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Classification of Nuclear Isomers

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A classification is made on the basis of the forbiddenness of the transitions from the known nuclear isomeric states. The resultant grouping is shown to be in agreement with the Weizsäcker theory of isomerism and with internal conversion theory. The predictions of internal conversion theory are also compared with the experimental values that are available for the isomers considered.

A NUCLEAR isomeric state may be defined as an excited state of a nucleus which exists for a measurable length of time ($\tau \geq 10^{-9}$ second). Since most nuclear excited states decay with much shorter half-lives, isomeric states must be characterized by having only highly forbidden transitions. Weizsäcker[1] first explained this forbiddenness by postulating that each transition had a large angular momentum change associated with it and that, therefore, only quanta of high multipole order could be emitted.

The effect of this high multipole order can be exhibited by considering the analogous system of an oscillating distribution of charge in classical electrodynamics. The total radiation, R, from such a distribution is given by:

$$R=(R_{E1})+(R_{M1}+R_{E2})+(R_{M2}+R_{E3})+\cdots, \quad (1)$$

where R_{Ei} is the rate of electric 2^i pole radiation and R_{Mi} is the rate of magnetic 2^i pole radiation. In this expansion, individual contributions which are of the same order of magnitude are bracketed into terms. Successive terms decrease rapidly if the dimension of the oscillator is small compared to the radiated wavelength. Therefore, the first contributing term essentially determines the total probability of radiation and a high forbiddenness corresponds to the vanishing of the first group of terms. It is convenient to designate this forbiddenness by l, the number of the first contributing term. A given l value corresponds to either electric 2^l or magnetic 2^{l-1} pole radiation.

A semiquantitative theory for the decay of isomeric states can be developed by extending these electrodynamic considerations to a nucleus in an excited state. The mean life of the state (for gamma-ray emission only) can be shown to be:

$$\tau_\gamma=\frac{3(l!)^2}{\rho^{2l}}\left(\frac{137}{W}\right)^{2l+1}\frac{\hbar}{\mathrm{mc}^2}, \quad (2)$$

where ρ = radius of the nucleus in units of e^2/mc^2, W = excitation energy in units of mc^2, and $\hbar/\mathrm{mc}^2 = 1.31 \times 10^{-21}$ second. The derivation of this relation involves various approximations. The crudest of these is that the nuclear matrix element for the transition depends solely on the size of the nucleus. Thus, Eq. (2) could be expected to give only an average value for τ_γ. Fluctuations about this average by factors of 10 or 100 may be anticipated by considering the results of the analogous procedure applied to atomic transitions. The derivation of Eq. (2) is based on the model of a single charge executing oscillations whose amplitude is equal to the nuclear radius. Other models yield formulas agreeing as to the dependence of τ_γ on ρ and W but differing as to numerical factors.

A comparison can be made between the experimentally determined lifetimes and those predicted by Eq. (2). Figure 1 shows the experimental data for 56 isomers; each isomer is exhibited as a point on a graph of $\log\tau_{\mathrm{exp}}$ as a function of $\log W$. Only those isomers that decay predominantly to another state in the same nucleus (i.e., generically related isomers) are included; of these, isomers living for less than 1 second are omitted since they are too few to define groups. (A plot of this type was first published by Wiedenbeck.[2]) While a grouping is in evidence, the dependence of the lifetime on energy is certainly not as is predicted by Eq. (2).

However, one would expect that the experimentally determined lifetime, τ_{exp}, would be less than the gamma-ray lifetime, τ_γ. This decrease in lifetime is caused by the competing process of internal conversion which tends to increase the total transition rate.[3] If this additional process is considered, the relation between the two lifetimes is given by

$$\tau_\gamma=\tau_{\mathrm{exp}}\left(1+\frac{N_e}{N_\gamma}\right), \quad (3)$$

where N_e/N_γ is the total conversion coefficient. Since the internal conversion coefficient depends on the atomic number, Z, τ_{exp} cannot be expressed as a function of W alone. Therefore, for a quantitative comparison, it is necessary to convert τ_{exp} to τ_γ for each isomer. (Wiedenbeck[2] did not make this type of point-by-point correction.)

