

APPENDIX. ANALYTIC EXPRESSIONS OF INTEGRALS USED IN THE TEXT

A straightforward integration gives the following expression for the normalization constant  $N$ :

$$N = -\frac{1}{2} \left( \frac{E\sqrt{3}}{\pi} \right)^{\frac{1}{2}} \left( \frac{b^2 + 10b + 1}{3b(b-1)^4} - \frac{2(b+1)\ln b}{(b-1)^5} \right)^{-\frac{1}{2}}$$

The overlap integral  $A(\mathbf{U})$  defined by (23) is equal to the following expression:

$$\begin{aligned} A(\mathbf{U}) &= (32\pi^3 N^2 / 3\sqrt{3} E^3 (b-1)^4) \\ &\times \left[ \frac{(b-1)^2}{12v} \frac{(v + \frac{2}{3}(b-1))(b-1)^2 \ln b}{8v^2} \right. \\ &+ \frac{v+4}{12} \left( 1 + \frac{4}{v} \right)^{\frac{1}{2}} \tanh^{-1} \left( 1 + \frac{4}{v} \right)^{-\frac{1}{2}} \\ &+ \left( \frac{v+4b}{12} - \frac{1}{2}(b-1) - \frac{b(b-1)^2}{v(v+4b)} \right) \left( 1 + \frac{4b}{v} \right)^{\frac{1}{2}} \\ &\left. \times \tanh^{-1} \left( 1 + \frac{4b}{v} \right)^{-\frac{1}{2}} + \frac{(2(b-1)^2 + (b-5)v - v^2)}{12v^2} \right] \end{aligned}$$

$$\begin{aligned} &\times (v^2 + 2(b+1)v + (b-1)^2)^{\frac{1}{2}} \\ &\left. \times \tanh^{-1} \frac{(v^2 + 2(b+1)v + (b-1)^2)^{\frac{1}{2}}}{v+b+1} \right] \end{aligned}$$

where  $v = \mathbf{U}^2/E$ .

Finally  $B(\mathbf{P}, \mathbf{Q})$  defined by (20) is equal to

$$\begin{aligned} B(\mathbf{P}, \mathbf{Q}) &= (2NN_a\pi^2/E^2(b-1)^2) \\ &\times \left[ \frac{1}{Q} \left( \tan^{-1} \frac{(\eta-\epsilon)Q}{(\gamma+\eta)(\gamma+\epsilon)+Q^2} - \tan^{-1} \frac{(\eta-\epsilon)Q}{(\alpha+\eta)(\alpha+\epsilon)+Q^2} \right) \right. \\ &\left. + \frac{(b-1)E}{2\eta} \left( \frac{1}{(\alpha+\eta)^2+Q^2} - \frac{1}{(\gamma+\eta)^2+Q^2} \right) \right] \end{aligned}$$

where

$$\eta = (bE + \frac{3}{4}P^2)^{\frac{1}{2}}, \quad \epsilon = (E + \frac{3}{4}P^2)^{\frac{1}{2}}$$

In particular, when  $Q=0$ ,

$$\begin{aligned} B(\mathbf{P}, 0) &= (2NN_a\pi^2/E^2(b-1)^2) \\ &\times \left[ (\eta-\epsilon)((\gamma+\eta)^{-1}(\gamma+\epsilon)^{-1} - (\alpha+\eta)^{-1}(\alpha+\epsilon)^{-1}) \right. \\ &\left. + ((b-1)E/2\eta)((\alpha+\eta)^{-2} - (\gamma+\eta)^{-2}) \right] \end{aligned}$$

### Internal Conversion of Gamma-Ray Transitions in the $L$ -Subshells\*†

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(Received April 11, 1952)

A number of internally converted  $\gamma$ -transitions have been investigated with high resolution  $\beta$ -spectrographs and the relative intensities of conversion electron lines from the three  $L$ -subshells obtained. Generalizations are made for  $L$ -conversion as related to multipole order. The magnetic transitions investigated are converted in the  $L_I$ - and  $L_{III}$ -shells, the ratio  $L_{III}/L_I$  increasing with increasing multipole order. Less can be said about electric transitions; but  $L$ -conversion takes place mainly in the  $L_{II}$ - and  $L_{III}$ -shells for the transitions studied.

#### INTRODUCTION

THE improved resolution with which internal conversion electron spectra are being examined and the eventual availability of accurate theoretical values for the  $L$ -subshell conversion coefficients make it of interest to determine what can be learned about  $\gamma$ -ray transitions from  $L$ -subshell internal conversion electrons. There are cases where a  $K/L$  ratio or  $K$ -shell internal conversion coefficient is not obtainable with sufficient accuracy or may not give a unique assignment of multipole order by comparison with the theoretical values.<sup>1</sup> Particularly, if the  $\gamma$ -transition is of too

low an energy to convert in the  $K$ -shell, some other criterion would be needed.

In the medium and heavy elements it is usually not difficult to compare the intensities of the electron lines from the internal conversion of the  $L_I$ -,  $L_{II}$ -, and  $L_{III}$ -shells. In certain cases, even electrons from different  $M$ -subshells have been resolved by us. In the course of experiments, data have been obtained for a number of internally converted transitions which have been classified as to multipole order by the analysis of Goldhaber and Sunyar.<sup>2</sup> It has become apparent that the relative conversion in the various subshells indeed depends upon the multipolarity of the  $\gamma$ -transition.

