ABSTRACT

NUCLEAR SPECTROSCOPIC STUDIES OF NEUTRON-DEFICIENT, ODD-MASS RARE EARTHS NEAR THE N=82 CLOSED SHELL

By

Richard Earl Eppley

The decay schemes of Gd^{149}, Gd^{145g+m} and Sm^{141g+m} have been studied by γ- and β-ray spectroscopy in an effort to elucidate their nuclear properties. Also, a search for Dy^{147g+m} has been conducted, although no decay scheme can be proposed.

γ-ray spectroscopic techniques including Ge(Li) singles, Ge(Li)-NaI(Tl) coincidence, and Ge(Li)-Ge(Li) megachannel, 2-dimensional coincidence techniques have been employed. Where appropriate, electron spectra have been obtained by use of Si(Li) surface barrier detectors.

Twenty-five γ rays have been attributed to the electron-capture decay of 9.4-day Gd^{149} and incorporated in a decay scheme having 13 levels with energies of 0, 149.6, 459.9, 496.2, 574.2, 666.0, 748.2, 794.8, 812.4, 875.8, 933.3, 939.1, and 1097.3 keV. The isomeric decay of Gd^{145m} has been characterized as having a half-life of 85±3 seconds and an $M4$ transition of 721.4±0.4 keV. A direct ε/β^+ branch has also been determined as accounting for 4.7% of the decay from this level. Gd^{145g} is reported to have 38 γ rays associated with its decay, 26 of which (accounting for ≈97% of the total γ intensity) are incorporated into the decay scheme. This

decay scheme has 19 levels placed at 0, 329.5, 808.5, 953.4, 1041.9, 1567.3, 1599.9, 1757.8, 1845.4, 1880.6, 2203.3, 2494.8, 2642.2, 2672.6, 3236.0, 3259.6, 3285.6, 3623.8, and 4411.3 keV. Sm^{141g} and Sm^{141m} appear to have 9 and 32 associated γ rays, respectively. Levels in Sm^{141g} have been placed at 0, 196.6, 403.9, 728.3, 858.5, 1292.7, 1495.6, and 2005.0 keV. Levels in Sm^{141m} have been placed at 0, 196.6, 628.6, 804.5, 837.1, 974.0, 1108.1, 1167.2, 1313.2, 1414.8, 1834.0, 1983.1, 2063.5, 2091.6, 2119.2, and 2702.4 keV. The half-lives of Sm^{141g} and Sm^{141m} have been measured to be 11.3±0.3 and 22.1±0.3 minutes respectively.

Spin and parity assignments for the nuclear levels encountered in this investigation are based on log *ft* values, relative γ intensities, and transition multipolarities (where internal-conversion-electron intensities were available). Two intense transitions in Eu^{145}, which account for ≈72% of the β decay from Gd^{145g}, are interpreted as being three-quasiparticle states. Also, there are 6 states in Pm^{141}, lying between the energies of 1414.8 and 2702.4 keV, which appear to be three-quasiparticle states analogous to those reported recently for Pr^{139}. Mechanisms are suggested for these three-quasiparticle types of decay in terms of simple shell-model transitions and nucleon rearrangements.

Several families of nuclear isomers are compared with each other in terms of their level energies and reduced *M*4 transition probabilities. These include the *Z*=82, *N*=81, *N*=79, *Z*=50, *Z*=49, and *N*=49 families. High multipolarity electromagnetic transitions from these isomers are considered to be quite "pure" single-particle

transitions. The experimentally measured transition probabilities are compared to these single particle predictions in an effort to determine the degree to which this is true. Finally, levels in the odd-mass N=82 and Z=63 nuclides are traced as far as possible in an effort to understand nuclear behavior in this region better.

NUCLEAR SPECTROSCOPIC STUDIES OF NEUTRON-DEFICIENT, ODD-MASS RARE EARTHS NEAR THE N=82 CLOSED SHELL

By

Richard Earl Eppley

A THESIS

Submitted to
Michigan State University
in partial fulfillment of the requirements
for the degree of

DOCTOR OF PHILOSOPHY

Department of Chemistry

1970

ACKNOWLEDGEMENTS

I wish to thank Dr. Wm. C. McHarris not only for suggesting this region of study but also for guidance and help in all phases of the ensuing investigation, including the preparation of this thesis.

I also wish to thank Dr. W. H. Kelly for his help and advice. His suggestions on the performance of experiments and analysis of data are greatly appreciated.

Dr. H. G. Blosser and Mr. H. Hilbert have assisted with the operation of the Michigan State University sector-focused cyclotron. As with any large, complicated machine, it had its temperamental moments at which times their special touch was necessary for a successful run.

The members of our research group have all contributed in one way or another to the successful completion of this thesis research. Dr. D. Beery, Mr. J. Black, Mr. W. Chaffee, Mr. J. Cross, Mr. R. Doebler, Mr. R. Firestone, Mr. G. Giesler, Mr. R. Goles, Mr. K. Kosanke, Mr. R. Todd, and Dr. R. Warner have all helped in this work. I particularly wish to thank Dr. D. Beery for his help in the early stages of this project. Mr. R. Todd and Dr. R. Warner have shared jointly in the Sm^{149} research. In addition they have provided me with many hours of discussion on the research and other things philosophical.

Dr. P. Plauger, Mr. R. Au, Mr. and Mrs. W. Merritt and the cyclotron computer staff have been helpful in the writing of programs and with the operation of the computer.

