INTERNAL CONVERSION IN THE L-SUBSHELLS

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Summary

The relative intensities of conversion-electron lines from the three L-subshells have been determined for a number of γ-transitions, and the results compared with the theoretical values of Gellman, Griffith, and Stanley (1952). Experiment has been shown to be in accord with theory.

Calculations of theoretical K/L ratios indicate that the empirical curves of Goldhaber and Sunyar (1951) may need to be modified, especially for low Z. For a given Z^2/E, the K/L ratio increases with decreasing atomic number.

I. Introduction

Accurate theoretical values of the internal conversion coefficients of electric and magnetic multipole radiation for the L-subshells would be of great assistance in the identification of transitions, and in particular of those which, owing to their low energy, do not convert in the K-shell.

Conversion coefficients for the K-shell have been computed for a wide range of energies, Z-values, and multipolarities by Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) in the relativistic case with the unscreened Coulomb field acting on the electron, and for a more restricted range, but including screening, by Reitz (1950).

Calculations of L-shell conversion coefficients have been made by Hebb and Nelson (1940), Tralli and Lowen (1949), and more recently by Gellman, Griffith, and Stanley (1952). The results of Gellman, Griffith, and Stanley are of particular interest, as they have been computed for the three L-subshells for E1, E2, and M1 radiations, using relativistic wave functions for the atomic electrons but not taking into account the effects of screening. As these results are the most complete at present available, it is of interest to determine the degree of reliability which may be attached to them.

An experimental investigation of the relative L-subshell conversions has been made by Mihelich (1952), whose results in specific cases support those of Gellman, Griffith, and Stanley (1952). It should be pointed out that the latter's calculations indicate certain crossings of the L-subshell conversion curves at particular values of Z and E. In the case of E2 transitions, for example, at high Z the L_I conversion can be more important than the L_{II} conversion. This

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was already pointed out by Mihelich in a note added in proof after Gellman, Griffith, and Stanley's paper had appeared, but nevertheless the generalization for all electric transitions implied by Mihelich in the abstract of his paper has often since been misinterpreted.

In the present paper, results of investigations of several additional transitions are presented, and these are compared with Gellman, Griffith, and Stanley's values, both for relative $L_I : L_{II} : L_{III}$ ratios and for absolute conversion coefficients. In order that a valid comparison may be made, it is desirable that the nature of the transition involved be known beforehand. Several transitions have been selected, for which the L-subshell conversion lines can be clearly resolved, and the identification of the transition has been made independently on the basis of lifetime, K-conversion, and K/L ratio.

II. Identification of Transition Multipolarities

${}^{181}_{73}$Ta : 133 keV

This transition arises in the $2 \cdot 2 \times 10^{-5}$ sec isomeric state of ^{181}Ta following the β-decay of ^{181}Hf, and has been quoted by Goldhaber and Hill (1952) as E2. The theoretical radiation lifetimes τ_γ, calculated by Weisskopf (1951) and expressed in nomograph form by Montalbetti (1952), are $1 \cdot 4 \times 10^{-11}$ sec for M1 and $1 \cdot 4 \times 10^{-7}$ sec for E2. The experimental value, calculated from $\tau_\gamma = T_{\frac{1}{2}}(1+\alpha_{\text{total}})/\ln 2$ by employing a measured total conversion coefficient of $\sim$1 (Hedgran and Thulin 1951 ; Chang-Yun Fan 1952), is $\sim 6 \cdot 5 \times 10^{-5}$ sec, which, although long, is in reasonable agreement with an E2 assignment, and seems to rule out an appreciable M1, E2 mixture. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are $1 \cdot 91$ for M1, $0 \cdot 50$ for E2. Experimental values of $0 \cdot 34$ (Chang-Yun Fan 1952) and $0 \cdot 49$ (Hedgran and Thulin 1951) indicate a pure E2 transition. The experimental K/(L+M) ratio obtained by both these authors is $0 \cdot 61$, in excellent agreement with the empirical ratio of Goldhaber and Sunyar (1951) of $0 \cdot 6$ for E2.

The absence of any M1 component is unexpected in view of the spin assignments quoted by Goldhaber and Hill (1952), but the transition is probably to be identified as pure E2.

