

²²M. H. Brennan and A. M. Bernstein, *Phys. Rev.* **120**, 927 (1960).

²³N. B. Gove, in *Nuclear Spin Parity Assignments*, edited by N. B. Gove and R. L. Robinson (Academic, New York, 1966), p. 83.

²⁴J. B. Ball and C. B. Fulmer, *Phys. Rev.* **172**, 1199 (1968).

²⁵J. McDonald and S. Malmskog, private communication.

²⁶J. B. Ball, R. L. Auble, and P. G. Roos, *Phys. Rev. C* **4**, 196 (1971).

²⁷M. E. Bunker, B. J. Dropesky, J. D. Knight, and J. W. Starner, *Phys. Rev.* **127**, 844 (1962).

²⁸T. Yamaya, Y. Nakagome, Y. Hiratate, S. M. Lee, S. Iwasaki, T. Tohei, and S. Morita, *Nucl. Phys.* **A126**, 449 (1969).

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Decays of the Even-Even Lead Isomers: ^{202m}Pb and ^{204m}Pb

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The decays of the 9^- isomers, ^{202m}Pb and ^{204m}Pb , have been investigated using Ge(Li) detectors in a variety of singles and coincidence configurations. Improved conversion coefficients were obtained for two of the transitions following the decay of ^{204m}Pb , but no new, weak transitions were seen. Considerable improvement and enlargement of the ^{202m}Pb decay scheme was possible. We have been able to make unambiguous multipolarity assignments for 15 of the 18 transitions following its decay and to place all 18 in a consistent decay scheme. States in ^{202}Pb were established (following the 90.5% isomeric-transition decay) at 0 ($J^\pi=0^+$), 960.70 (2^+), and 1382.82 (4^+), 1623.1 (4^+), 1915.2 (4^+), 2040.3 (5^-), and 2169.8 keV (9^-). Those in ^{202}Tl (following the 9.5% electron-capture decay) lie at 0 (2^-), 490.47 (4^-), 950.19 (7^+), 1098.7 (6^+ , 7^+ , 8^+), 1340.1 (8^+), 1552.1 (8^+ [9^+]), and 1675.6 keV (9^+ , 8^+). Interpretation is made concerning the nature of the states in ^{202}Pb and even-even Pb isotopes and also in ^{202}Tl and odd-odd Tl isotopes on the basis of the simple shell-model plus pairing forces.

I. INTRODUCTION

The Pb isomers were the first case of even-even isomerism to be observed. Besides being interesting in their own right, they also provided (and still provide) an advance opportunity to study some of the lower-lying states in the Pb isotopes, states that presumably should be amenable to explanation in simple shell-model terms. ^{204m}Pb and ^{202m}Pb , especially, have proven useful for building up skeletal level schemes for ^{204}Pb and ^{202}Pb before these could be studied by transfer reactions or the much more complicated decays of ^{204}Bi and ^{202}Bi . In the present work we reexamine the decays of ^{204m}Pb and ^{202m}Pb , using Ge(Li) γ -ray detectors for the first time. We then correlate our findings with those from other types of experiments when possible and examine the trends in the even-even Pb isotopes not too far from stability. We are also able to make arguments concerning

the structures of some of the odd-odd ^{202}Tl states populated by the electron-capture (ϵ) decay branch of ^{202m}Pb .

The decay scheme of 66.9-min ^{204m}Pb has been the subject of many previous studies, beginning with Sunyar *et al.* in 1950.¹ An excellent summary of the intervening work is to be found in Hyde, Perlman, and Seaborg² and thus will not be repeated. It should be mentioned, however, that the decay scheme remains essentially that proposed by Fritsch in 1956.³ Our study was aimed chiefly at finding weak, previously unobserved transitions. None was found, but we were able to obtain better values for some of the internal-conversion coefficients, using a unique single-crystal Ge(Li) "conversion-coefficient spectrometer" developed in this laboratory.⁴

The existence of an isomer associated with ^{202}Pb was first reported by Maeder and Wapstra in 1954.⁵ They produced this isomer by bombarding

natural thallium with 25-MeV deuterons to induce the reaction $^{203}\text{Tl}(p, 3n)^{202m}\text{Pb}$. The half-life of this activity was set at 3.5 ± 0.1 h. In further studies using NaI(Tl) scintillation spectrometers and conversion-electron detectors, Maeder *et al.*⁶ assigned five transitions to the decay of this nuclide and proposed a decay scheme having four excited states in ^{202}Pb at 421, 1384, 2042, and 2171 keV. Among the ambiguities remaining in this study, no clear indication of the correct ordering of the 963-421-keV γ -ray cascade (the intense cascade connecting the ground state and the first two excited states) was found. Their prediction of the relative position of these two transitions had to be based, therefore, on a comparison with the then postulated systematics of the first excited states in other even-even nuclei. The $E2$ multipolarity given these transitions established spin and parity assignments of 2^+ and 4^+ for the first two excited states. The feeding of the 1384-keV second excited state by a 788-keV $E5$ isomeric transition and a 658-keV- $E1$ -128-keV- $E4$ cascade led to a 9^- assignment for the isomeric state and a 5^- assignment for the state at 2042 keV.

In 1955 Bergström and Wapstra⁷ supported these findings and began the investigation of the ϵ -decay branch. They reported the existence of three transitions converted in ^{202}Tl , having energies of 460, 389, and 401 keV. In the following year McDonnell *et al.*⁸ conducted an extensive investigation of both branches populated in the decay of ^{202m}Pb . Besides determining more accurate values for the transition energies, this group was able to establish the ordering sequence of the 961- and 421-keV transitions. By comparing the relative intensities of these two transitions in the decay of ^{202m}Pb and ^{202}Bi , the ordering previously postulated by Maeder *et al.* was reversed and the 961-keV transition was shown to be the transition between the ground and first excited states. This group also added a second 4^+ excited state. This state, fed from the metastable state by a 547.2-keV $E5$ transition, was placed at 1623.8 keV. An $M1$ transition of 240.3 keV was also reported and attributed to the transition between the two 4^+ states. This work indicated a 9.5% ϵ branching ratio, with the following sequence of levels in ^{202}Tl being populated: 2^- (0 keV), 4^- (490.4 keV), 7^+ (950.2 keV), 6^+ (1099.1 keV), and 8^+ (1340.1 keV). A further level at 1675.7 keV, feeding the 1340.1-keV level by means of a 335.6-keV $M1$ transition, was also tentatively suggested. With the exception of the 2.0 ± 1.5 -nsec half-life of the 1384-keV state in ^{202}Pb measured by Johansson, Alväger, and Zuk⁹ in 1959, the decay scheme as proposed by McDonnell *et al.* has remained unaltered until the present study.