Help has also been given by Mr. R. N. Mercer and his staff in the cyclotron machine shop, and by Mr. W. Harder in the cyclotron electronics shop.

I also wish to thank Mrs. Ina Samra for her help with the typing of this thesis as well as the typing of several reports for publication.

The National Science Foundation, U. S. Atomic Energy Commission and Michigan State University have provided much of the financial assistance for this research.

Finally, I thank my wife Sara for her help and encouragement during these past few years. I also thank her for just plain putting up with me during this hectic time.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Figure

CHAPTER I

INTRODUCTION

"...In the development of atomic physics the interaction of atoms with electromagnetic radiation has been of paramount importance for the understanding of atomic structure. In the case of nuclei the interaction has not been so important a tool, since, unlike the atomic case, the wavelengths of interest are so short that they cannot be measured by the usual optical devices. The rather indirect methods which must be employed make the energy determination quite inaccurate compared with spectroscopic standards, and the available resolution is low. Furthermore, in most cases the radiation process is only one of many competing processes (such as particle emission or sometimes even beta-decay) and its probability is correspondingly lower.

For these reasons the study of gamma-rays from nuclei has remained on a rather rudimentary level..." (Blat52).

This was written in 1952, not so very long ago in terms of years but a very long time ago in terms of technological development. This thesis has been possible precisely because such a statement about γ-ray spectroscopy is no longer valid.

In the early 1960's, when solid state detectors became available, energy resolution was dramatically improved to the point where, for the first time, individual transitions could be resolved

in complex γ-ray spectra. Needless to say, a whole new dimension was then opened for the study of nuclear properties. γ-ray spectroscopy immediately became an important tool for these investigations.

The present investigation is a good example of the unpredictableness of scientific research. One begins a research project looking for answers to predetermined questions and, as in the present case, finds a few answers and many more unanswered questions. Also, one is apt to end up investigating certain phenomena which were unrealized at the outset of the study. The topics of this thesis evolved in just such a manner.

The original intent was to study a sequence of neutron-deficient odd-mass rare earth isotopes in an effort to elucidate their decay schemes in as much detail as possible. This information would be of great value for the testing of existing nuclear models as well as possibly suggesting a means of, at least semi-emperically, being able to describe in a uniform fashion the change in the various levels as one goes from permanently deformed nuclei to spherical nuclei near closed shells. Gadolinium was chosen as an element satisfying this criterion, as well as possessing neutron-deficient isotopes in the desired spherical region which would be obtainable with our energy limitations (p_{max} = 45 MeV and He^{3}_{max} = 70 MeV).

With this intent work was completed on the decay schemes of Gd^{149}, Gd^{145m}, and Gd^{145g}. However, with our discovery of an 85-

second isomer in Gd^{145}, we became interested in studying other N=81 isotones (six of which were already known). Preliminary calculations, based on the known $M4$ N=81 isotones, indicated that Dy^{147m} should be of a half-life that we could handle by our counting techniques. Thus, a series of experiments was initiated at Yale University in an effort to identify this species.

At about this same time, states were observed in Nd^{139m} (McH69j,Bee69d) which were characterized as three-quasiparticle states. These states are quite uncommon but where found to be very useful for determining nuclear properties of relatively high-lying states. Out of the Nd^{139m} work came predictions for similar states in Sm^{141m} and Nd^{137m}.

Consequently, the nuclear species studied during the course of this thesis work were Gd^{149}, Gd^{145g}, Gd^{145m}, Sm^{141g}, Sm^{141m}, and Dy^{147}.

Chapter II describes the many types of experimental set-ups used during the course of this work. The latter part of the chapter also describes three chemical techniques, used primarily for Gd^{149}, for separating the Gd fraction of the target material from contaminents of Eu and Sm.

Since we are fortunate in having two computers at our disposal, an SDS Sigma-7 and a DEC PDP-9, data analysis could be accomplished both in the quantity and in the quality needed for this work. Three of the most useful data reduction codes used in this laboratory are described in Chapter III.

The heart of this thesis, the experimental results, comprise Chapter IV. During this work, certain "discoveries" proved to be particularly interesting. As already mentioned, this investigation began with Gd^{149}. The fairly complete study we made of the Eu^{149} levels populated by the decay of this species will undoubtedly prove valuable for any future theoretical considerations of this region. Eu^{149} is in the twilight zone. In § 4.1. we report the observation of more levels than can be incorporated in a simple shell model picture, while, at the same time, the levels are such that they cannot be fit into rotational structures as would be expected of a permanently deformed nucleus. There is presently no nuclear model which can adequately account for such a situation.

As opposed to higher mass Gd isotopes, Gd^{145g} decays primarily to two states at 1757.8 and 1880.6 keV. These states, in turn, decay predominantly directly to the ground state of Eu^{145}. We report over 30 γ transitions accompanying the decay of Gd^{145g} with the above mentioned two transitions accounting for $\approx$63% of the total γ intensity. This anomaly occurs, most likely, because the $s_{1/2}$ state drops down to become the ground state (replacing the $d_{3/2}$ state). Another interesting facet of this work was the observation of an $\varepsilon/\beta+$ branch from the isomeric decay of Gd^{145}. Of the N=81 isomers which have been studied previously, only Sm^{143} appears to have a β-decay branch. From such branchings one can directly arrive at occupation probabilities for the state involved.