${}^{186}_{76}$Os : 137 keV

This transition arises in the 8×10^{-10} sec isomeric state of ^{186}Os following the β-decay of ^{186}Re, and has been quoted by Goldhaber and Hill (1952) as E2. The theoretical radiation lifetimes (Weisskopf 1951 ; Montalbetti 1952) are $1 \cdot 3 \times 10^{-11}$ sec for M1 and 9×10^{-8} sec for E2. The experimental value, employing a measured total conversion coefficient of $\sim$1 (Metzger and Hill 1951 ; Steffen 1951), is $\sim 2 \cdot 3 \times 10^{-9}$ sec, which is in satisfactory agreement with pure E2, though it may indicate some admixture of M1. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are $2 \cdot 17$ for M1, $0 \cdot 43$ for E2. Experimental values of $0 \cdot 35$ (Metzger and Hill 1951) and $0 \cdot 37$ (Steffen 1951) indicate an E2 transition with no M1 admixture. The empirical K/L ratios cannot be used for identification in this case, since the E2 curve has been drawn utilizing this transition. The observed K/L ratio is 0·6 (Metzger and Hill 1951; Steffen 1951). It seems reasonable to identify this transition as pure E2.

$^{191}_{77}$Ir : 129 keV

This transition follows the β-decay of ^{191}Os, and has been quoted by Goldhaber and Hill (1952) as an M1+E2 mixture. No lifetime has been measured. The theoretical radiation lifetimes (Weisskopf 1951; Montalbetti 1952) are $1{\cdot}7\times10^{-11}$ sec for M1 and $1{\cdot}0\times10^{-7}$ sec for E2. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are 3·0 for M1, 0·49 for E2. The experimental value of 1·36 (Kondaiah 1951; Swan and Hill 1952) indicates a 0·42 : 0·58 mixture of M1 : E2 transitions. The empirical K/L ratios of Goldhaber and Sunyar (1951) are 7·5 for M1 and 0·36 for E2. The observed value of 2·2 (Kondaiah 1951; Swan and Hill 1952) indicates a 0·64 : 0·36 mixture of M1 : E2. While the evidence does not agree accurately on the relative M1 : E2 mixture, it is clear that this transition does involve a mixing of M1 and E2 radiations.

$^{198}_{80}$Hg : 411 keV

This transition follows the β-decay of ^{198}Au, and has been identified by Hill and Mihelich (1950) as E2. No lifetime has been measured, though the observations of Graham and Bell (1951), Moon (1951), and Bell, Graham, and Petch (1952) indicate a half-life of $\sim10^{-11}$ sec. The theoretical radiation lifetimes (Weisskopf 1951; Montalbetti 1952) are 5×10^{-13} sec for M1 and 4×10^{-10} sec for E2. The experimental value, employing a total conversion coefficient of $\sim0{\cdot}1$, is $\sim2\times10^{-11}$ sec, which is consistent with pure E2. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are 0·17 for M1, 0·032 for E2. The observed values of 0·04 (Plesset 1942), 0·025 (Peacock and Wilkinson 1948), and 0·03 (Siegbahn and Hedgran 1949) indicate a pure E2 transition. The empirical K/L ratios cannot be used in identification, for the reasons given under ^{186}Os. The observed value is 2·1 (Hill and Mihelich 1950). The transition may be identified as probably pure E2.

$^{199}_{80}$Hg : 159 keV

This transition arises in the $2{\cdot}4\times10^{-9}$ sec isomeric state of ^{199}Hg, following the β-decay of ^{199}Au, and has been quoted by Goldhaber and Hill (1952) as E2. The theoretical radiation lifetimes (Weisskopf 1951; Montalbetti 1952) are 1×10^{-11} sec for M1 and 5×10^{-8} sec for E2. The experimental value, employing a measured total conversion coefficient of 0·6 (Sherk and Hill 1951), is $5{\cdot}5\times10^{-9}$ sec, which is in satisfactory agreement with pure E2, though it may indicate some admixture of M1 radiation. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are 2·24 for M1, 0·28 for E2. The experimental value of 0·19 (Sherk and Hill 1951) indicates that no M1 is present. The empirical K/L ratios cannot be used in identification, for the reasons given under ^{186}Os. The observed values are 0·8 (Beach, Peacock, and Wilkinson 1949), 0·87 (Hill and Mihelich 1950), and 0·6 (Sherk and Hill 1951). The transition may be identified as probably pure E2.