II. SOURCE PREPARATION

The sources for our ^{204m}Pb study were prepared by bombarding separated isotope ^{206}Pb (97.2% enriched, obtained from Oak Ridge National Laboratory) with 30-MeV protons from the Michigan State University sector-focused cyclotron to induce the reaction $^{206}\text{Pb}(p, 3n)^{204}\text{Bi}$. Typically, 100-mg targets were bombarded with a 1- to 2- μA beam for approximately one-half hour. The ^{204}Bi , which has a half-life of 11.2 h, decays by electron capture to excited states in ^{204}Pb , and $\approx 13\%$ of its decay populates the metastable state.¹⁰ After bombardment, the ^{204}Bi activity was separated from the Pb target material by dissolving the target in 6 N HNO_3 and precipitating the Pb with fuming HNO_3 . It was then taken up in 6 N HCl and loaded onto a heated (1.5-mm-diam by 5-cm-long Dowex 1×8 200-mesh) anion-exchange column. The ^{204m}Pb activity was then eluted with 0.3 M HCl at the end of successive 1-h intervals.¹¹ The few drops of activity obtained in this manner were vaporized through 3-mm-diam masks onto a 0.25-mil Pt backing for use in the conversion-coefficient spectrometer. Using this method, many sources could be obtained from a single bombardment because of the 11.2-h half-life of ^{204}Bi .

Our ^{202m}Pb sources were prepared by bombarding natural thallium foils and enriched ^{203}Tl (70% ^{203}Tl , obtained from Oak Ridge National Laboratory) with a 16-MeV proton beam from the Michigan State University cyclotron. The induced reaction $^{203}\text{Tl}(p, 2n)^{202m}\text{Pb}$ produced the desired activity in good yield with a 7-10-min bombardment at a beam current of 0.5 μA . Chemical separations were performed on approximately half of the sources used. All sources were allowed to age for a period of 2 to 3 h before counting in order to minimize contamination from shorter-lived activities.

III. EXPERIMENTAL METHODS

The single-crystal Ge(Li) conversion-coefficient spectrometer used in our measurements on ^{204m}Pb was manufactured in this laboratory. The detector was a planar drift device with a drift depth of ≈ 7 mm and an active volume of ≈ 0.4 cm^3 . It was mounted in a conventional dipstick cryostat with a window of 0.25-mil Havar foil. The design and operation of this conversion-coefficient spectrometer has been discussed previously in greater detail by Gruhn *et al.*⁴

The γ rays from the decay of ^{204m}Pb and ^{202m}Pb were studied using Ge(Li) detectors with photopeak efficiencies of 2.5 and 10.4% at 1332 keV, both

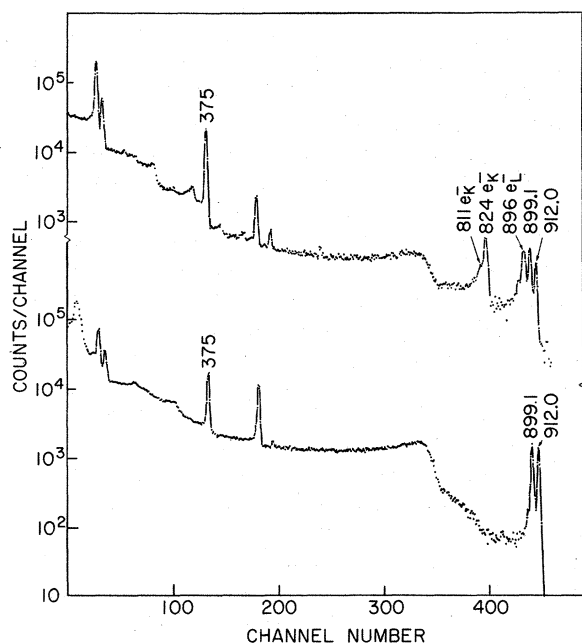


FIG. 1. ^{204m}Pb spectra obtained with the Ge(Li) conversion-coefficient spectrometer. Only lines from ^{204m}Pb decay are labeled. Top: spectrum showing both γ rays and electrons. Bottom: spectrum taken through an Al absorber to remove the electrons.

with an optimum resolution of ≈ 2.1 keV at that same energy. In the singles runs, a PDP-9 computer interfaced with a 4096-channel analog-to-digital converter served as the multichannel analyzer for data collection. The energies and intensities of the transitions were then determined by means of a spectrum-analysis routine employing a live-display computer program.¹² For details of the methods of data reduction and analysis used, see, for example, Doebler, McHarris, and Kelly.¹³

For coincidence measurements the 10.4% detector mentioned above was coupled with a 4.6% efficient Ge(Li) detector having an optimum resolution of 1.9 keV at an energy of 1332 keV. The addresses of coincidence events from both sides of the system were processed and listed on magnetic tape in pairs. The coincidence spectra were

later recovered off line as gated slices. The basic format of the two-dimensional "megachannel" Ge(Li)-Ge(Li) spectrometer system used has been previously presented.^{13,14}

IV. ^{204m}Pb DECAY

The relatively straightforward decay scheme of ^{204m}Pb remains essentially that proposed by Fritsch³ in 1956. However, recent studies in our laboratory¹⁰ on ^{204}Bi decay have shown a number of new states in ^{204}Pb that lie below the 2185.4-keV metastable state. Using the larger (including the 10.4% efficient) Ge(Li) detectors now available, an attempt was made to determine if these states might be weakly populated in the decay of ^{204m}Pb . Although we were unable to detect any new transitions, we did measure directly several conversion coefficients, viz., those for the 899.1-keV $E2$ and 912.0-keV $E5$ transitions, using the unique single-crystal Ge(Li) conversion-coefficient spectrometer described in Sec. III.

Two spectra of ^{204m}Pb taken with the Ge(Li) conversion-coefficient spectrometer are shown in Fig. 1. The upper spectrum contains contributions from both γ rays and electrons, while the lower spectrum, taken with an Al absorber placed between the source and the detector, contains only the γ rays. By hand stripping the lower from the upper spectrum, the areas of the 899.2- and 912.0-keV γ -ray and conversion electron peaks were found. The K conversion coefficients could then be determined easily using the calculated areas and an efficiency curve for our spectrometer obtained by correlating theoretical conversion coefficients¹⁵ with γ -ray and electron-conversion peak intensities of several well-known standards (cf. Ref. 4).

The experimentally determined α_K for both transitions and the K/L ratio for the $E5$ transition are given in Table I along with the theoretical values of Hager and Seltzer.¹⁵ Agreement between the measured and theoretical values is seen to be good. (No reliable K/L ratio for the 899.1-keV $E2$ transition could be extracted because of the weakness of the L lines and the complexity of that portion of the spectrum.)

