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Preface

The year 1968 marks the completion of the first three years of nuclear chemistry research at Michigan State University. During these three years the nuclear chemistry group has grown from nothing into a rather large and active group. The year 1968 also marks the awarding of the first AEC contract (beginning 1 March) to our group, and this Annual Report contains résumés of the current status of research performed under this contract.

Our nuclear chemistry research program centers around the MSU 50-MeV Sector-Focused Cyclotron in the MSU Cyclotron Laboratory, part of the Department of Physics. The nuclear chemistry group has had a most successful and close relationship with the various nuclear physics groups, and, indeed, most of our financial, as well as moral, support thus far has come from the Cyclotron Laboratory. Thus, this Annual Report should be considered somewhat as a supplement to the Cyclotron Laboratory Annual Report, for without the aid of that laboratory with their NSF grant, little of the research reported herein could have been accomplished. We express special thanks to Dr. W. H. Kelly and his nuclear spectroscopy group, with whom we have had the closest association, and also to Dr. C. H. Gruhn, whose Ge(Li) detector fabrication is very much appreciated.

The reports that follow, some about projects essentially completed or in print, some about projects barely begun, should give one a general idea of the types of research we have been carrying out. Most of the work this past year has been in the field of β - γ spectroscopy, although our research program is broader than just this; some of our on-line and in-beam projects, as well as nuclear reactions studies, are now in their beginning stages. Also, most of the projects reported on in this Report fit into programs of larger overall scope; an idea of some of these can be obtained by planning at the Ph.D. thesis topics listed on p. 69.

The nominal cut-off date for the reports that follow was 1 November. For the most part, the reports were written by the graduate students involved -- the quality of these students is evidenced by their

writing, especially considering that in many cases this was their initial experience with professional scientific reporting. Much has been accomplished, of course, since the cut-off date -- for example, we produced Gd^{144} and discovered Gd^{143} only a few days later. However, as the intent of this Report is to present a survey of our work, both completed and in progress, we have produced the Report as it stands, with the understanding that it is a status report and some of its contents will likely change before final publication.

The cover design is by K. Kosanke, and the typing was done by Mrs. I. Samra.

I. Decay of Gd^{149}

R. Eppley

The decays of odd-mass gadolinium isotopes are being studied in an effort to understand better the systematic trends of the excited states in this region. This region is of particular interest since the nuclei exhibit properties from those of spherical nuclei near the closed shell ($N=82$) to the properties of deformed nuclei ($N \geq 87$). The higher mass gadolinium isotopes have been quite thoroughly studied. Consequently, we began our investigation with Gd^{149} .

Gd^{149} was produced in the MSU cyclotron by the $Eu^{151}(p,3n)Gd^{149}$ reaction with a proton beam energy of 28 MeV. Both natural Eu and separated Eu^{151} have been used for the bombardments. Chemical reduction as well as ion exchange techniques [1,2] have been used for separating the Gd fraction from other active contaminants (mostly Eu).

Twenty-three gamma-transitions have been ascribed to the decay of Gd^{149} and the energies and intensities have been determined. These values are listed in table 1. A gamma-ray singles spectrum is shown in figure 1; this spectrum was taken with a 7cm³ coaxial Ge(Li) detector (constructed in this laboratory) having a resolution of 2.5 keV FWHM for the Cs^{137} peak. The three peaks at 963, 993, and 1013 keV, as well as a peak at 1083 keV, have been reported by other authors as being part of the Gd^{149} decay scheme. However, these peaks appear to have a much shorter half-life than the other transitions. They are prominent immediately after the bombardment but have all but disappeared by two to three days after the bombardment. Consequently, they have not been included in the decay scheme. It is interesting to note, however, that they are not removed from the Gd fraction during the ion exchange process.

For use in calculating the internal conversion coefficients, the electron intensities of Harnatz and Handley[3] have been used. The electron and photon intensities were normalized relative to one another by assuming the theoretical M2 conversion coefficient for the 346.5 keV transition. The theoretical conversion coefficients used were those of Sliv and Band.[4] The γ -ray and electron intensities together with the assigned multipolarities are given in table 1.

Several γ - γ coincidence measurements have been performed. These

utilized a Ge(Li) detector to record the coincidence spectra in conjunction with either a 3" x 3" NaI(Tl) detector or an 8" x 8" split annulus detector [5] used to set the coincidence gates. One of the resulting spectra, using a 150 keV gate, is shown in figure 2. In this spectrum the 947.7, 788.6, 663.3, 645.2, 516.4, and 346.5 keV γ -transitions are enhanced, as compared to the singles intensities. This suggests that these transitions feed the 149.6 keV level.

Since there is no measurable positron emission, the electron capture decay endpoint must be estimated. This was accomplished as shown in figure 3. This method is similar to that of Way and Wood.[6] The plot is of all experimentally known decay energies versus the neutron number for the decay pairs that have the same mass number as the pair to be estimated. These points fall on a straight line, thus yielding a good estimate of the endpoint energy. For Gd^{149} the use of this method results in a value of 1320 keV for the electron capture energy.

The decay scheme is based on coincidence runs, intensity balances for each level, and sums and differences checks. The decay scheme is shown in figure 4, together with the level spins and parities where known, the $\log ft$ values, and the percentage decay to each level.

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TABLE 1

Transition Data for Gd^{149}

Energy	Photon Intensity	K-conversion Intensity	Total Intensity	Experimental I.C.C. (K)	Multipole Order
K-X-rays	-	-	577	-	-
149.6	233.4	118.7	369.6	0.51	M1
214.5	0.81	-	0.81	-	-
252.3	1.08	0.13	1.21	0.12	M1
260.5	5.79	0.14	5.94	2.49×10^{-2}	E1
272.0	14.60	1.66	16.49	1.14×10^{-2}	E1
298.5	126.7	10.2	138.1	8.08×10^{-2}	M1
346.5	≈ 100.0	≈ 19	122.3	≈ 0.19	M2
405.5	3.68	0.031	3.71	8.43×10^{-3}	E1
459.9	2.42	0.069	2.49	2.85×10^{-2}	M1
478.7	0.95	0.011	0.96	1.13×10^{-2}	M1+E2
496.4	7.21	0.226	7.48	3.09×10^{-2}	E3
516.4	11.05	0.202	11.29	1.83×10^{-2}	M1
534.2	13.20	0.243	13.48	1.84×10^{-2}	M1
645.2	5.93	0.017	5.95	2.80×10^{-3}	E1
663.3	1.09	0.015	1.11	1.41×10^{-2}	M1 or E3
666.2	3.93	0.025	3.96	6.35×10^{-3}	E2
748.2	34.57	0.068	34.64	1.96×10^{-3}	E1
788.6	29.55	0.014	29.59	4.82×10^{-3}	M1+E2
812.4	0.55	-	0.55	-	-
875.8	0.62	0.0039	0.62	6.32×10^{-3}	(M1)
933.3	2.21	-	2.21	-	-
939.1	8.99	0.042	9.04	4.62×10^{-3}	M1
947.7	3.67	0.0059	3.67	1.62×10^{-3}	E1

Fig. I-1. Singles γ -ray spectrum of Gd^{149} recorded with a 7-cm³ Ge(Li) detector.

Fig. 1-2. Spectrum of γ -rays from Gd^{149} decay observed in coincidence with the 150-keV region. [3"x3" NaI(Tl) gate, 7-cm³ Ge(Li) signal]

Fig. I-3. Graphical estimates of Q_E 's for the odd-mass Gd isotopes near Gd^{149} .

DECAY SCHEME - Gd^{149}

$64Gd^{149}_{85}$

$Q_{\alpha} \approx 1320$

Fig. I-4. Gd^{149} decay scheme.

$63Eu^{149}_{86}$

II. The Decay of Gd^{147}

R. Eppley

As a part of the complete study of the decays of the odd-mass neutron-deficient Gd isotopes, through which we hope to follow the behavior of states in the Eu isotopes all the way from the closed neutron shell into the deformed region, we have also begun the study of Gd^{147} decay. Gd^{147} has been prepared by the $Eu^{151}(p,3n)Gd^{147}$ reaction, using a proton beam energy of 40 MeV. We are currently determining the γ -ray energies and intensities by calibration with well-known standards. Electron intensities will be measured with the MSU 6-gap β -ray spectrometer.[1] From these intensity measurements, internal conversion coefficients can be calculated and compared with the theoretical values to help determine the transition multipolarities. Some γ - γ coincidence runs have been made, and more are planned, both single- and two-dimensional, to help with the energy-level placements in the decay scheme.