$^{203}_{81}$Tl : 279 keV

This transition follows the β-decay of ^{203}Hg, and has been identified as E2 (Saxon 1948 ; Wilson and Curran 1951). No lifetime has been measured, though Deutsch and Wright (1950) have shown the half-life to be shorter than 3×10^{-9} sec. The theoretical radiation lifetimes (Weisskopf 1951 ; Montalbetti 1952) are 2×10^{-12} sec for M1 and 3×10^{-9} sec for E2. The observed upper limit to the half-life is too long to allow of any differentiation between M1 and E2. The theoretical K-conversion coefficients of Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) are 0·535 for M1, 0·079 for E2. The experimental values of 0·18 (Saxon 1948 ; Wilson and Curran 1951) and 0·15 (Thulin, personal communication from I. Bergström, 1953), using a mean of 0·16, indicate a 0·24 : 0·76 mixture of M1 : E2. The empirical K/L ratios of Goldhaber and Sunyar (1951) are 7·7 for M1 and 1·3 for E2. The observed values of 3 (Saxon 1948 ; Slätis and Siegbahn 1949*a*, 1949*b*), 3·7 (Wilson and Curran 1951) and 3·5 (Thulin, personal communication from I. Bergström, 1953), using a mean of 3·5, indicate a 0·40 : 0·60 mixture of M1 : E2, a lower K/L ratio giving a smaller percentage of M1. It is clear that this transition does involve a mixing of M1 and E2 radiations.

III. Experimental Data

The relative L-subshell conversion ratios of ^{181}Ta, ^{186}Os, ^{191}Ir, and ^{203}Tl were measured using a photographic 180° magnetic spectrograph of 0·1 to 0·2 per cent. resolution, and are given in Table 1. The values tabulated for ^{198}Hg and ^{199}Hg are those of Hill and Mihelich (1950), originally assigned to conversion in the L_I- and L_{III}-shells, but later by Mihelich (1952) to L_{II} and L_{III}.

The figures for the theoretical L-subshell conversion were obtained from Gellman, Griffith, and Stanley's (1952) table by interpolation and extrapolation. For each multipolarity and value of the atomic number Z a log-log plot of the conversion coefficient *v.* γ-energy, k, in units of mc^2, was prepared. The conversion coefficients so obtained for the particular k were plotted against Z on a log-linear scale, and interpolation made for the particular Z.

The theoretical K/L ratio is the ratio of Rose's K-conversion coefficient to Gellman, Griffith, and Stanley's total L-conversion coefficient. Since it can be assumed that the true K-conversion is given accurately by Rose's K-conversion coefficients (Rose, Goertzel, and Perry 1951 ; Rose *et al.* 1951), in comparing this theoretical ratio with either the observed or empirical K/L ratio, one is in effect comparing theoretical and observed absolute total L-conversion. Alternatively, an absolute L-conversion coefficient may be obtained from K-conversion and K/L ratio data, and values have been tabulated in Table 2 for the E2 transitions. In this table " experimental " values have also been calculated

TABLE 1

THEORETICAL AND EXPERIMENTAL L-CONVERSION COEFFICIENTS AND K/L RATIOS

Con-verting Nucleus	Transition Energy (keV)	Theoretical L-Subshell Conversion						Observed Ratio			Theoretical K/L Ratio		Empirical K/L Ratio		Observed K/L Ratio
		M1			E2										
		L_I	L_{II}	L_{III}	L_I	L_{II}	L_{III}	L_I	L_{II}	L_{III}	M1	E2	M1	E2	
^{181}Ta	133	0·34 340 :	0·018 18 :	0·001 1·0	0·064 0·21 :	0·36 1·2 :	0·30 1·0	0·20 :	1·22 :	1·00	5·3	0·69	7·5	0·6	0·61
^{186}Os	137	0·42 420 :	0·025 25 :	0·001 1·0	0·058 0·17 :	0·40 1·14 :	0·35 1·0	small :	1·24 :	1·00	4·9	0·54	7·5	0·6	0·6
^{198}Hg	411	0·028 280 :	0·0018 18 :	0·0001 1·0	0·0060 2·73 :	0·0060 2·73 :	0·0022 1·0	— :	2·5* :	1·00	5·7	2·3	7·8	2·6	2·1
^{199}Hg	159	0·405 405 :	0·027 27 :	0·001 1·0	0·042 0·192 :	0·294 1·34 :	0·219 1·0	small :	1·6 :	1·00	5·2	0·51	7·5	0·6	0·6
^{191}Ir	129	0·52 370 :	0·033 24 :	0·0014 1·0	0·069 0·147 :	0·58 1·23 :	0·47 1·0	4·9 :	1·75 :	1·00	5·4	0·44	7·5	0·36	2·2
^{203}Tl	279	0·086 308 :	0·0075 27 :	0·00028 1·0	0·013 0·81 :	0·030 1·88 :	0·016 1·0	2·85 :	2·08 :	1·00	5·7	1·34	7·7	1·3	3·5