TABLE I. ^{204m}Pb conversion data.

Transition energy (keV)	Multipolarity	α_K		K/L	
		Experimental	Theoretical (Ref. 15)	Experimental	Theoretical (Ref. 15)
899.1 \pm 0.1	$E2$	0.0072 \pm 0.0022	0.0065
912.0 \pm 0.1	$E5$	0.0545 \pm 0.0045	0.046	1.66 \pm 0.25	1.73

The decay scheme of ^{204m}Pb is shown in Fig. 2, where it is compared with the decays of the other even-even Pb metastable isomers.

V. ^{202m}Pb DECAY

A. Level Placements

A ^{202m}Pb γ -ray singles spectrum is presented in Fig. 3. 18 γ rays having a half-life comparable to that of ^{202m}Pb were observed, and only these have been labeled. The energies and intensities of these γ rays are given in Table II, where they are compared with the conversion-electron results obtained by McDonnell *et al.*⁸ The listed uncertainties in energies are a reflection of three factors: the uncertainty in the energies of the calibration standards used, the height of a particular photopeak above the background level, and the reproducibility of our measurements over many independent spectra. The values of the relative transition intensities have been averaged over several spectra, and the deviations presented are based on the reproducibility of these intensities and the

uncertainties in our experimentally determined efficiency curves. Of the transitions in this table, those with energies of 124.75, 211.92, 417.3, 601.95, and 954.5 keV have not been reported previously. Of these, the 417.3-keV transition was seen to fit into the existing decay scheme between the 2040.3- and 1623.1-keV levels in ^{202}Pb . The existence of such a transition had been predicted by McDonnell *et al.*,⁸ but no experimental verification was available at that time. Using energy sums and differences we were able to incorporate the remaining four transitions by adding two new levels, one at 1915.2 keV in ^{202}Pb and one at 1552.1 keV in ^{202}Tl . These two levels have been included in the decay scheme given in Fig. 2. In this decay scheme all energies are given in keV and (total) transition intensities are given in percent of the total ^{202m}Pb disintegrations. A word on the intensities: Two of the major transitions, at 960.70 and 129.1 keV, have larger than usual errors associated with their intensities, the 960.70-keV one because of contaminant peaks falling near it in the γ -ray spectrum and the 129.1-

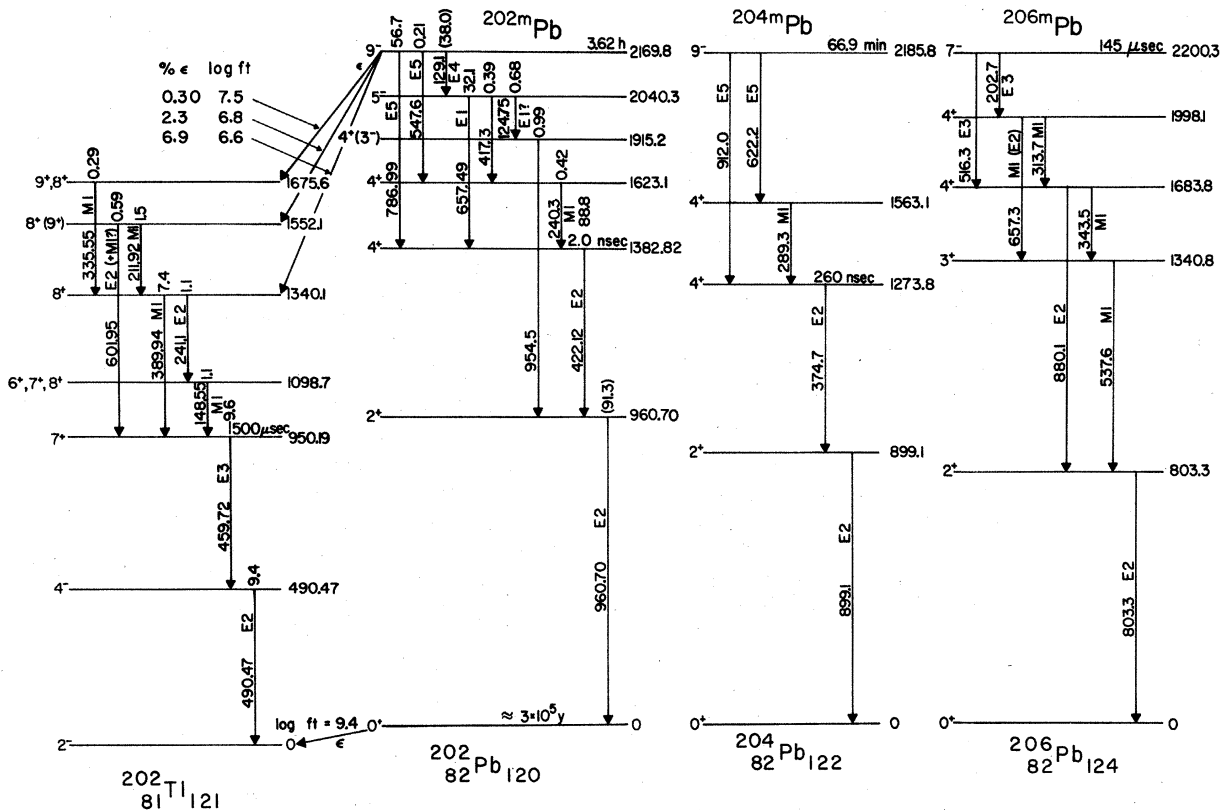


FIG. 2. Decay schemes of the even-even Pb metastable isomers. The decay scheme of ^{202m}Pb results from this work; that of ^{204m}Pb , from Ref. 3 and this work; and that of ^{206m}Pb , from D. E. Alburger and M. H. L. Pryce, Phys. Rev. 95, 1482 (1954). All energies are given in keV and the (total) transition intensities in the ^{202m}Pb decay scheme are given in percent of the disintegrations of the parent state.

TABLE II. Energies and relative intensities of transitions following the decay of ^{202m}Pb .

