Internal Conversion Coefficients of γ Rays in Pu^{239}

G. T. EWAN, J. W. KNOWLES, AND D. R. MACKENZIE
Chalk River Laboratories, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada
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The K-conversion coefficients of the predominantly $M1$ γ rays of 209.7, 228.2, and 277.9 kev following the β decay of Np^{239} have been measured using a curved crystal γ-ray spectrometer and a lens type β-ray spectrometer. The measured K-conversion coefficients are 1.76 ± 0.30, 1.60 ± 0.16, and 1.16 ± 0.12, respectively. A limit of less than 10% has been placed on the admixture of $E2$ in these radiations from a study of the K/L and $L_{\mathrm{I}}:L_{\mathrm{II}}:L_{\mathrm{III}}$ conversion ratios. It is found, for $Z=94$, that the measured conversion coefficients show better agreement with the $M1$ predictions of Sliv who allowed for finite nuclear size than with Rose *et al.* who assumed a point charge nucleus. In addition, the K-conversion coefficients of the 254.7-, 285.5-, 316.2-, and 334.4-kev γ rays of Pu^{23}, have been measured to be 0.88 ± 0.32, 0.11 ± 0.02, 0.027 ± 0.005, and 0.025 ± 0.005, and the L_{I} and L_{II} conversion coefficients of the 106.4-kev γ ray to be 0.062 ± 0.007 and 0.071 ± 0.007, respectively.

I. INTRODUCTION

THE effect of a finite nuclear volume on the calculation of internal conversion coefficients has recently been estimated by Sliv *et al.*[1] Their calculated K-conversion coefficients differ significantly from those of Rose *et al.*,[2] who assumed a point nucleus. The ratio of the values calculated by Sliv to those calculated by Rose is shown in Fig. 1, where this ratio for a series of multipolarities is plotted as a function of Z for a γ ray of energy 255 kev (0.5 mc^2). The ratio, which is nearly unity at low values of Z, decreases with increasing Z, and is particularly small for $M1$ transitions at high Z. Recent experiments have indicated that measured conversion coefficients differ significantly from the calculated coefficients of Rose. Measurements of the $K:L_{\mathrm{I}}:L_{\mathrm{II}}$ conversion coefficient ratios of the 279-kev γ ray of Tl^{203} by Nordling *et al.*[3] show this disagreement. Direct evidence that the K-shell internal conversion coefficients for $M1$ transitions are less than those predicted by Rose, has been shown by Nielsen *et al.*[4] for the predominantly $M1$ transitions of Bi^{214} and McGowan *et al.*[5] for transitions in Ta^{181}, Au^{197}, and Tl^{203}.

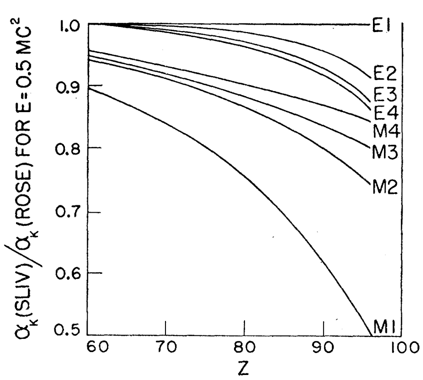

FIG. 1. Ratio of K-conversion coefficients calculated by Sliv to those calculated by Rose for γ ray of energy 255 kev (0.5 mc^2).

In the present experiments measurements were made of the conversion coefficients of the predominantly $M1$ γ rays of energies 209.7, 228.2, and 277.9 kev which occur in Pu^{239} following the β decay of Np^{239}. These γ rays are all transitions from the 5/2+ level at 285.6 kev to members of the ground state rotational band as shown in Fig. 2.[6] In addition, measurements are made of the K-conversion coefficients of other γ rays occurring in the decay of Pu^{239}. Measurements of the conversion electrons were made using a double lens β-ray spectrometer. Only relative measurements were made and their intensities are quoted in terms of the total intensity of the K-conversion electrons. Gamma-ray intensity measurements were obtained using a carefully calibrated two meter curved crystal spectrometer of the DuMond type, and were measured relative to the total intensities of those K x-rays of Pu which result from internal conversion. The total intensity of these K x-rays, when corrected for fluorescent yield, gives a measure of the total intensity of the K-conversion electrons and enables the two sets of measurements to be normalized to each other and hence the conversion coefficients of the γ rays to be determined.