Reference:

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III. The Decay of Gd^{145}

R. Eppley

Only three investigations of this isotope have been published up to this time.[1-3] For our investigation, Gd^{145} has been prepared by the $Sm^{144}(\alpha, 3n)Gd^{145}$ and $Sm^{144}(He^3, 2n)Gd^{145}$ reactions. Alpha beams of 42 MeV and He^3 beams of 30 MeV were used in the bombardments. The singles spectrum for the gamma transitions is given in figure 1. It is interesting to note that the two transitions with the largest intensities are at the relatively high energies of 1758 ± 1 and 1881 ± 1 keV. γ - γ coincidence runs are planned to help confirm the existing skeletal decay scheme and to be able to place the remainder of the observed γ -transitions.

We will soon be able to move sources in and out of the beta-ray spectrometer rapidly without breaking the vacuum. This will be necessary in order to obtain conversion electron spectra, due to the short half-life of this isotope (24 min.).

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Fig. III-1. Singles γ -ray spectrum of 25-min Gd^{145g} .

IV. A New $N = 81$ Isomer: Gd^{145m}

R. Eppley

During the investigation of Gd^{145} , an isomeric state was observed with a half-life of 85 ± 3 seconds and an energy of 721.4 ± 0.4 keV. This activity was produced by the same reactions as for the Gd^{145g} activity. The γ -spectrum is shown in figure 1. The weaker peaks that are present in the spectrum are from transitions in the daughter nucleus, Eu^{145} .

For the half-life determination, the spectrum was collected by utilizing the Sigma-7 computer at the Cyclotron Laboratory. The pulse height signals, from a 7cm^3 Ge(Li) detector and associated electronics, were fed into a 4096-channel ADC which is interfaced directly to the Sigma-7. This method allowed us to print out the 600-800 keV region of the spectrum at fifteen second intervals on the computer line printer. The isomeric peak on each print out was corrected for dead-time, the background subtracted, and the net peak integrated. A log plot was made of the difference in the net counts for each interval and plotted versus the time of the interval. The experimental points were least-squared and from the resulting straight line the half-life was determined.

The determination of the internal conversion coefficient was carried out by simultaneously measuring the gamma-ray transition intensity and the internal conversion electron intensity. The gamma-ray intensity measurement was made with a Ge(Li) detector and the electron intensity was measured by using a plastic scintillator (Pilot B, $1\frac{1}{4}'' \times 1\frac{1}{2}''$). The conversion electron spectrum is shown in figure 2. After suitable efficiency calibrations of the detectors, the K-conversion coefficient was determined to be approximately 0.1. Comparison of this value to the theoretical values of Sliv and Band would indicate an $M4$ transition.

There is supporting evidence for this assignment. The shell model predicts a transition ($h_{11/2} \rightarrow d_{3/2}$) of this multipolarity. Also, the measured half-life is consistent with the Weisskopf estimate for an $M4$ transition. This isomeric state is the seventh known in the series of nuclei with eighty-one neutrons. A plot of these nuclei in the form of energy versus proton number is shown in figure 3. Since $M4$ transitions are generally regarded as being relatively "pure" single-particle transitions, they should be useful in studying the systematic changes of the single-particle states involved. At present these systematic changes, such as the changes in the radial matrix elements and the configuration interactions, are under investigation.

FIG. IV-1. γ -ray spectrum of 84.8-sec Gd^{145m} . The 721.4-keV γ is the $M4$ isomeric transition. The unlabelled γ 's all result from the decay of 25-min Gd^{145g} .

Fig. IV-2. Electron spectrum of 84.8-sec Gd^{145m} taken with a Pilot B scintillator. This spectrum shows the conversion electrons from the 721.4-keV $M4$ isomeric transition riding above the β^+ continuum from Gd^{145g} decay. Using Cs^{137} as a standard, α was determined to be ≈ 0.1 .

Fig. IV-3. Energies of the known isomeric states in $N=81$ nuclei.

V. Decay of Yb^{167}

R. Goles

The (p,t) reaction is expected to show slight similarities to α -decay[1] in that it involves the selective plucking out of an ordered nucleon group from a nucleus. Consequently, it should be interesting to examine this reaction in terms of α -decay theory. The extensive theory of Rasmussen and Mang, which works well in the heavy element region should also be applicable to α -decay in the deformed rare earths, so we decided to study the (p,t) reaction on some of these. Tb^{159} , Ho^{165} , and Tm^{169} seemed the most likely candidates, so we have initiated our study by re-examining the levels in the daughter of the $\text{Tm}^{169}(\text{p,t})\text{Tm}^{167}$ reaction through the decay of Yb^{167} . As we become familiar with the levels and rotational bands in Tm^{167} with the aid of this tested technique, we shall proceed to the (p,t) reaction.

In 1954, 18.5-hr Yb^{167} was first produced.[2] It has subsequently been investigated by several workers.[3-7] The most recent reports employed electron and γ -ray spectroscopy as well as γ - γ coincidence measurements in order to propose a partial decay scheme consistent with seventeen observed transitions. Essentially all work to date has made use of magnetic electron spectrometers and NaI(Tl) scintillation γ -ray detectors. Since the γ -ray spectrum in the Yb^{167} decay would be expected to be more complex than reported based on comparisons with the level schemes of well-known neighboring odd thulium isotopes, it was thought that it would be profitable to re-investigate the decay of Yb^{167} with high-resolution Ge(Li) γ -ray detectors.

To this end, extensive γ -ray measurements of the decay $\text{Yb}^{167} \rightarrow \text{Tm}^{167}$ have been made with particular emphasis on γ -rays of energy greater than 500 keV. Because of the much higher resolution of the Ge(Li) detectors, it was possible to observe details of the γ -ray spectra that previously were either unnoticed or incorrectly interpreted. Specifically, over 70 additional low intensity γ -rays have been identified and associated with the decay of Yb^{167} , with 31 of these gamma rays having energies between 1300 and 1700 keV.

The aim of the present work then is to extend upon and to characterize further the rotational energy levels of Tm^{167} through use of conversion electron, γ - γ coincidence, and particle scattering experiments.

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VI. Pb^{203m} Decay and $M4$ Transition Probabilities in Neutron-Deficient Lead Isotopes

R. Doebler

The decay of the 6.1-sec isomer Pb^{203m} has been the subject of a number of investigations[1-5] using conversion electron and NaI(Tl) spectrometers. These studies found only a single transition of about 825 keV, which was assigned an $M4$ multipolarity. The transition was assumed to be from a $13/2^+$ state to a $5/2^-$ ground state, similar to those in the other odd-mass isomers of Pb. However, the reduced transition probability, which for $M4$'s appear to be remarkably regular, was abnormally large. Stockendal[1] suggested the possibility of an additional 5-keV transition from the isomeric state to a state at 820-keV which is populated in the electron capture decay of Bi^{203} . The spin and parity of this state has been reported both as $7/2^-$ [6] and $9/2^-$. [1] If the level were $9/2^-$, the 5-keV transition would be an $M2$ and could possibly partially de-excite the 825-keV $13/2^+$ level. Since the 5-keV transition would be difficult to observe directly, we decided to look for the 820-keV transition that should follow it.

Separated isotope Pb^{206} was bombarded with 40 MeV protons from the Michigan State University Sector-Focused Cyclotron to produce Bi^{203} ($t_{1/2}=12h$) through the reaction $Pb^{206}(p,4n)Bi^{203}$. The Bi^{203} was chemically separated from the Pb target, taken up in 6M HCl and loaded onto an anion-exchange column. The Pb^{203} was eluted with 0.3M HCl at the rate of two drops every 6 sec for a 2-h period, each drop being immediately transferred to a 7-cm³ Ge(Li) γ -ray detector. With a resolution of 2.7 keV FWHM for the 825.2 keV γ from Pb^{203m} , we could easily resolve the 820.1- and 825.2-keV γ 's from Bi^{203} as shown in fig. 1a.

