

GAMMA RAY SPECTROSCOPIC STUDIES
OF MEMBERS OF THE A=141 DECAY
CHAIN

By

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ABSTRACT
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The techniques of beta and gamma ray spectroscopy have been applied to a study of the decay schemes of $^{141m+g}\text{Sm}$ and ^{141}Pm . The goal of this study was to increase our understanding of nuclear structure and to obtain information about energy level systematics in these members of the A=141 decay chain below the N=82 closed shell.

Ge(Li) singles, Ge(Li)-Ge(Li) two parameter coincidence and Ge(Li)-NaI(Tl) (anti) coincidence spectrometers were used to identify and establish the sequence of gamma ray transitions. The decay schemes were determined and comparisons are made with previous studies as well as the results of recent reaction work.

Forty-seven transitions have been identified with the beta decay of ^{141m}Sm and 40 of these have been incorporated in a decay scheme involving 16 levels in the daughter ^{141}Pm . These levels are 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, 2119.0, and 2702.4 keV. The lifetime of the state at 628.6 keV in ^{141}Pm was measured to be 700 ± 20 ns and the E3 transition from this level to the ground

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state was found to exhibit an enhancement of 1.6 spu. The decay of ^{141}gSm was observed to populate levels at 403.9, 438.4, 728.0, 1292.7, 1495.7, 2004.5, and 2037.7 keV in ^{141}Pm .

Of the 50 transitions observed in the decay of ^{141}Pm , 43 of these have been placed in a level scheme containing 23 levels. The levels identified are at 0, 193.8, 1223.3, 1345.8, 1564.8, 1596.8, 1820.4, 1897.1, 1968.0, 2066.4, 2073.7, 2109.6, 2145.2, 2246.6, 2265.3, 2303.4, 2354.4, 2388.4, 2430, 2506, 2620, 2805, and 2986 keV.

Limits on the possible spin assignments of the levels identified, have been made from calculated log ft values and observation of transitions to levels of known spins. A three-quasiparticle multiplet of high-spin, odd parity states was identified between 1.4 and 2.7 MeV in ^{141}Pm . The decay characteristics of the multiplet suggest the possibility of obtaining information about its structure from shell model calculations using a limited basis.

A survey has been made of the energy level systematics and properties of low-lying states in the N=80, 81, and 82 odd mass isotones. These observations have led to some predictions and suggestions for future experiments.

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Chapter I

Introduction

A measure of our current understanding of nuclear structure is demonstrated by the ability of current nuclear models to explain observed properties of nuclei and to predict other properties previously unobserved. The origin and development of these models involves the observation of these same nuclear state properties. In this interplay of theory and experiment, both the quality and quantity of the available experimental data serve to test the existing models and aid in the development of new and improved ones.

Radioactive decay studies and nuclear reaction studies complement one another as basic tools of nuclear research. Their ability to determine significant nuclear state properties serves to test and advance our knowledge of nuclear structure. By means of beta and gamma ray spectroscopy, this investigation seeks to increase our current knowledge, and, where possible, to improve the existing information in the field of radioactive decay.

In the past few years there have been many radioactive decay studies of odd mass isotopes in the $50 \leq Z \leq 65$, $62 \leq N \leq 82$ region of the nuclidic chart (Ti 67)(NDS)(NSA). These studies attempt to characterize the states in the daughter as completely as possible by comparing their results with

those of reaction studies and theoretical calculations. Recently there have also been a number of nuclear reaction studies on odd mass nuclei in and near the $N=82$ closed shell. These involved particle transfer studies such as $(d, {}^3\text{He})$, $({}^3\text{He}, d)$, (p, d) , (d, p) , (p, t) , inelastic scattering experiments such as (α, α') , (d, d') , (p, p') and in-beam gamma experiments such as $(n, n'\gamma)$, $(\alpha, 2n\gamma)$