E_γ (keV)	This work I_γ	McDonnell <i>et al.</i> Transition E (keV) ^a		Relative conversion- line I^b
		Converted in Pb	Converted in Tl	
124.75 ± 0.09	1.1 ± 0.3
129.1 ± 0.2	0.08 ± 0.03	129.3 ± 0.1	...	690 ± 50 ($\sum L$)
148.55 ± 0.15	0.45 ± 0.15	...	148.9 ± 0.1	17 ± 3
211.92 ± 0.07	1.5 ± 0.3	...	212.1 ± 0.5 ^c	2.6 ^c (L_I)
240.8 ± 0.1 ^d	2.2 ± 0.3 ^d	{ 240.3 ± 0.1	4.0 ± 0.3
			241.1 ± 0.1	2.5 ± 0.2
335.55 ± 0.10	0.45 ± 0.10	...	335.6 ± 0.2	1.3 ± 0.1
389.94 ± 0.07	12.4 ± 1.0	...	389.9 ± 0.2	28 ± 3
417.3 ± 0.2	0.8 ± 0.2
422.12 ± 0.06	172 ± 9	422.1 ± 0.2	...	66 ± 5
459.72 ± 0.07	17.3 ± 1.0	...	459.8 ± 0.2	9 ± 1 ($\sum L$)
490.47 ± 0.07	18.4 ± 1.0	...	490.4 ± 0.3	1.7 ± 0.2 ($\sum L$)
547.6 ± 0.3	0.25 ± 0.08	547.2 ± 0.3	...	1.1 ± 0.1 ($\sum L$)
601.95 ± 0.08	1.2 ± 0.1	...	602.1 ^c	0.25 ^c
657.49 ± 0.06	65 ± 3	657.6 ± 0.3	...	4.4 ± 0.4
786.99 ± 0.06	≡ 100	787.2 ± 0.4	...	≡ 100
954.5 ± 0.2	2.0 ± 0.4
960.70 ± 0.15	184 ± 15	964.4 ± 0.4	...	13 ± 1

^a The deviations in these energies were calculated by us, using the precision (5 parts in 10^4) claimed by McDonnell *et al.*

^b Relative intensities are for the K conversion lines unless otherwise specified.

^c For these two transitions we assigned lines from McDonnell *et al.*'s table of unassigned conversion lines, using our photon energies as a guide. The estimate on the precision in energy for the 212.1-keV transition is based on the scatter in their L_I , M_I , and N_I energies. No meaningful estimate of the precision in energy for the 602.1-keV transition or for the intensity of either transition could be made.

^d In our measurements we were unable to delineate between the 240.3- and 241.1-keV γ rays emitted in the decay of ^{202m}Pb . The energy and relative intensity given here are for the total doublet peak.

keV one because of uncertainties in the electron data (only the L_I line was seen). Because of this relatively large uncertainty in the intensity for the 960.7-keV transition, we used the average of the 459.72- and 490.47-keV intensities vs the sum

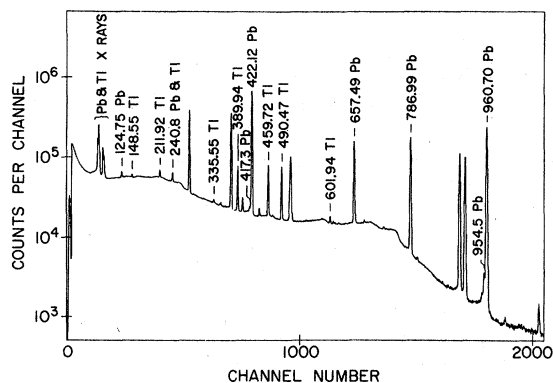


FIG. 3. ^{202m}Pb singles γ -ray spectrum taken with a 10.4% efficient Ge(Li) detector. Only the γ rays that we attribute to ^{202m}Pb decay are labeled. Because of the low intensity of the 129.1- and 547.6-keV transitions, they are not labeled in this figure.

of the 422.12- and 954.5-keV intensities to obtain 9.5% ϵ and 90.5% isomeric transition decay. Adding up the ϵ decay as inferred from γ -ray feedings yielded only 9.2%, so these ϵ feedings were renormalized to 9.5%. Otherwise, all intensities shown on the decay scheme are strictly experimental. The $\log ft$ values were calculated on the assumption¹⁶ that the ground state of ^{202}Tl lies ≈ 50 keV below that of ^{202}Pb .

To test the consistency of these new assignments a γ - γ coincidence experiment was performed. A summary of the results of this experiment is given in Table III, where it can be seen that most of the previous placements are corroborated. The integral coincidence spectra and five of the gated coincidence spectra of particular interest are presented in Fig. 4. Parts (b) and (c) show clearly the existence of a 124.75-954.5-keV cascade. Our assignment of this cascade to ^{202}Pb rather than ^{202}Tl was substantiated when spectra gated on the Pb K x rays increased the relative intensities of these peaks with respect to those of transitions known to occur between levels in ^{202}Tl . From parts (e), (f), and (g), the 211.92-keV γ is seen to

TABLE III. Summary of the γ - γ coincidence results for ^{202m}Pb .

Gate energy (keV)	γ rays in coincidence (keV)
124.7	954.5, 960.7
148.6	211.9, 241.1
211.9	241.1, 389.9
≈ 241	148.6, 211.9, 417.3
335.6	241.1, 389.9
389.9	211.9, 335.6
422.1	240.3, 657.5, 787.0, 960.7
459.7	490.5
490.5	459.7
657.5	129.3, 422.1, 960.7
787.0	422.1, 960.7
954.5	124.7
960.7	124.7, 240.3 (weak), 422.1, 657.5, 787.0, 954.5

feed the 1340.1-keV level in ^{202}Tl , which deexcites by either a 389.94- or a 241.1-keV transition. No coincidence data were available for the 601.95-keV transition, because of the 0.5-msec half-life of the 950.19-keV state in Tl. By gating on the Tl K x rays, however, it was possible to place both the 211.92- and 601.95-keV transitions in the ϵ branch of ^{202m}Pb decay.

B. Spin and Parity Assignments

Combining our work and that of McDonnell *et al.*,⁸ we have unambiguous multipolarity assignments for all but three of the 18 γ transitions that follow ^{202m}Pb decay. Assuming the 422.12-keV transition from the second to the first excited state of ^{202}Pb to be a pure $E2$ transition, we normalized our photon data to their electron data, using the calculated α_K of Hager and Seltzer. In Table IV

