^{119m}

J. C. Bowe* and P. Axel
Physics Department, University of Illinois,† Urbana, Illinois
(Received August 6, 1951)

The photons from the second transition in Sn^{119m} were detected by using two proportional counters in coincidence. These photons have an energy of 23.8±0.6 kev and could be differentiated from In x-rays only by coincidence measurements. The fraction of the second transitions which emits photons is 0.13±0.03, corresponding to a total conversion of 7.3±1.7. The slow neutron activation cross section of Sn^{118} was found to be 0.01 barn.

INTRODUCTION

A 250-DAY isomer in Sn^{119} was first reported and correctly assigned by Mihelich and Hill.[1] Their original report established the presence of conversion electrons from a gamma-ray of about 69 kev and they postulated a second unobserved transition. Nelson, Ketelle, and Boyd[2] confirmed the existence of this isomer, reporting a half-life of 279 days and an energy of 64 kev. The postulated presence of the second transition was confirmed by Scharff-Goldhaber, *et al.*[3] They determined a total transition energy (for the two cascade radiations) of 85±4 kev, indicating that the gamma-ray energy of the second transition is 19±5 kev.

A more precise and direct determination of the energy of this second transition was carried on independently by Hill and ourselves. Using a beta-ray spectrograph Hill[4] observed the L and M conversion electrons from a 24.2-kev transition. The identification of the lower energy conversion electrons was made even though they were strongly absorbed in the source and were partially obscured by the presence of Auger electrons of the same energy. Hill's intensity measurements, which included large corrections, indicated about equal numbers of conversion electrons from both transitions. Hill also redetermined the energy of the isomeric transition as 65.3 kev.

Our measurements were carried out before any direct evidence for the second transition was available. We observed the unconverted gamma-rays from the second transition and quantitatively distinguished them from both the In and Sn x-rays which were present. The unconverted gamma-rays were observed in coincidence with x-rays from the first transition; quantitative analysis of these coincidence data gave the conversion coefficient.

SOURCE

The Sn^{119m} source was made available to us by R. D. Hill of the University of Illinois. It had been produced by a five-month slow neutron bombardment of enriched Sn^{118}.[5] Although the sample contained only about 0.2 percent Sn^{112} and 91 percent Sn^{118}, the x-ray activity due to Sn^{113} was larger than that of Sn^{119m}. The actual ratio of the Sn^{113} to Sn^{119m} x-rays in this source after five months of bombardment was 1.7. This implies that the slow neutron activation cross section of Sn^{118} is less than 1 percent of Sn^{112}; the calculation of this cross section is given below.

EQUIPMENT

Two identical proportional counters were used, each made from 6-inch diameter brass tubing. A 175 mg/cm² circular Be window 1 inch in diameter permitted the photons to enter with a minimum of attenuation. The counters were 20 inches long and contained 9.2 liters of a mixture of 90 percent argon and 10 percent methane at atmospheric pressure. They were operated with a battery voltage of about 2500 volts corresponding to a gas multiplication of about 10^3.

The voltage pulses were amplified using a non-overloading linear amplifier with a rise time of 0.8 microsecond and a clipping time of 4 microseconds. The gain of the amplifier was 7000.

The pulses from each counter were sorted by a single channel pulse-height selector. These selectors use vacuum diodes as discriminators and a 6AS6 for an anticoincidence tube. The output of each selector was a blocking oscillator pulse which was applied to the coincidence circuit.

The coincidence circuit had four independent channels each of which had a different adjustable delay and an adjustable resolving time.[6] The multichannel delay feature was used to constantly verify that the resolving time was sufficiently large to insure 100 percent detection efficiency of true coincidences. With a resolving time of 1.85 microseconds the first three channels

* Now at Argonne National Laboratory, Chicago, Illinois. The material in this paper was submitted to the University of Illinois in partial fulfillment of the requirements for the Doctor of Philosophy degree.

† This research was supported in part by the joint program of the ONR and AEC.

[1] J. W. Mihelich and R. D. Hill, Phys. Rev. **79**, 781 (1950).

[2] Nelson, Ketelle, and Boyd, Oak Ridge National Laboratory Report ORNL 685, 1950.

[3] Scharff-Goldhaber, der Mateosian, Goldhaber, Johnson, and McKeown, Phys. Rev. **83**, 480 (L) (1951).