This region is bounded by the magic numbers of $Z=50$ protons and $N=82$ neutrons. Nuclei with these magic numbers have tightly bound or relatively inert spherical cores of protons ($Z=50$) or neutrons ($N=82$). As one moves far from these spherical cores, it is expected the nuclei will acquire a permanently deformed shape. The transitional region between these extremes is characterized by a change in the nature of the levels, produced by the increasing admixture of various combinations of shell model orbitals and intermixed collective effects. As a result, there are a greater number of low-lying levels suggesting a softer nucleus. In the transitional region the line of stability runs approximately through the nuclei ${}_{51}^{121}\text{Sb}_{70}$, ${}_{53}^{127}\text{I}_{74}$, ${}_{55}^{133}\text{Cs}_{78}$, and ${}_{57}^{139}\text{La}_{82}$. These nuclei, along with the large number of stable nuclei in the $Z=50$ and $N=82$ closed shells, are conveniently located for systematically probing part of the transitional region by means of the $({}^3\text{He}, xn)$ and (p, xn) reactions.

The beta and gamma ray spectroscopy group at MSU is currently conducting a systematic study of the region between the $Z=50$ and $N=82$ closed shells. In the process of conducting such a study it is useful to begin with a knowledge of levels in the nuclei close to the line of stability. As more information becomes available regarding these levels, it will be convenient to move away from stability in successive steps toward the region of permanent deformation.

During the course of earlier investigations in this region, the study of ^{139m}Nd resulted in the identification of a three-quasiparticle multiplet of low-lying, high-spin states in ^{139}Pr (Bee 69b). The existence of such states is rare, and, because they can be characterized in rather simple shell model terms, they yield a significant amount of information about nuclear structure. As a result of the information to be gained, it seemed worthwhile to attempt to identify these three-quasiparticle levels in other nuclei in the vicinity of ^{139}Pr . It was suggested by McHarris et al. (McH 69) that the decays of ^{141m}Sm and ^{137m}Nd would populate similar multiplets in the daughters ^{141}Pm and ^{137}Pr . The ready accessibility of ^{141}Sm by means of the reaction $^{142}\text{Nd}(^3\text{He}, 4n)^{141}\text{Sm}$ ($Q=-27.3$ MeV) suggested it as the next system to be studied.

A satisfactory study of ^{141m}Sm necessitates a knowledge of other members of the $A=141$ decay chain as they appear in the sequence $^{141m+g}\text{Sm} \rightarrow ^{141}\text{Pm} \rightarrow ^{141m+g}\text{Nd} \rightarrow ^{141}\text{Pr}$ (stable). The

decays of ^{141m}Nd and ^{141g}Nd have been previously reported by Beery et al. (Bee 68). However, little was known about the 20.6m ^{141}Pm and 22.1m ^{141m}Sm activities other than their half-lives and the isomer ^{141g}Sm had not been identified at all prior to initiating this investigation. In light of the existing situation, a concurrent study of the decays of $^{141m+g}\text{Sm}$ and ^{141}Pm was undertaken. The study of ^{141}Sm was initiated through a joint effort with R. Eppley. Some preliminary results concerning ^{141}Sm appear in (Epp 70a).

The decay of ^{141}Pm is of interest because the daughter ^{141}Nd is one neutron removed from the N=82 closed shell. As a result, the low-lying states are expected to be of a fairly pure single-particle nature, (i.e. neutron hole states). During the course of this investigation several studies have appeared directed at locating the neutron hole states in the N=81 nuclei by means of pickup reactions such as (p,d) and (d,t) on N=82 nuclei. As a consequence, it has been possible to compare the ^{141}Pm decay study with these data.

During the course of the ^{141m}Sm investigation, a delayed state was observed in ^{141}Pm at 628.6-keV. This state is suggested to decay via M2 and E3 transitions of 431.8- and 628.6-keV, respectively. Lifetime measurements indicate the E3 transition is enhanced. This observed enhancement, coupled with a study of systematic trends in the N=82 nuclei, suggests a series of interesting experiments to be performed on odd proton odd mass nuclides in the region.