The values for τ_γ must be calculated by using theoretical conversion coefficients since the experimental internal conversion data are inadequate. The calculations can be made for each nucleus if the energy of the metastable state is known and if a tentative l value

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[1] C. F. von Weizsäcker, Naturwiss. **24**, 813 (1936).

[2] M. L. Wiedenbeck, Phys. Rev. **69**, 567 (1946).

[3] H. M. Taylor and M. F. Mott, Proc. Roy. Soc. **A142**, 215 (1933).

assignment is made on the basis of Fig. 1. Because of the approximate nature of the formula for τ_γ, the difference between electric and magnetic conversion coefficients is not significant; for simplicity, only the electric conversion coefficients are used. The correction factors for the 56 isomers range from 1.04 at the highest energy to 7.8×10^7 at the lowest energy. The values of τ_γ are plotted as a function of energy in Fig. 2. Not only is the resultant grouping much more definite, but also there is qualitative agreement with regard to the predicted energy dependence (i.e., $\tau_\gamma \propto W^{-(2l+1)}$).

A more quantitative check may be made on the absolute value of τ_γ by taking account of the value of ρ^{2l} for each isomer. Equation (2) indicates that the product $\tau_\gamma\rho^{2l}$ depends only on the transition energy. Except for the expected variation in nuclear matrix elements, this product is directly calculable from Eq. (2) without any adjustable parameters. Figure 3 shows the isomers on a plot of $\log\tau_\gamma\rho^{2l}$ as a function of $\log W$. (The value of the nuclear radius was taken as $r=1.5A^{\frac{1}{3}}\times10^{-13}$.) The unadjusted theoretical lines are drawn in for $l=3$, 4, 5, and 6. The agreement between the $l=4$ and $l=5$ lines and the experimental points is seen to be quite good; the experimental points all fall within the anticipated factor of 100 of the theoretical lines. Pb^{204}, which is the only even-even isomer on this graph, seems to fit the $l=6$ group. (The point that is shown for Pb^{204} was calculated by assuming $l=5$; for $l=6$, $\tau_\gamma\rho^{2l}$ would increase by a factor of 10.)

The existence of distinct groups corresponding to $l=4$ and $l=5$ is not shown by a superficial examination of Fig. 3, since the conversion correction which is based on an assumed l value tends to produce a grouping. (The conversion correction increases with l, especially at low energy.) However, the following three considerations tend to substantiate the existence of these groups: (a) Groups are already in evidence in Fig. 1, particularly in the significant region above 200 kev where internal conversion corrections are very small. (b) The grouping of Fig. 1 would be accentuated even if the same l value were used to calculate the conversion correction for all points. Therefore, the assumed l value is not responsible for artificially inducing a grouping. (c) The most striking evidence for the existence of two groups is the semiquantitative agreement which is exhibited in Fig. 3.

Once the existence of groups is established, the maintenance of these can be used as the criterion for assigning l values in the few questionable cases. For example, in the case of the most doubtful member of the $l=5$ group, Te^{131}, a reassignment to $l=4$ would shift the point down 0.76 unit on the logarithmic plot of Fig. 2. Similarly, reassignment of Tc^{99} and Cs^{134} to the $l=5$ group would shift these points up by 0.83 and 0.88. These changes would obviously not fit the above criterion since the isomers concerned would then be further separated from either group.