[4] R. D. Hill, Phys. Rev. **83**, 865 (1951).

[5] The enriched isotope was obtained from the Y-12 Plant, Oak Ridge, Tennessee. It was irradiated by special arrangement with the Isotopes Division of the AEC, Oak Ridge, Tennessee.

[6] We wish to thank Dr. Sherman Frankel, of the University of Pennsylvania, who designed and tested the pulse-height selector and the multichannel coincidence circuit.

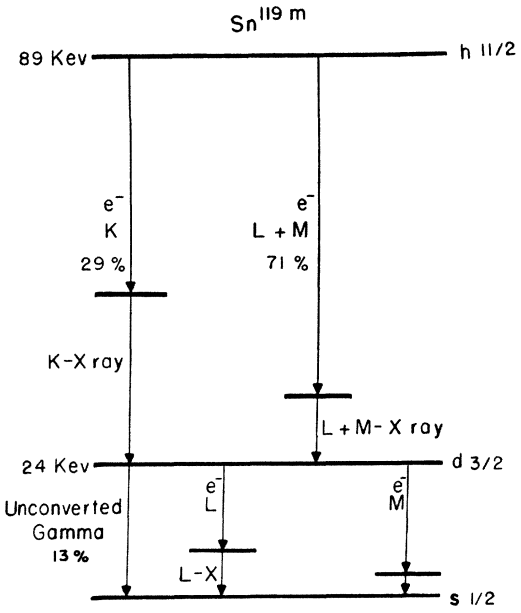

FIG. 1. Decay scheme of 280-day Sn^{119m}.

(0, 0.25, 0.60 microsecond delays) always counted the same number of coincidence pulses; the fourth channel was delayed by 1.70 microseconds and it counted fewer coincidence pulses.

A scintillation spectrometer with a 16-mm-thick NaI crystal and a RCA 5819 phototube was used. The pulses from the phototube were amplified and analyzed by a pulse height selector using the same type of equipment discussed above.

OUTLINE OF EXPERIMENTAL PROCEDURE

Several different measurements and analyses were necessary in order to obtain quantitative data on the second transition in Sn^{119m}. Since these measurements are interdependent, an outline of the entire investigation will be given before the individual measurements are described.

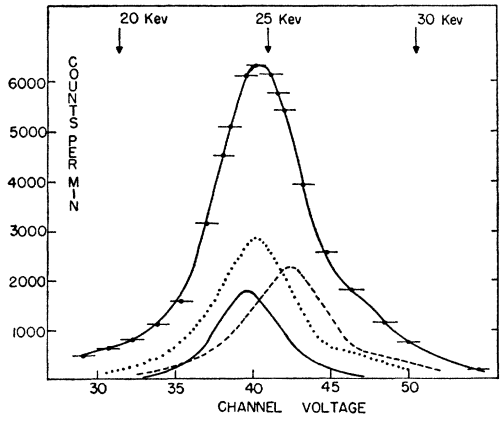

FIG. 2. Proportional counter photon spectrum of mixed Sn^{113} Sn^{119m} source. The breakdown into three components is shown: dotted curve = In x-ray; dashed curve = Sn x-ray; solid curve = 24 kev gamma-ray.

The decay scheme of Sn^{119m} is shown in Fig. 1. One major complication in the experimental determination of this scheme is the proximity in energy of the 24-kev gamma-ray to the Sn x-ray (25.2 kev). Another more limiting condition is the presence of In x-rays (24.2 kev) originating from Sn^{113}. These In x-rays completely mask the gamma-ray.

The pulse height distribution due to the photons shows a broadened peak but no detailed structure in the region of the In and Sn x-rays. The existence of the gamma-ray was shown qualitatively by the detection of coincidences between the photons contributing to the low energy part of this peak and those in the high energy part.

Quantitative data on the structure of the broadened peak were derived from three different experiments:

1. The contribution of the Sn x-ray was found by using a palladium absorber and a proportional counter detector. (Palladium absorbs the Sn x-ray more strongly than it absorbs either the Sn γ-ray or the In x-ray.)

2. To determine the contribution of the In x-rays we made use of the fact that these x-rays come from the decay of Sn^{113}. Since there is also a 390-kev gamma-ray emitted by Sn^{113} (from the In^{113m} daughter), this gamma-ray was used to indicate how much Sn^{113} was present. For this purpose, both the Sn^{119m} source and a "pure" Sn^{113} source[7] were investigated with a NaI crystal scintillation spectrometer.