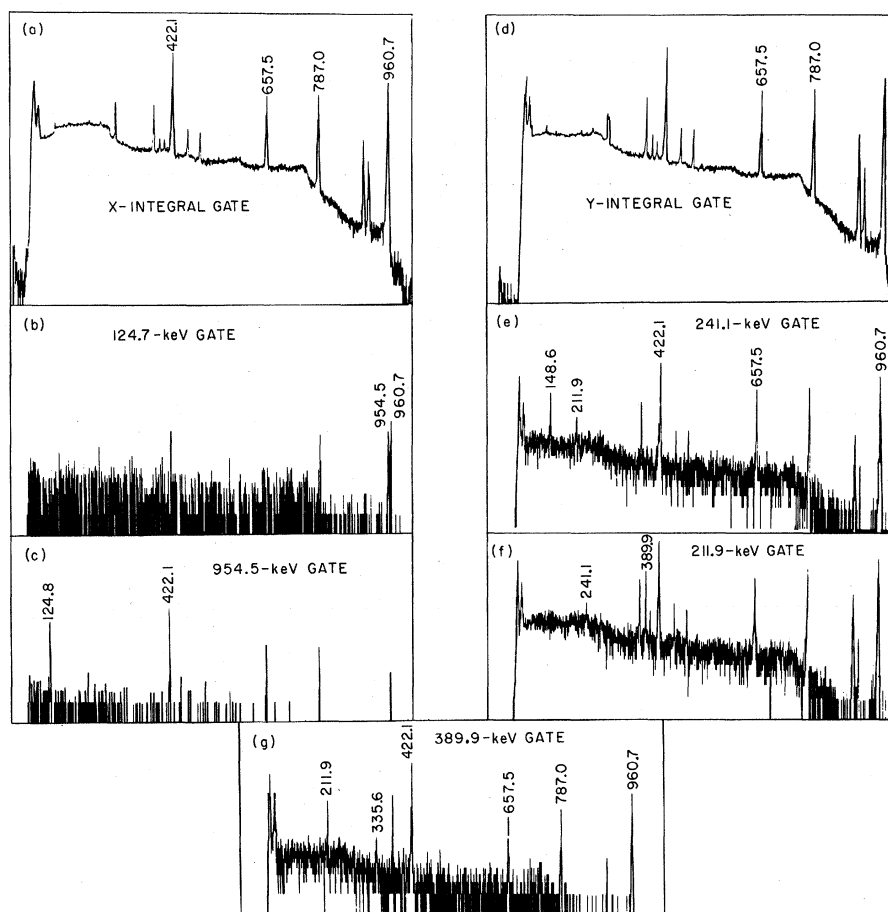


FIG. 4. Two-dimensional "megachannel" γ - γ coincidence spectra for ^{202m}Pb decay. Part (a) shows the total spectrum seen by the 4.6% efficient detector (the X detector) in coincidence with all events gated by the 10.4% efficient detector (the Y detector); similarly, part (d) shows the total spectrum seen by the Y detector in coincidence with all events gated by the X detector. The other five parts [(b), (c), (e)-(g)] show five gated slices (background subtracted), using the X detector for the gates and the Y detector for the displays.

we show our multipolarity assignments, based for the most part on α_K 's, and compare them with the similar assignments made by McDonnell *et al.*, which were based primarily on K/L and L subshell ratios. In all cases where both groups were able to make unambiguous assignments, the results are in agreement. We also show our α_K values in Fig. 5 plotted as points against the calculated values of Hager and Seltzer. As the deviation between the theoretical curves for Pb and Tl is small in comparison to the uncertainty in our experimentally determined α_K 's, we have simply plotted our experimental α_K values against the calculated conversion-coefficient curves for Pb. These will be used further, especially with respect to $M1/E2$ mixing ratios, when we discuss the states in Sec. VII.

1. States in ^{202}Pb

Our findings are consistent with the previous spin-parity assignments by McDonnell *et al.* for the states in ^{202}Pb , viz., 0^+ (0 keV), 2^+ (960.70), 4^+ (1382.82), 4^+ (1623.1), 5^- (2040.3), and 9^- (2169.8). Although we can add arguments to theirs to corroborate many of these assignments, it is not necessary to detail these here.

The additional state at 1915.2 keV can have its assignments narrowed down even though the multipolarities of the 124.75- and 954.5-keV transitions passing in and out of it are not known as well as most of the other transitions. The ordering of this cascade is made unambiguous, incidentally, because the opposite ordering would require a moderately high-spin state at 1085.5 keV, which

TABLE IV. ^{202m}Pb multipolarity assignments.

E (keV)	Exp. α^a	Theor. α (Ref. 15) ^b	Assignment ^c	
			This work	McDonnell <i>et al.</i> (Ref. 8)
Transitions in ^{202}Pb				
124.75	...	$\left\{ \begin{array}{l} 2.0 \times 10^{-1} E1 \\ 4.4 \times 10^{-1} E2 \end{array} \right\}$	$E1?$ ^d	...
129.07	$\approx 7.0 \times 10^2$ ($\sum L$) ^e	3.7×10^2	High multipole ^e	$E4$
240.3	...	6.7×10^{-1}	...	$M1$
417.3	...	1.2×10^{-2}	$E1?$...
422.12	$\approx 3.0 \times 10^{-2}$	3.0×10^{-2}	"Pure $E2$ "	$E2$
547.6	3.5×10^{-1}	3.9×10^{-1} ^f	$E5$	$E5$
657.49	5.3×10^{-3}	4.6×10^{-3}	$E1$	$E1$
786.99	7.9×10^{-2}	8.6×10^{-2} ^f	$E5$	$E5$
954.5
960.70	5.6×10^{-3}	5.8×10^{-3}	$E2$	$E2$
Transitions in ^{202}Tl				
148.55	2.9	2.4	$M1$	$M1$
211.92	1.4×10^{-1} (L_1)	1.3×10^{-1}	$M1$...
241.1	...	$\left\{ \begin{array}{l} 1.1 \times 10^{-1} E2 \\ 6.1 \times 10^{-1} M1 \end{array} \right\}$...	$E2$
335.55	2.3×10^{-1}	2.5×10^{-1}	$M1$	$E1? M1?$
389.94	1.8×10^{-1}	1.7×10^{-1}	$M1$	$M1$
459.72	4.1×10^{-2} ($\sum L$)	4.6×10^{-2}	$E3$	$E3$
490.47	7.3×10^{-3} ($\sum L$)	6.5×10^{-3}	$E2$	$E2$
601.95	1.6×10^{-2}	$\left\{ \begin{array}{l} 1.4 \times 10^{-2} E2 \\ 5.2 \times 10^{-2} M1 \end{array} \right\}$	$E2 (+M1?)$...

^a α_K except where noted.

^b Calculated values are for the K shell (except where the experimental α is for the L shell) and for the assigned multipolarity except where a choice is indicated.

^c For the most part the assignments of McDonnell *et al.* are based on K/L or L subshell ratios — for more details consult their Tables 1 and 2. We agree with most of their assignments and list an assignment of our own only when we have independent information, usually α_K .

^d This tentative $E1$ assignment is made on the basis of its being the only one that gives a reasonable intensity balance in and out of the 1915.2-keV level.

^e The uncertainty in this Exp. α_K is so large that we do not use it to make an assignment.

^f From L. A. Sliv and I. M. Band, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1965).

on the basis of systematics (cf. Ref. 10) is quite unlikely. A tentative $E1$ assignment for the 124.75-keV transition is the only one consistent with the intensity balance in and out of the 1915.2-keV state. This, in conjunction with the 954.5-keV transition to the 2^+ 960.70-keV state, would limit the assignment to 4^+ . Even allowing the full range of $E1$, $M1$, or $E2$ multipolarities for the two γ rays yields just 4^+ or 3^- for the state.