On the basis of the agreement achieved, it is possible

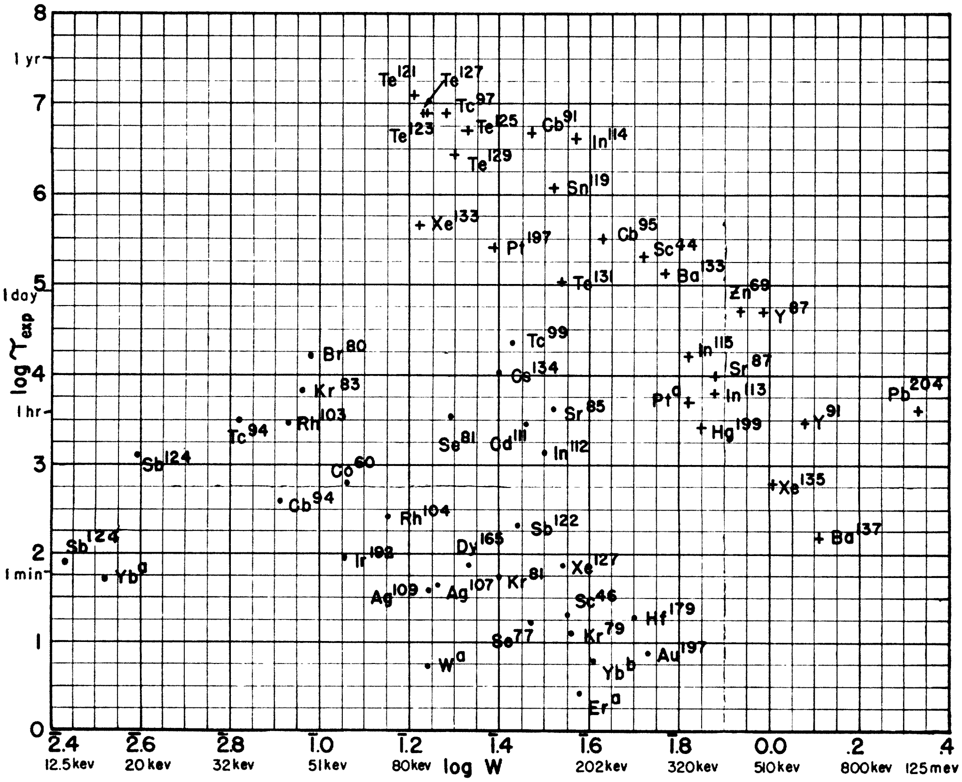

FIG. 1. Comparison between experimentally determined lifetimes of isomers and those predicted by Eq. (2).

to draw several conclusions. The existence of the $l=4$ and $l=5$ groups tends to substantiate Weizsäcker's explanation of the existence of isomeric states. The semiquantitative agreement shown in Fig. 3 further indicates the qualitative validity of internal conversion theory. Finally, the specific l value assignments seem valid and may be useful in the evaluation of the proposed "shell" models of nuclei which purport to describe the detailed nature of isomeric transitions.

Notes added in proof.—(1) Xe^{133} was erroneously listed as an isomer; it should be deleted from the three figures. (2) The energy of Hf^{179} has been redetermined so that $\log W=\bar{1}.47$. This changes the ordinates on the three figures to 1.3, 2.62, and 6.45. (3) Four new isomers should be added to the 55 remaining in the three figures. The values for the abscissa and for the ordinates appropriate for the three figures for each of these isomers are, respectively: Co^{58} $\bar{2}.65$; 4.55, 9.8, and 12.35; Ta^{182} $\bar{1}.55$; 2.95, 3.95, and 7.8; Xe^{131} $\bar{1}.51$; 6.0, 7.8, and 12.2; and Ba^{135} $\bar{1}.75$; 5.0, 5.6, and 9.95.

INTERNAL CONVERSION DATA

In principle, it should be possible to provide a further check on the l value assignments by comparing experimental internal conversion data with the theoretical predictions. For this purpose, independent checks might be afforded by both the absolute value of the K conversion coefficient and the ratio of K conversion to L conversion. However, the present status of both experiment and theory makes such checks impractical.*

One limitation has been the lack of adequate theoretical calculations caused by the computational difficulties involved. Recently, Rose and his collaborators[4] have made some relativistic calculations using the Mark I Relay Calculator at Harvard. Thus far, these calculations are available only for K conversion (both electric and magnetic) and only in the energy range above 150 kev. It is, therefore, necessary to use calculations based