In apparent confirmation of the three-quasiparticle predictions discussed above, Sm^{141m} does exhibit 6 β-allowed states lying

between 1414.8 keV and 2702.4 keV. These states have properties similar to those in Nd^{139m} with one notable exception. The three-quasiparticle states in Nd^{139m} have a plethora of interconnecting transitions; these states in Sm^{141m} have not a single observed interconnecting transition. Five of these six states in Pm^{141} decay principally to an $11/2^-$ state at 628.6 keV. The sixth state, however, completely bypasses the $11/2^-$ state, preferring to decay to the ground and 1st excited state instead. In § 4.4.11. a mechanism will be proposed for this three-quasiparticle type of decay.

The decay of Sm^{141g} still remains as a partly unsolved problem. There are two intense transitions, at 403.9 keV and 438.2 keV, neither of which can be ruled out as belonging to this ground state decay. The puzzle is that one of them, the 403.9-keV state, exhibits a half-life of 11.3 minutes, while the other level has a measured half-life of 10.0 minutes. We have evidence both for and against both of these transitions being from levels in Pm^{141}. Other than this the decay of Sm^{141g} seems to be straightforward, decaying largely to the Pm^{141} ground state.

The identification of Dy^{147g} and Dy^{147m} still remains a challenging problem. We began a search (primarily for the Dy^{147m} species) after our experimental determination of the decay properties of Gd^{145m}. The only feasible means of producing this species is with a C^{12} beam on either a Nd^{142} or a Sm^{144} target. Because of the beam requirement we repaired to Yale University where their heavy ion linear accelerator was made available for our use. §4.5. will describe our results to date. We have evidence from several types of experiments

which indicates that we do produce Dy^{147}. We do have seemingly conclusive evidence that there is no Dy^{147m} $M4$ transition (at least not in our useable half-life range) with a measurable internal-conversion intensity. This could well mean that Dy^{147m} de-excites predominantly via an ε/β^+ branch.

Chapter V sketches the derivation of the single-particle transition probability expressions for various multipole transitions, beginning with the Maxwell equations. These expressions, first expressed by Weisskopf, are by far the most frequently used estimates against which to compare experimentally measured transition half-lives. The purpose of the chapter is not to present a mathematically rigorous derivation but rather to give a beginner a feeling for what is invloved in such a derivation.

The final chapter, Chapter VI, is primarily concerned with the trends in families of nuclear isomers, both neutron and proton, around the 50 and 82 closed shells. For example, the N=79 and N=81 isotones show marked energy dependence on Z while the isomers of the lead isotopes (Z=82) are quite similar in energy and consequently show a constancy in their reduced transition probabilities. The trends of other isomeric families will be compared to these two seemingly different types of behavior.

Inasmuch as possible the energy levels of nuclei in the Gd-Eu-Sm region will be traced as functions of energy. The available information on much of this region is quite sketchy and few conclusions can be drawn.

CHAPTER II

EXPERIMENTAL EQUIPMENT AND PROCEDURES

All of the nuclear species studied in the present investigation decay by positron emission and/or electron capture. A variety of techniques, discussed in this chapter, were used to construct decay schemes based on the study of these decay characteristics.

This chapter is divided into three main parts. The first, consisting of § 2.1. through § 2.3, describes the electronics set-ups for the γ-ray and conversion-electron data taken at Michigan State University. The apparatus used at Yale University, in conjunction with the Dy^{147} experiments, is briefly described in § 2.4. § 2.5 describes the chemical separation techniques used principally for the isolation of the Gd^{149} activity.

2.1. γ-Ray Spectrometers

As mentioned in Chapter I, the Ge(Li) γ-ray detector has been principally responsible for the resurgence of the study of complex nuclear structures by measurements of the γ-ray de-excitations. Hand in hand with the development of these solid state detectors has been the development of vastly improved electronics. In just the approximately $3\frac{1}{2}$ years since the beginning of this study, we have gone from a γ-ray spectrometer system having a resolution of ≈5.6 keV FWHM for the 1333-keV γ of Co^{60}, a detector efficiency of <<1%, and an analyzer having 1024 channels of memory to our present system: a detector resolution of 1.9 keV FWHM for Co^{60}, a detector efficiency of 3.6% (relative to a 3×3-in. NaI(Tl) detector 25 cm. from a Co^{60} source), and the capability of obtaining spectra up to 8192 channels in length.

2.1.1. Ge(Li) Singles Spectrometer

Our γ-ray spectrometer is really a "system" that includes, besides the Ge(Li) detector and bias supply, a charge sensitive FET preamplifier, a spectroscopy amplifer, an analog to digital converter (ADC), and some type of memory or storage unit and associated spectrum readout. The majority of the electronic equipment is modular so that equipment from various manufacturers can be used together for a particular experiment.

A typical spectroscopy charge-sensitive preamplifer integrates and shapes the charge output from the detector and presents a tail pulse (rise time ≈25 nsec, decay time ≈50 μsec) to the main amplifier.

The spectroscopy amplifiers which we use have considerable versatility. Shaped unipolar or bipolar output pulses with 0.25-μs widths are available for timing purposes or for input to an ADC. These amplifiers have highly linear amplification responses and include such features as adjustable pole-zero cancellation and base-line restoration.

The pole-zero feature permits precise elimination of undershoots on the amplifier pulse after the first differentiation. This becomes important at high counting rates, since if the undershoot saturates, the amplifier will be blocked not only for the time of the primary pulse, but also for the duration of the undershoot. Also, succeeding pulses which may fall into the undershoot will have an apparent area smaller than the actual area, thus causing a loss in resolution.

