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Statistical-Theory Calculation of Charge Distribution in Fission*

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The nuclidic mass formula developed recently by the authors is employed to calculate the most probable charge Z_p of fission products. The predicted values of Z_p for thermal-neutron fission of U^{235} and spontaneous fission of Cf^{252} differ from the experimental values by a root-mean-square deviation of 0.35 and 0.55 charge unit, respectively, the computer calculation being made for charge values varying 0.05 charge unit at a time. These values may be compared with the root-mean-square deviations between two existing sets of experimental results on U^{235} and on Cf^{252}, which are 0.34 and 0.50, respectively. The root-mean-square deviations of the predicted Z_p values for many other schemes of charge division are calculated for comparison. The width of the charge-distribution curve of the primary fission fragments (before prompt neutron emission) in the peak-yield regions, calculated by the statistical theory (standard deviation of Gaussian $\sigma=0.51$), agrees with the empirical value of Ferguson and Read ($\sigma=0.55$) within 8%. Fine structure of Z_p due to the shell effect is noticed.

I. INTRODUCTION

THE statistical theory of nuclear fission[1] provides theoretical predictions on a variety of interesting phenomena of the fission process. The detailed calculation of the theory, nevertheless, depends on the availability of accurate nuclear data of several kinds, which are still incomplete because of the complication of the effects of the nuclear shells. The necessary data include the nuclidic masses of the primary fission products. In the last few years, the authors have developed a nuclidic mass formula[2] with an accuracy of the order of 1 MeV for the 842 known masses of nuclides close to the beta-stability line. While some calculations require a higher degree of accuracy, the formula provides a reasonable basis for a detailed calculation of the charge distribution in fission even though the uncertainty of the calculated masses of the fission fragments is expected to be larger than that of the nuclides closer to beta-stability line. This calculation is significant because charge distribution is essentially determined by nuclidic masses not involving the other nuclear data and therefore is not encumbered by the incompleteness of our knowledge on nuclear data. Thus, the theory and the mass formula provide definite predictions which may be compared with experimental results as a test of the theory. This is not so in mass, kinetic energy, and prompt neutron distributions where more unknowns are involved.

Inasmuch as the most probable charge Z_p of the primary fission products can be determined empirically to an accuracy of 0.1 charge unit, experimenters feel that the information may be used to check the accuracy of various mass formulas. In this work, of course, it is not possible to check the statistical theory and the mass formula at the same time. However, applying different mass formulas in the same model calculation, it is possible to draw useful conclusions concerning the various mass formulas.

The formulation of the statistical theory and the application to charge distribution have been reported previously.[1] Together with the mass formula, we are in a position to calculate the relative yields of all primary fission products (before prompt neutron emission) of the same mass number. These are not directly comparable with the experimental independent yields which are yields of fission products after the emission of the prompt neutrons (before beta decay). On the other hand, experimental information is usually stated in the form of the most probable charge Z_p as a function of the mass number A of the fission product, and the width or the standard deviation σ of the Gaussian curve representing charge distribution of fission products of a given mass number. Therefore, instead of calculating the independent yields, we choose to calculate Z_p and σ so that the results may be compared directly with the empirical values.

II. THE WIDTH OF THE CHARGE-DISTRIBUTION CURVE

The half-width at half maximum of charge-distribution curves, calculated according to the statistical theory, in the peak regions of the mass-yield curve of thermal neutron fission of U^{235} has been reported to be 0.6 charge unit ($\sigma=0.51$) previously.[1] The use of the new mass formula does not change this figure. This

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value compared unfavorably with the then known empirical value of the width in the peak regions which was about 0.8 or 1.0.[1] However, it should be kept in mind that the theoretical width is that of the primary fission products and the empirical width is that of the final fission products after prompt neutron emission; the two are not directly comparable. Preliminary calculation[3] of the independent yields of shielded nuclei based on statistical theory, taking into account the effect of prompt neutron emission, showed reasonable agreement with experimental values. This result indicates that the larger empirical width is essentially due to the effect of prompt neutron emission. Thus the difference should not be taken as evidence against the statistical theory. As a matter of fact, there is evidence that the width of the primary fission products is narrower than that of the final fission products. Ferguson and Read[4] have used a Gaussian curve to represent the charge distribution of the primary fission product with the σ value equal to 0.55, which compares favorably with the theoretical value of 0.51. Thus the theory is reasonably satisfactory as far as the prediction of the shape of the distribution curve is concerned.