* There appears to be only the L_{II} line present. However, the L_I and L_{II} lines would barely be resolved on account of the relatively high γ-energy.

using the theoretical K-conversion coefficient as, in general, K/L ratios may be determined with greater accuracy than K-conversion coefficients, particularly if the decay scheme is complex.

TABLE 2

L-CONVERSION COEFFICIENTS OF E2 TRANSITIONS

Converting Nucleus	L-Conversion Coefficient		
	Theoretical	$\frac{\alpha_{K\ expt.}}{(K/L)_{expt.}}$	$\frac{\alpha_{K\ th.}}{(K/L)_{expt.}}$
^{181}Ta	0·72	0·80	0·82
^{186}Os	0·81	0·60	0·73
^{198}Hg	0·014	0·014	0·015
^{199}Hg	0·56	0·32	0·47

IV. DISCUSSION

E2

The values given in Tables 1 and 2 indicate that for the pure E2 radiations investigated Gellman, Griffith, and Stanley's (1952) values of L-conversion coefficients are in accord with experiment. The observed relative L_{II}-conversion of ^{186}Os and ^{199}Hg may be in even better agreement with theory if one considers that the L_{II}-intensity has been increased by a small contribution from the weak L_I-conversion line. However, the case of ^{198}Hg is more difficult to explain. Here the measured L_{II}/L_{III} ratio is 2·5, in agreement with theory, but no L_I-line has been observed. This line should, according to theory, be of intensity comparable with that of the L_{II}-line. However, with the present resolution of $\sim$0·2 per cent., a broader line somewhere between the L_I and L_{II} positions might be expected with approximately the observed intensity. Regarding the absolute L-conversion coefficient, in ^{186}Os and ^{199}Hg, where the K-conversion is lower than the theoretical value, it is apparent from Table 2 that this has the effect of similarly reducing the calculated experimental L-conversion coefficient.

M1+E2

Due to the uncertainty in the relative M1 and E2 transition intensities of M1+E2 mixtures, it is more profitable to calculate the relative intensities from the observed and theoretical L-conversion ratios ; by comparison with mixtures determined by alternative means, the reliability of the theoretical L-conversion coefficients may be decided. For the 129-keV transition in ^{191}Ir, the observed L_I/L_{III} ratio and Gellman, Griffith, and Stanley's values indicate a 0·8 : 0·2 mixture of M1 : E2, which gives an L_{II}/L_{III} ratio of $\sim$1·4, in fair accord with the experimental ratio of 1·75. The agreement with either a 0·42 : 0·58 or a 0·64 : 0·36 mixture, by K-conversion and K/L ratios respectively, is not satisfactory, but merely serves to confirm existence of the mixture. It is unlikely that the poor agreement results from the neglect of (M+N)-conversion in the calculation of total transition intensity. For the 279-keV transition in ^{203}Tl, the observed L_I/L_{III} ratio indicates a 0·35 : 0·65 mixture of M1 : E2, giving an L_{II}/L_{III} ratio of 2·04, in excellent agreement with the experimental ratio of 2·08. Satisfactory agreement also exists with mixtures determined by K-conversion and K/L ratios.

In calculating mixtures, it should be pointed out that we have assumed strictly " linear " mixtures of the mixing multipoles, proportional to the total transition probabilities, that is, including K- and L-electron conversion transitions as well as unconverted γ-ray transitions. The effect on the mixtures of including (M+N)-conversion transitions is slight, amounting to only 1 or 2 per cent., as is the effect of using the K/L ratios discussed in Section V, in place of the empirical values.