At the present time there exists little in the way of theoretical calculations with which to compare the experimental results presented here. Muthukrishnan and Kromminga have performed a shell model calculation of the three-quasiparticle odd parity states in ^{139}Pr by using several phenomenological potentials and limiting the basis states to those suggested by McHarris *et al.* (McH 69) for the three-quasiparticle multiplet. The results obtained on the energies of the states were in rough agreement with experiment. However, the theoretical transition probabilities did not agree very well with experiment. Preliminary calculations of energies of states below 2 MeV in ^{141}Nd have been carried out very recently by Reehal using the pairing-plus-quadrupole Hamiltonian of Kisslinger and Sorensen (Ree 71), reasonable agreement was obtained. The recent shell model calculations of Wildenthal on the N=82 isotones appear very promising. It is hoped that in the near future these calculations might be extended to the N=81 and N=80 isotones. Moderately successful calculations might be performed using a rather limited basis such as the one proposed for the three quasiparticle multiplets in ^{141}Pm and ^{139}Pr .

Chapter II describes in a general way the techniques involved in decay scheme construction and contains a brief description of the experimental apparatus and techniques along with a discussion of the methods of data analysis.

Chapter III describes in some detail the steps involved in the systematic identification of the transitions associated with the radioactive decay of members of the $A=141$ chain below the $N=82$ closed shell.

Chapter IV describes the experimental results of this study. The decay of ^{141m}Sm involved the identification of 47 gamma rays and the placement of 16 levels in the daughter ^{141}Pm populated by this decay. A set of six odd-parity levels between 1.6 and 2.2 MeV were identified as members of a three-quasiparticle multiplet having the configuration

$$[(\pi d_{5/2})^3(v d_{3/2})^{-1}(v h_{11/2})^{-1}]_{J^-}.$$

The properties of these states are compared with the corresponding ones in ^{139}Pr . The half-life of the delayed $11/2^-$ state at 628.6-keV in ^{141}Pm was measured and the partial half-lives of the 628.6-keV E3 and 431.8-keV M2 transitions were determined. The decay scheme of ^{141g}Sm resulted in the placement of an additional 7 levels in the daughter ^{141}Pm and the identification of 12 transitions belonging to this decay.

The study of the decay of ^{141}Pm resulted in the placement of 23 levels in ^{141}Nd and the identification of 50 transitions associated with this decay. A comparison is made between the levels of ^{141}Nd populated via the beta decay of ^{141}Pm and levels identified in ^{141}Nd by means of the reaction

studies (p,d) and (d,t) on ^{142}Nd (Jol 71), (Cha 71), and (Fos 71). These results are compared with very preliminary theoretical calculations by Reehal (Ree 71) using the pairing-plus-quadrupole Hamiltonian of Kisslinger and Sorensen.

In Chapter V the systematic behavior of some nuclear properties in the $N=82$ region are discussed. The characteristics of the three-quasiparticle multiplet in ^{141}Pm are discussed and compared with similar states in other nuclei in and near the $N=82$ closed shell.

Chapter II

Experimental Methods and Apparatus

As one moves further away from the region of β -stability the identification of radioactive isotopes and the construction of their decay schemes becomes increasingly difficult. The difficulties arise principally because the half-lives become shorter and the reactions used to produce the various desired isotopes are not as clean. The limitations on studying a specific activity depend on several factors such as (1) the types of bombarding beams available, (2) the energies of these beams, (3) the specific nuclear reactions desired, (4) the stable isotopes available for targets, (5) the location of these isotopes with respect to the line of beta-stability, (6) the half-lives of the desired activity and the competing activities, and (7) the data acquisition facilities available.

The isotopes considered in this study were produced by either (p, xn) or $(^3\text{He}, xn)$ reactions ($x \leq 4$) on stable isotones in the $N=82$ closed-shell. The complex decay characteristics of these isotopes required the use of the latest highest quality electronic equipment such as high resolution Ge(Li) detectors (~ 2.0 -keV FWHM for the 1332.4-keV transition in ^{60}Co), FET preamplifiers, low noise amplifiers with Gaussian shaping, base-line restoration, dc-coupling, pile-up rejection and significant (anti-) coincidence circuitry

as well as the PDP-9 and XDS Sigma-7 computers and their available data acquisition programs.