Comparison may be made with certain available theoretical calculations, in addition to the  $K$ -shell values of Rose *et al.*<sup>1</sup> for energies above 150 kev. Gell-

\* A preliminary report of this work was given at the Chicago American Physical Society meeting in October, 1951, J. W. Mihelich and E. L. Church, Phys. Rev. **85**, 733 (1952).

† Research carried out at Brookhaven National Laboratory under the auspices of the AEC.

<sup>1</sup>Rose, Goertzel, Harr, Spinrad, and Strong, Phys. Rev. **83**, 79 (1951).

<sup>2</sup>M. Goldhaber and A. W. Sunyar Phys. Rev. **83**, 906 (1951).

man, Griffith, and Stanley<sup>3</sup> have calculated conversion coefficients (neglecting screening) for the  $L_I$ -shell for  $Z=92, 84,$  and  $49$  for electric and magnetic dipoles and electric quadrupole. Reitz<sup>4</sup> has calculated  $K$ -shell conversion coefficients for the same multipole orders and same energy range ( $k \sim 0.1$  to  $k \sim 0.5$ ). It is convenient to compare these two sets of calculations. In order to utilize these data, we have made interpolations of  $[\log \alpha$  (or  $\log \beta$ ) vs  $Z$ ]. On this semilog plot, the points do not fall far from a straight line for both  $K$ - and  $L_I$ -shells for the  $M1$  and  $E1$  cases. For the  $E2$  case, the points fall on a sharply curving line and here the interpolated values are very approximate. Comparisons were made of the ratio of the  $K$ -conversion coefficient of Reitz divided by the  $L_I$ -conversion coefficient of Gellman *et al.* with the experimental  $K/L_{\text{total}}^{2,5}$  ratio for various multipole orders. For magnetic dipole transitions, the agreement is good implying that the  $L_I$ -coefficients of Gellman *et al.* are reasonably good if there is not much conversion in the  $L_{II}$ - and  $L_{III}$ -shells. The latter point is in accord with experiment (see below).

For electric dipole transitions, the theoretical  $K/L_I$  ratio varies between 6 and 9 for atomic numbers between  $Z=40$  and  $Z=80$  and transition energies of  $k=0.1$  to  $k=0.5$ . As yet, there is no well-established electric dipole transition with which to compare these ratios. Hulme<sup>6</sup> has made calculations for an electric dipole transition of 362 keV and finds that  $K/L_{\text{tot}} \sim 7$  and the ratio of  $L_I:L_{II}:L_{III}$ -conversion is 1:0.0086:0.049. It is of interest to note that for the 47-keV transition in RaD, Cranberg<sup>7</sup> finds the  $L_I:L_{II}:L_{III}$ -ratio to be 1.0:0.09:0.019. However, Cranberg gives the  $L$ -conversion coefficient as 16 by comparing the number of  $L$ -electrons to the total number of disintegrations. By referring to the coefficients of Gellman *et al.* the values of the  $L_I$ -conversion coefficient for magnetic dipole and electric dipole are  $>10.0$  and  $\sim 0.25$ , respectively. From these data, a magnetic dipole is definitely indicated.

For electric quadrupole transitions  $K/L_I$  theor is much larger than the experimental  $K/L_{\text{total}}$ , requiring a large amount of conversion in the  $L_{II}$ - and/or  $L_{III}$ -shells to be consistent. This is in agreement with our experiments on  $L_I:L_{II}:L_{III}$ -ratios. In addition, Goodrich, in a private communication to Bowe and Axel,<sup>8</sup> finds that when the formulas of Hebb and Nelson<sup>9</sup> are separated to show the  $L_I$ -contribution explicitly, an agreement within a factor of two with the value of Gellman is obtained.

Tralli and Lowen<sup>10</sup> have published results of calcula-

<sup>3</sup> Gellman, Griffith, and Stanley, Phys. Rev. **80**, 866 (1950).

<sup>4</sup> J. R. Reitz, Phys. Rev. **77**, 10 (1950).

<sup>5</sup> Nuclear Data, National Bureau of Standards Circular No. 499 (1950).

<sup>6</sup> H. R. Hulme, Proc. Roy. Soc. (London) **A138**, 643 (1932).

<sup>7</sup> L. Cranberg, Phys. Rev. **77**, 155 (1950).

<sup>8</sup> J. C. Bowe and P. Axel, Phys. Rev. **84**, 939 (1951).

<sup>9</sup> M. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).

<sup>10</sup> N. Tralli and I. S. Lowen, Phys. Rev. **76**, 1541 (1949).

TABLE I. Comparison of theoretical ( $K/L_I$ ) ratios with experimental ( $K/L_{\text{tot}}$ ) ratios.