2. States in ^{202}Tl

Previous assignments for these states have been considerably less certain, so a discussion of each is in order here.

(a) *Ground state.* The ground state has been established rather convincingly as a 2^- state, based on its decay to ^{202}Hg .¹⁷ This assignment is consistent with systematics of the region, for all the odd-odd Tl isotopes between ^{194}Tl and ^{204}Tl appear to have this same 2^- ground state.

(b) *490.47- and isomeric 950.19-keV states.* The unambiguous assignments of the 490.47- and 459.72-keV transitions as $E2$ and $E3$, respectively, by both McDonnell *et al.* and ourselves rather strongly imply assignments of 4^- and 7^+ for these two states. That this is indeed a stretched cascade is indicated by the lack of appreciable $M1$ admixture in the $E2$ (in other odd-odd and odd-mass Tl isotopes $M1$'s tend to predominate over $E2$'s when they are possible¹³) and by the absence of any crossover transition. This sequence, $7^+ \rightarrow 4^- \rightarrow 2^-$, is the same as in the decay of other odd-odd Tl metastable isomers.¹⁸

(c) *The three capture states at 1340.1, 1552.1, and 1675.6 keV.* The $\log ft$ values for ϵ decay to

these three states places those decays into a "hindered" allowed or "normal-speed" first-forbidden classification. As first-forbidden transitions tend to compete favorably with allowed transitions in this nuclear region, one looks for them, and the $M1$ and/or $E2$ character of the γ transitions to the 7^+ state confirms the positive parity of these states. Thus, the ϵ decay points toward 8^+ , 9^+ , or 10^+ assignments.

The 389.94-keV $M1$ transition to the 7^+ state limits the assignment for the 1340.1-keV state to 8^+ . The apparent $M1$ admixture in the 601.95-keV $E2/M1$ transition to the 7^+ state also limits the assignment for the 1552.1-keV state to 8^+ ; the 211.92-keV $M1$ is consistent with this. If one does not believe the $M1$ admixture, 9^+ is also possible. The 335.55-keV $M1$ transition to the 8^+ 1340.1-keV state limits the assignment for the 1675.6-keV state to 9^+ or 8^+ .

(d) *1098.7-keV state.* The 148.55-keV $M1$ transition from this state to the 7^+ 950.19-keV state limits its assignments to 6^+ , 7^+ , or 8^+ . The existence of a 241.1-keV $E2$ transition feeding this state from the 8^+ 1340.1-keV state is consistent with this. McDonnell *et al.* chose 6^+ on the basis that the $M1$ and $E2$ were stretched transitions, but this is not necessarily true, so we retain all three possibilities.

VI. STATES IN EVEN-EVEN Pb ISOTOPES

$^{204}\text{Pb}_{122}$ and $^{202}\text{Pb}_{120}$ lie four and six neutron holes away from the ^{208}Pb doubly closed shell. Thus, full-fledged shell-model calculations for states in these nuclei are few and cumbersome, and one has to rely considerably on less exact methods, such as those of Kisslinger and Sorensen.¹⁹ It is beyond the scope of this paper to go into the details of the various calculations; yet it is very instructive to consider some of the more qualitative aspects of the states in these nuclei. This we do, using as guides the work of Kisslinger and Sorensen along with the calculations of True and Ford²⁰ for ^{204}Pb and Harvey and Clement²¹ for the lighter isotopes.

A. Metastable States

Using the simplification of only two uncoupled neutron holes, the 9^- states in both ^{204}Pb and ^{202}Pb can have only $[(\nu i_{13/2})^{-1}(\nu f_{5/2})^{-1}]_{9^-}$ as their major components. Other two-hole 9^- components, $[(\nu i_{13/2})^{-1}(\nu f_{7/2})^{-1}]$ and $[(\nu i_{13/2})^{-1}(\nu h_{9/2})^{-1}]_{9^-}$, lie high enough to preclude serious configuration mixing. More complex states and core-coupled components involving, say, an $i_{13/2}$ neutron hole with the 3^- octupole vibration are certainly possible and very likely present, although, as we

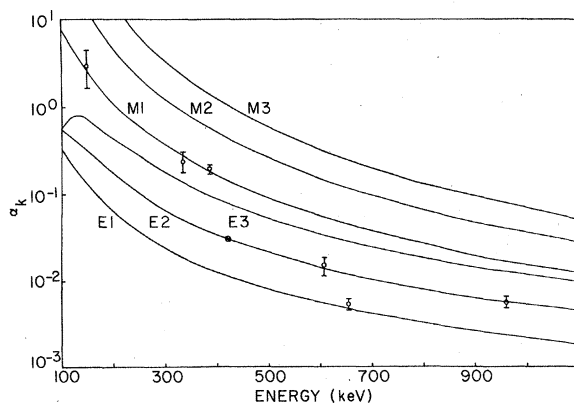


FIG. 5. Experimental α_K conversion coefficients (points) for the ^{202}Pb and ^{202}Tl γ transitions plotted against the theoretical curves (for Pb) obtained from the work of Hager and Seltzer (Ref. 15). The data were normalized assuming the 422.12-keV transition to be pure $E2$.

shall see below, the $E5$ isomeric transitions are remarkably good single-particle transitions, which can be taken to imply fairly simple configurations for the 9^- states.

Serious attempts have been made to locate 9^- metastable states in ^{200}Pb and ^{198}Pb , with both fast off-line¹³ and in-beam²² spectroscopy. None was found. However, 7^- metastable states were found²² in ^{202}Pb , ^{200}Pb , and ^{198}Pb at 2208, 2154, and 2142 keV, respectively. These are probably much more complex states and consist mostly of the configuration-mixed components, $[(\nu i_{13/2})(\nu p_{1/2})^{-1}]_{7^-}$, $[(\nu i_{13/2})^{-1}(\nu p_{3/2})^{-1}]_{7^-}$, and $[(\nu i_{13/2})^{-1}(\nu f_{5/2})^{-1}]_{7^-}$, similar to the 7^- state in ^{206}Pb . The lack of 9^- metastable states in ^{200}Pb and ^{198}Pb can be explained by the fact that the $p_{3/2}$ neutron-hole state is falling¹⁹ with respect to the $f_{5/2}$ state, so the 7^- states fall below the 9^- states.