II. EXPERIMENTAL RESULTS

For the β-ray spectrometer measurements, sources were prepared from samples of U^{238} depleted in U^{235}. These were irradiated for approximately one day in the NRX reactor, and the Np^{239} separated from the uranium and fission products by an ion exchange column. Suitably thin sources for use in the spectrometer were prepared by sublimation of the purified Np^{239} oxide on to 200 $\mu g/cm^2$ aluminum foil over an area one-half mm in diameter. The internal conversion spectrum was studied in a double lens β-ray spectrometer operated at

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[2] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).
[3] Nordling, Siegbahm, Sokolowski, and Wapstra, Nuclear Phys. **1**, 326 (1956).
[4] Nielsen, Nielsen, and Waggoner, Nuclear Phys. **2**, 467 (1956).
[5] F. K. McGowan and P. H. Stelson, Phys. Rev. **103**, 1133 (1956).
[6] Hollander, Smith, and Mihelich, Phys. Rev. **102**, 740 (1956).

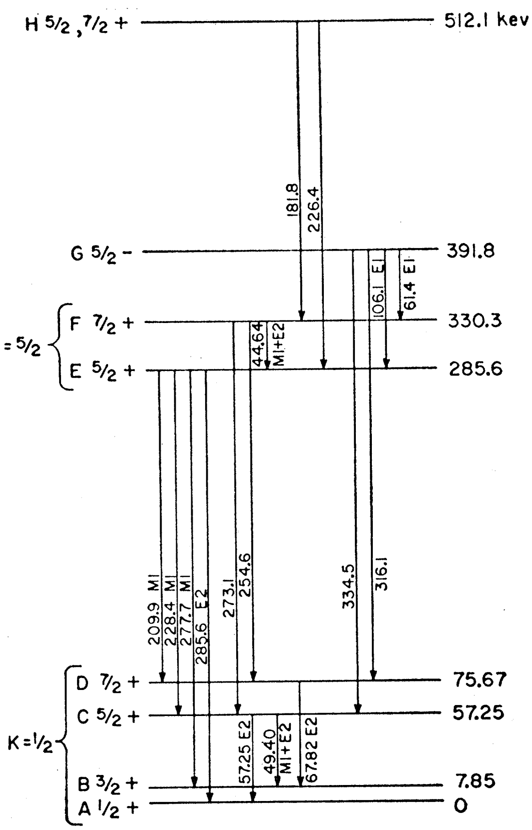

FIG. 2. Level scheme of Np^{239} proposed by Hollander *et al.*[6]

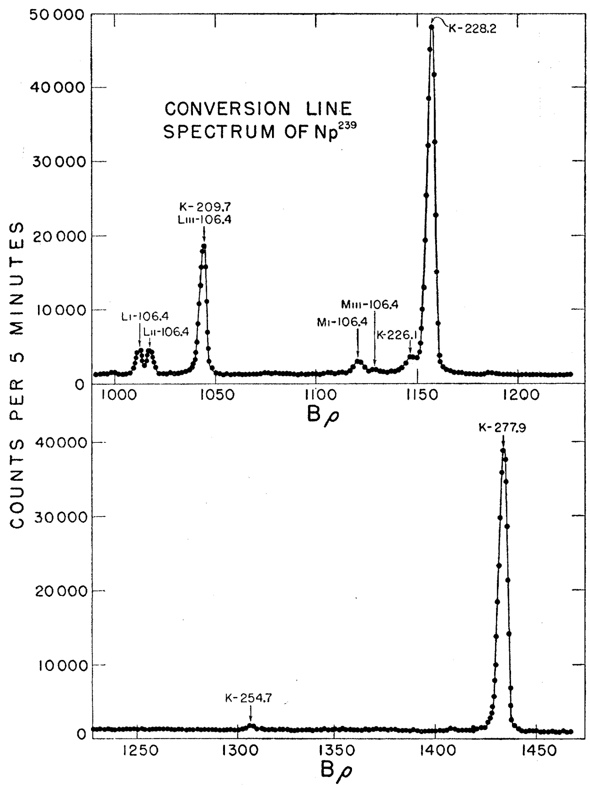

FIG. 3. Section of conversion line spectrum of Np^{239} showing K-conversion lines of 209.7-, 228.2- and 277.9-kev γ rays. The statistical error is smaller than the size of the points.

0.35% resolution in momentum. A section of the spectrum is shown in Fig. 3. The conversion lines were observed to decay with a half-life of 2.3 days, confirming that they were all associated with Np^{239} decay. The results of the conversion electron measurements are listed in Table I. The intensities are quoted relative to that of the total intensity of the K-conversion lines.