Multipole order	Nucleus	$E$ (keV)	$(K/L_I)$ theor	$(K/L_{\text{tot}})$ exp
$E1$		none known	6-9	...
$M1$	Xe <sup>131</sup>	80	6.7	7.0
	Tc <sup>99</sup>	140	$\sim 8.0$	7.3
	Te <sup>121</sup>	213	6.5	7.3
	Te <sup>123</sup>	159	7.7	7.6, 8.2
	Te <sup>125</sup>	35.4	$\sim 7.5$	7.3
	Hg <sup>199</sup>	209	6.2	5.4
	Cd <sup>111</sup>	247	$\sim 8$	5.12
$E2$	Er <sup>166</sup>	80.8	$\sim 5$	0.10
	Yb <sup>170</sup>	84.8	$\sim 4.5$	0.14
	Ta <sup>181</sup>	134	8.0	0.5
	Os <sup>186</sup>	137	7.5	0.6
	Hg <sup>199</sup>	159	7.5	0.6
	Pb <sup>204</sup>	374	$\sim 3.2$	2.0

tions estimating the ratio of conversion in the  $L_{III}$ - and  $L_I$ -shells for magnetic multipole orders one to five.  $L_{II}$ -conversion is said to be less than 5 percent of the other two. In short, their curves indicate that the ratio  $\beta_{L_{III}}/\beta_{L_I}$  increases with  $Z^2/E$  and increasing multipole order. Our experiments are in accord with this.

Table I lists some comparisons between available theoretical data and experimental values for  $E1$ ,  $M1$ , and  $E2$  transitions.

#### EXPERIMENTAL DATA

Permanent magnet photographic spectrographs of 20-cm maximum radius of curvature using x-ray film as a detector were employed. An instrument of this type is ideal for an investigation of this sort, due to the high resolution available, the integrating action of the film, and the permanence of the magnetic field. Magnets of various field strengths were employed, depending on the energy of the conversion electrons. For electrons of  $<90$  keV, a 54-gauss magnet was used, giving an energy dispersion of  $\sim 0.4$  keV/mm. The magnets were calibrated against the conversion electron lines of I<sup>131</sup>, Au<sup>198</sup>,<sup>12</sup> and W<sup>187</sup>.<sup>13</sup> The group of  $L$ -lines lie close together on the film; hence their relative energies may be determined with considerably more accuracy than their absolute energies. The magnitudes of the differences of the  $L$ -shell energy levels may be seen by referring to Fig. 1, where the x-ray absorption edge energies in keV are plotted against  $Z$ . These values are obtained from Siegbahn's book<sup>14</sup> by converting  $\nu/R$  values to keV using up to date values of physical constants.<sup>15</sup> One sees that in the rare earth region, the  $L_I-L_{II}$  and  $L_{II}-L_{III}$  separations are about the same; this makes the

<sup>11</sup> Following the custom of abbreviation for  $\alpha_K/\alpha_L$ , etc., ratios, we shall designate the ratio of conversion coefficients in two subshells as  $L_I/L_{III}$ , etc.

<sup>12</sup> Lind, Brown, Klein, Muller, and DuMond, Phys. Rev. **75**, 1544 (1949).

<sup>13</sup> J. W. M. DuMond, 16th Quarterly Report California Institute of Technology NP 3142 (1951).

<sup>14</sup> M. Siegbahn, *Spektroskopie der Roentgenstrahlen* (Julius Springer, Berlin, 1931).

<sup>15</sup> Hill, Church, and Mihelich, *A Table of Critical X-Ray Absorption Energies*, privately circulated.

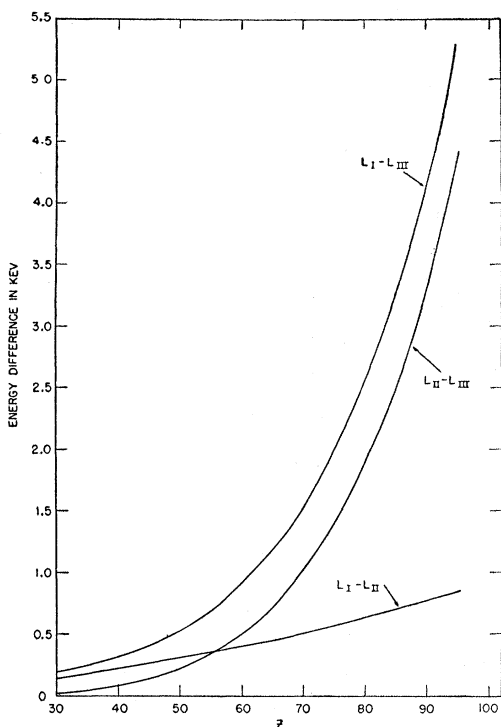


FIG. 1. Differences between  $L$  subshell x-ray critical absorption energies vs atomic number. The points on the curve correspond to the energy separation of conversion electrons from the designated shells.

assignment of a doublet  $L$ -line as  $L_I L_{III}$  or  $L_{II} L_{III}$  simple. In the region of high  $Z$ , where the  $L_I-L_{II}$  difference is small compared to  $L_{II}-L_{III}$  or  $L_I-L_{III}$ , there might be cases where a doublet could not uniquely be described as  $L_I L_{III}$  or  $L_{II} L_{III}$ , if an instrument of marginal resolving power was being employed.

The position of the lines relative to one another can be determined to within  $\pm 0.15$  mm on a good photographic plate, which means an energy difference uncertainty (in a 54-gauss magnet) of the order of 0.06 kev. The consistency of the energy sums obtained shows that this accuracy is indeed obtained. In addition, the small difference in energy means that the response of the film is essentially linear over the  $L$ -electron group. In addition, source self-absorption effects are of the same magnitude for the different  $L$ -lines. Hence, photometric densitometry allows a measure of the electron line intensities. In certain cases, where  $Z$  is not high and the energy of the transition is large enough to require a higher field magnet, clear resolution of the lines is not obtained; however, a study of the line shape will often allow one to decide whether the  $L$ -line is complex and if so, whether or not it is an  $L_I L_{III}$  or  $L_{II} L_{III}$  doublet.