Base-line restoration also improves resolution at high count rates by restoring the undershoot of the amplifier signals to a DC baseline after all other shaping has been accomplished. The improved resolution is accomplished by the reduction of pile-up distortion caused by undershoot.

In this laboratory spectra can be collected in a variety of analyzers. Of principle use for the present research has been a Nuclear Data ND-2200 4096-channel analyzer, a PDP-9 computer interfaced to a Northern Scientific NS-625 dual 4096-channel ADC, and our Sigma 7 computer interfaced to four Northern Scientific 8096-channel ADC's. The NS-625 has a 40-MHz digitizing rate and the NS-629 has a 50-MHz digitizing rate.

2.1.2. Ge(Li)-NaI(Tl) Coincidence Spectrometers

Coincidence spectra have played an important role in the present research. Since the vast majority of nuclear excited states have very short half-lives as compared with our ability to measure them, coincidence units with resolving times on the order of nanoseconds can be used to study those transitions which are in fast or "prompt" coincidence with one another. In this manner, coincidence spectra are a useful tool for determining the relationships of the observed γ-transitions and their interconnections with the deduced nuclear energy levels.

Several types of single parameter coincidence spectra have proved to be useful. Both prompt and delayed coincidence spectra have been used, utilizing a Ge(Li) detector for one leg of the timing circuit. In addition, anti-coincidence spectra and 511-511-keV (pair) coincidence spectra have been obtained by use of a Ge(Li) detector in conjunction with an 8×8-in. split annulus. Each of these will be described in the following sections.

2.1.2.A. Coincidence Spectrometer using Ge(Li) *vs* 3×3-in. NaI(Tl)

Before the advent of Ge(Li)-Ge(Li) coincidence techniques, which will be described in § 2.1.2.C, NaI(Tl) scintillators coupled to photomultiplier tubes provided the principle means of obtaining timing pulses for coincidence spectra. Such a set-up is shown in Figure 1. In addition to the equipment needed for the collection of a singles spectrum, equipment is needed to provide timing information. As shown in this figure, a 3×3-in. NaI(Tl) detector was used for this purpose. The pulses from the photomultiplier were fed to a spectroscopy

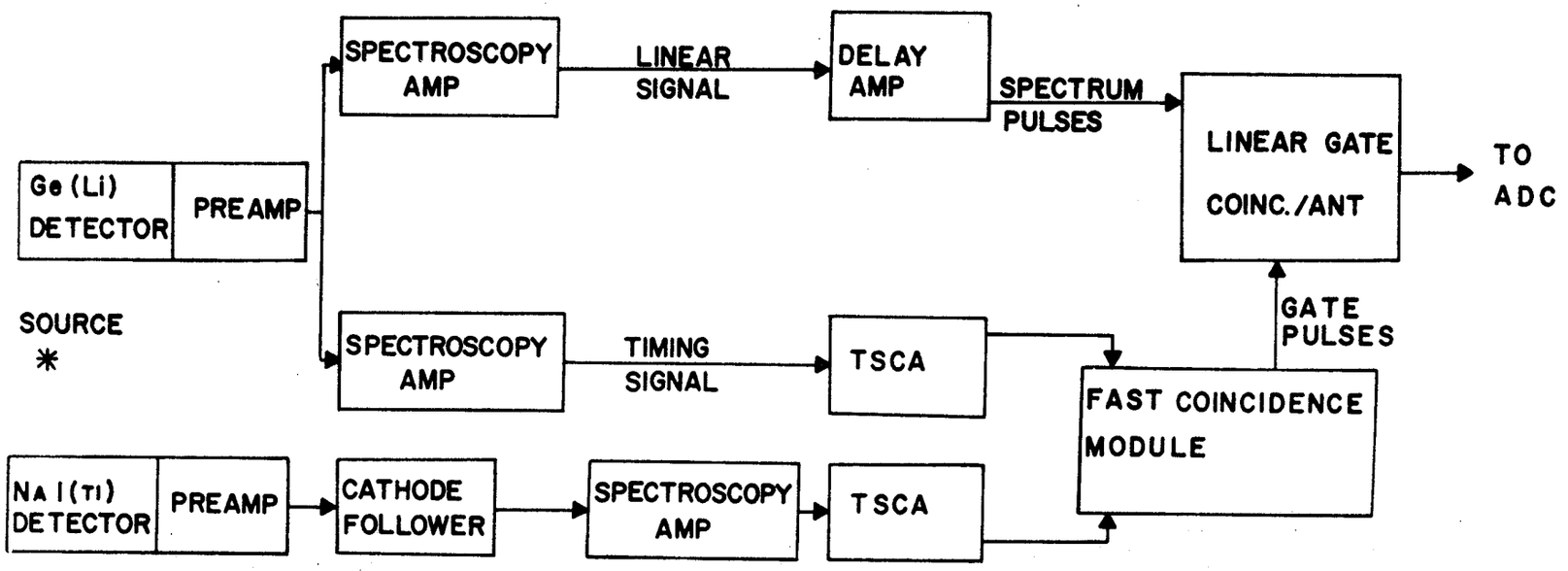

Figure 1. Schematic illustration of the Ge(Li)-Nai(T1) coincidence apparatus.

amplifer through a cathode follower, which is used simply as a buffer for impedance mismatch between the photomultiplier and amplifier. The NaI(Tl) signals are then presented to a timing single channel analyzer (TSCA). The TSCA, in turn, is used first to determine the energy gate and then to output a logic pulse to a fast coincidence unit. In this manner timing pulses are output only for pulses which fall in the voltage range selected by the TSCA.