The sources used in the curved crystal spectrometer were prepared by irradiating a thin sheet of U^{238} metal (depleted in U^{235}) 4×10^{-2} cm thick and 0.7×2.3 cm^2 area. Before irradiation the uranium was sealed in a thin aluminum holder. After irradiation the uranium and the aluminum holder were mounted directly in the crystal spectrometer. The observed spectrum is shown in Fig. 4. The resolution is determined by the geometric source thickness and is about 0.75% in energy at 150 kev. The γ rays and x-rays were observed to decay with a half-life of 2.3 days, confirming that the Pu

TABLE I. Experimental and theoretical K-conversion coefficients for Pu^{239} gamma rays.

γ ray (kev)	Conversion line int. I_K	I_{LI+LII}	I_{LIII}	Quantum int. I_γ	K-conversion coefficients Expt.[a]	Theor. Sliv	Rose	I_{LI+LII}/I_{LIII}	Limit on $E2$ admixture for $M1$ γ rays
106.4		6.7±0.5	2.3±1.7[b]	50.4±2.5					
209.7	15.9±2[b]	3.5±0.3	0.03±0.1	9.0±0.7	1.76±0.30	2.58 ($M1$)	4.70 ($M1$)	115±40	10%
226.1	2.2±0.7								
228.2	44.7±1.5	9.4±0.5	0.065±0.015	28.0±1.4	1.60±0.16	2.05 ($M1$)	3.74 ($M1$)	145±35	10%
254.7	0.50±0.05			0.57±0.2	0.88±0.32	{1.48 ($M1$)	2.75 ($M1$)		
						0.10 ($E2$)	0.11 ($E2$)		
273.1	0.27±0.07								
277.9	36.1±1.5	7.5±0.5	0.05±0.01	31.1±1.5	1.16±0.12	1.17 ($M1$)	2.22 ($M1$)	150±30	10%
285.5	0.15±0.015			1.4±0.2	0.11±0.02	0.084 ($E2$)	0.089 ($E2$)		
316.2	0.090±0.015			3.3±0.3	0.027±0.005	0.029 ($E1$)	0.029 ($E1$)		
334.4	0.11±0.015			4.4±0.4	0.025±0.005	0.026 ($E1$)	0.026 ($E1$)		

[a] The errors assigned to the K-conversion coefficients make allowance for a possible systematic error in the normalization of the measurements made with the two instruments.
[b] These lines were not resolved and had combined intensity 18.2±1. The intensity has been assigned to L_{III} (106.4 kev) assuming Rose's L-conversion coefficients and assuming that the gamma ray is $E1$. The errors assigned to the two lines make allowance for a large error in this assumption.

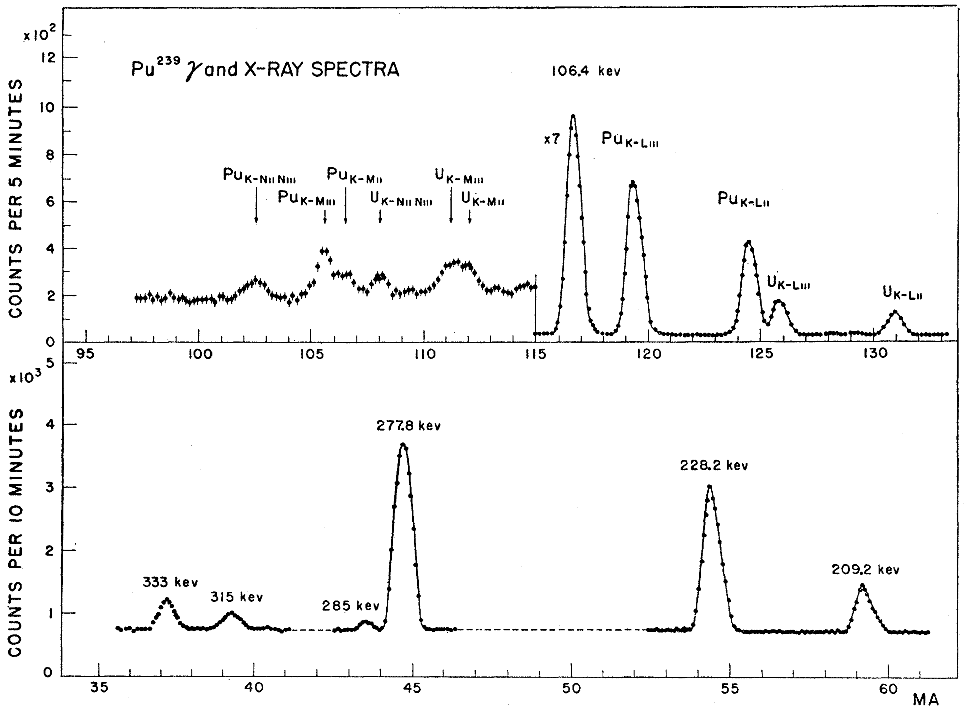

FIG. 4. Section of γ-ray and x-ray spectra of Pu^{239}, showing the 209.7-, 228.2- and 277.9-kev γ rays and the internal K-conversion x-rays. The statistical error, where not indicated, is smaller than the size of the points.