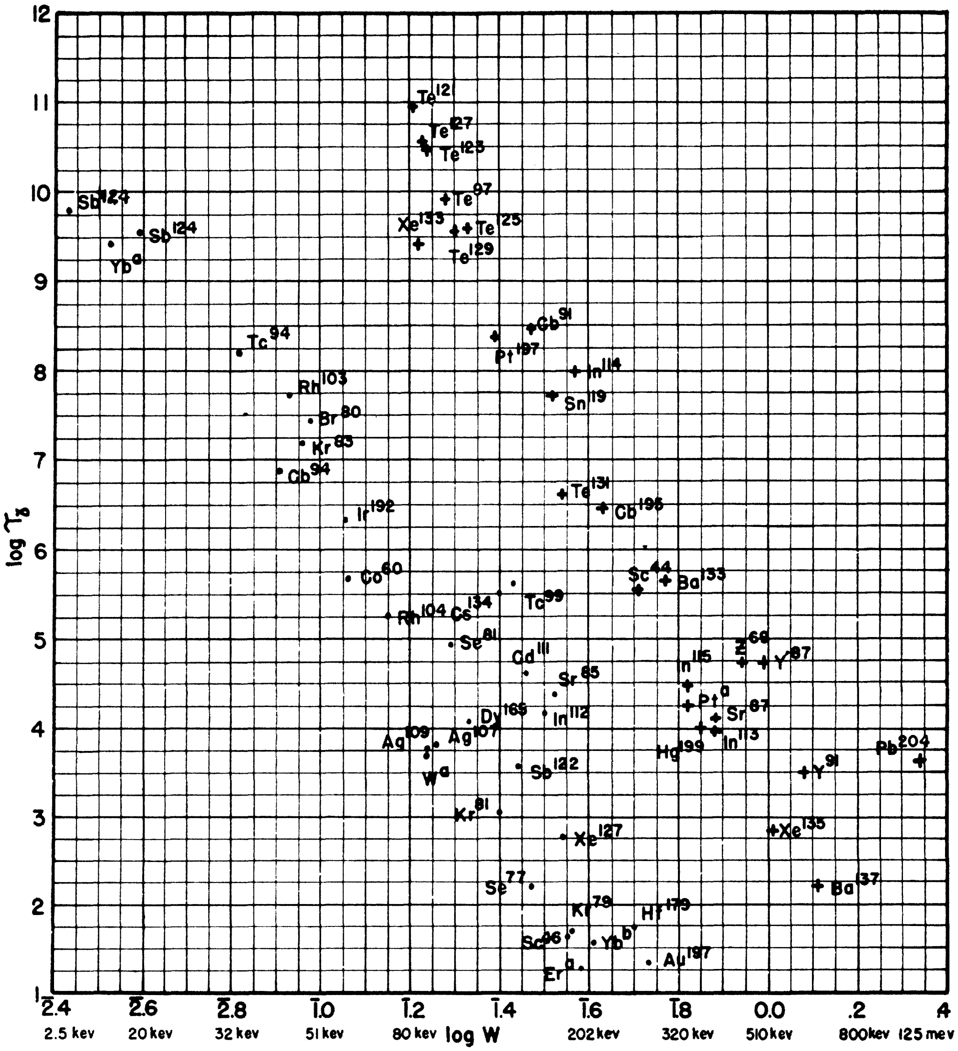

FIG. 2. Values of the calculated γ-ray lifetimes of isomers as a function of the energy.

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* It should be emphasized that the theoretical uncertainties which prevent a quantitative check of internal conversion theory are too small to have an appreciable effect on the grouping of Figs. 2 and 3.

[4] Rose, Goertzel, Spinard, Harr, and Strong, report privately circulated.