Sources generally were a thin layer of fine radioactive powder (activated in the Brookhaven reactor) on a Scotch tape strip about 15 mm  $\times$  0.4 mm. The sources had an average density of the order of 1.0 mg/cm<sup>2</sup>.

The following is a discussion of the data obtained on the  $L$ -conversion of certain transitions of various multipole orders which are believed to be well established.

### M1

For the several cases investigated (as well as those published by others), it is apparent that the  $L_I$ -conversion electron lines are far more intense than the other two. No  $L_{II}$ - or  $L_{III}$ -electrons were observed in any of these cases, and a conservative upper limit of  $L$ -conversion for  $L_{II}$  and  $L_{III}$  compared to  $L_I$  is 10 percent. Table II lists the data for the  $M1$  (as well as the other multipole orders).<sup>‡</sup>

Caution is required in the assigning of an  $L$ -line where only one is visible, as the  $K-L$  energy is large enough that errors due to small inaccuracies in field calibration and correction for film expansion may be sufficient to make a definite assignment as either  $L_I$ - or  $L_{II}$ -electrons uncertain.

### M2

No  $L$ -conversion data have been obtained yet for this multipole transition. Of interest is the 150-kev transition in  $\text{Lu}^{177}$  following the 1.5-hr  $\beta^-$  activity of  $\text{Yb}^{177}$ . McGowan<sup>16</sup> has obtained a  $K/L$  ratio of 3 using a scintillation spectrometer; this datum along with the lifetime<sup>16</sup> of  $1.3 \times 10^{-7}$  makes the assignment of  $M2$  likely. The activity obtained by thermal neutron ac-

TABLE II. Experimental intensities of  $L$ -conversion lines for various multipole order transitions.

Multi-pole order	Nucleus	Magnetic transitions		Remarks
		$E$ (kev)	$L_I/L_{III}$	
M1	$\text{Xe}^{131}$	80	large ( $>10$ )	$L_{II}$ and $L_{III}$ not observed
	$\text{Tc}^{99}$	140	large ( $>10$ )	$L_{II}$ and $L_{III}$ not observed
	$\text{Sn}^{119}$	24		$L_{II}$ and $L_{III}$ not observed
	$\text{Te}^{123}$	159	large ( $>10$ )	$L_{II}$ and $L_{III}$ not observed
M3	$\text{Br}^{80m}$	49	1.0	
M4	$\text{Te}^{123m}$	89	0.5	$L_{II}$ if any, of low intensity
	$\text{Te}^{125m}$	109	$\sim 1$ (estimated)	
	$\text{Sn}^{117m}$	155	$\sim 1$	
Multi-pole order	Nucleus	Electric transitions		Remarks
		$E$ (kev)	$L_I:L_{II}:L_{III}$	
E1	See text			
E2	$\text{Er}^{168}$	80.8	$\approx 0.1:0.7:1$	$L_I$ intensity estimated
	$\text{Yb}^{170}$	84.8	$\sim 0.1:0.8:1$	$L_I$ intensity estimated
	$\text{Hg}^{198}$	411	2.5:1	
	$\text{Hg}^{199}$	159	$\sim 1.6:1$	
E3	$\text{Dy}^{165m}$	109	$\sim 1.5:1$	Reinterpretation of Caldwell's data
	$\text{Cs}^{134m}$	127.6	$\sim 1:1$	Unresolved. Rough estimate of $L_{II}/L_{III}$ ratio
E4	$\text{In}^{114m}$	191.6	...	Probably $L_{II}$ and $L_{III}$ but unresolved. Also, possibly weak $L_I$

<sup>‡</sup> Note added in proof: Recent theoretical results of Gellman, Griffith, and Stanley (see reference 29) extended to the  $L_{II}$  and  $L_{III}$  shells obtain the following  $L_I:L_{II}:L_{III}$  ratio for an  $M1$  transition ( $k=0.23$ ,  $Z=84$ ): 1:0.081:0.0018. In addition, comparison may be made of  $K/L_{tot}$  ratios as obtained from the data of Reitz and Gellman *et al.* with the experimental values. For high atomic number, the theoretical  $K/L$  ratio for  $M1$  transitions is about 4 or 5. Experimental data for the 286-kev transition in  $\text{Tl}^{203}$  indicate a  $K/L$  ratio of 4.8 for an  $M1$  transition (see reference 2).

<sup>16</sup> F. K. McGowan, Oak Ridge National Laboratory Report No. 952 (1951), p. 104.

tivation is not high; attempts to obtain a measurable spectrum by inserting several fresh sources through a vacuum lock and integrating the exposure on a single photographic plate have so far been unsuccessful.

The number of examples in which the  $L$ -lines may be resolved is restricted by the energy resolution requirement that the transition energy not be too great compared to the shell excitation energies.

### M3

A transition believed to be  $M3$  is the 49-keV transition in the decay of  $\text{Br}^{80m}$ . A source was made from neutron irradiated potassium bromate by a Szilard-Chalmers separation and the conversion spectrum obtained.<sup>17</sup> The  $L$ -conversion is in the  $L_I$ - and  $L_{III}$ -shells, and the  $L_I/L_{III}$  ratio is  $1.0 \pm 0.25$ . The only other  $M3$  transition for which  $L$ -lines have been resolved is that in the 19-sec  $\text{Hf}^{179m}$  (161 keV). Keller and Burson<sup>18</sup> report two  $L$ -lines which are either an  $L_I L_{III}$  or  $L_{II} L_{III}$  pair.