B. Pairs of 4^+ States

Pairs of 4^+ states are populated in both ^{202}Pb and ^{204}Pb , and there are some interesting parallels and inconsistencies concerning these states. The $E5$ transition probabilities to these states are listed in Table V and compared with the single-particle estimates calculated using Moszkowski's equations.²³ In both nuclei the stronger transition, i.e., the one to the lower 4^+ state, has a transition probability remarkable close to the calculated value. This lends credence to the fact that these are nearly single-particle transitions. In both cases, also, the weaker, lower-energy $E5$ is retarded over the simple $E^{11/2}$ power factor by an additional factor of about 5. The simplest inference is that the ^{204}Pb 1273.8-keV state and the ^{202}Pb 1382.82-keV state are similar in nature, as are the ^{204}Pb 1563.1-keV state and the ^{202}Pb 1623.1-keV state.

It is tempting to think of the lower 4^+ states as consisting primarily of 4^+ members of the two-phonon quadrupole vibrational states, while the upper 4^+ states contain more two-particle character. This is an oversimplification, however, and cannot be pushed too far, and all of the states

are undoubtedly highly admixed and quite complex in nature. With respect to the lower states, in particular, there are conflicting pieces of data. First, the $4^+ \rightarrow 2^+$ $E2$ transition probabilities – the $E2$'s are not enhanced. For ^{202}Pb and ^{204}Pb we calculate²³ single-particle estimates of 0.6 and 0.8 nsec for the half-lives, to be compared with the experimental 2.0 and 260 nsec. The ^{204}Pb transition, in particular, is highly hindered, implying that the 4^+ state is not collective. (The 2^+ state is most likely a simple one-phonon quadrupole vibration, cf. Ref. 19.) Second, (p, t) transfer data – although complete angular distributions and analyses are not available, enough $^{206}\text{Pb}(p, t)^{204}\text{Pb}$ data do exist both at 22 MeV²⁴ and at 30 MeV²⁵ to show that this reaction populates the lower 4^+ state strongly and the upper 4^+ state almost not at all. Recent evidence^{26,27} shows quite clearly that the (p, t) reaction tends to favor populating collective states both in deformed and in single-closed-shell nuclei. This implies some collective nature for the 1273.8-keV state in ^{204}Pb .

Actually, both of the lower 4^+ states probably contain some collective components in their wave functions but not enough to insure the enhancement of the $E2$ transitions – and the factor of 100 difference in retardation of these transitions still needs explaining. (Note, however, that $E2$ transitions in general are not very enhanced in this region.) The only important low-lying two-particle configurations that can contribute are $[(\nu f_{5/2})^{-2}]_{4^+}$ and $[(\nu f_{5/2})^{-1}(\nu p_{3/2})^{-1}]_{4^+}$. If these were concentrated in the lower and upper states, respectively, the differences in the $E5$ transition probabilities are, qualitatively at least, in the correct direction; however, a thorough understanding of these states awaits much more sophisticated shell-model calculations.

C. Position of Ground State

The 2^+ first excited states of the even-even Pb isotopes have been established quite definitely as one-phonon quadrupole vibrational states,^{2,19} so we will not dwell on this here. However, a peculiarity in the positions of these states is readily seen from Fig. 2. As one moves *away* from the closed $N=126$ shell, the energies of the first excited states *increase*, contrary to what might be expected from arguments of the type that the nucleus gets softer with respect to vibrations as one moves farther away from closed shells. And these states continue to rise: The first 2^+ state lies at 1027 keV in ^{200}Pb and at 1064 keV in ^{198}Pb (Refs. 22, 28, and 29). A number of theorists, including those referenced in this paper, have duplicated this trend. This usually comes out of

TABLE V. $E5$ transition probabilities.

Nucleus	E_γ (keV)	$t_{1/2}$		Retardation
		Calc. ^a	Exp.	
^{204}Pb	912.0	39.8 min	67.7 min	1.7
	622.2	44.5 h	417 h	9.4
^{202}Pb	786.99	3.47 h	6.70 h	1.9
	547.6	190.6 h	1810 h	9.5

^a Reference 23. Since all are $9^- \rightarrow 4^+$ transitions, no statistical corrections were made.

complex calculations, however, where it is difficult to isolate as to specific cause. Perhaps over simply, but instructively, this can also be explained as a straightforward result of the increase in strength of the pairing force in moving away from the closed $N=126$ shell. Thus, it is really the ground state that is dropping in the lighter isotopes. Assuming a constant coupling constant (strength) for the pairing force here, we see that it is indeed weaker in ^{206}Pb than in the lighter isotopes. In ^{206}Pb , only the low-multiplicity $p_{1/2}$ orbit is available below the shell closure to participate in the neutron pairing stabilization. As one moves to the lighter isotopes, more and more orbits in higher multiplicity states become available, especially the $f_{5/2}$ orbits and near or below $A=200$ the $i_{13/2}$ orbits. Thus, as the single-closed proton shell tends to hold the nucleus rigid or spherical, the increasing neutron pairing lowers the ground states.

D. Remaining States

The only remaining states are the $4^+(3^-)$ 1915.2- and 5^- 2040.3-keV states in ^{202}Pb . About the former we can say little, but the latter should contain the component $[(\nu f_{5/2})^{-1}(\nu i_{13/2})^{-1}]_{5^-}$. This is consistent with the $E4$ transition probability from the 9^- state to this state. The single-particle estimate (corrected for conversion) for its half-life is about 2.8 h, which compares well with the (approximate) experimental value of 8.6 h. This is a relatively small retardation for an $E4$

transition, consistent with a simple change in coupling of the $f_{5/2}$ and $i_{13/2}$ neutron holes.

VII. STATES IN ODD-ODD ^{202}Tl

Unfortunately, it is extremely difficult, if not impossible, to make meaningful shell-model assignments for the odd-odd states of ^{202}Tl . This comes about for two reasons. First, the competing proton orbits, as determined from the neighboring odd-mass Tl isotopes, lie very close together. Thus, throughout this region, although the $s_{1/2}$ orbit remains the ground state, the $d_{3/2}$ and $d_{5/2}$ orbits lie only a few hundred keV away. Second, and worse, the competing neutron orbits not only lie close together, but also one obtains different predictions for the lowest-lying ones when considering the $N=119$ isotones vs the odd-mass Pb isotopes. A thorough discussion of these problems is given in Ref. 13, where the states in ^{200}Tl were being considered; most of it applies to ^{202}Tl and will not be repeated here.

In Table VI we show the magnitude of the problem. Here we list the possible configurations that can contribute to odd-odd ^{202}Tl states, using only the three most likely proton orbits and the four most likely neutron orbits. All of these odd-odd configurations should lie well below 1 MeV. The problem is worst, of course, for the low-spin states. When we tried to assign a major configuration to the 2^- ground state of ^{200}Tl (cf. Ref. 13), we were unable to choose between $[(\pi s_{1/2})(\nu f_{5/2})^{-1}]_{2^-}$ and $[(\pi s_{1/2})(\nu p_{3/2})^{-1}]_{2^-}$. And there we had additional information to work with, such as the transitions connected with the 0^- first excited state at 147.63 keV. In considering the 2^- ground state of ^{202}Tl , we arrive again at the same two configurations and are unable to decide between them.