3. The most accurate data on the breakdown of the composite peak into In x-ray, 24-kev Sn gamma-ray, and Sn x-ray, were obtained from coincidence experiments. Coincidences were recorded between pulses of different amplitude from two proportional counters. These coincidence data were used to quantitatively analyze and readjust the breakdown of the composite peak. Coincidence data were also taken with a palladium absorber; these data also confirmed the validity of the final breakdown.

PHOTON SPECTRUM

The photon spectrum of the mixed Sn^{113}—Sn^{119m} source is shown in Fig. 2. The final analysis of this peak into its three components is also shown.

The amount of the In x-ray was determined from the intensity of the In^{113} gamma-ray in the mixed source, using the crystal data described subsequently. The exact shape and position of the In x-ray peak was obtained by observing an In^{114m} source with a proportional counter. This source was stronger than and provided more precise data than the Sn^{113} source. However, the Sn^{113} source was also investigated and showed a similar peak due to a pure In x-ray.

To estimate the amount of Sn x-ray, a pulse-height

[7] We are indebted to Professor R. B. Duffield for the loan of this source. Although it was a neutron-bombarded Sn sample which had been enriched in Sn^{122}, the only photons present were due to Sn^{113}.

distribution curve was taken with a palladium absorber between the mixed source and the proportional counter. This curve is shown in Fig. 3 together with the original distribution. In addition, Fig. 3 shows the contribution of the Pd x-rays from the absorber. To help with the analysis of this curve, a pulse height distribution was taken with a palladium absorber and a source of pure In x-rays.

A pulse-height distribution curve was also taken with a rhodium absorber. These data showed that all the radiations in the composite peak were absorbed in the K shell of rhodium; this places a lower limit on the gamma-ray energy of 23.2 kev (the K binding energy in rhodium).

SCINTILLATION SPECTROMETER RESULTS

A NaI crystal scintillation spectrometer was used to determine the amount of Sn^{113} in the mixed $Sn^{119m}-Sn^{113}$ source. The scintillation spectrometer pulse height distribution curve for the mixed source is shown by the solid line in Fig. 4. In the same figure, the data for a pure Sn^{113} source are shown normalized (i.e., multiplied by a factor of 2.7) so that the two gamma-ray intensities are equal. The dashed curve in the low energy region indicates the amount of In x-rays which accompanies the 390-kev gamma-rays.

The Sn^{119m} source shows no evidence for any photons other than those in the x-ray peak and at 390 kev. The region of 85 kev was investigated with particular care and an upper limit of 0.5 percent can be placed on any gamma-ray branch of that energy in Sn^{113}. This gamma-ray had been reported by early investigators but had not been seen in the recent study of Thomas, *et al.*[8]

COINCIDENCE EXPERIMENTS

A preliminary qualitative study established the existence of coincidences in a $Sn^{119m}-Sn^{113}$ source and showed no coincidences in the pure Sn^{113} source. For quantitative results, three separate sets of coincidence data were taken. For each of these three sets of data, the pulses from one proportional counter (counter 1) were selected in six different three volt channels covering the pulse height range from 30 to 48 volts. The pulse distribution from counter 1 had its peak at 40 volts and had a full width at half-maximum of 6.6 volts (a 6.0 volt full width would be expected for a monoenergetic radiation). The differences between the three sets of data involved the second proportional counter.

Set I: The pulse height selector of counter 2 was adjusted to select the lower $\frac{1}{3}$ of the pulses in the peak. The resultant coincidence rate was plotted as a function of the channel level selected in counter 1; these data are shown by the circled points in Fig. 5. There is a peak coincidence rate at a voltage close to that of a Sn x-ray. Thus, the fixed channel can be interpreted as emphasizing the gamma-ray.

Set II: The pulse height selector of counter 2 was moved to include only the upper $\frac{1}{3}$ of the pulses in the peak. The resultant coincidence rate is shown by the points enclosed in squares in Fig. 5. In this case the fixed channel emphasizes the Sn x-ray and therefore the peak in the coincidence distribution comes at the energy of the gamma-ray, 24 kev.