III. THE MOST PROBABLE CHARGE Z_p

The most probable charges $Z_{p,L}$ and $Z_{p,H}$ for a pair of primary fission fragments of mass numbers A_L and A_H (L and H stand for light and heavy) are determined, according to the statistical theory, by finding the charge values that maximize the total excitation energy of the pair of fragments (E_E). The excitation energy is given by

$$\begin{aligned} E_E &= M - M_c(Z_L,A_L) - M_c(Z_H,A_H) \\ &\qquad - c_m Z_L Z_H - D_L - D_H \\ &= M - E_p - D_L - D_H \\ &= E_R - c_m Z_L Z_H - D_L - D_H\,, \end{aligned} \qquad (1)$$

where M is the mass of the fissioning nucleus; $M_c(Z,A)$ is the mass of the fission fragment (measured at a characteristic level, essentially the nuclidic mass without the pairing energy term) with an atomic number Z and mass number A; c_m is a charge-independent but mass-dependent parameter in the kinetic-energy (of fission fragments) term; D is the deformation energy of the fission fragment; E_P is the total potential energy; E_R is the total energy release. The maximizing process leads to nonintegral values of $Z_{p,L}$ and $Z_{p,H}$ which represent the position of the peak of the charge distribution curve.

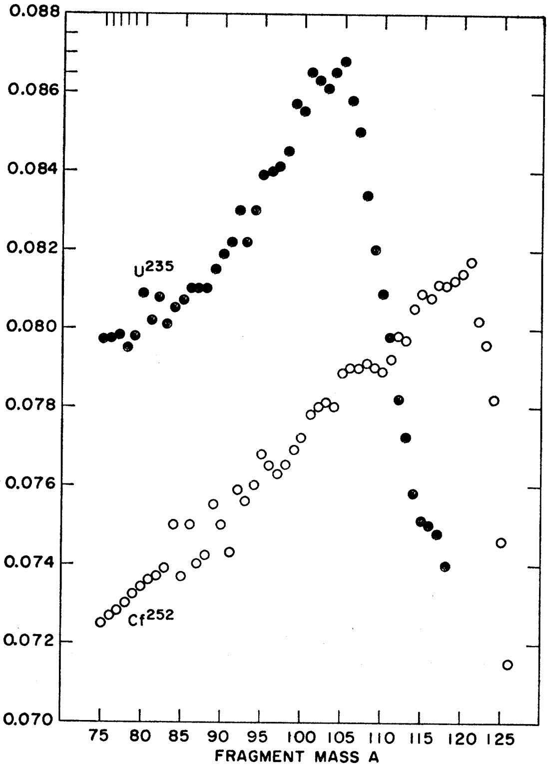

FIG. 1. Parameter c_m of the kinetic energy term $c_m Z_1 Z_2$ of U^{235} and Cf^{252} as a function of the mass number A of fission fragments.

We have calculated the Z_p values of thermal neutron fission of U^{235} and spontaneous fission of Cf^{252}. The mass formula developed by the authors[2] with the pairing-energy term dropped was used for calculating M_c. The formula is particularly suitable for the present purpose because its value of M_c is a continuous function of Z and A, and the maximization may be carried out with fractional Z values. Many mass formulas do not give mass values for fractional Z because discontinuous terms are introduced to account for the shell effect on nuclidic mass. The values of c_m for U^{235} and Cf^{252} are deduced empirically from the fission-fragment kinetic-energy data of Apalin *et al.*,[5] Whetstone,[6] and Schmitt *et al.*,[7] and are plotted in Fig. 1. The deformation energy as a function of A is taken from a previous calculation[8] on the kinetic energy and prompt neutron distributions, which takes into account the nuclear shell effect on nuclear deformation. However, for a given mass number A the variation of deformation energy with respect to charge Z, which enters the maximization of Eq. (1), is assumed to be given according to the liquid-drop model.[1] The deformation energy term may thus be expressed as follows:

$$D_i = \xi(A_i)[0.01A_i^{2/3} - 0.000128Z_i^2/A_i^{1/3}]_{i=L,H}\,. \qquad (2)$$

The parameter ξ as a function of A is constructed from the results of Ref. 8 and is plotted in Fig. 2. Since the variation of the deformation energy can be shown to be