### M4

In agreement with Tralli and Lowen<sup>10</sup> who predict an increasing amount of  $L_{III}$ -conversion relative to  $L_I$  for increasing magnetic multipole order,  $M4$  transitions are  $L$ -converted in the  $L_I$ - and  $L_{III}$ -shells. Several  $M4$  spectra have been obtained with the  $L_I$ - and the  $L_{III}$ -lines resolved. In the case of the  $\text{Te}^{123}$  (88 keV) transition the  $L_{III}/L_I$  ratio is  $2.0 \pm 0.3$ ; for this value of  $Z^2/E$  the Tralli and Lowen graph gives  $\sim 1.8$ . Figure 2 is the graph of Tralli and Lowen with our experimental points on magnetic transitions. It is remarkable that the agreement is so good since the Pauli approximation they employed is known to be poor at the origin (nucleus).

### E1

As stated previously, no experimental data on  $L$ -shell conversion are yet available for this multipole order.

### E2

Electric quadrupole transitions are relatively common and considerable data have been obtained. As stated above, available theoretical estimates suggest that the  $L$ -conversion is mainly in the  $L_{II}$ - and  $L_{III}$ -shells. For several low energy ( $\sim 85$  keV) transitions in the rare earths ( $Z \sim 68$ ),<sup>19</sup> the ratio of the intensities of the  $L_{II}$ - and  $L_{III}$ -lines is about 0.8. The  $E2$  159-keV  $\gamma$ -ray in  $\text{Hg}^{199}$  has an  $L_{II}/L_{III}$  ratio of 1.6 while the 411-keV transition in  $\text{Hg}^{198}$  has an  $L_{II}/L_{III}$  ratio of 2.5.<sup>20</sup> If one considers the  $K/L_{\text{total}}$  ratios for these two

<sup>17</sup> Thanks are due Dr. Joan Welker of the Brookhaven Chemistry Department for performing the Szilard-Chalmers separation.

<sup>18</sup> H. Keller and S. B. Burson, Phys. Rev. **83**, 62 (1951).  
<sup>19</sup> J. W. Mihelich and E. L. Church, Phys. Rev. **85**, 690 (1952); J. W. Mihelich, unpublished.

<sup>20</sup> R. D. Hill and J. W. Mihelich, Phys. Rev. **79**, 275 (1950). Here the  $L$ -lines of both transitions had been designated as  $L_I$  and  $L_{III}$ . However, the author has remeasured the 159- $L$  con-

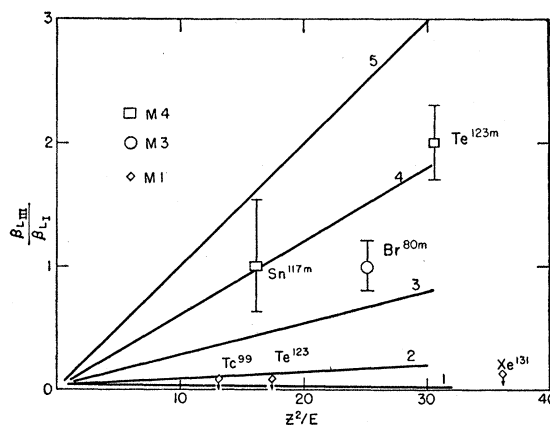


Fig. 2. The theoretical values of Tralli and Lowen for the ratio of conversion coefficients in the  $L_{III}$ - and  $L_I$ -shells for magnetic transitions of various multipole order. Their calculations were made in the Pauli approximation and are valid for low  $Z$  and low energy. Our experimental  $L_{III}/L_I$  ratios are shown for comparison.

transitions, along with their  $K$ -conversion coefficients and their  $L_{II}/L_{III}$  ratios one sees that the conversion coefficient of the  $L_{II}$ -shell decreases less rapidly with increasing transition energy than does that for the  $L_{III}$ -shell.

Only an estimate is possible for the  $L_I$ -conversion. For the low energy transitions  $\text{Er}^{166}$  and  $\text{Yb}^{170}$ , a rough value of the intensity of the  $L_I$ -line as compared to the  $L_{II}$  is about or less than 0.1.<sup>19</sup>

It must be pointed out that these results are for  $E2$  transitions occurring in high  $Z$  nuclei ( $Z > 65$ ).§

### E3

Data on  $E3$  transitions are meager, only isolated cases having been studied with sufficient resolution. One of the first was that of the 109-keV (1.25 min) isomeric transition in  $\text{Dy}^{166m}$  which was examined by Caldwell.<sup>21</sup> He was able to distinguish two  $L$ -lines which he labeled  $L_I$  and  $L_{III}$  with an intensity ratio of 1.5. However, if one adds x-ray absorption energies

version lines under better conditions and finds that they are  $L_{II}$  and  $L_{III}$ . The 411- $L$  lines could not be sufficiently resolved. However,  $L_{II}-L_{III}$  conversion electrons are assumed since this is a well established  $E2$  transition and all evidence indicates  $L_{II}-L_{III}$  conversion for this multipole order.