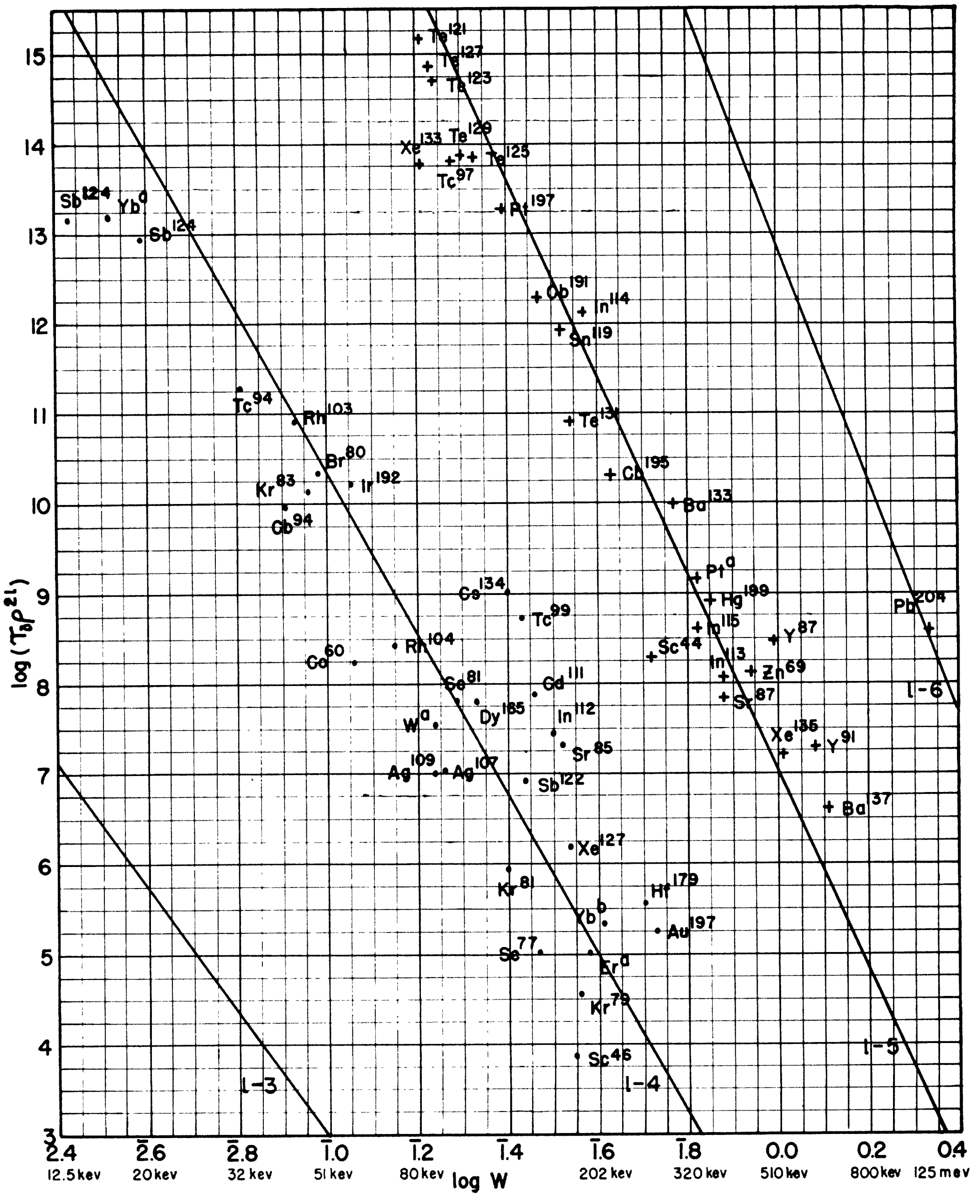

FIG. 3. Values of log ($\tau_\gamma\rho^{2l}$) as a function of log W.

on simpler approximations when dealing with either K conversion coefficients for energies below 150 kev or any L conversion coefficients.

There are two sources for the approximate K conversion coefficients. A non-relativistic theory of electric K shell conversion was developed by Hebb and Uhlenbeck[5] and Dancoff and Morrison;[6] the calculations from this theory are available in the tabulation of Hebb and Nelson.[7] A comparison of these theoretical predictions with those of Rose (in the region above 150 kev) shows that the approximate results are always low and that they improve as the energy decreases. However, even at energies as low as 150 kev, errors as large as 70 percent exist. The magnetic K conversion coefficients can be estimated from Drell's calculations,[8] which give the ratio of the magnetic K to electric K conversion. Comparison with Rose's data shows that these approximate results are also in error; Drell's ratio is always low and may be in error by a factor as large as 3.

The approximate calculations for L shell conversion coefficients have been made by Hebb and Nelson[6] for the electric case and by Lowen and Tralli[9] and Drell[10] for the magnetic case. Although the calculations for the K and L shells are similar, the deviation of the derived

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[5] M. H. Hebb and G. E. Uhlenbeck, Physica **5**, 605 (1938).

[6] S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939).

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

[8] S. D. Drell, Phys. Rev. **75**, 132 (1949).

[9] I. S. Lowen and N. Tralli, Phys. Rev. **75**, 529 (1949).

[10] S. D. Drell, private communication.

K to L ratio from the true value is difficult to estimate; the possibility of considerable discrepancies cannot be excluded.