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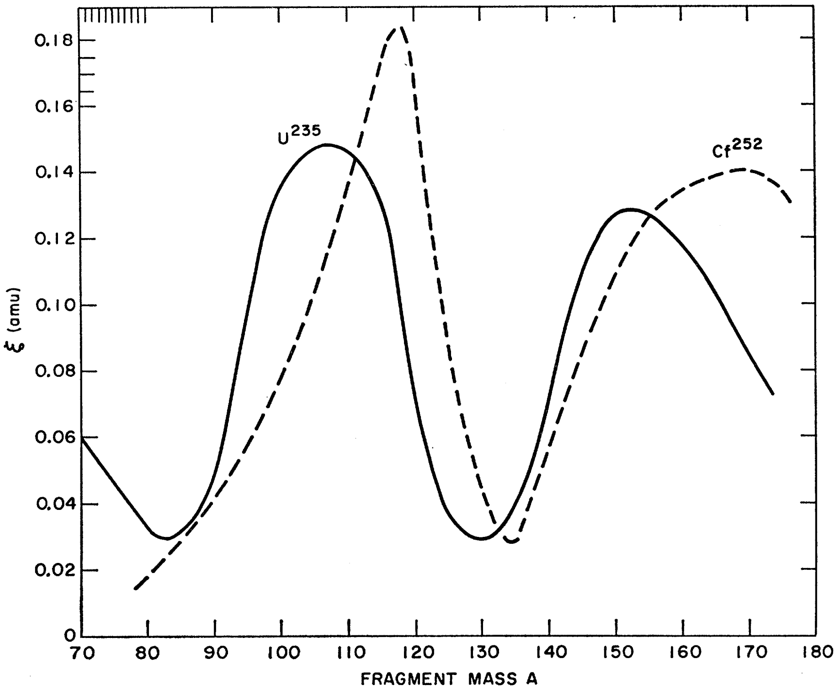

FIG. 2. Parameter ξ of the deformation energy expression of Eq. (2) of U^{235} and Cf^{252} as a function of the mass number of fission fragments.

rather small, the above approximate treatment seems adequate for the purpose of maximizing Eq. (1). The maximum of excitation energy (MEE) is found by using Argonne's IBM-1620-II and CDC-3600 computers. The computation is made with Z varying 0.05 unit apart and thus the value of Z_p obtained is expected to be associated with an amount of uncertainty of 0.05 charge unit.

For the purpose of comparison, similar calculations were carried out to obtain Z_p values determined by minimizing the total potential energy (MPE) and by maximizing the total energy release (MER).

The results of calculation are presented graphically. The function $Z_{pi}-A_i(Z_H+Z_L)/(A_H+A_L)$ is plotted in Figs. 3 and 4 together with experimental results for U^{235} and Cf^{252}, respectively. To bring out the fine features, the graphs are plotted with the ordinates expanded tenfold with respect to the fragment-mass number scale. It may not be meaningful to compare the general trend on such a plot; it is more useful to compare the root-mean-square deviations (RMSD) of the results of various predictions from the experimental values. The following is such a comparison.

Our calculated results are compared with (1) previous MER and MEP calculations[4,9] based on the mass formulas of Cameron[10] and Seeger[11]; (2) other charge division schemes based on the hypotheses of unchanged charge density (UCD),[12] equal charge displacement (ECD),[12] Apalin *et al.*,[9,13] Armbruster,[14] and Present[4,15]; and (3) the experimental results of Wahl *et al.*[16] and Strom *et al.*[17] on U^{235} and of Glendenin and Unik,[18] Kapoor *et al.*,[19] Dolce *et al.*,[20] and Wahl *et al.*[16] on Cf^{252}. In the ECD calculation, the nonintegral values of Z_A, the charge of the most stable isobar, are taken from the average values of Dewdney[21] and Hillman.[22] Apalin's charge-distribution function,[13] $Z_L=Z_HA_L/A_H+5.9-0.05A_L$, is that of unchanged charge density plus a linear function of A_L. Armbruster's semiempirical calculation[14] is based on maximum energy release between the saddle point and scission. Present's scheme is

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TABLE I. Root-mean-square deviations (in charge units) of various Z_p values from experimental values.