x-rays are associated with the observed Pu^{239} γ rays. To obtain the relative intensities of the observed γ rays, corrections were applied for efficiency of reflection of the quartz crystal, absorption of γ rays in the source material and flight path, and variation of sensitivity with energy of the NaI detector. The reflectivity of the curved quartz crystal as a function of energy between 40 and 511 kev was obtained to a precision of $\pm 3\%$ by measuring the direct and diffracted γ rays and x-rays following the β decay of Sm^{153}, Tb^{160}, and Cu^{64}. These sources were found to be suitable for calibration since their spectra are characterized by a few strong γ rays. The γ-ray absorption in the flight path consisting of quartz, air, and aluminum window in the NaI detector, was less than 10% and was calculable. Since the γ-ray absorption in the source was large it had to be known with precision comparable to the γ-ray intensity measurements. The absorption was estimated for all γ rays and also was measured experimentally for the strong γ rays and x-rays by comparing the Pu^{239} pulse-height spectrum of the direct radiation from the thick source with that from a thin source having negligible γ-ray absorption.

The γ-ray intensity measurements in Table I are quoted relative to the total intensity of K-conversion electrons. This was done by measuring the γ rays relative to the total intensity of the Pu K x-rays. The total intensity of these x-rays, when corrected for fluorescent yield, is a direct measure of the number of K-shell vacancies caused by internal conversion. The conversion electron measurements were normalized to the sum of the observed K-conversion line intensities given in Table I. It was not possible to observe directly K-conversion electrons below 30 kev. However, the contribution below this energy is probably negligible since no unassigned conversion lines were observed which could be attributed to the L or M conversion of γ rays between 125 kev (the K absorption edge of Pu) and 155 kev, and no unconverted γ rays were observed in this energy range.

III. DISCUSSION OF RESULTS

The measured conversion coefficients of the 209.7-, 228.2-, and 277.9-kev γ rays given in Table I can be compared with theory only if their multipolarities are known. For these γ rays the multipolarities were determined from measurements of the K/L and $(L_{I}+L_{II})/L_{III}$ conversion ratios. The latter ratio is more sensitive for predominantly $M1$ γ rays in this region, and was used to place the limits quoted in this paper. The values of the $(L_{I}+L_{II})/L_{III}$ ratios have been calculated for various multipole orders on the basis of Rose's conversion coefficients and are shown for $M1$ and $E2$ multipolarities as a function of energy in Fig. 5. For higher multipole orders in this energy range, the maximum value of the ratio is 20, which is considerably less than the observed values. This implies that only admixtures of $M1$ with other multipoles will give agreement with experiment. Of these, the most probable multipole admixture is $E2$.[7,8] The calculated $(L_{I}+L_{II})/$

[7] The decay scheme for Pu^{239}, Fig. 2, represents the 228.2-kev γ ray as a transition with no change of parity between two states of the same nonzero spin. In elements of high Z, a low-energy $E0$ transition may compete favorably with $M1$ and $E2$ transitions.[8]

L_{III} ratio, for a 5% $E2$ admixture is consistent with experiment as shown in Fig. 5. The dependence of the estimate of $E2$ admixture on Rose's theory does not seriously affect the comparison of experimental K-conversion coefficients with theory, unless variations of either the L_{I} or L_{III} conversion coefficients are greater than a factor of two. If $\alpha_{L\text{I}}$ is less than Rose's prediction by a factor of two, and $\alpha_{L\text{III}}$ remains constant, the percentage of $E2$ would be reduced from approximately 5% to 2%. A corresponding decrease of $\alpha_{L\text{III}}$ by a factor of two greater than that for $\alpha_{L\text{I}}$ would predict an admixture of $E2$ of 10%. This latter decrease is still not large enough to affect the conclusions drawn in this paper. A comparison of measured conversion coefficients with theory is shown in Fig. 6. Theoretical curves are