The l value assigned by the correlation of Fig. 3 can indicate either only electric 2^l pole radiation or a combination of electric 2^l and magnetic 2^{l-1} pole radiation. This ambiguity exists because of the selection rules which depend on parity considerations. If the transition occurs between two energy levels which have an angular momentum difference of S, then the multipole order of the radiation must be at least S. If the parity change is $(-)^S$, then electric 2^S pole radiation can occur and the forbiddenness, $l=S$. In this case the transition is termed parity allowed and the radiation is essentially electric in nature. On the other hand, if the parity change is $(-)^{S+1}$, electric 2^S pole radiation cannot occur. Both magnetic 2^S and electric 2^{S+1} pole radiation can occur with probabilities which are of the same order of magnitude and $l=S+1$. In these parity forbidden cases, the relative amounts of electric and magnetic radiation are not predictable.

Despite this uncertainty, it might still be possible to check the theory if adequate experimental data were available on both the internal conversion coefficient and the K to L ratio. The procedure for carrying out this check would consist of using the lifetime to fix the l value. It would then often be possible to use one of the two pieces of internal conversion data to fix the relative amounts of electric and magnetic radiation. The other experimental determination would then provide an independent check of these assignments.

Unfortunately, the available experimental data are inadequate for this type of analysis. There are very few internal conversion coefficients that have been measured with sufficient precision. In many cases, the gamma-rays have not even been observed; in the other cases, the precision is limited by the lack of information on the relative efficiencies for detecting gamma-rays and electrons. While many K to L ratios have been determined, most of these were investigated with early beta-ray spectrographs that were incapable of giving precise ratios. The broad line shapes, the weak sources and the unknown efficiency characteristics of the detectors all limited the precision.

The data that have been reported for the isomers in the $l=4$ and $l=5$ groups are collected in Table I. The theoretical values that are listed were calculated from the sources which have been discussed. The experimental K to L ratios marked with asterisks are crude estimates based on published spectrograms. Although

TABLE I. Data for the isomers in the $l=4$ and $l=5$ groups.

				K to L Ratio			Conversion coefficient		
					theoretical				
Z	Element	A	kev	experimental	Electric	Magnetic	Experimental	$\alpha_K+\alpha_L$	$\beta_K+\beta_L$
					$l=4$				
27	Co	60	59	4±0.5	2.6	7.0			
34	Se	81	99	4	2.6	7.2			
35	Br	80	49	2*	0.59	4.5			
36	Kr	79	187	10*	4.6	8.5			
36	Kr	81	127	4*	3.1	7.6			
36	Kr	83	46	1	0.43	4.0			
41	Cb	94	41.4	.31	0.11	2.8			
43	Tc	99	136	4*	1.8	6.8			
47	Ag	107	94	.92±.06; 1	0.58	4.7	99; 75^{+200}_{-45}	145	44
47	Ag	109	88	1.01±.06	0.49	4.4	19±3	214	37
48	Cd	111	148	10*	1.4	6.3			
54	Xe	127	175	5*	1.2	6.2			
55	Cs	134	150	.64	0.55	5.4			
66	Dy	165	109	0.08	0.10	2.8			
79	Au	197	273	3.4	0.51	4.8			
					$l=5$				
21	Sc	44	270	8±4	19.5	9.2	0.07±0.012; 0.5	0.72	0.15
30	Zn	69	440				0.01 to 0.1	0.091	0.056
38	Sr	87	386	6 to 7	5.6	8.3	0.15	0.32	0.58
43	Tc	97	97	2	0.45	4.0			
49	In	113	390	5.4	3.0	7.2	2.3±0.6	0.47	0.58
49	In	114	190	1.0±0.1	1.03	5.4	>9	23	14
49	In	115	340	5.0±0.5	2.64	6.8	1±0.3	0.88	0.97
52	Te	121	82	0.75	0.07	1.8			
52	Te	123	88.5	0.68	0.10	1.9			
52	Te	125	109	1.5	0.21	3.0			
52	Te	127	86	0.75±0.25	0.088	1.9			
52	Te	129	102	1.0	0.16	2.9			
52	Te	131	177	2	0.69	4.8			
54	Xe	135	520				0.2	0.15	0.24
56	Ba	133	300	3.18	1.33	6.1	2.4±0.6	2.25	2.8
56	Ba	137	663	4.8±0.3	3.9	7.9	0.12	0.068	0.11
78	Pt	197	126	$\ll 1$	0.01	1			
80	Hg	199	362	2.3*	0.41	4.4			

the accuracy of the experimental K to L ratios is often quite poor, it seems safe to assume that the listed values correctly indicate which component is larger. In the case of the reported internal conversion coefficients, the general accuracy claim is only one of order of magnitude.