For the other states with definite J^π assignments, the various configurations obtainable from Table VI are consistent with the known transition probabilities, but we are again unable to choose among them. (Also, there should be considerable configuration mixing in this nucleus with its myriad of close-lying states.) An experiment such as $^{203}\text{Tl}(p, d)$ is needed before definite assignments can be made.

Postscript: Recently, Hanser²⁹ in his work on the decay of ^{202}Bi has determined the multipolarity of the 954.5-keV transition to be $E2$. This confirms our tentative assignment of 4^+ for the 1915.2-keV state and eliminates the 3^- possibility.

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TABLE VI. Possible configurations for producing some low-lying odd-odd states in ^{202}Tl .

J^π	$(\pi\nu)$ configurations
0^-	$(s_{1/2}p_{1/2}) (d_{3/2}p_{3/2}) (d_{5/2}f_{5/2})$
1^-	$(s_{1/2}p_{1/2}) (s_{1/2}p_{3/2}) (d_{3/2}f_{5/2}) (d_{3/2}p_{1/2})$ $(d_{3/2}p_{3/2}) (d_{5/2}p_{3/2}) (d_{5/2}f_{5/2})$
2^-	$(s_{1/2}f_{5/2}) (s_{1/2}p_{3/2}) (d_{3/2}p_{1/2}) (d_{3/2}f_{5/2})$ $(d_{3/2}p_{3/2}) (d_{5/2}p_{1/2}) (d_{5/2}p_{3/2}) (d_{5/2}f_{5/2})$
3^-	$(s_{1/2}f_{5/2}) (d_{3/2}f_{5/2}) (d_{3/2}p_{3/2}) (d_{5/2}f_{5/2})$ $(d_{5/2}p_{3/2}) (d_{5/2}p_{1/2})$
4^-	$(d_{3/2}f_{5/2}) (d_{5/2}p_{3/2}) (d_{5/2}f_{5/2})$
4^+	$(d_{5/2}i_{13/2})$
5^-	$(d_{5/2}f_{5/2})$
5^+	$(d_{3/2}i_{13/2}) (d_{5/2}i_{13/2})$
$6^+, 7^+$	$(s_{1/2}i_{13/2}) (d_{3/2}i_{13/2}) (d_{5/2}i_{13/2})$
8^+	$(d_{3/2}i_{13/2}) (d_{5/2}i_{13/2})$
9^+	$(d_{5/2}i_{13/2})$

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¹A. W. Sunyar, D. Alburger, G. Friedlander, M. Goldhaber, and G. Scharff-Goldhaber, *Phys. Rev.* **83**, 906 (1951).

²E. K. Hyde, I. Perlman, and G. T. Seaborg, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Englewood Cliffs, N. J., 1964), Chap. 10.

³A. R. Fritsch, University of California, Lawrence Berkeley Laboratory Report No. UCRL-3452, 1956 (unpublished).

⁴C. R. Gruhn, R. R. Todd, C. J. Maggiore, W. H. Kelly, R. E. Doebler, and Wm. C. McHarris, *Nucl. Instr. Methods* **75**, 109 (1969).

⁵D. A. Maeder and A. H. Wapstra, *Phys. Rev.* **93**, 1433 (1954).

⁶D. A. Maeder, A. H. Wapstra, G. J. Nijgh, and L. Th. M. Ornstein, *Physica* **20**, 521 (1954).

⁷I. Bergström and A. H. Wapstra, *Phil. Mag.* **46**, 61 (1955).

⁸J. A. McDonell, R. Stockendal, C. J. Herrlander, and I. Bergström, *Nucl. Phys.* **3**, 513 (1957).

⁹B. Johansson, T. Alväger, and W. Zuk, *Arkiv Fysik* **14**, 439 (1959).

¹⁰J. B. Cross, Michigan State University Report No. COO-1779-33, 1970 (unpublished); J. B. Cross, Wm. C. McHarris, and W. H. Kelly, to be published.

¹¹R. E. Doebler, Michigan State University Report No. COO-1779-42, 1970 (unpublished).

¹²MOIRAE, a program developed for the Michigan State University Cyclotron Laboratory Sigma-7 computer by

R. Au and G. Berzins.

¹³R. E. Doebler, Wm. C. McHarris, and W. H. Kelly, *Phys. Rev. C* **2**, 2422 (1970).

¹⁴G. C. Giesler, Wm. C. McHarris, R. A. Warner, and W. H. Kelly, *Nucl. Instr. Methods* **91**, 313 (1971).

¹⁵R. S. Hager and E. C. Seltzer, *Nucl. Data A4*, 1 (1968).

¹⁶W. D. Myers and W. J. Swiatecki, Lawrence Berkeley Laboratory Report No. UCRL-11980, 1965 (unpublished).

¹⁷W. H. G. Lewin, J. Bezemer, and C. W. E. Van Eijk, *Nucl. Phys.* **62**, 337 (1965).

¹⁸R. M. Diamond and F. S. Stephens, *Nucl. Phys.* **45**, 632 (1963).

¹⁹L. S. Kisslinger and R. A. Sorensen, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.* **32**, No. 9 (1960).

²⁰W. W. True and K. W. Ford, *Phys. Rev.* **109**, 1675 (1957).

²¹T. F. Harvey and D. M. Clement, *Nucl. Phys.* **A176**, 592 (1971).

²²E. J. T. Burns and J. E. Draper, *Bull. Am. Phys. Soc.* **36**, 1146 (1971); E. J. T. Burns, Ph.D. thesis, University of California, Davis, 1971 (unpublished).

²³S. A. Moszkowski, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1965); *Phys. Rev.* **89**, 474 (1953).

²⁴G. E. Holland, C. W. Whitten, N. Stein, and D. A. Bromley, in *Contributions, International Conference on Properties of Nuclear States, Montreal, Canada 1969*, edited by M. Harvey *et al.* (Presses de l'Université de Montréal, Montréal, Canada, 1969), p. 288.

²⁵C. R. Gruhn and Wm. C. McHarris, Michigan State University, unpublished data, 1967.

²⁶R. J. Ascutto, N. K. Glendenning, and B. Sørensen, *Phys. Letters* **34B**, 17 (1971).

²⁷R. W. Goles, Michigan State University Report No. COO-1779-56, 1971 (unpublished); R. W. Goles, R. A. Warner, Wm. C. McHarris, and W. H. Kelly, to be published.

²⁸J. M. Dairiki, University of California, Berkeley, Report No. UCRL-20412 (unpublished).

²⁹A. Hanser, *Nucl. Phys.* **A146**, 241 (1970).