		U^{235}			Cf^{252}			
Z_p function	Nuclidic mass formula	Wahl	Storm	Wahl and Strom	Glendenin	Kapoor	Glendenin and Kapoor	Dolce
Number of data points		$n=30$	$n=13$	$n=43$	$n=26$	$n=26$	$n=52$	$n=8$
RMSD of predicted values from experimental values								
UCD		0.53	0.52	0.53	0.51	0.70	0.61	0.92
ECD		0.43	0.49	0.45	0.47	0.30	0.40	0.61
Apalin		0.40	0.30	0.37	0.62	0.43	0.54	0.37
Armbruster		0.44	0.32	0.41	0.78	0.67	0.72	0.41
Present		0.77	0.58	0.72				
MER	Cameron	0.67	0.62	0.66	0.95	0.64	0.82	0.17
	Seeger	0.36	0.29	0.34	0.71	0.47	0.61	0.39
	Wing-Fong	0.49	0.39	0.46	0.79	0.52	0.66	0.22
MPE	Cameron	0.39	0.35	0.38				
	Seeger	0.39	0.32	0.37				
	Wing-Fong	0.41	0.24	0.36	0.54	0.63	0.58	0.66
MEE	Wing-Fong	0.39	0.23	0.35	0.52	0.57	0.55	0.67
RMSD of one set of experiment from another								
Wahl from Strom ($n=13$)[a]				0.34				
Kapoor from Glendenin ($n=9$)							0.50	

[a] There are two Z_p values for the fragment mass 136.8.

based on maximum energy release with the fission fragments assumed to have a nonuniform proton density.[15] The experimental results of Wahl *et al.*[16] for U^{235} and Cf^{252} and those of Strom *et al.*[17] are based on the experimental values of the independent yields of individual fission products. The experimental results for Cf^{252}, except those of Wahl *et al.*,[16] are all obtained by measuring the energy distribution of the prompt K x rays emitted by the primary fission products.[18,19,20] Wahl *et al.*'s results have been corrected for the prompt neutron numbers by using the more recent results of Apalin *et al.*[23] for U^{235} and of Bowman *et al.*[24] for Cf^{252}. Experimental results of upper and lower limits of Z_p are not included here. We decided not to include for comparison the experimental results of Armbruster *et al.*[25] and Konecny *et al.*[26] on U^{235}, which are based on the counting of the number of beta particles of the fission fragments, because of the unresolved discrepancy between their results and the radiochemical ones,[27] in spite of the fact that the statistical-theory prediction agrees better in the general trend with the results of Armbruster *et al.*

The comparison of our results with the above-mentioned ones, in terms of the RMSD of the predicted Z_p values from the experimental values, which makes it possible to compare point by point instead of the rather vague general trend, is summarized in Table I. The continuous curves of Figs. 3 and 4 are convenient for interpolations for fractional mass numbers which are employed in presenting most experimental results. Table I lists the RMSD of the existing calculated values of Z_p from the experimental values of comparable fragment masses. In the lower part of Table I, we also list the RMSD of one experiment from the other in both U^{235} and Cf^{252} (corrected for the small mass difference of $\Delta Z=0.4\Delta A$). In U^{235}, Wahl *et al.*'s 13 points (two points at $A=136.8$) that have comparable points in Strom *et al.*'s results deviate from the latter with a RMSD of 0.34. In Cf^{252}, Glendenin and Unik's nine points that have comparable points in Kapoor *et al.*'s results deviate from the latter with a RMSD of 0.50. These may be taken as the range of uncertainty of the experimental results. The RMSD of the statistical-theory prediction from the experimental results is 0.35 for U^{235} and 0.55 for Cf^{252}, exceeding the experimental uncertainty by only 0.01 and 0.05 unit, respectively. Since the computer calculation is carried out

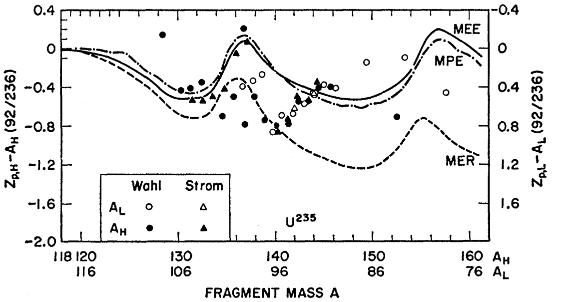

FIG. 3. The most probable charge Z_p of thermal neutron fission of U^{235} calculated according to the statistical theory with the authors' mass formula (MEE) compared with the experimental results of Wahl *et al.* (circles) and Strom *et al.* (triangles). Dashed and dotted-dashed curves show Z_p calculated according to maximum energy release (MER) and minimum potential energy (MPE), respectively, with the authors' mass formula.