§ Note added in proof: The theoretical  $L$  ratio (see reference 29) for an  $E1$  transition ( $k=0.23$  and  $Z=84$ ) is:

$$1:0.41:0.395.$$

For  $E2$  transitions, when the energy is low ( $k < 0.25$ ) and atomic number large ( $Z > 60$ ), the theoretical values indicate that  $L_I$  conversion is less than 10 percent of  $L_{II}$  or  $L_{III}$  conversion. For low  $Z$  and large energy,  $L_I$  conversion becomes important. For  $Z=49$  and  $k=0.2$ , the conversion coefficients for the three  $L$ -shells is about equal ( $\sim 0.1$ ). For  $Z=49$  and  $k=0.4$ , the  $L$  ratio is:

$$1:0.4:0.45.$$

In Table I where comparison was made of the theoretical  $K/L_I$  ratio with the experimental  $K/L$  ratio, the values for  $\text{Cd}^{111}$  were  $\sim 8$  and 5.12, respectively. This was close agreement as compared with the case for  $E2$  transitions in heavy elements. The results of Gellman *et al.* explain this behavior. The theoretical  $K/L$  ratio is  $\sim 4.5$ .

<sup>21</sup> R. L. Caldwell, Phys. Rev. **78**, 407 (1950).

TABLE III. Estimate of  $L$ -subshell conversion coefficients for certain  $E2$  transitions.

$\gamma$ -transition	$(K/L)$	$\left(\frac{L_{II}}{L_{III}}\right)$	$\alpha_K$	$\alpha_L$ total	$\alpha_{L_I}$	$\alpha_{L_{II}}$	$\alpha_{L_{III}}$
84.8 keV (Yb <sup>170</sup> )	0.15 <sup>a,b</sup>	0.8 <sup>b</sup>	1.4 <sup>c,d</sup>	9.3	~0.3	4.1	5.1
159 keV (Hg <sup>199</sup> )	0.8 <sup>a</sup>	1.6 <sup>a</sup>	0.03 <sup>f</sup>	0.4	~0.04	0.246	0.154
411 keV (Hg <sup>198</sup> )	3.0 <sup>a</sup>	2.5 <sup>a</sup>	0.01 <sup>f</sup>	0.01	~0.001	0.0072	0.0029

<sup>a</sup> See reference 5.  
<sup>b</sup> See reference 19.  
<sup>c</sup> See reference 3.  
<sup>d</sup> See reference 4.  
<sup>e</sup> See reference 20.  
<sup>f</sup> See reference 1.

believed to be the best available at the present time,<sup>15</sup> the assignment  $L_{II}$  and  $L_{III}$  is preferable.

We have examined the (127.6±0.3 keV)  $E3$  transition in Cs<sup>134m</sup> and believe that the  $L$ -line is complex and, although complete resolution was not possible, that it is an  $L_{II}L_{III}$  pair. The intensities of the two lines are of the same order of magnitude.

The 57.3-keV transition in Ir<sup>192m</sup> (1.5 minutes) is believed to be either  $E3$  or  $M3$ .<sup>2</sup> Caldwell<sup>21</sup> observed two  $L$ -conversion lines and reported them as  $L_I$  and  $L_{III}$ . However, an analysis of Caldwell's data, obtained with a thick source, shows that the energy difference of these two lines is not sufficiently well measured to definitely indicate whether the pair is  $L_I$  and  $L_{III}$  or  $L_{II}$  and  $L_{III}$ . The experimental energy difference is 1.9 keV; x-ray absorption energy differences for  $L_I$ - $L_{III}$  and  $L_{II}$ - $L_{III}$  are 2.2 and 1.6 keV, respectively. Goldhaber, Muehlhause, and Turkel<sup>22</sup> have shown by critical absorption the existence of  $L$  x-rays arising from the filling of holes in the  $L_{III}$ -shell. A clear-cut decision as to whether the lower energy  $L$ -line is  $L_I$  or  $L_{II}$  should indicate whether the transition is  $M3$  or  $E3$ , respectively.

#### E4

One case has been examined, that of In<sup>114m</sup>,<sup>23</sup> which has been well established as an  $E4$ . The low atomic number and relatively high energy (191.5±0.5 keV) make a clear-cut decision difficult. It is believed that  $L$ -conversion takes place mainly in the  $L_{II}$ - and  $L_{III}$ -shells.

The internal conversion spectrum consists of three lines:  $K$ ,  $L_{III}$ , and  $M_V$ ; the experimental energy difference between the  $K$ - and  $L$ -lines is 24.33 keV. The energy difference expected if the  $L$ -line were  $L_{II}$  or  $L_{III}$  would be 24.00 or 24.20 keV, respectively. The  $L_{III}$  assignment is favored.

#### $M1 + E2$ Mixtures

As pointed out by Goldhaber and Sunyar,<sup>2</sup> the only mixtures expected are those of  $E2$  and  $M1$ . We have investigated certain transitions including the 70- and 104-keV  $\gamma$ -transitions in Eu<sup>153</sup> following the  $\beta$ -decay of Sm<sup>153</sup> (47 hr). Here the  $K/L$  ratios which we obtain

<sup>22</sup> Goldhaber, Muehlhause, and Turkel, Phys. Rev. **71**, 372 (1947).

<sup>23</sup> R. M. Steffen, Phys. Rev. **83**, 166 (1951).