Set III: The pulse height selector of counter 2 was kept so that it included the upper $\frac{1}{3}$ of the pulses and a palladium absorber was included between the source and counter 2. Since the palladium strongly absorbs the Sn x-ray, this fixed channel, although at a relatively high voltage, emphasized the gamma-ray. The peak of the coincidence distribution shifted back to the position of the Sn x-ray. Since the fraction of the coincidences absorbed varied from 0.6 to 0.8, the statistics obtained on the individual points were relatively poor. Thus, although these data were checked semiquantitatively, the lack of precision made it impossible to use them to determine the amount of 24-kev gamma-ray.

The quantitative analysis of the first two sets of data was based on the equations given below. The following symbols will be used:

$N \equiv$ number of Sn^{119m} nuclei decaying per unit time.
$E \equiv$ probability of detecting a radiation (including solid angle and efficiency).

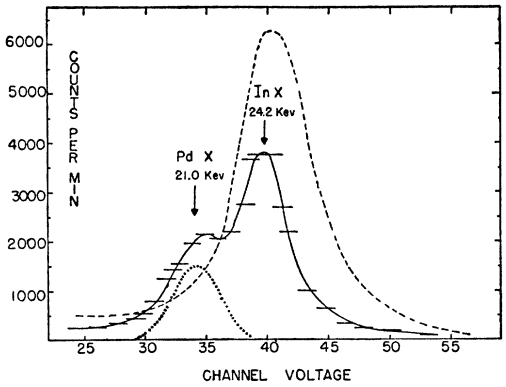

FIG. 3. Proportional counter spectrum with palladium absorber. The curve without absorber is shown dashed. The analyzed contribution of Pd x-rays is given by dotted curve.

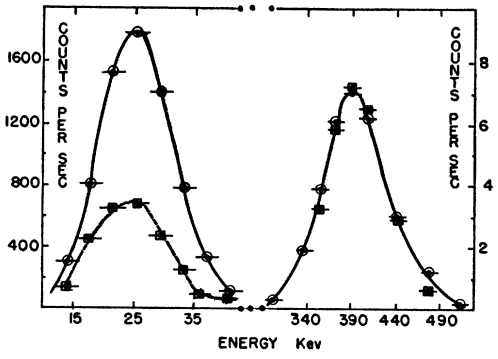

FIG. 4. NaI crystal spectrum of Sn^{113} and mixed $Sn^{113}-Sn^{119m}$ sources. Mixed source data given by circled points. Normalized Sn^{113} data shown by points enclosed in squares.

[8] Thomas, Haynes, Broyles, and Thomas, Phys. Rev. 82, 961 (L) (1951).

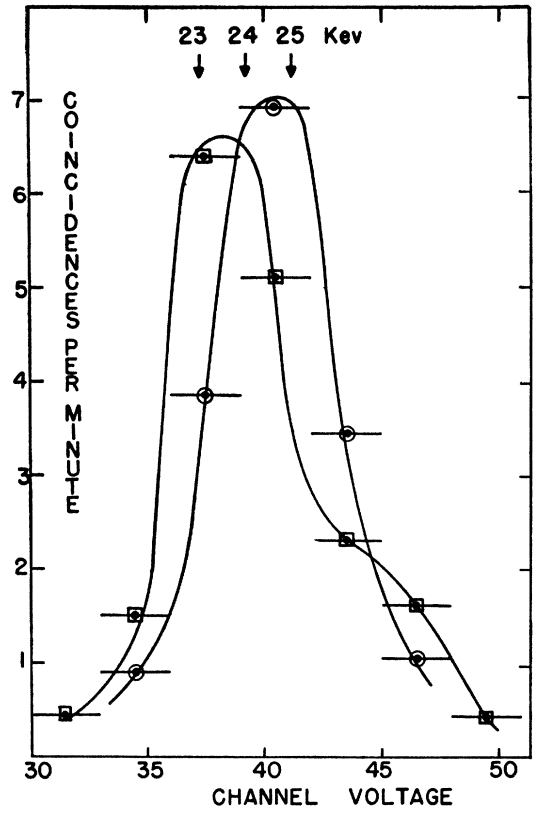

FIG. 5. Proportional counter coincidence spectrum of Sn^{119m}. The two curves represent two different channel levels in the counter with fixed bias. The circled points correspond to coincidences with lower $\frac{1}{3}$ of photon peak; the points enclosed in squares show coincidences with the upper $\frac{1}{3}$.

$r \equiv$ ratio of the number of In x-rays to Sn x-rays detected by proportional counter. r was determined as 1.5 at the time of the coincidence measurements.