The spectrum of the Pb^{203m} after separation, shown in fig. 1b, clearly shows a peak at 820.1 keV. The areas of the two peaks obtained after stripping and correction for the detector efficiency yield relative γ -ray intensities of $10.6 \pm 1.0\%$ and $89.4 \pm 1.0\%$ for the 820.1- and 825.2-keV γ 's respectively. After correcting for internal conversion, the $13/2^+$ state is found to decay 8% by the 5.1-keV transition and 92% by the 825.2-keV $M4$ transition. Our proposed decay scheme for Pb^{203m} is shown in fig. 2. The previous uncertainty in the spin assignment for the 820.1-keV state can be removed; The 5.1-keV transition is $M2$, making the state $9/2^-$.

The squares of the radial matrix elements for the $i_{13/2} \rightarrow f_{5/2}$ $M4$ γ -transition in the odd-mass Pb isomers were calculated using Moszkowski's approximations.[7] These values are listed in table 1, along with those calculated previously by Stockendal.[1,2] The $|M|^2$ value for the Pb^{203m} transition now fits quite nicely in line with the others, indicating that this transition behaves regularly, as one would expect for $M4$ transitions.

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- 9] R. Stockendal, Ark. Fys. 17, 553 (1960).

Table 1

Radial Matrix Elements for $M4$ γ -Transitions
in Odd-Mass Pb Isotopes

$ M ^2$	A	197	199	201	203	205	207
Stockendal [1]		3.8 ± 0.5	3.8 ± 0.2	3.5 ± 0.2	4.3 ± 0.1	---	3.7 ± 0.1
Present Calc. ^a		3.6 ± 0.4	3.7 ± 0.2	3.5 ± 0.2	3.8 ± 0.2	$(18 \pm 2)^b$	3.6 ± 0.1

^aWe have used our own experimental data for the Pb^{203m} calculation and the most recent half-life values tabulated in ref.[8] for the other calculations.

^bThe value of $|M|^2$ for Pb^{205m} is inexplicably large, perhaps because of the presence of two unresolved 1014-keV transitions, and is included only for completeness. See ref. [9]

Fig. VI-1. The γ -ray spectrum of: a) Bi^{203} (unavoidably containing some Bi^{204} , which has the same half-life); b) Pb^{203m} after chemical separation from its Bi^{203} parent.

Fig. VI-2. The decay scheme of Pb^{203m} .

VII. The Electron Capture Decay of Pb^{200}

R. Doebler

The decay of Pb^{200} has been the subject of several investigations[1-3], which have made use of NaI(Tl) scintillation and magnetic conversion electron spectrometers. Because the decay scheme of Pb^{200} was incomplete, we felt that it would be profitable to study this isotope using Ge(Li) detectors.

Pb^{200} was produced in high yield using 37 MeV protons from the Michigan State University Sector-Focused Cyclotron to induce the $Tl^{203}(p,4n)Pb^{200}$ reaction on natural thallium foil. The samples were aged from 1-3 days and the lead activity chemically separated from the Tl and Hg contaminants.

Figure 1 shows the γ -ray singles spectrum obtained in a 36 hour period using a 7 cm³ Ge(Li) detector with a resolution of 2.7-keV FWHM on the 661-keV γ -ray of Cs^{137} . To reduce the Tl^{200} contamination, which was constantly growing in, a new sample, free of Tl, was prepared every hour over the 36 hour period. Fourteen γ -transitions belonging to the decay of Pb^{200} were found, having the energies and intensities listed in table 1. Of these, the 377.8- and the 525.4-keV transitions had not been reported previously.

The placement of the transitions in the level scheme had already been fairly well determined by Wierhed[1]; however, we decided to check this by γ - γ coincidence experiments using a 3"x3" NaI(Tl) detector in coincidence with the 7 cm³ Ge(Li) detector. We are also in the process of using two Ge(Li) detectors to perform a two-dimensional coincidence experiment of 1024x1024 channels which should yield all possible coincidence relationships in one experiment. The results of our integral and anticoincidence runs are shown in figures 2 and 3; and relative intensities are listed in table 1 for comparison with the singles run. The relative intensities are based on the 450.5-keV transition, an unfed ground state transition, assigned the value of 100.

From the integral and anticoincidence data it appears that all the γ -rays are in coincidence except the 450.5-, 525.5, and 605.3-keV transitions, which are therefore assumed to be unfed ground state transitions. It also appears that a large part of the Pb^{200} decay populates the 0- first excited state.

To obtain multipolarity assignments for the transitions we calculated conversion coefficients using the conversion electron data of Wierhed[1]

and our intensities from the singles run. In order to normalize the two sets of data we assumed that the 147-keV γ -transition from the 0- first excited state to the 2- ground state was a "pure" $E2$ transition. That this is a reasonable assumption is confirmed by Aström and Co-workers[4]. We then assumed that the theoretical conversion coefficients of Sliv and Band[5] are good approximations for this $E2$ transition. The 289.1- and 289.9-keV γ 's could not be resolved by our system. However, by calculating the conversion coefficient for the sum of the two transitions we obtain strong evidence for the assignment of an $M1$ multipolarity to both transitions.

Table 2 shows the multipolarity assignments which we propose for those transitions where both conversion electron and γ -ray intensities are available. These multipolarity assignments agree with those of Wirhed and co-workers for those transitions they were able to assign.

It is of interest to note that in only one transition is there a significant amount of $M1$, $E2$ mixing and this is for the 257.2-keV transition which is $61 \pm 13\%$ $E2$. This lack of $E2$ admixture would seem to indicate that the levels in Tl^{200} populated in the decay of Pb^{200} are relatively pure single particle states with little contribution from collective effects.

Our proposed decay scheme is shown in figure 4. The spin and parity assignments are based on our multipolarities, and they agree with the assignments of Wirhed and Herrlander[1] for those levels for which the latter were able to make assignments.

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Table 1

Energies and Relative Intensities of Gamma Rays
from the Decay of Pb^{200}

Measured energies (keV)	Relative intensities		
	Singles	Integral γ - γ coincidence	Anticoincidence
109.5 $\pm$ 0.1	14.1 $\pm$ 0.3	370 $\pm$ 10	11.8 $\pm$ 1
142.3 $\pm$ 0.2	106 $\pm$ 3	5210 $\pm$ 200	53.8 $\pm$ 6
147.6 $\pm$ 0.1	1185 $\pm$ 30	22,500 $\pm$ 1000	1270 $\pm$ 200
161.3 $\pm$ 0.2	11.3 $\pm$ 0.7	760 $\pm$ 50	7.5 $\pm$ 2
235.6 $\pm$ 0.1	145 $\pm$ 3	8210 $\pm$ 400	93.3 $\pm$ 10
257.2 $\pm$ 0.1	149 $\pm$ 3	5930 $\pm$ 300	113 $\pm$ 10
268.4 $\pm$ 0.1	130 $\pm$ 4	7140 $\pm$ 400	86.6 $\pm$ 9
289.7 $\pm$ 0.2	90 $\pm$ 3	2190 $\pm$ 200	75.6 $\pm$ 9
302.8 $\pm$ 0.2	5.8 $\pm$ 1	155 $\pm$ 20	4.5 $\pm$ 2
315.6 $\pm$ 0.3	7.6 $\pm$ 1	285 $\pm$ 30	5.2 $\pm$ 2
377.8 $\pm$ 0.4	1.0 $\pm$ 0.4	-	-
450.5 $\pm$ 0.2	$\approx$ 100	$\approx$ 100	$\approx$ 100
457.8 $\pm$ 0.3	3.65 $\pm$ 0.1	24.2 $\pm$ 3	2.5 $\pm$ 1
525.4 $\pm$ 0.2	12.5 $\pm$ 0.2	-	11.9 $\pm$ 2
605.3 $\pm$ 0.2	16.6 $\pm$ 0.3	22 $\pm$ 5	17.9 $\pm$ 3