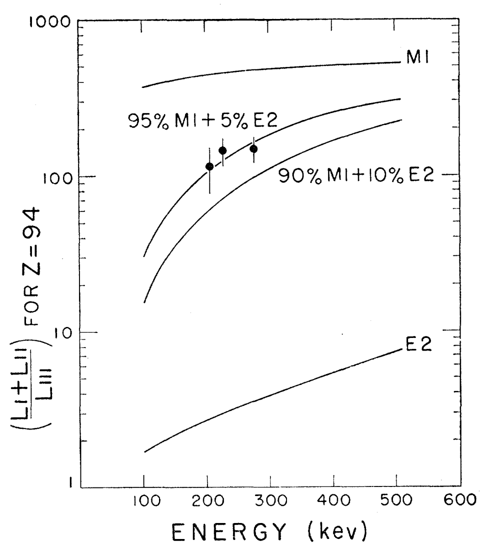

FIG. 5. Comparison of experimental $(L_{\text{I}}+L_{\text{II}})/L_{\text{III}}$ conversion ratio with theoretical values for $M1$ and $E2$ multipolarities calculated on the basis of Rose's L-conversion coefficients.

given for pure $M1$ and for a 10% $E2$ admixture. The measurements lie well below the theoretical predictions of Rose.

In addition to the above measurements, K-conversion coefficients were measured for the 255-, 285-, 316-, and 334-kev transitions. From the measured conversion coefficients, the multipolarity of the 285-kev γ ray is interpreted as $E2$, of the 316- and 334-kev γ rays as $E1$ and of the 255-kev γ ray as an $M1+E2$ admixture on the basis of either theory. The experimental precision here is not sufficient to choose definitely between the theories.

The measurements of the L_{I} and L_{II} conversion coefficients of the 106.4-kev γ ray are $\alpha_{L\text{I}}=0.062 \pm 0.007$ and $\alpha_{L\text{II}}=0.071\pm0.007$. This γ ray would be interpreted as $E1$ with some admixture of $M2$ on the basis of the L_{I} and L_{II} conversion coefficient calculations of Rose. However, if one assumes sufficient $M2$ admixture to agree with the measured L_{I} conversion coefficient, the expected value for $\alpha_{L\text{II}}$ would be 0.026, which is considerably lower than that observed. Thus it is clear that Rose's point-nucleus calculations do not account for the observed conversion coefficients.

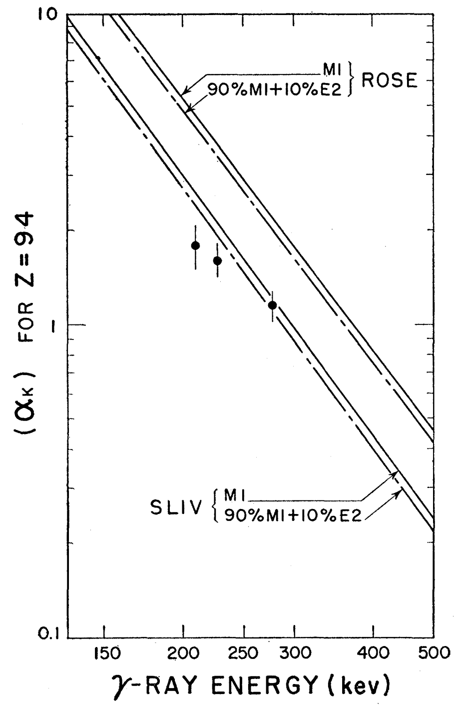

FIG. 6. Comparison of experimental conversion coefficients with theoretical values of Rose and Sliv for pure $M1$ γ rays and a 90% $M1$+10% $E2$ mixture.

The results of the present experiments show that K-conversion coefficients of $M1$ γ rays of energies 209.7, 228.2, and 277.9 kev at $Z=94$ are considerably lower than those calculated by Rose assuming a point nucleus. The measurements lie closer to the K-conversion calculations of Sliv. In addition, the K-conversion measurements of the 209.7- and 228.2-kev γ rays, even allowing for 10% $E2$ admixture, appear to be smaller than the predictions of Sliv. This suggests that conversion coefficients, as well as depending on the finite nuclear size, may well be model-dependent, as postulated by Weneser and Church.[9]

However, admixtures of $E0$ cannot account for both the observed $(L_{\text{I}}+L_{\text{II}})/L_{\text{III}}$ ratio and the observed K-conversion coefficient of the 228.2-kev γ ray if one assumes Rose's theory to be correct.

Compared to $E2$, admixtures of higher multipole orders are improbable. In addition, the $(L_{\text{I}}+L_{\text{II}})/L_{\text{III}}$ experimental ratio places such a limit on the admixture of multipoles higher than $E2$, that it could not significantly influence the $M1$ K-conversion coefficients.

[8] E. L. Church and J. Weneser, Phys. Rev. **103**, 1035 (1956).

[9] J. Weneser and E. L. Church, Phys. Rev. **104**, 1383 (1956).