Similarly, a timing signal must be derived from the Ge(Li) linear signal. This is accomplished by the use of a second TSCA which outputs a logic signal to the coincidence unit. The function of the fast coincidence unit is to output a logic signal only when a Ge(Li) and NaI pulse arrive within a specified time interval of one another. For most of our coincidence work this resolving time (2τ) was set at 100 nsec.

The majority of coincidence runs utilized an external linear gate. A logic pulse from the fast coincidence unit opens the gate for a pre-determined length of time, allowing any linear signal present at the input during that time to pass through the open gate and on to the ADC. In this manner, only pulses from γ-ray transitions occurring within the specified resolving time are allowed to pass through the linear gate and be digitized by the ADC. The delay amplifier shown in Figure 1 is solely for the purpose of delaying the linear signal to compensate for the delay incurred by the timing signals while they are proceeding through the coincidence circuits. The delay can be adjusted to insure that the linear singnals and coincidence timing signals arrive simultaneously at the gate.

2.1.2.B. Ge(Li) vs 8×8-in. NaI(Tl) Split Annulus

Several types of coincidence experiments can be carried out using the split annulus for one of the detectors. One-half of the annulus can be used as a simple NaI(Tl) detector. In addition to the usual prompt and delayed coincidence and anti-coincidence experiments that can be accomplished with the annulus are Compton suppression experiments. The annulus can also be used as a pair spectrometer for obtaining β^+ feedings and double escape spectra. For the present research, the annulus was used primarily for anti-coincidence experiments and as a pair spectrometer.

While coincidence spectra show the relationship of γ-rays which are related to each other, anti-coincidence spectra enhance those transitions which are either transitions to the ground state from levels populated primarily by electron capture with little or no γ-ray feeding from above, or transitions from a level with a half-life considerably longer than the resolving time of the apparatus, i.e., transitions from isomeric levels. The electronics set-up using the annulus in an anti-coincidence experiment is shown in Figure 2. A 3×3-in. NaI(Tl) detector is positioned in the top of the annular tunnel in order to increase the solid angle subtended by the detectors. The Ge(Li) detector is positioned in the bottom of the annular tunnel, facing the 3×3-in. NaI(Tl) detector at the top. The source is placed between these two detectors, approximately in the middle of the tunnel. The electronics used is very similar to that used in the coincidence experiments which have been described in § 2.1.2.A. above The main difference is that now there are three timing signals