Table II
Transition Data for Pb^{200}

Energy keV	Photon intensity	Conversion intensity	Experimental conversion coefficient	Theoretical conversion coefficient	Multiple order
109.5	14.1	K 22	5.7	5.8	M1
		L_I 4.2	1.1	0.86	M1
		L_{II} 0.42	0.11	0.092	M1
142.3	106	K 69	2.39	2.5	M1
		$L_I[L_{II}]$ 14.2	0.49	0.45	M1
147.6	1185	K ≈ 100	≈ 0.35	0.35	E2
		$L_I[L_{II}]$ 137	≈ 0.405	0.405	E2
		L_{III} 90	≈ 0.27	0.27	E2
161.3	11.3	K 4.5	1.46	1.8	M1
235.6	14.5	K 26.7	0.68	0.64	M1
		$L_I[L_{II}]$ 4.6	0.12	0.11	M1
257.2	149	K 10.1	0.249	0.50	M1
				0.09	E2
		$L_I[L_{II}]$ 2.8	0.069	0.086	M1
				0.049	E2
		L_{III} 0.5	0.012	0.00064	M1
				0.019	E2
268.4	130	K 18.4	0.520	0.44	M1
		$L_I[L_{II}]$ 3.1	0.088	0.078	M1
289.1	90	K 3.7	0.38	0.36	M1
289.9		K 5.4			
302.8	5.8	K 0.70	0.44	0.31	M1
		$L_I[L_{II}]$ 0.12	0.076	0.056	M1
315.6	7.6	K 0.68	0.33	0.28	M1
		$L_I[L_{II}]$ 0.20	0.097	0.051	M1
450.5	≈ 100	K 4.4	0.16	0.11	M1
		$L_I[L_{II}]$ 0.76	0.028	0.011	M1
		M 0.16	0.0059	0.0055	M1
457.3	3.65	K 0.40	0.40	0.3	M1
605.3	16.6	K 0.24	0.053	0.053	M1
		L 0.040	0.009	0.009	M1

Fig. VII-1. Singles γ -ray spectrum of a purified Pb^{200} source. This spectrum was collected for ca. 1 day, with a thallium separation being performed every hour in order to reduce the Tl^{200} that grows in. Impurity peaks are labelled as such; the remaining peaks appear to result from Pb^{200} decay.

Fig. VII-2. Pb²⁰⁰ integral coincidence spectrum. All γ -rays above the x-rays were included in the gate. Only Pb²⁰⁰ γ -rays are labelled.

Fig. VII-3. Pb^{200} anti-coincidence spectrum. The 7-cm³ Ge(Li) was placed inside the tunnel of an 8"x8" NaI(Tl) split annulus and run in an anticoincidence mode both with the annulus and with a 3"x3" NaI(Tl) detector placed at the other end of the tunnel.

Fig. VII-4. The decay scheme of Pb^{200} .

VIII. Decay Studies on Bi²⁰⁵

K. Kosanke

Previous investigators [1,2] have reported a total of 84 transitions in the decay of Bi²⁰⁵; these were observed in internal conversion studies. Of these 84, one or more electron intensities have been reported for only 33 transitions.[3,4,5] Previous NaI(Tl) γ-ray spectrum studies[1,4] have resulted in the reporting of 23 γ-ray intensities. Of these only 15 have been tied to a single transition. The result was that for only 12 transitions were both electron and γ-ray intensities reported and thus conversion coefficients determined. Multipolarities, of varying degrees of certainty, have been suggested for 32 of the transitions.[1,2,5] Currently, the decay scheme has 31 levels with 22 being rather definite.[1,6]

Present work in this lab on Bi²⁰⁵ is pursuing three objectives. One, securing more complete and more definite multipolarity assignments for the transitions already known and more definite log *ft* values for the electron capture decay through a more complete collection of γ-ray intensities. Improved resolution of Ge(Li) γ-ray detectors (figure 1 shows a Bi²⁰⁵ singles spectrum, run 90 hours, FWHM of 2.7 keV for the 703.4 keV photopeak) and stripping techniques (figure 2 shows the 560 to 583 keV region of figure 1 with the background subtracted and peaks stripped) has allowed the determination of γ-ray energies and intensities for 74 of the transitions. The current results of intensity determinations is given in Table I.

Two, a search of the region above 1 MeV for unreported transitions. There is evidence suggesting a number of weak new transitions with energies above 1.5 MeV.

Three, many coincidence measurements have been made utilizing Ge(Li) detectors. These have already contributed information about a number of new levels and have cleared up many of the previously uncertain levels. We are currently in the process of setting up a two-dimensional Ge(Li)-Ge(Li) coincidence experiment, similar to that already used in this laboratory [7] for Nd^{139m} and Pb²⁰⁰, which should complete our coincidence work. This experiment utilizes the Sigma 7 computer to store coincident spectra from two 4096-channel ADC's on magnetic tape - the tape is sorted by the computer later. The completed decay scheme should be forthcoming shortly after the analysis of this experiment.

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Table 1. Current Status of Bi²⁰⁵ γ-ray Intensity Results.

γ-ray Energies from Vegor's Conversion Electron Experiments (keV)	Relative γ-ray Intensities	
	This Work	Ref. 1
26.2	-	-
45.4	-	-
90.0	-	-
115.1	0.26	-
122.9		-
128.7	1a	-
185.1	0.38	±0.7
205.8	0.14	-
221.2	0.14	-
236.0		-
237.2	12.20b	
248.9	-	-
260.5	-	-
262.9	14.97b	15.7±1.5
282.3	17.12b	16.0±1.0
284.2		
297.2	a	-
310.4		-
313.0	10.53b	-
349.6	1.66	2.7±0.7
361.6	0.16	-
421.8	-	.35±0.2
493.8	1.13	3.5±1.0
498.8	0.63	-
511.7	2.94	6.3±1.5
550.1	8.78	39.0±5.0
561.7	0.15	
570.7	22.9	
574.1	1.97	
576.3	0.96	
580.0	16.5	6.0±0.8
627.0	1.77	

Table 1. continued

634.4	-	-
688.7	0.62	-
703.4	100.0	100.0
710.8	-	-
712.6	-	-
717.5	0.90	-
720.5	0.37	-
723.5	0.44	-
729.5	0.19	-
766.1DE	-	-
745.0	113.8b	-
759.0	-	13.0±3.0
751.2	15.96b	-
780.8	1.49	-
795.6	0.30	-
806.1	0.62	-
813.8	1.47	-
828.1	0.97	-
859.9	1.21	-
872.0	1.34	15.5±2.0
890.0	2.28	-
894.9	1.99	-
910.9	-	10.6±2.0
913.4	15.41	-
971.9	0.85	-
987.8	55.6	63.0±3.0
1002.4	1.92	-
1013.8c	-	-
1014.4c	13.46	-
1043.7	25.3	28.0±5.0
1048.2	-	-
1062.7	0.1	-
1072.4	0.90	-
1128.1	8	-
1190.0	8.09	7.8±1.5

Table 1. continued

1199.8	0.69	-
1208.3	1.89	-
1264.7		-
1777.7SE	]1.22	
1351.7	-	≈3.0
1863.9SE	]3.80b	
1439.0	0.63	-
1500.0	1.28	
1552.2	4.57	]4.5±1.5
1615.4	8.51	7.5±2.0
		-
1721.0	]0.28b	-
1766.1	124.5	
1768.9	-	]115.0±10.
1777.7	14.2	
1863.9	23.6	
1905.9	9.35	]36.0±5.0
1925.1	-	-
2567.8	0.22	-
2610.0	0.09	-

a) Intensity data incomplete at this time.

b) Stripping will resolve multiple peak into its components.

c) From ref. 2.

Fig. VIII-1. Singles γ -ray spectrum of a chemically separated Bi^{205} sample. Data were collected for 90 hr to produce this spectrum. The detector used was our 7-cm³ Ge(Li) detector, which was manufactured at MSU.

Fig. VIII-2. Portion of the Bi^{205} γ -ray spectrum in the 560-580 keV region, showing the stripping that was necessary even for the relatively high resolution (≈ 2.5 keV FWHM) obtained in Fig. VIII-1. The energies shown are corroborated by the conversion electron energies of ref. [1].

IX. The Decay of Iron-53

J. Black

Iron-53, an 8.5 minute nuclide decaying by both positron emission and electron capture, was first produced by a $\text{Cr}(\alpha, n)$ reaction on naturally occurring chromium.[1] Consequent investigations have been made using a number of methods, including scattering and (p, α) reactions.[2-5] Since modern techniques of gamma spectroscopy with semiconductor detectors have made it possible to study short-lived nuclides with greater ease and good resolution, it was decided to reinvestigate the decay of 8.5 minute Fe^{53} .