( $K/L_{70}=3.5^{+2.5}_{-1.0}$ ;  $K/L_{104}=6.5\pm 1.0$ ) are lower than would be expected for magnetic dipoles. Lifetime considerations rule out  $M2$  or  $E3$  multipole orders. The  $L$ -electron spectra consist of  $L_I$ ,  $L_{II}$ ,  $L_{III}$ -lines in the intensity ratios 1, ~0.1, ~0.1, respectively. The presence of the  $L_{II}$ - and  $L_{III}$ -electrons to this amount may indicate an admixture of  $E2$ . It is possible that accurate determinations of the  $L$ -line intensities may be of aid in deciding proportions of such mixtures. This could be another criterion for studying these interesting mixtures, in conjunction with  $K/L$  ratios,  $K$ -conversion coefficients, and angular correlation data, all of which are more or less sensitive to the degree of mixture.

#### DISCUSSION

Admittedly, the data presented are not complete in the disclosure of trends with energy and atomic number. What has been shown is the fact that here is a means of identifying or confirming multipole assignments.

In general, it appears that for magnetic multipoles, conversion takes place in  $L_I$ - and  $L_{III}$ -shells with the  $L_I/L_{III}$  ratio decreasing with increasing multipole order. For electric transitions where  $Z > 65$ , the  $L_{II}$ - and  $L_{III}$ -shells are preferred. Not enough data is available to predict any variation of the  $L_{II}/L_{III}$  ratio with multipole order. For the two  $E2$  transitions in Hg, it was pointed out that the  $L_{II}/L_{III}$  ratio decreased with decreasing energy.

If one takes the  $K/L_{total}$  ratios, the theoretical  $K$ -conversion coefficients, and our  $L$ -ratios, an estimate may be made of the absolute conversion coefficient of the  $L$ -subshells for certain multipole orders. For electric quadrupole transitions, values may be obtained for the  $L_{II}$ - and  $L_{III}$ -shells, and comparison made with the  $L_I$ -shell values of Gellman *et al.* These data for transitions of various energies are shown in Table III. These  $L_I$ -shell values are very approximate due to the difficulty of interpolation between  $Z$  values. However, the relative magnitudes of the coefficients of the three shells is consistent with the experimental data. ¶

The two transitions in Hg are of particular interest since they are of quite different energy but occur in nuclei of the same atomic number. For these two points

TABLE IV. Estimates of  $L$ -subshell conversion coefficients for  $M3$  and  $M4$  transitions.  $L_{II}$ -conversion is assumed to be negligible.

Multipole order	$\gamma$ transition	$(K/L)$	$\left(\frac{L_I}{L_{III}}\right)$	$\beta_K$	$\beta_{total}$	$\beta_{L_I}$	$\beta_{L_{III}}$
$M3$	49 keV (Br <sup>80m</sup> )	5.3 <sup>a</sup>	1.0	100 <sup>b</sup>	~20	~10	~10
$M4$	88.5 (Te <sup>120m</sup> )	0.68 <sup>a</sup>	0.5	620 <sup>b</sup>	910	303	607

<sup>a</sup> See reference 5.  
<sup>b</sup> See reference 2.

¶ Note added in proof:—The possibility of these transitions being  $E1$  may not be definitely excluded.

¶ Note added in proof:—Interpolated values taken from the results of Gellman *et al.* (see reference 29) for  $\alpha^2_{L_{II}}$  and  $\alpha^2_{L_{III}}$ , respectively, are: (84.8 keV) 1.35 and 1.35; (159 keV) 0.36 and 0.25. The experimental values appear to be larger than the theoretical coefficients for which screening had not been taken into account.

it would appear that conversion coefficients for all three  $L$ -shells fall off with increasing energy. One should expect this behavior to be rather regular.

For magnetic transitions,  $M3$  and  $M4$ , estimates may be made for the coefficient for the  $L_I$ - and  $L_{III}$ -shells (Table IV). For  $M1$  transitions, at least for the energies and atomic number of nuclei investigated,  $L$ -conversion is predominantly  $L_I$ .

Obviously, when a nucleus undergoes a transition of a given multipolarity, the probability of a given  $L$ -orbital electron being ejected is strongly dependent upon the  $l$  and  $j$  values of the electron since the two  $L_I$ -electrons are  $s_{\frac{1}{2}}$ , the two in the  $L_{II}$ -shell are  $p_{\frac{3}{2}}$ , and the four in the  $L_{III}$ -shell are  $p_{\frac{3}{2}}$  electrons.

It is of interest to note that in the case of  $\text{Os}^{191}$  (15-day) there is a 42-keV transition (probably  $E2$ ) which, of course, does not convert in the  $K$ -shell. The  $L$ -conversion occurs in the  $L_{II}$ - and  $L_{III}$ -shells, the ratio  $L_{II}/L_{III}$  being  $\sim 0.8$ . In addition, the following  $M$ -subshell electrons are resolved:  $M_{II}$ ,  $M_{III}$ , and  $M_{IV}$  and/or  $M_V$ . The intensity ratio for these three lines is 0.8:1.0:0.2. The first three  $M$ -subshells have the same " $l$ " and " $j$ " values as the three  $L$ -shells, while  $M_{IV}$  and  $M_V$  are  $d_{\frac{3}{2}}$  and  $d_{\frac{5}{2}}$  electrons.