Section 2.1 describes in a general way the pattern followed in the acquisition of the experimental data and the construction of parts of a nuclear decay scheme. Section 2.2 discusses the two types of gamma ray detectors used in this study and compares the factors influencing their resolution. A description of the various gamma ray spectrometers used in this investigation is also included. In Section 2.3 some of the methods used in the data analysis are broadly outlined. A detailed description of the logical identification of the members of the $A=141$ decay chain considered in this study is deferred to Chapter III. Peculiarities pertaining to a specific experiment are included in the discussion of the experimental results in Chapter IV.

2.1 Decay Scheme Construction

One of the primary objectives of this study has been the incorporation of the significant experimental results into a consistent description of the manner in which an isotope decays and a comparison of these results with theories and other data such as reaction studies. The important features of such a study involve proper identification of

the activity to be studied, determination of the half-life of the parent activity, precise measurement of the energies of the transitions, beta and gamma transition intensities, electron capture disintegration energies, calculation of $\log ft$ values for beta decay to each level in the daughter, and spin and parity assignments. There are a wide variety of experimental techniques available and each isotope has special characteristics requiring variations in the method of approach.

The placement of energy levels may be suggested initially by energy sums and gamma-ray intensity balances. Additional evidence for level placement comes from coincidence evidence such as 511-511-keV-gamma coincidences, anti-coincidence, prompt and delayed coincidence results, as well as other accessible parameters. As one moves further from stability, the energy available for decay increases generally resulting in an increase in the number of transitions to be placed. In addition to this difficulty, the half-life generally decreases, and, as it becomes shorter, it makes Ge(Li)-Ge(Li) 2-dimensional coincidence experiments more difficult. As a result such problems make the interpretation of the data more difficult.

After placing the energy levels, the determination of beta and gamma transition intensities follows. Then, providing the EC decay energies are known, the $\log ft$ values for decay to each state in the daughter can be calculated.

This involves a rather lengthy but generally routine sequence. A description of the procedure to be followed in doing these calculations appears in (Bee 69a) and (Epp 70a). Computer programs are available which calculate per cent feeding to each level, electron capture to positron ratios and log ft values. The program used in this study was DECAY SCHEME written by D. Beery for use on the Sigma-7 computer and modified by R. Epply for use on the PDP-9 computer (Epp 70a).

2.2. Gamma-Ray Spectrometers

In the following study we have used two different photon detectors: NaI(Tl) scintillation spectrometers and Ge(Li) solid state spectrometers. The processes involved in the detection of the energy deposited in the NaI(Tl) crystal and the Ge(Li) crystal are quite different. As a result the energy resolution of the two types of spectrometers differs markedly. As a means of comparison the energy resolution of a very good NaI(Tl) system is ~ 60 keV full-width-half-maximum (FWHM) for the 1332-keV γ -rays of ^{60}Co . The energy resolution of a typical Ge(Li) system is 2.0- to 3.0-keV FWHM for these same ^{60}Co γ -rays. The reason for the difference in resolution can be understood from the following brief description of the processes involved.

Each spectrometer relies on the fact that incident radiation deposits some portion of its energy in the crystal. In the NaI(Tl) spectrometer the deposited energy appears as visible and ultraviolet light. This light is detected by a photomultiplier tube optically coupled to the NaI(Tl) crystal. This produces a voltage pulse proportional to the intensity of the incident light that is in turn proportional to the energy deposited in the crystal. In the Ge(Li) spectrometer the incident radiation creates hole-electron pairs that are collected by an electric field across the crystal. The result is a current pulse proportional to the energy deposited by the radiation; this is converted to a voltage pulse by the preamplifier for purposes of pulse shaping etc.