The Fe^{53} sources were produced by the reaction: ${}_{25}\text{Mn}^{55}(p, 3n){}_{26}\text{Fe}^{53}$ - a relatively unexplored method of production. Manganese metal (Fisher, powder, 99.94%) was bombarded with 34 MeV protons from the Michigan State University Sector-Focused Cyclotron. Typically, $\approx 0.5\text{g}$ targets were bombarded with a 2 μA beam for 3 minutes.

The singles spectrum of Fe^{53} (figure 1) was obtained by using a 7-cm³ Ge(Li) detector built in this laboratory, a Canberra 1408C FET preamplifier and 1417 amplifier, a Northern Scientific 4096 Channel ADC, and the SDS Sigma 7 computer. A total of twenty-four γ -rays were observed. These are listed in Table 1, along with their intensities and probable assignments. Note that both Mn^{54} and Mn^{52} were produced together with the Fe^{53} .

In addition to the singles data, the Fe^{53} investigation has included the following γ -ray studies using the Ge(Li) detector in conjunction with an 8"x8" NaI(Tl) split annulus: anticoincidence, integral coincidence, and coincidence with the 377.9 γ -ray. The assignment of 7/2- for the spin and parity of the ground state of Fe^{53} is in agreement with the nuclear shell model predictions and also experimental evidence.[6] In addition, the first excited state at 377.9 keV is probably 5/2 $\pm$ in agreement with the other odd nuclei in the $f_{7/2}$ shell region. Theoretically, the second excited state should have the character 3/2-, [7] which would also be in agreement with other nuclides in this part of the table of isotopes. A number of observed γ -rays have not been placed, and now are awaiting further experimental verification.

The positron spectrum of Fe^{53} is also of considerable interest. Previous work has shown three β^+ groups with end-point energies of $\approx 1.6, 2.4$, and 2.8 MeV.[8] A reinvestigation of this positron spectrum is now underway. Additional beta and gamma studies of Fe^{53} using a chemically separated target are being carried out. Our present, partial decay scheme is shown in figure 2.

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Table 1
Energies and Relative Intensities of Gamma-Rays
Observed in Fe^{53} Spectra

Measured γ -ray energy (keV)	Relative intensity	Assignment
319.2	5.47×10^{-2}	$\text{Cr}^{51} (?)$
377.9	1.00	Fe^{53}
412.3	-	DE 1434.3
511.01	8.60	-
701.09	5.83×10^{-2}	Not Assigned
669.28	8.32×10^{-3}	NA
743.6	2.01×10^{-2}	Mn^{52}
788.2	5.86×10^{-3}	DE 1811.4
835.3	5.16×10^{-1}	Mn^{54}
846.3	1.25×10^{-1}	NA
935.0	2.63×10^{-2}	Mn^{52}
961.9	9.67×10^{-3}	NA
1011.4	6.17×10^{-2}	Fe^{53}
1318.7	1.63×10^{-2}	DE 2340.7
1330.0	7.52×10^{-2}	Fe^{53}
1434.3	6.58×10^{-1}	$\text{Fe}^{53} + \text{Mn}^{52}$
1621.2	3.60×10^{-3}	Fe^{53}
1729.5	3.81×10^{-3}	NA ^a
1791.4	4.36×10^{-3}	NA ^a
1811.4	4.50×10^{-2}	NA ^a
2104.6	2.56×10^{-3}	NA ^a
2113.8	2.48×10^{-2}	NA ^a
2311.8	2.69×10^{-3}	Fe^{53}
2340.7	2.16×10^{-2}	Fe^{53}

^aThere is some evidence that these could be ground-state transitions in Mn^{53} , but we are deferring placing them until further experimental verification of their belonging to Fe^{53} decay has been carried out.

Fig. IX-1. γ -ray spectrum of 8.5-min Fe^{53} . This spectrum was recorded with the 7-cm³ Ge(Li) detector. Repeated bombardments were made for a total counting period of approximately 20 hr. See the text for a discussion of the γ -ray assignments.

THE DECAY SCHEME OF Fe^{53}

Fig. IX-2. Preliminary decay scheme for Fe^{53} .

X. Decay of Zn^{63}

G. Giesler

The Zn^{63} isotope has been studied by many authors since it was first reported by Bothe et al.[1] in 1937. Until recently, the most precise work had been by fitting spectra obtained from NaI(Tl) detectors to the more precisely measured levels obtained from charged particle scattering. As a result, there was a need for a high resolution re-examination of the Zn^{63} decay with Ge(Li) detectors.

In figure 1, the result of counting 12 samples is shown. The decay scheme obtained from this spectrum is shown in figure 2. These energies agree well within experimental error with those of Mazari et al.[2], and Bachner et al.[3], and the decay scheme compares well with one proposed by DeFrenne et al.[4]. Several differences between this work and that of DeFrenne are seen, however. One notable difference is at about 3040 keV. We see an intense γ -ray at 3045 keV and possibly a very weak one about 3030 keV, while DeFrenne reports a 3032 keV γ and a weaker 3042-keV γ . We have levels at 2095 keV and 2717 keV that they do not see, and they have levels at 2394 keV and 2768 keV that we have not seen. We place the 1392 keV transition as coming from the 2063 keV level, whereas they have two possible positions for it. We also see γ -rays at 1123, 1374, 1928, 2027, 2050, and 2187 keV that are placed as interstate transitions, but we do not see one reported at 1700 keV.

The three γ -rays at 1150, 1796, and 2012 keV have not been placed in the decay scheme. Mazari et al.[2] report a level at 2012 keV that could produce this γ -ray, which has not been seen by other authors. The remaining two γ -rays cannot be fitted as either ground state transitions or as transitions between two excited states. No levels at these two energies have been reported in scattering. These two could perhaps be only a peculiarity in statistics or could possibly be from an unidentified source.

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Fig. X-1. Singles Y-ray spectrum of Zn^{63} .

Fig. X-2. Decay scheme of ^{63}Zn .

XI. Decay of Zn^{62}

G. Giesler

Since the discovery of Zn^{62} in 1948 by Miller et al.[1], many authors have reported work on this isotope. Within the last two years, three authors [2-4] have reported γ -ray studies using Ge(Li) detectors. The 682 keV γ -ray is the highest energy seen by any of these authors, even though they looked for higher energy transitions. Charged particle scattering has excited eleven levels in Cu^{62} above 682 keV that could be populated by the decay of Zn^{62} . Because we are searching for very short lived Ga^{62} , which we expect to be a delayed α -emitter (see the accompanying proposal to the AEC), we need a thorough knowledge of the decay characteristics of Zn^{62} , including whether or not it populates any of these higher-lying levels.

Several measurements of Zn^{62} decay have been performed. A typical singles spectrum is shown in figure 1. The area below 700 keV agrees with Roulston et al.[2] and Antman et al.[3], although the 507 keV γ could not be resolved. Bakhru[4] reported a transition at 682 keV, which neither we nor the other authors saw, and did not report the 638 keV γ that we did see. None of the authors reported any transitions above 700 keV.

Six of the eight γ -rays we see above 700 keV have been identified, as shown in figure 1. Those from K^{40} and Tl^{208} come from room background. The decay of the Cu^{62} daughter to Ni^{62} produces three known γ -rays. The 1115 keV γ comes from Zn^{65} produced from Cu^{65} present in the natural Cu foil target and is the only possible contaminant. All other contaminants are removed by allowing the sample to decay for about 12 hours after irradiation, then separating the Zn from the Cu target by ion exchange.

This leaves two unidentified γ -rays, one at 1279 keV and one at 1430 keV. These have appeared in several different runs, indicating that they are part of either Cu^{62} or Zn^{62} decay. In Ni^{62} there are no levels below Cu^{62} decay energy that are spaced either 1297 keV or 1430 keV apart. In Cu^{62} levels at 1285 keV and 1432 keV are seen in $Ni^{61}(He^3,d)$ [5] scattering, as well as by other methods. This is strong evidence that the two unknown γ -rays are from Zn^{62} decay. Further work is now being performed to identify these γ -rays completely and find any others present. In particular, a much larger (≈ 40 cm³) Ge(Li) detector is being drifted at the laboratory so that we can look more carefully for weak higher-energy transitions.

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FIG. XI-1. γ -ray spectrum of Zn^{62} .