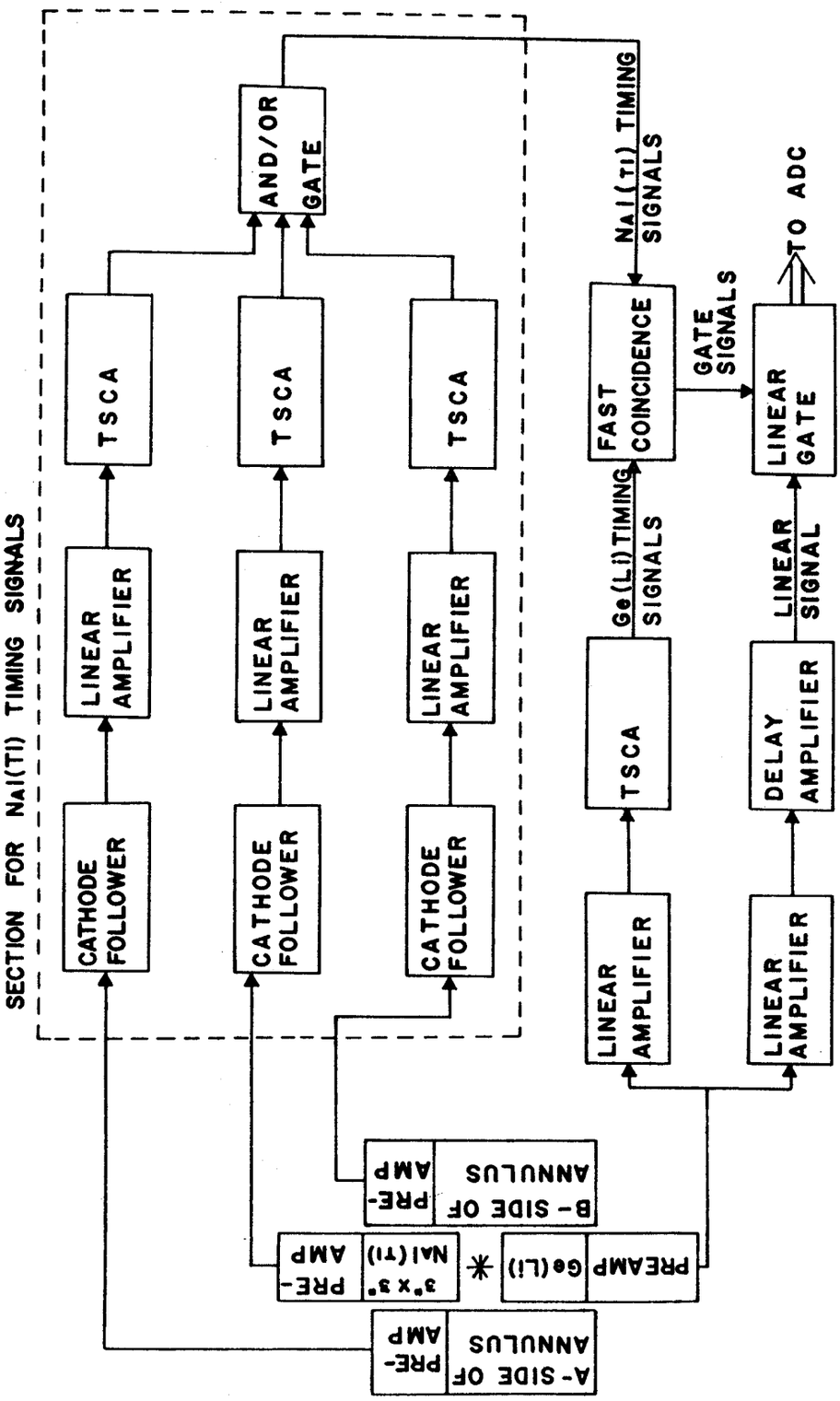

Figure 2. Schematic illustration of the anticoincidence apparatus. This same apparatus is used for obtaining pair spectra after the elimination of the 3×3-in. NaI(Tl) detector leg and AND/OR gate.

necessary instead of just one. These timing signals must be combined in some way so that the fast coincidence unit is presented with a single logic pulse (no matter whether a signal is present in only one leg or a combination of the legs). For this purpose we have used an ORTEC universal coincidence module, shown in Figure 2 as an AND/OR gate. The number of coincidence requirements is set to 1 on this module to effect the desired single, well shaped output pulse. In this mode, resolving time for the unit has no meaning and does not have to be set. The only relevant resolving time consideration is that associated with the fast coincidence module, which was commonly set to 100 nsec for these experiments.

Another use for the annulus was as a pair spectrometer for determining the β^+ feeding of levels. In this mode of operation the resulting spectra show enhancement of those γ-rays involved in prompt coincidence with levels which are fed by β^+ emission. Of course, double-escape peaks are also greatly enhanced in these spectra. The apparatus shown in Figure 2 can be employed, with the exception that the 3×3-in. NaI(Tl) detector and associated electronics are not used. Since we are now interested in a triple coincidence requirement, i.e., a coincidence output pulse only when a timing signal from the Ge(Li) detector and both halves of the annulus occur within the resolving time of the fast coincidence module, the AND/OR gate is not needed. Signals from each half of the annulus, as well as the Ge(Li) timing signals, are routed directly to the fast coincidence unit, which is set for the triple-coincidence requirement. With these exceptions and different settings of the linear gate, the experimental set-up remains the same as for the anticoincidence experiment.

2.1.2.C. Ge(Li)-Ge(Li) Megachannel, 2-Dimensional Coincidence Spectrometer

With the availability of larger volume Ge(Li) detectors came the technique of 2-dimensional coincidence experiments. The ultimate aim of such experiments is to obtain coincidence data for any and all peaks or regions with a single run. (In practice the data obtained usually fall somewhat short of this goal.)

Even though Ge(Li) detectors are still quite inefficient with respect to available NaI(Tl) detectors, they compensate for this in other ways. Due to the Ge(Li) resolution, peak to Compton ratio, etc., good coincidence data can be obtained with fewer counts per channel than necessary with NaI(Tl). The electronics used in this laboratory for the 2-dimensional coincidence experiments is diagrammed in Figure 3. This is a double coincidence set-up where we want the linear signal to be recorded from each of the detectors, hence the fast coincidence unit transmits a logic pulse to two linear gates, one for each detector. The two detectors presently used for these runs have efficiencies of 2.5% and 3.6%.

Coincidence events are collected on magnetic tape by use of a task called EVENT which runs under the JANUS monitor system of our Sigma-7 computer. The data obtained under this system are a 2-dimensional integral coincidence array.

Each linear gate transmits a linear signal when opened by a coincidence event. These two linear signals are input to separate ADC's, operated in a synchronous mode, which are interfaced to the computer. The encoded coincidence pair is then stored on magnetic