$f \equiv$ fluorescence yield of Sn x-ray ($f=0.8$).[9]

$a \equiv$ fractional occurrence of K conversion in 65-kev transition ($a=0.29$).[4]

$b \equiv$ fractional occurrence of unconverted 24-kev gamma-ray.

$C \equiv$ fraction of the pulses from one type of radiation included in a particular channel width at a given setting.

Superscripts are used to denote the counter; the counter with the fixed broader channel level is designed as counter 2. Subscripts are used for the type of radiation; 1 denotes the Sn x-ray, 2 the 24-kev Sn γ-ray, and 3 the In x-ray.

The number of pulses in a given pulse height selector channel from counter 1 is

$$n^1 = N[afE_1{}^1C_1{}^1 + bE_2{}^1C_2{}^1 + rafE_1{}^1C_3{}^1],$$

where only the C values vary with the channel width and channel level. Similarly, the number of coincidences are

$$n^{12} = Nafb[E_1{}^1C_1{}^1E_2{}^2C_2{}^2 + E_1{}^2C_1{}^2E_2{}^1C_2{}^1].$$

If these two equations are combined and the values for a, f, r and E_2/E_1 are substituted, one finds

$$n^{12}/n^1 = bE_1{}^2[C_1{}^1C_2{}^2 + C_1{}^2C_2{}^1]/ [0.88(C_1{}^1 + 1.5C_3{}^1) + 4.35bC_2{}^1].$$

Experimental data from eleven different coincidence points were used to determine b. The probability of counter 2 detecting a Sn x-ray emitted by the source was known from an independent set of measurements to be (1.9±0.2) percent. The values for the constants, C, were determined by using the breakdown shown in Fig. 2. In the course of estimating the experimental error several extreme breakdowns were used. In addition, estimates were made of the possible variation of channel level and channel width during the coincidence runs.

The resultant value of b is found as $b=0.13\pm0.03$. The statistical accuracy and the internal consistency are well within this assigned error which includes a reasonable allowance for possible systematic errors. The best value of the total conversion coefficient is 6.7, with a possible range from 5.6 to 9. However, in view of the size of the possible error, the value of the conversion coefficient can be most conveniently expressed as 7.3±1.7.

DISCUSSION OF RESULTS

The experimental measurements give only the total conversion coefficient, including electron contributions from the L, M and N shells. However, the theoretical calculations available are only for the L shell.

The only experimental data on the electron contributions from different shells are those of Hill.[4] He reports that (72±8) percent of the electrons originate in the L_I subshell while the others originate in the M_I shell.

The only two sets of conversion calculations applicable to this problem are those of Gellman, Griffith, and Stanley[10] and those of Hebb and Nelson.[11] The calculations of Gellman, *et al.* are more accurate; they use relativistic expressions but neglect screening. However these have been made only for the L_I shell. The calculations of Hebb and Nelson are valid only for the internal conversion of electric multipoles. These calculations are made on the basis of poorer, nonrelativistic formulas and the contributions of the individual L subshells are not given.

The comparison of the experimental and theoretical values are shown in Table I.

A comparison of the theoretical results of Gellman, *et al.* with the combined experimental results of Hill and ourselves favors an assignment of magnetic dipole to the 24-kev transition. However, this assignment

[9] H. Tellez-Plasencia, J. phys. et radium **10**, 14 (1949); Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).

[10] Gellman, Griffith, and Stanley, Phys. Rev. **80**, 866 (1951).

[11] M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).

cannot be based solely on the conversion data now available. Experimentally, it is possible that some L_{II} or L_{III} conversion electrons are obscured in the Auger electron background.

Theoretically, the conversion contribution from the L_{II} and L_{III} shells is not known for magnetic radiation. Also, the effects of screening on the L_I conversion are not known. For electric radiation, a comparison of the calculations of Gellman, *et al.*, with those of Hebb and Nelson indicate that the predominant conversion contribution comes from the L_{II} and L_{III} shell. Although Hebb and Nelson's calculations are approximate, the approximation is expected to be accurate at low energy. Furthermore, Goodrich[12] has separated the formulas of Hebb and Nelson to show the L_I contribution explicitly. Goodrich finds that for electric quadrupole radiation, the approximate calculation gives values which are within a factor of two of those of Gellman, *et al.* below 75 kev in energy. If these calculations prove to be correct when the more complete L conversion tables of Rose and his collaborators become available, the experimental data presented in this paper will give an unambiguous assignment of magnetic dipole to the 24-kev transition.‡

The assignment of the spins and parities to the levels in Sn^{119} can also be based on more indirect evidence from both the shell model of Mayer[13] and the analogous two step transitions in Sn^{117m},[1] Te^{121m},[14] Te^{123m} [14] and Te^{125m}.[15] These assignments have already been suggested[1,3] without knowledge of the conversion coefficient.