XII. Es²⁵² α Decay and Coriolis Mixing

Wm. McHarris

(in cooperation with I. Ahmad and P. R. Fields,
Argonne National Laboratory)

The decay of Es²⁵² is of particular interest because it is one of the very few nuclei that decays across a closed shell, here the subshell at $N = 152$. This means that the favored α decay is forced to populate states lying much higher in Bk²⁴⁸ than would ordinarily be populated by α decay. Also, as Es²⁵² is analogous to Es²⁵²[1] in that it is a high-spin species, Coriolis mixing and distortion of the bands seen in Bk²⁴⁸ would be expected.

An Es²⁵² sample was prepared by bombarding a 75 μ g Cf²⁵² target with 14 MeV deuterons for 5 days in the Argonne Cyclotron. The α spectra were taken (with Si detectors) at Argonne, but because of the MSU γ -detectors and the experience of this author with odd-odd actinides, a joint project was worked out, in which much of the γ -spectroscopy and analysis was performed here. A publication on our findings should be forthcoming early in 1969, so only a few brief remarks are made here.

We have found that 20% of the Es²⁵² decay is by electron capture to what appear to be two quasi-particle states just above 1 MeV in Cf²⁵²[2]. The ground state of Es²⁵² is a 7+ state - the triplet coupling of a 7/2+ [633+] proton and a 7/2+ [613+] neutron. The corresponding state in Bk²⁴⁸ lies at 593 keV, so the favored α group has an energy of 6.024 MeV; this group is only 1% abundant. The ground-state band in Bk²⁴⁸ appears to be a $K\pi=8-$ band - the triplet coupling of the 7/2+ [633+] proton with a 9/2- [734+] neutron. Surprisingly, the hindrance factors for decay to the 8-, 9-, and 10- members of this band, at 0, 72, and 151 keV, respectively, are only 30, 100, and 240. Also, the band is severely distorted, with $\hbar^2/2\mathcal{J}$ being only 4.00 keV (based on the 8- to 9- spacing). There is also a large positive B in the rotational energy equation,

$$E_I = E_0 + \frac{\hbar^2}{2\mathcal{J}} I(I+1) + BI^2(I+1)^2,$$

making the apparent K of the band less than it really is.

We are reasonably sure of the assignments of this ground-state band,

because it is not possible to fit the α -decay intensities with any lower spin assignments. Also, the $7/2^+$ [633 $\dagger$] proton state originates with the spherical $i_{13/2}$ state and has been characterized[1] as a state that causes severe distortions of the even-parity bands in Bk^{250} through Coriolis coupling. In the ground-state band of Bk^{248} we have this same proton state *plus* the $9/2^-$ [734 $\dagger$] neutron state, which originates with the spherical $j_{15/2}$ state and should have even larger Coriolis matrix elements. Thus, one can account for both the distortion and the relatively low hinderance factors for unfavored α decay. The states in Bk^{248} are mixed considerably by Coriolis coupling. We are presently looking into the calculations in more detail.

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XIII. The PDP-9 Computer

Wm. McHarris

Reflecting the need of the nuclear chemistry group for a large, versatile multichannel analyzer, the major purchase of the year was a PDP-9 computer, whose principal purpose will be to serve as such an analyzer. This computer, complete with multichannel analyzer software, was purchased at essentially the same cost as an elaborate hard-wired analyzer. The PDP-9 arrived in the middle of October, was operational within a day and a half, and, aside from irritating and recurring problems with the paper-tape punch, is being used routinely for data collection and, to a somewhat lesser extent, for data analysis.

Our system is made up of the basic PDP-9, which consists of an 8192 18-bit word machine, with teletype and fast paper tape punch (50 cps) and optical reader (300 cps), plus a type NH14B nuclear interface to a Northern Scientific type NS-620 dual 12-bit ADC and a type 34C oscilloscope interface. The teletype on our computer is an ARS 33, with (slow) paper tape punch/reader so that one can prepare source tapes off-line.

The nuclear interface allows one to collect either one-dimensional (up to 4096 channels) or two-dimensional spectra in two modes: An AC (accumulator) mode, in which the data are feed into the central processor under program control; this mode is useful when one needs to make decisions about data - for example, if one were distinguishing between, say, (p,d) and (p,t) reactions in a counter telescope system and desiring to collect each experiment in its proper memory location. Also, an MB+1 (or memory buffer plus one) mode, in which the data are fed into memory through the data channel; this mode is useful when high count rates are involved, for only three memory cycles (3 μ sec) are required to transfer a datum into memory. With very slight modification (possible in the laboratory here), the interface will be able to operate in a "LIST" mode, as well; this should be most useful when we extend our two-dimensional work (see §XV) to the PDP-9.

At present the oscilloscope interface drives a Tektronix RM503 oscilloscope with P-7, a long duration phosphor. This oscilloscope will remain in use, primarily for the taking of photographs. However, our group has ordered a Tektronix 611 storage scope, which is scheduled to arrive in

February, at which time we will trade this scope with the Fairchild 737A scope, which is now in use on the Sigma 7 computer. The Fairchild scope was found to be too demanding in time-sharing usage, and a storage scope that did not require updating of nonessentials was desired. Thus, because we plan only the most rudimentary background-foreground time sharing with the PDP-9, the exchange will be mutually beneficial.

The PDP-9 was furnished with a complete set of multianalyzer software, which will be discussed in §XIV, but it can, and has, served also as a very "available" small computer per se. It came implemented with two sets of software: *basic*, based on FORTRAN II, and *advanced*, based on FORTRAN IV. A number of routine, but semi-essential data processing programs, such as M^4 matrix element calculations and Fermi-Kurie calculations have already been performed on the PDP-9.

XIV. Multichannel Analyzer Programs

O. McHarris

To utilize the PDP-9 most fully and efficiently, we deemed it advisable to benefit from the courses offered by Digital Equipment Corp. on programming and maintenance of the computer. These courses were donated by DEC (the PDP-9 being an AEC discounted machine), and all travel and living expenses (~\$1000) for the two-week programming and three-week maintenance course were borne by the McHarris family budget.

The PDP-9 came furnished with quite good multianalyzer programs, and these have furnished us with a firm basis on which to build our changes and improvements. Four basic programs were furnished: 1) single parameter, four 1024-channel data regions; 2) single parameter, one 4096-channel data region; 3) dual parameter, $2^n \times 2^m$, where $2^n \times 2^m = 4096$; and 4) energy-calibration program. Our first task was to make the output of the PDP-9 intelligible to the Sigma 7, which is used for plotting and more sophisticated analysis. To do this the programs were altered so as to punch out in IBM (odd-parity) format rather than in ASCII. The tapes thus can be converted to cards with an IBM 826 tape-to-card converter, and the cards fed into the Sigma 7.

Also changed is the overflow routine, enabling us to obtain more than the $2^{18}-1$ maximum number of events in a channel. Currently in the process of being altered are the energy-calibration routines, to enable us to use least-squares polynomial energy, background, and peak fits. The next projects are 1) to write an "ad hoc double-precision" program, in which one can set aside extra words to be incorporated into double precision for only those channels that actually overflow, these not having to be known ahead of time; and 2) a tape-lising program for two-dimensional spectroscopy, to allow us to use more than 4096 channels (cf. §XV).

XV. Two-Dimensional γ -Ray Spectroscopy

R. Doeblner

In the elucidation of a nuclear decay scheme, coincidence experiments (integral, gated, and anti) are generally the most time consuming. Until recently each coincidence experiment had to be run separately and therefore required the repeated use of the cyclotron, electronic coincidence apparatus, and detectors. With the introduction of two-dimensional γ -ray spectroscopy, it is now possible to obtain almost all coincidence information from one experimental run. It also offers the added features of background subtraction from gated coincidence regions and the convenience of a single gain for all of the coincidence runs.

A program, "Task Event", has been written for the MSU Cyclotron Sigma 7 Computer under the "janus" time sharing supervisor by Douglas Bayer, a physics graduate student. Figure 1 shows the apparatus in flowsheet form used in a typical two-dimensional coincidence experiment. The coincidence apparatus used is similar to that used for a conventional integral coincidence experiment; however, unlike the conventional experiment, the two-dimensional experiment records the output of both detectors following each coincidence event. When two photons are detected in coincidence, the linear gate is opened allowing the output from detector 2 to go to the x side of the dual 4096 channel ADC, while the output from detector 1 goes to the y side of the ADC. With the dual ADC in the coincidence mode the x and y sides must be in coincidence before the two signals will be accepted. The dual ADC has been interfaced to the Sigma 7 computer and the program "Task Event" reads the data from both sides of the ADC through the interface. The events are then stored in a dedicated buffer in the computer until the buffer fills, whereupon the contents of the buffer are written on magnetic tape. The two coincidence events are recorded by channel number in one word of memory in the computer and on the magnetic tape; the x -side being recorded in bits 0-15 and the y -side in bits 16-31. Since the events are written to tape, this program cannot be used for count rates greater than ≈ 500 coincidence events per second. However, this has been quite sufficient for all coincidence runs we have attempted. A 2400 foot tape hold $\approx 1.3 \times 10^6$ coincidence events (one word = 1 coincidence event).