A study of the data in Table I shows that the experimental results do not contradict the theory nor the l value assignments. In general, the theoretical values can be made to agree with the experimental values by assuming appropriate arbitrary quantities of electric 2^l and magnetic 2^{l-1} radiations. However, the significance of this agreement is very questionable since the lack of precision and the existence of the arbitrary adjustment make other l values equally acceptable.

The data in Table I do show one characteristic of isomers which may be significant. A large majority of the transitions have K to L ratios which indicate that magnetic radiation occurs (i.e., most of the transitions are parity forbidden). Because of the large difference between electric and magnetic K to L ratios, this conclusion seems relatively sound in spite of the possible uncertainties. This apparent predominance of parity forbidden transitions, if substantiated by future experiments and more trustworthy theoretical calculations, might further aid in evaluating the "shell" models of nuclei.

Note added in proof.—(4) Additional internal conversion data have appeared on Cd^{111}, Hf^{179}, Ta^{182}, Ba^{137}, Pt^{197}, Hg^{199}, and Pt^{a}. These new data are mostly values of the K to L ratio. With the exception of Ta^{182} and Cd^{111}, these isomers seem to have parity forbidden transitions.

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Experiments on the Effect of Atomic Electrons on the Decay Constant of Be^7. II

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A comparison of the decay constants of Be^7 in beryllium oxide and in beryllium fluoride has given $\lambda(BeO)-\lambda(BeF_2)=(+1.375\pm0.053)10^{-3}\lambda(BeO)$ thus showing a definite effect of the chemical binding on the radioactive decay constant.

IN previous papers on this subject[1] we pointed out the possibility of altering the half-life of Be^7 by chemical binding and we compared the decay constants of beryllium metal and beryllium oxide.[2] This experiment was not too conclusive because the effect $\lambda(BeO)-\lambda(Be)=(-3.0\pm1.8)10^{-4}\lambda(Be)$ was only about 1.7 times the standard deviation of the measurement and for this reason we decided to try another pair of compounds, and to use stronger samples. Our choice was beryllium fluoride and beryllium oxide and this time we have observed a definite effect, namely $\lambda(BeO)-\lambda(BeF_2)=(+1.375\pm0.053)10^{-3}\lambda(BeO)$.

The apparatus used was the same as that described in reference 2, the only change being that we enclosed the ionization chambers by a thermally insulating box. This provision kept the temperature of the two chambers equal to each other and effectively reduced the tendency of ambient temperature gradients to alter the relative sensitivity of the chambers. This effect of the temperature gradients was inconvenient, although it did not affect the results of the previous experiments because samples were interchanged between the chambers every 20 minutes. Also the positioning of the sample was carefully rechecked in order to have the sources in a position in which they would give a maximum of ionization. Small displacements from this position affect the sensitivity of the chamber only by a term quadratic in the displacement.

The Be^7 was obtained from a proton bombardment of lithium with the 60-inch Crocker cyclotron. The beryllium was purified as follows.

The bombarded lithium was dissolved in dilute hydrochloric acid containing approximately 20 milligrams of beryllium. Carriers of cadmium, cobalt, copper, nickel and zinc were added and the beryllium was precipitated by excess ammonium hydroxide from a hot solution. After cooling, the precipitate of beryllium hydroxide was washed two times by dilute ammonium hydroxide. The precipitate was redissolved in dilute nitric acid and precipitation by ammonium hydroxide after the addition of the carriers was repeated four times.

The last by-product of added carriers had a small amount of activity, but on adding to it normal beryllium and reseparating no activity was found in the fraction containing the carrier and all went with the inactive beryllium, showing that the activity was due to Be^7 and not to isotopes of the carrier. At this point an

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[1] E. Segrè, Phys. Rev. **71**, 274 (1947).

[2] E Segrè and C. Wiegand, Phys. Rev. **75**, 39 (1949).