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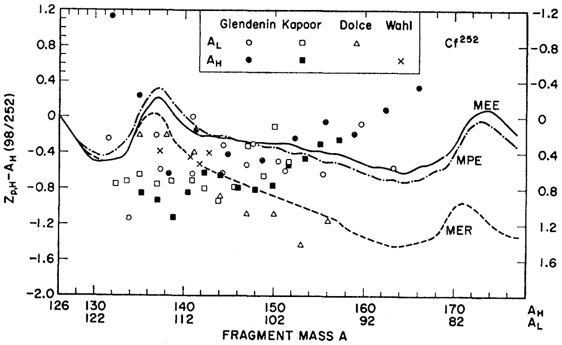

FIG. 4. The most probable charge Z_p of spontaneous fission of Cf^{252} calculated according to the statistical theory with the authors' mass formula (MEE) compared with the experimental results of Glendenin and Unik (circles), Kapoor *et al.* (squares), Dolce *et al.* (triangles), and Wahl *et al.* (cross). Dashed and dotted-dashed curves show Z_p calculated according to maximum energy release (MER) and minimum potential energy (MPE), respectively, with the authors' mass formula.

with Z varying 0.05 charge unit at a time, it may be concluded that the statistical theory predicts Z_p within the limits of the existing uncertainties.

One observes from Table I as well as Figs. 3 and 4 that disregarding the recent results of Dolce *et al.*,[20] the prediction of the statistical theory based on our mass formula agrees with the experimental results better than that based on the assumption of maximum energy release, slightly better than that based on the assumption of minimum potential energy, and better than those of all the five other schemes of charge division based on *ad hoc* assumptions (except ECD in Cf^{252}). One may surmise from the above conclusions that the excitation energy is a more significant quantity in the fission process than the total energy release (and perhaps also the total potential energy) as the statistical theory claims. The results of Dolce *et al.*, contradicting the other two experiments of the same kind, favor the assumption of MER.

Concerning the various mass formulas, we notice that the RMSD values of our mass formula are comparable with those of Seeger's while those of Cameron's are larger than ours. What this means in terms of the relative accuracy of the three formulas is difficult to say. On the other hand, in the symmetric fission region where the Z_p function is rather insensitive to the choice of theory of charge division, the comparison of the three Z_p based on the three mass formulas with experimental results does provide a check on the relative accuracy of the formulas in this mass region. In this comparison, the result of our mass formula in U^{235} is closer to the experimental data than the other two formulas [4,9]

IV. FINE STRUCTURE OF Z_p AND SHELL EFFECT

It has been assumed tacitly that Z_p is a more or less smooth function of A, and this has been the basis for extrapolation in many previous works. In Figs. 3 and 4 the calculated curves based on statistical theory exhibit dips at mass 132, followed by a rapid rise reaching a peak at mass 137. They show the existence of fine structure over the smooth trend, and the origin of this fine structure may easily be traced to the effect of nuclear shells. Fission fragments in the neighborhood of $_{50}Sn_{82}^{132}$ (closed-shell configuration of 50 protons and 82 neutrons) are formed preferably according to the statistical theory because of higher internal excitation as a result of the lower nuclear mass; thus the dip in the neighborhood of mass 132. The existence of irregularities (perhaps traceable to shell effect) over a general smooth trend is a familiar pattern in fission phenomena; we have seen it in mass distribution, in kinetic-energy distribution, and in prompt neutron distribution. There is every reason to expect the same in charge distribution.