A program for recovering the coincidence data from the magnetic

tape in the desired form has been written to run under "Janus" by Dwight Beery, another physics graduate student. Figures 2 and 3a show the integral coincidence spectra obtained by Beery in the study of $\text{Nd}^{139\text{g}}$ [1] using a 3"x3" NaI(Tl) detector for the x-side and a 7 cm³ Ge(Li) detector for the y-side. Figure 3b shows the results of gating on the 404 KeV region of the NaI(Tl) side with no background subtraction. The gated region is shown by crosshatching in figure 2. By using the "Event Recorder" program for extracting the coincidence data from the magnetic tape, any gate may be selected, and provision is made for background subtraction.

Two-dimensional γ -ray spectroscopy can be made even more powerful by using two Ge(Li) detectors. We have made a successful preliminary attempt using 2.5 cm³ and 7 cm³ detectors. However because of the low efficiency of the 2.5 cm³ detector, the statistics were poor. We are presently waiting for the completion of a larger ~35 cm³ detector before attempting further experiments of this nature.

Reference

- 1] D. B. Beery, W. H. Kelly, and Wm. C. McHarris, Phys. Rev., to be published.

Fig. XV-2. Integral coincidence spectrum of $\text{Nd}^{139\text{g}}$ obtained using the coincidence data from the NaI(Tl) side of a 2-dimensional coincidence experiment.

Fig. XV-3. a) Integral coincidence spectrum of Nd^{139g} obtained using the coincidence data from the Ge(Li) side of a 2-dimensional coincidence experiment. b) 404 KeV gated coincidence spectrum obtained from 2-dimensional coincidence experiment using a 3"x3" NaI(Tl) detector and a 7 cm³ Ge(Li) detector.

XVI. Chemical Separation of Gd from Eu and Sm

R. Eppley

In the Gd decay scheme investigations it is often necessary to isolate chemically the Gd isotope under investigation from the radioactive Eu and Sm daughter products and other active species produced as by-products in the bombardment.

Ion exchange with α -hydroxyisobutyric acid[1] works well. However, the procedure requires at least two to three hours to carry out. For the separation of isotopes having short half-lives ($t_{1/2}$ for Gd^{145} is 24 min.), a faster method must be used. It is fortuitous that both Eu and Sm possess meta-stable +2 oxidation states while Gd exists only in the +3 state. The standard oxidation potentials are listed in table 1.

A strong reducing agent should transform Eu^{+++} to Eu^{++} , leaving the Gd in the +3 state, and thus a separation could be effected by precipitating Eu^{+2} as the sulfate. Hypophosphorous acid was tried as the reducing agent. Since H_3PO_2 is hygroscopic as well as being unstable, it must be prepared as needed. The method chosen for the preparation and purification is due to Klement[2]. This method employs the reaction,

(Several other methods have been published[3]). The NaH_2PO_2 was passed through the H^+ form of a cation exchange column (Dowex 50-X4). The resulting solution of H_3PO_2 is dehydrated by evaporation on a water bath and then placed in a vacuum chamber over P_2O_5 and kept there until all of the water is removed. The acid is then recrystallized. This method yields a product with a purity of approximately 98%. It was found that the H_3PO_2 could be kept for a long period of time by keeping it in a closed container at dry ice temperature.

We are primarily interested in separating Gd from Eu contaminants. That is, we want to reduce Eu^{+++} to Eu^{++} . The reaction is

where the half-reactions are

and

therefore,

$$E_{\text{cell}}^\circ = E_{\text{red}}^\circ - E_{\text{ox}}^\circ = 0.16\text{V}.$$

For europium,

$$E_{\text{Eu}} = E_{\text{Eu}}^\circ + \frac{RT}{nF} \ln \frac{a_{\text{Eu}^{+3}}^2}{a_{\text{Eu}^{+2}}^2},$$

and for H_3PO_2 ,

$$E_{\text{Acid}} = E_{\text{Acid}}^\circ + \frac{RT}{nF} \ln \frac{a_{\text{H}_3\text{PO}_3}}{a_{\text{H}_3\text{PO}_2}} \cdot a_{\text{H}^+}^2$$

$$= E_{\text{Acid}}^\circ + \frac{RT}{nF} \ln \frac{a_{\text{H}_3\text{PO}_3}}{a_{\text{H}_3\text{PO}_2}} - 0.0591 (\text{pH}).$$

Consequently, the cell reaction can be written as

$$E_{\text{cell}} = (E_{\text{Eu}}^\circ - E_{\text{Acid}}^\circ) + 0.0591 (\text{pH})$$

$$+ \frac{RT}{2F} \ln \frac{a_{\text{Eu}^{+3}}^2}{a_{\text{Eu}^{+2}}^2} \frac{a_{\text{H}_3\text{PO}_2}}{a_{\text{H}_3\text{PO}_3}}.$$

Finally,

$$E_{\text{cell}} = 0.16 + 0.0591 (\text{pH}) + \frac{0.0591}{2} \ln \left(\frac{a_{\text{Eu}^{+3}}^2}{a_{\text{Eu}^{+2}}^2} \frac{a_{\text{H}_3\text{PO}_2}}{a_{\text{H}_3\text{PO}_3}} \right).$$

As a result of this analysis, it is seen that as the pH increases, there is a better chance for the reaction to take place, all else being constant.

A reduction attempt was first carried out at a low pH (≈ 1), with a solution containing equal parts of Eu and Cd. H_2SO_4 was added in order to precipitate any Eu^{++} that might be formed. However, no precipitate was visible, indicating that the reduction had not taken place. Reductions

were attempted by raising the pH in small increments. Up to a pH of about 4, no precipitation occurred. However, a complicating factor is that as the pH is raised above four, hydroxide precipitation of the rare earths begins to occur. Both Gd and Eu begin to precipitate at the same pH and at this point the reduction process could no longer be checked.

At this time it looks doubtful that this reduction technique is feasible.

Table 1*

$Gd(s) \rightleftharpoons Gd^{3+}(aq) + 3e^-$	$\frac{E_{298}^{\circ}}{+2.40}$
$Eu(s) \rightleftharpoons Eu^{3+}(aq) + 3e^-$	+2.41
$Sm(s) \rightleftharpoons Sm^{3+}(aq) + 3e^-$	+2.41
$Eu^{2+}(aq) \rightleftharpoons Eu^{3+}(aq) + 1e^-$	+0.43
$Sm^{2+}(aq) \rightleftharpoons Sm^{3+}(aq) + 1e^-$	+1.55

*T. Moeller, *The Chemistry of the Lanthanides*, Reinhold Publishing Corp., New York, 1963.

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E°

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- 2] R. Clement, *Z. Anorg. Allg. Chem.* 260, 267 (1949).
- 3] G. Brauer, *Handbook of Preparative Inorganic Chemistry*, (Academic Press, New York, 1963), Vol. 1, 2nd. edition.

XVII. A Fast Target Changer

G. Giesler

The study of isotopes with half lives on the order of seconds requires a rapid method of moving the sample from the irradiation site to a detector. This changer should not involve opening the beam line to air pressure when changing a sample, and should be able to hold several samples. As we are studying isotopes whose half-lives are predicted to be small fractions of a minute, it was necessary that it take only a few seconds to move the sample to the detector. Since the changer would be located in a hot area, it must be controlled remotely. Reproducibility of positioning a sample must be within ± 0.25 in. because of the size of the beam and the sample.