Thus, the present experimental report quantitatively establishes the existence of the 24-kev gamma-ray. Furthermore it strongly supports the level assignments. The conversion data will provide a check of conversion theory when more adequate calculations become available. They now seem to indicate that in Sn^{119} the 24-kev magnetic dipole conversion is predominantly in the L_I shell.

CALCULATION OF Sn^{118} CROSS SECTION

To calculate the neutron absorption cross section of Sn^{118} relative to Sn^{112} the following data were used:

1. The relative number of x-rays due to Sn^{113} (and its daughter In^{113m}) compared to those of Sn^{119m}, after five months of slow neutron bombardment, was experimentally determined as 1.7.

2. The fraction of Sn^{112} in the bombarded sample was about 0.2 percent, while Sn^{118} was 91 percent.

TABLE I. Conversion coefficients of 24-kev gamma-ray.

A. Experimental

Total conversion all shells	L_I conversion using Hill's data
7.3±1.7	5.3±1.4

B. Theoretical

	Electric multipole		Magnetic multipole
	Hebb and Nelson complete L shell	Gellman, *et al.* L_I shell	Gellman, *et al.* L_I shell
Dipole	1.16	0.92	4.7
Quadrupole	200	3.5	

3. The half-life of Sn^{113} was taken as 112 days;[16] that of Sn^{119m} as 279 days.

4. The electron capture disintegration of Sn^{113} results in a K shell hole 71 percent of the time. This figure is based on the experimental L to K electron capture ratio of 0.42 determined by Thomas, *et al.*;[8] this experimental figure is used although the theoretical L to K ratio[17] is 0.11.

5. The Sn^{113} daughter, 1.7-hour In^{113m} has a K to L ratio[18] of 5.4 and a theoretical K conversion coefficient[19] of 0.5.

6. K conversion electrons are emitted by Sn^{119m} in 29 percent of the isomeric transitions.[4]

Using these data, the cross section for the activation of Sn^{119m} is 1/110 that for Sn^{113}. This large difference in activation cross section does not necessarily imply a comparable difference in the total capture cross section. When Sn^{118} captures a slow neutron, the compound nuclear state formed probably has a low spin. The radiations from this state probably lead predominantly to the ground state of Sn^{119} (spin 1/2) rather than the isomeric level, Sn^{119m} (probable spin 11/2).

Using the value of 1.1 barns[20] for the activation cross section of Sn^{112}, the absolute cross section of Sn^{118} for the activation of Sn^{119m} is 0.01 barn.

ACKNOWLEDGMENTS

We wish to thank Mr. Robert Baer who proficiently helped in taking and analyzing the data.

We are also grateful to Professor R. D. Hill for many enlightening discussions as well as for his cooperation in loaning us the source.

We are indebted to Dr. Sherman Frankel of the University of Pennsylvania for discussions and aid pertaining particularly to electronic problems. Dr. Frankel also designed and tested the pulse height selector and the multichannel coincidence circuit.

[12] R. F. Goodrich, University of Illinois, private communication.

‡ *Note added in proof.*—Additional evidence for the predominance of L_I conversion in magnetic dipole transitions has been given by Mihelich and Church, Bull. Am. Phys. Soc., Chicago, Illinois, October 24–27, 1951, p. 37. They also point out that electric quadrupole transitions give rise to mainly L_{II} and L_{III} conversion.

[13] M. G. Mayer, Phys. Rev. **78**, 16 (1950).

[14] Katz, Hill, and Goldhaber, Phys. Rev. **78**, 9 (1950).

[15] J. C. Bowe and P. Axel (to be published).

[16] C. M. Nelson and G. E. Boyd, Oak Ridge National Laboratory Report ORNL 499, 1949.

[17] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 540 (L) (1949).

[18] J. L. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940).

[19] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[20] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947).