V. Theoretical K/L Ratios

Since it appears from Table 2 that there are reasonable grounds for considering that the absolute magnitudes of the total L-shell conversions given by Gellman, Griffith, and Stanley (1952) are approximately correct, it would be valuable for experimental purposes to have plots of the K/L ratio for M1 and E2 transitions. It will be recalled that Gellman, Griffith, and Stanley computed

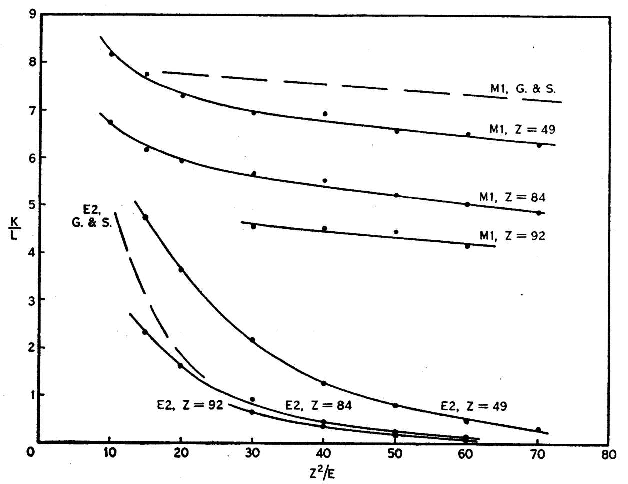

Fig. 1.—Theoretical K/L ratios for M1 and E2 transitions for different values of Z. The curves of Goldhaber and Sunyar (1951), G. & S., are included for comparison. These curves may also be compared with those of Hebb and Nelson (1940), and Tralli and Lowen (1949), calculated for Z=35.

the L-conversion coefficients for the same Z-values (49, 84, and 92) as used by Reitz (1950) in the latter's computation of K-conversion coefficients. The K/L ratios plotted in Figure 1 have been calculated from the Reitz and Gellman, Griffith, and Stanley values and, following the usual practice, have been plotted *v.* Z^2/E (E in keV).

It will be noticed that, while the K/L ratio curves show the accepted trends of M1 and E2 transition, there is a significant difference between the curves of different Z-values. In view of the fact that Goldhaber and Sunyar (1951) gave only one curve for each multipole transition, it is at first sight strange that the behaviour suggested by the curves of Figure 1 has not been seen earlier. These authors did point out that it would be very likely that the exact K/L ratios would depend on Z and E in a complicated manner, and that low Z points are sometimes higher, and high Z points lower, than the average empirical K/L ratio curve. However, especially for the E2 curve where there seemed to be adequate data, inspection of Goldhaber and Sunyar's curve shows that most examples were of high Z. For this reason their variations are within the bounds of the E2 curves in Figure 1. There was really only one instance, of ^{111}Cd, which was of intermediate Z, and this point was at one end of their curve and therefore did not show a distinct departure. For the M1 curve there is also accumulating evidence that the single curve drawn by Goldhaber and Sunyar is not fully representative. Their line, drawn mainly through points of the tellurium isomer transitions, is probably intermediate between the true curves for elements of low Z like ^{88}Rb, for which there is some evidence of the high M1 K/L ratio of $\sim$8 (Thulin 1952), and curves for high-Z elements like ^{199}Hg, for which the K/L ratio is $\sim$5 (Sherk and Hill 1951).

Comparison of the present curves with those of Hebb and Nelson (1940) and Tralli and Lowen (1949), which were calculated by the approximate method for $Z=35$, further supports the general trend of increasing K/L ratio with decreasing Z.

VI. Conclusion

Both the results presented in this paper and those of Mihelich (1952) indicate that the agreement between the experimental and theoretical L-subshell conversion coefficients may be considered to be very good. Until more complete computations of L-subshell conversion coefficients are available, those already published by Gellman, Griffith, and Stanley (1952) may be used with considerable confidence for confirmation or identification of γ-transition multipolarities.

Further, the L-shell conversion coefficients of Gellman, Griffith, and Stanley may be used in conjunction with the K-shell coefficients of Reitz (1950) or Rose, Goertzel, and Perry (1951) and Rose *et al.* (1951) to calculate K/L ratios, which may be used in preference to those of Goldhaber and Sunyar (1951), particularly for transitions taking place in the lighter elements.

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