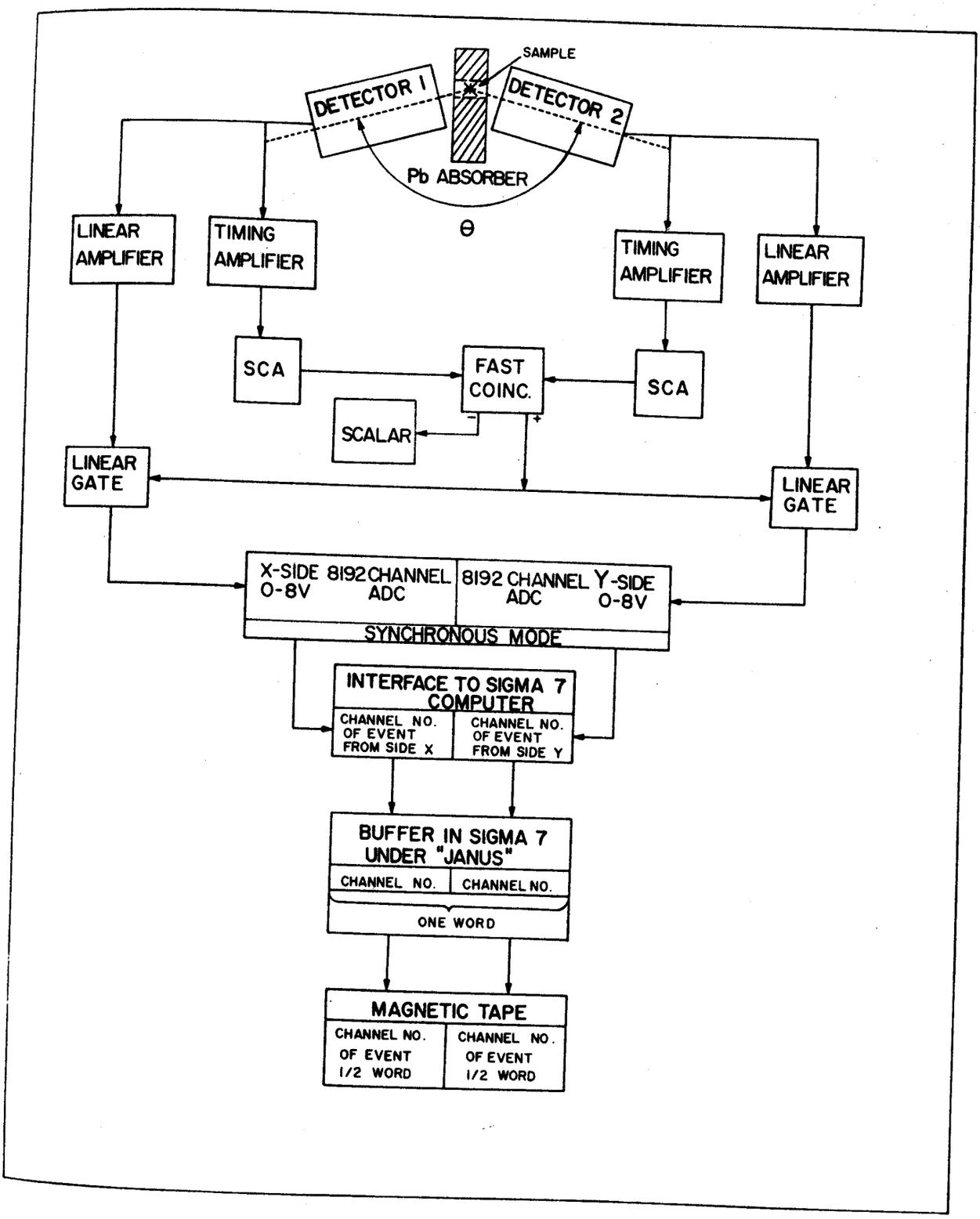

Figure 3. Schematic diagram of the megachannel 2-dimensional apparatus.

tape as one word, the first half of the word being the address from detector 1 and the second half of the word being the address associated with detector 2. The lead absorber shown in the diagram between the detectors is used to reduce γ-ray scattering between the two detectors. Without the absorber spurious peaks appear in the coincidence spectra. These scattering phenomena are discussed in (Gie70)

The data thus stored on the magnetic tape represent information on all peaks in coincidence with all other peaks. All that remains is to strip these 2-dimensional data by taking the appropriate slice representing a particular peak on energy region. These gated specta are recovered off-line on the Sigma-7 computer through use of the program EVENT RECOVERY (event). This code allows us to search the tape for a miximum of 10 gates per single pass of the tape. Background subtraction can be effected for any or all of the gates on the same pass.

All coincidence runs used in the present work utilize a 4096×4096 array. The system operates satisfactorily at counting rates up to ≈2000 coincidence events per second.

2.2. Conversion-Electron Spectrometer

For the conversion electron spectra of Gd^{145m} we utilized a 100-μ thick Si(Li) detector. This detector was mounted in the usual γ-ray cryostat and cooled to methanol-dry ice temperature (≈77°C) for operation. A 1/4-mil Havar window was used for this detector. A 200-V bias was used for operation of the detector. A typical resolution of ≈5 keV on the Cs^{137} *K*-conversion peak was obtained, thus allowing

separation of the *K*- and *L*- conversion electron peaks. In addition to the required bias supply, an ORTEC 109A preamplifier and 451 spectroscopy amplifier were used in conjunction with an analyzer. To utilize this detector for the determination of internal conversion coefficients required the simultaneous counting of the source with both the conversion-electron detector and Ge(Li) γ-ray detector. Normalization between the detectors (efficiency corrections) was obtained through the counting of standards having known (and measurable) conversion coefficients, Cs^{137} and Bi^{203} being the most suitable. Conversion electron spectra of Gd^{145m} are shown in the experimental section.

2.3. X-Ray Spectrometer

X-ray spectra also provide a powerful method of studying nuclear decay. With the resolution presently available it is possible to determine unambiguously which elements are present in particular sources. Coincidence experiments using an x-ray detector will be discussed in § 2.4. The x-ray detector in use at Michigan State is a Si(Li) detector with a 150 Å gold surface barrier contact. This ORTEC detector has a warranted resolution of 275 eV at 6.4 keV, counting through a 5 mil Be window The detector active diameter is 6mm, with a sensitive depth of 3mm. This detector is operated at a bias of 1500 V at liquid N_2 temperature. An integral part of the detector is an ORTEC model 117 preamplifier with a cooled FET first stage. The preamp is directly attached to the cryostat to reduce capacitance.