A simple system fitting these requirements has been developed. It is an aluminum disc one foot in diameter with twelve aluminum arms equidistant about the circumference, each extending to a 3 ft radius. Each time a sample is changed, the disc rotates $5/12$ of a revolution. The changer can be started remotely, either manually or automatically when the beam is turned off. When changing samples, the disc is turned at about 8 RPM by a "Slosyn" motor, and stopped precisely by a U shaped aluminum plate mounted on a solenoid. The samples are mounted on a clip at the end of each arm, and are positioned reproducibly within ± 0.2 in. The whole assembly is movable vertically to adjust for beam-to-detector distance and is mounted on wheels for easy movement. This system does not require any changes to the existing equipment used to irradiate samples.

In a search for 33 second Ga^{63} , the sample changer has shown its value. We have seen γ -rays at 650 and 1060 keV corresponding to those predicted by Birstein, et al.[1] These γ -rays were produced from a $\text{Zn}^{64}(\text{p},\text{n})$ reaction on natural zinc, and the decay of the Zn^{63} daughter was also seen to substantiate the production of the Ga^{63} . Further work is in progress to obtain the complete decay scheme of Ga^{63} .

Reference

- 1] L. Birstein, M. Harchol, A. A. Jaffe, and A. Tsukrovitz, Nucl. Phys. 84, 81 (1966); L. Birstein, Ch. Drory, A. A. Jaffe, and Y. Zioni, Nucl. Phys., A97, 203 (1967).

XVIII. Cyclotron On-Line Mass Separation

K. Kosanke

As one moves away from the line of beta stability, greatly increasing decay energies allow the excitation of many more states through beta decay than is possible near beta stability. Thus, the amount of information obtainable through γ -ray and conversion electron spectroscopy is also greatly increased, making it quite desirable to extend γ -ray and conversion electron spectrum analysis into this region. However, the added decay energy also has the effect of greatly decreasing β -decay half-lives, making physical and chemical separation and identification procedures very difficult.

The use of isotope separators is an obvious way of combating this problem, and the use of on-line separators is an especially powerful technique. We expect to have an on-line mass separator operational in early 1969. Figure 1 shows the location for the mass separator in the experimental areas of the cyclotron building, and figure 2 is a block diagram of the system to be used.

The heart of the system is a Granville-Phillips (Finnigan) residual gas analyser[1] with slight mechanical modification. This is an electrostatic quadrupole mass filter, capable of separating masses of 1 part in 750 for charged particles having energies less than 100 eV. Both particle velocity independence (in Z direction) of the mass transmitted through the separator and the stability of the separator makes the application of this system to isotopic separation quite attractive.

The most severe difficulty that we foresee is the requirement that the nuclear recoils, initially with energies up to ~ 2 MeV (see table 1), be degraded to energies less than 100 eV for separation to be effected in the quadrupole. We have considered three methods of accomplishing this: passing the recoils through extremely thin foils,[2,3] passing the recoils through a He thermalizer[4], diffusion of the recoils through heated graphite[5]. Upon consideration of energy resolution, transit time, efficiency, and simplicity of construction, we have tentatively decided to use a He thermalizer.

Separated isotopes from the quadrupole can be studied using any of several techniques: γ -ray spectrum analysis using solid state detectors, internal conversion electron spectrum analysis using solid state detectors or eventually using the MSU six-sector beta spectrometer on-line, and α and β decay energies using solid state detectors. Also, many of the product

Nuclides would lie close enough to the line of particle instability that decays such as beta-delayed proton emission[6] or α emission[7] might be observed.

Sometimes the identification of mass number is not enough to identify a species uniquely, such cases as when the reactions $(\tau, p\alpha)$ and $(\tau, (x+1)n)$ are competing reactions. In these instances the MSU six-sector beta spectrometer could be used to determine atomic number by looking at K-conversion electron energies.

Clearly this collection of equipment (MSU sector-focused, isochronous cyclotron, on-line electrostatic isotope separator, MSU six-sector beta spectrometer) is uniquely well suited for studying nuclides far from the line of beta stability.

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- 1] For a description of the principles of electric quadrupole mass separation, see, e. g., W. Paul, H. P. Reinhard, and U. von Zahn, Z. Phys. 152, 143 (1958).
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- 5] I. A. Bernas, et. al., Ark. Fys., 36, 77, (1967).
- 6] P. L. Reeder, A. M. Poskanzer, R. A. Esterlund, and R. McPherson, Phys. Rev., 147, 781 (1966).
- 7] See paper XI in this report.

Table 1

Some of the Considerations Necessary in the Design
of Experiments Using the Isotope Separation System.

MSU Cyclotron beams ^a	Maximum cyc. beam currents ^b	Ag ¹⁰⁹ Target		Pb ²⁰⁸ Target	
		Possible reaction	Recoil energy ^c	Possible reaction	Recoil energy ^c
30 MeV p	~20 μ A	(p,4n)	0.28 MeV	(p,4n)	0.15 MeV
50 MeV p	~20 μ A	(p,7n)	0.48 MeV	(p,6n)	0.25 MeV
50 MeV He ³	~3 μ A	(He ³ ,9n)	1.45 MeV	(He ³ ,9n)	0.74 MeV
76 MeV He ³	~10 μ A	(He ³ ,12n)	2.28 MeV	(He ³ ,11n)	1.14 MeV

^aIt is expected that the MSU cyclotron will also be producing C¹², N¹⁴, and O¹⁶ beams shortly.

^bIndications are that beam currents will be limited by our ability to cool our targets and not by the cyclotron itself.

^cAssumes isotropic emission of neutrons and no energy degradation in the target material.

Fig. XVIII-1. Physical layout of the experimental areas at the MSU cyclotron, showing the position of the quadrupole mass filter on the 0° beam line in the β -spectrometer room.

Fig. XVIII-2. Block diagram of the scheme for mass separation on-line with the quadrupole mass filter.

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Publications Involving Members of the
Nuclear Chemistry Group

I. Papers in Print

1. "High Resolution Gamma-Ray Spectroscopic Studies of the Decays of 2.6-Hour Nd^{141g} and 60-Second Nd^{141m} ", D. B. Beery, W. H. Kelly, and Wm. C. McHarris, Phys. Rev. 171, 1283 (1968).
2. " $^{203}\text{Pb}^m$ Decay and $M4$ Transition Probabilities in Neutron-Deficient Lead Isotopes", R. E. Doebler, Wm. C. McHarris, and C. R. Gruhn, Nucl. Phys. 120, 489 (1968).
3. "The States of ^{117}Sn ", D. B. Beery, G. Berzins, W. B. Chaffee, W. H. Kelly, and Wm. C. McHarris, Nucl. Phys. 122, ... (1968).

II. Papers Submitted to Journals or to Be Submitted in the Very Near Future

4. "A Rare-Earth Szilard-Chalmers Reaction", Wm. C. McHarris, Submitted to J. Chem. Ed.
5. "The Decay Chain Nd^{139m} , Nd^{139g} , and Pr^{139} ", D. B. Beery, W. H. Kelly, and Wm. C. McHarris, submitted to Phys. Rev.
6. "The Decay of Gd^{149} ", R. E. Eppley, Wm. C. McHarris, and W. H. Kelly, submitted to Phys. Rev.
7. "The Electron Capture Decay of Pb^{200} ", R. E. Doebler and Wm. C. McHarris, submitted to Phys. Rev.
8. "A Single-Crystal Ge(Li) Conversion Coefficient Spectrometer", C. Maggiore, C. R. Gruhn, R. Todd, and Wm. C. McHarris, submitted to Nucl. Inst. and Meth.
9. "A New $N=81$ Isomer: Gd^{145m} ", R. E. Eppley, D. B. Beery, W. H. Kelly, and Wm. C. McHarris, to be submitted to Phys. Rev.
10. "The Decay of Fe^{53} ", J. N. Black, Wm. C. McHarris, and W. H. Kelly, to be submitted to Phys. Rev.

III. Abstracts and Papers Presented at Meetings

1. The Electron Capture Decay of Gd^{149} , R. E. Eppley and Wm. C. McHarris, American Chemical Society San Francisco Meeting, 1-5 Apr 1968, paper 072.
2. "High-Resolution Gamma-Ray Spectroscopic Studies of the Decays of 2.6-h Nd^{141g} and 60-sec Nd^{141m} ", D. B. Berry, Wm. C. McHarris,

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 8. "The Michigan State University 50-MeV Sector-Focused Cyclotron" (Invited Paper), Wm. C. McHarris, Symposium on "Major National Nuclear Chemistry Facilities", American Chemical Society Minneapolis Meeting, 14-18 April 1969.