It is possible to assign multipolarity of transition from data on x-ray emission spectra of radioisotopes, particularly in the very heavy elements, since if a detector of sufficient resolution is employed, the relative number of  $L_I$ ,  $L_{II}$ , and  $L_{III}$  holes may be determined. Barton, Robinson, and Perlman<sup>24</sup> have studied the  $L$  x-ray spectra of certain transuranic radioisotopes. In the case of plutonium x-rays arising from  $L$ -shell vacancies due to internal conversion of a 43-keV transition<sup>25</sup> following the  $\alpha$ -decay of  $\text{Cm}^{242}$ , they observe that transitions to the  $L_{II}$ -level are relatively twice as abundant for the internal conversion spectrum as for the x-ray spectrum to be expected from external electron bombardment, after normalization of the data for  $L_{III}$ -transitions. In addition, certain transitions involving the  $L_I$ -level seen in moderate abundance in the electron bombardment source are missing ( $< 20$  percent). One is not able to tell what the increase in  $L_{III}$ -transitions is for the internal conversion case. If  $L_{II}$ - and  $L_{III}$ -levels have been depleted by the internal conversion process, an electric  $\gamma$ -transition is indicated.

This conclusion is strengthened by the results of Freedman, Jaffey, and Wagner<sup>26</sup> on the  $\beta$ -decay of  $\text{Np}^{238}$ , which leads to excited states of the same  $\text{Pu}^{238}$ . In addition to  $\gamma$ -rays of 1.03 and 0.983 MeV, four conversion lines of low energy are seen. Table V lists their conversion line energies and intensities along with their<sup>26</sup> assignments which indicate three transitions of low energy. Applying extrapolated<sup>15</sup> x-ray absorption en-

TABLE V. Conversion electron lines of  $\text{Pu}^{238}$ . Energies and intensities are from Barton *et al.* (see reference 24).

Electron energy keV	Relative intensity	Previous assignment	Our assignment
20.8	71	43-L	43.1-L <sub>II</sub>
24.7	37	47-L	42.8-L <sub>III</sub>
37.4	27	42-M	43.0-M <sub>II</sub>
41.9	6.0	47-M	43.0-N <sub>III</sub>

ergies, the four lines fit well the assignment  $L_{II}$ ,  $L_{III}$ ,  $M$ , and  $N$  for a 43-keV transition in  $\text{Pu}$ . The existence of  $L_{II}$ - and  $L_{III}$ -conversion in the reported relative intensities and the existence of electron-x-ray coincidences<sup>26</sup> indicate an  $E2$  transition. It would appear that the 43-keV state is reached both by  $\beta$ -decay of  $\text{Np}^{238}$  and  $\alpha$ -decay of  $\text{Cm}^{242}$ . The regularity of  $2^+$  first excited states of even-even nuclei also is in line with the  $E2$  assignment.<sup>27</sup>

One is tempted to postulate the multipolarity of the low energy isomeric transition in  $\text{Am}^{241m}$ . O'Kelley, Barton, Crane, and Perlman<sup>28</sup> have studied the  $L$  x-rays arising from internal conversion and note that radiation due to  $L_{III}$ -holes is five times as prevalent as that due to  $L_{II}$ -holes. In the  $\text{Pu}^{238}$  case, which we postulated as  $E2$ , the  $L_{III}/L_{II}$  ratio was about one-half. Lifetime-energy considerations indicate the isomeric transition in  $\text{Am}^{241m}$  should be  $E3$  or  $M3$ . By comparing the  $L_{II}/L_{III}$  ratio to those of other  $E3$  transitions (of considerably smaller atomic number and greater energy) and extrapolating the Tralli and Lowen graph to a very high value of  $Z^2/E$ , the  $M3$  assignment is favored. For large  $Z^2/E$ ,  $L_I$ -conversion should be small. Possibly for magnetic transitions,  $L_{II}$ -conversion becomes more probable for high atomic number. For  $E3$ , more  $L_{II}$ -conversion than is observed would be expected.

Some further remarks may be made. Interpretation of angular correlation data involving  $L$ -conversion electrons will require knowledge of the kind of electrons being observed, that is, whether they are from the  $p_{\frac{3}{2}}$ ,  $p_{\frac{1}{2}}$ , or  $s_{\frac{1}{2}}$  levels. In addition, the determination of  $\gamma$ -ray transition energies from internal conversion lines may be made more accurately if one knows which  $L$ -subshell absorption energy to add to the energy of the  $L$ -conversion line (or lines).

After this paper was completed, theoretical values for the  $L_{II}$ - and  $L_{III}$ -shell conversion coefficients, neglecting screening, were published by Gellman, Griffith, and Stanley.<sup>29</sup> Our experimental results are in agreement with their theoretical ones.

Thanks are due Dr. M. Goldhaber of Brookhaven and Professor R. D. Hill of the University of Illinois for discussion. Mr. E. L. Church helped with the early phases of the work.

<sup>24</sup> Barton, Robinson, and Perlman, *Phys. Rev.* **81**, 208 (1951).

<sup>25</sup> G. D. O'Kelley, University of California Radiation Laboratory Report No. 1243, May (1951).

<sup>26</sup> Freedman, Jaffey, and Wagner, *Phys. Rev.* **79**, 410 (1950).

<sup>27</sup> G. Scharff-Goldhaber, *Phys. Rev.* **87**, 218 (1952).

<sup>28</sup> O'Kelley, Barton, Crane, and Perlman, *Phys. Rev.* **80**, 293 (1950).

<sup>29</sup> Gellman, Griffith, and Stanley, *Phys. Rev.* **85**, 944 (1952).