The useful efficiency range of the x-ray detector is from about 5 keV to <100 keV. This type of detector is also quite sensitive

to charged particles such as electrons and positrons. This is both an advantage and a disadvantage when obtaining spectra with high positron fluxes, such as the spectra obtained in this research. These charged particles cause detector output pulses of a much higher voltage than the x-ray pulses under consideration. As a consequence, the positron pulses are badly clipped by the amplifier, which cause a great deal of ringing on the pulses, and present a complex problem for the ADC. The ill effect of this situation shows up as a high noise background below approximately 10 keV. This problem can be partly alleviated by running the experiment in a self-gated coincidence mode. This allows the single channel analyzer to stop all pulses above the window level. This produces a cleaner spectrum but is not completely satisfactory since the problem is still present in the detector and can still have the adverse effect of lowering the system resolution.

A few attempts have been made at using various sized magnets for deflecting the positrons before they reach the detector. Unfortunately, no improvement has been realized. In addition, using a magnet requires removing the source to a greater distance from the detector. Due to the already extremely low efficiency of the detector, this becomes impractical.

2.4. Special Equipment for the Dy^{147} Experiments

The Dy^{147} experiments, described in Chapter 4, were carried out at Yale University using their Heavy Ion Linear Accelerator. While the detectors and electronics used for these experiments were standard items, most of the equipment was not the same as used for the other experiments reported in this thesis.

Four types of experiments were accomplished at Yale: electron singles, γ-ray singles, x-ray singles, and γ-ray versus x-ray coincidence experiments.

The search for the predicted *M*4 transition in Dy^{147} included looking for the expected conversion electrons. This was accomplished with the electron detector described in § 2.2. A 400-channel RIDL analyzer and an ORTEC 410 amplifier were used in conjunction with this detector.

For the γ-ray and x-ray experiments, the data were taken with a PDP-8/I computer interfaced to a dual Northern Scientific ADC.

γ-ray spectra were obtained with a Princeton Gamma-Tech Ge(Li) detector having a 2.3-keV resolution on the 1.33 MeV peak of Co^{60} and an efficiency of 6.5%. The detector was coupled to a Tennelec TC-137 preamplifier, ORTEC 451 amplifier, and the PDP-8/I.

The x-ray spectra were obtained with a Nuclear Equipment Corporation x-ray detector and preamplifier. The remainder of the system was the same as for the γ-ray detector.

X-ray versus γ-ray coincidence spectra were obtained using the x-ray and γ-ray detectors described above in conjunction with a standard fast coincidence circuit. Representative spectra collected at Yale are shown in § 4.5.

2.5. Chemical Separations

The rare earth group of elements (Z=57 through Z=71), in general, pose a real challenge for separation. These elements differ from one another mainly in the filling of inner 4*f* shell of electrons,

the outer electronic configuration being very similar for all of the members of this group. Consequently, they exhibit similar oxidation states and oxidation potentials - hence, the separation difficulty. All rare earth elements exhibit the characteristic +3 oxidation state. Only cerium has a relatively stable +4 oxidation state, with samarium, europium, thulium, and ytterbium exhibiting semi-stable +2 oxidation states.

For the present research, we needed separations of gadolinium from samarium and from gross amounts of europium. Gd^{149} was the only radioactive species involved in the present investigation with a half-life long enough to make some sort of chemical separation feasible. For the separation of Gd^{149} three different techniques were tried, two of them successfully. Two of these techniques utilized the fact that europium and samarium have 2+ oxidation states, while gadolinium exhibits only the 3+ state under the same conditions.

The first technique was that of HCl-Zn reduction. The second was an attempt at forming the 2+ europium species by use of hypophosphorous acid as the reducing agent. The third was that of ion-exchange. These are described in more detail below.

2.5.1. Zn-HCl Reduction

This procedure is similar to the one reported by R. N. Keller (Kel61). Targets usually consisted of about 100 mg of $Eu_2{}^{151}O_3$. After bombardment, these samples were dissolved in 3*M* HCl, keeping the volume to about 10-15 ml. This dissolution takes anywhere from a few minutes to an hour. Heating helps to speed up the process. The dissolved sample was then heated to boiling in a water bath and powdered

zinc added, nitrogen being bubbled through the solution during this entire period to prevent oxidation. The mixture was allowed to react in this manner for one hour.

At this point a few drops of H_2SO_4 was introduced to the solution for precipitation of $SmSO_4$ and $EuSO_4$, and the solution was allowed to cool. While cooling, the N_2 gas was still bubbled through the solution. When cool the mixture was centrifuged, the supernate decanted, and reduced in volume to allow transfer to a small glass vial for counting. This resulting solution should be free of europium and samarium contaminants. This solution could be used to run through the separation procedure a second time if itwas found that a complete separation was not obtained. We have found that one pass does separate Eu and Gd to the extent necessary to obtain a "clean" Gd γ-ray spectrum. The first separtion takes 2-3 hours, with each succeeding cycle requiring an additional 1 1/2 hours.

2.5.2. Hypophosphorous Acid Reduction

The HCl-Zn reduction technique discussed in the last section takes a minimum of two to three hours to perform, as does the ion-exchange method discussed in §2.5.3. A method was sought which would allow separation of species with shorter half-lives. Again, a reduction technique was sought which would allow the formation of Eu^{++} and Sm^{++} in the presence of Gd^{+++}. The relevant oxidation potentials are given in Table 1.

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 production and purification of