The results of Strom *et al.*[17] seem to indicate the existence of fine structure in charge distribution of U^{235} fission. The dip and the peak of the theoretical curve agree with their six points in the mass region of 131–137 quite closely. If we take their results exclusively, the RMSD of the statistical theory prediction is, as shown in Table I, only 0.23. This small RMSD lends strong support to the statistical theory. Concerning the discrepancy between Strom *et al.* and Wahl *et al.*, the most significant points under question are the Z_p values at masses 136.0, 136.8, and 137.2. The point at mass 136.0 of Strom *et al.* is constructed indirectly[17] and thus the other two points become crucial. Strom *et al.* derived a Z_p value of 53.51 for the fragment mass 137.2 from the charge-distribution curve of the mass-136 chain of the final fission products (using a value of 1.2 for the average number of prompt neutrons for mass 136), which is constructed on the basis of three yields: the cumulative yield[28] of I^{136} (yield of I^{136} including all its beta precursors), the cumulative yield[29] of Xe^{136}, and the independent yield[30] of Cs^{136} (yield of Cs^{136} excluding all its beta precursors). Wahl *et al.* obtained two Z_p values of 53.53 and 52.58 for mass 136.8 from the cumulative yield[28] of I^{136} and the independent yield of Cs^{136}, respectively, using an assumed Gaussian function for the charge distribution (a value of 0.8 for the average number of prompt neutrons[23] is used here). The resolution of the discrepancy of the Z_p value here involved thus depends largely on the accuracy of the yield of I^{136}, which becomes the crucial quantity in the whole argument. This yield was determined judiciously by Stanley and Katcoff[28] who noticed that their yield value was inconsistent with the ECD hypothesis, which would predict a yield about twice as large. They further noticed that the same discrepancy exists for the yield of I^{136} in thermal neutron fission of Pu^{239} and U^{233}. In the absence of any explana-

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tion for the low yield of I^{136}, the results in the three fission cases must be taken as evidence agsinst ECD and for the existence of fine structure. The unusual behavior of the I^{136} yield was noticed by Wahl[16] but not explained.

The fine structure of the Z_p curve is an interesting subject deserving further investigation experimentally. In particular, the predicted fine structure in Cf^{252} is not borne out in the existing experimental results. Investigation of independent yields of Pu^{239} and U^{233} in the fine-structure region may help clarify the situation.

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Relative Intensities of the *L*-Shell Internal-Conversion Lines of Pure $E2$ ($2+\rightarrow 0+$) Transitions in Rare-Earth Nuclei

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Accurate measurements of *L*-shell internal-conversion line intensity ratios are reported for four low-energy $E2$ transitions in ^{156}Gd, ^{160}Dy, ^{166}Er, and ^{170}Yb. The measurements were made using a 100-cm radius iron-free $\pi\sqrt{2}$ double-focusing β spectrometer at resolution settings of 0.03–0.09% in momentum. Different source and detector arrangements were used to explore for possible systematic effects. In all cases the experimental $L_{\mathrm{II}}/L_{\mathrm{III}}$ intensity ratios agreed with the corresponding theoretical ratios to $\leq 2\%$. The $L_{\mathrm{I}}/L_{\mathrm{II}}$ and $L_{\mathrm{I}}/L_{\mathrm{III}}$ experimental ratios are systematically larger than the theoretical values by an average of 5.4%.

INTRODUCTION

With the development of high-resolution, iron-free β spectrometers in the late 1950's,[1–7] and the ready availability of theoretical tabulations[8,9] of internal-conversion coefficients, the comparison of experimental and theoretical *L*-subshell conversion electron intensity ratios has become a powerful method[10] for establishing the multipolarities of electromagnetic transitions, and hence for the assignment of spins and parities to nuclear energy levels. The accuracy of this method of determining transition multipolarities depends critically on the accuracy of the tabulated conversion coefficients. The $2+\rightarrow 0+$ ground-state transitions in even-even nuclei provide one excellent experimental test of the accuracy of the calculated *L*-subshell ratios. These pure electric quadrupole transitions are highly enhanced and nuclear penetration effects are not expected to play a significant part[11] in their internal conversion.

At the 1965 International Conference on the Internal Conversion Process,[12] which was held at Vanderbilt University, two groups reported *L*-subshell ratios for $2+\rightarrow 0+$ transitions which were in marked disagreement with the calculated values of Rose[8] and of Sliv and Band.[9] Mladjenovic[13] and his co-workers from Belgrade, and Hamilton[14] and his co-workers from Vanderbilt reported measured $L_{\mathrm{I}}/L_{\mathrm{II}}$ and $L_{\mathrm{I}}/L_{\mathrm{III}}$ intensity ratios for ten pure $E2$ transitions in even-even rare-earth nuclei which were higher than the theoretical values by amounts ranging from 15–40%. Subsequent reexamination[15] of all of the accurate data available confirmed that there was some disagreement between theory and experiment but, perhaps more important,

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