The basic reason for the disparity in the energy resolution of the two systems is essentially statistical. The signal resolution in NaI(Tl) results from the large statistical fluctuations in the relatively small number of resultant photoelectrons emitted by the photocathode per given amount of energy deposited in the scintillating crystal. Ge(Li) solid state detectors employ direct collection of the ionization produced by the radiation. In Ge(Li) a hole-electron pair is produced for every 3-eV of energy (on the average) absorbed from the radiation. Correspondingly in NaI(Tl) on the order of 300 eV are required for every photoelectron produced.

Hence in the NaI(Tl) system the relatively large statistical fluctuation is due to the small number of electrons ejected from the photocathode of the photomultiplier, whereas in the solid state detector the primary energy is distributed over a far greater number of carriers and the relative statistical fluctuation is considerably reduced.

2.2.1 Singles Experiments

In the present study, singles experiments refer specifically to pulse height spectra taken with the aid of a single Ge(Li) spectrometer. In point of fact the Ge(Li) spectrometer is an electronic system composed of the following components: a) A lithium-drifted germanium crystal enclosed in a cryostat and cooled in liquid nitrogen, temperature (77°K), b) a high voltage bias supply and a charge-sensitive pre-amplifier with an associated field-effect transistor (FET)—the FET may be either cooled to $\sim 120^\circ\text{K}$ or operated at room temperature, c) a spectroscopy amplifier (for purposes of amplification and pulse shaping) with pole-zero cancellation, base-line restoration, and dc level adjustment, d) an analog-to-digital converter (ADC) and computer or multichannel analyzer (MCA), and e) a storage unit of some type with associated spectrum readout.

The performance characteristics of a particular Ge(Li) spectrometer are primarily determined by the Ge(Li) crystal

and the associated FET preamplifier. Such features as the volume of the Ge(Li) crystal, the manner in which the crystal is drifted (i.e. true-coaxial, trapezoidal or wrap-around, or planar), the capacitance of the crystal, and the operating bias all play interrelated parts in determining the final resolution of the system. The cooled FET will usually result in a lower noise system and as a result yields somewhat better resolution as compared to a room temperature FET preamplifier. The volume and shape of the crystal will play important parts in determining the efficiency and peak-to-Compton ratio for a particular detector. In general a larger volume crystal results in increased counting efficiency (related to the size) but this increase in volume usually results in a decrease in the resolution produced by an increase in crystal capacitance and poorer charge collection.

Other experimental features which are important in achieving satisfactory resolution are amplifier shaping constants, proper pole-zero cancellation, base-line restoration and correct dc-level adjustment. These can be determined rather quickly by using an oscilloscope and observing pulse-shapes and dc-levels at appropriate places in the system. The use of base-line restoration is strongly dependent on the counting rate and usually is best determined empirically.

The characteristics of several different Ge(Li) spectrometers used in the current gamma-ray studies are listed in Table I.

2.2.2. Coincidence Experiments

In the course of this study the character of the overwhelming majority of nuclear states observed suggested they had lifetimes $\ll 100$ nanoseconds (ns). Consequently it proved beneficial to use two or three singles spectrometers in various coincidence configurations to perform anticoincidence, prompt coincidence and delayed coincidence experiments. The standard procedure employed in a typical decay scheme study is to determine a) a singles spectrum, b) an anticoincidence spectrum, and c) coincidence spectra.

Table I

Eff.*	Shape	Peak-to Compton	Res. FWHM 1332-keV ⁶⁰ Co	Manufacturer
0.4%	Trapezoidal	4.5:1	3.4 keV	MSU Cyclotron Lab
2.5%	Trapezoidal	12:1	2.2 keV	Nuclear Diodes
3.6%#	True Coaxial	18:1	2.0 keV	Ortec
4.5%	True Coaxial	24:1	1.9 keV	Ortec
10.4%	True Coaxial	36:1	2.2 keV	Nuclear Diodes

*With respect to the 1332 keV peak of ⁶⁰Co at 25 cm from a 7.6 x 7.6-cm NaI(Tl) detector.

#This detector was damaged by an electronic malfunction and replaced by the manufacturer with the 4.5% detector.

The techniques involved are routine and are discussed in greater detail in (Bee 69a) and (Epp 70a).

A "classical" pattern of experiments was used in the study of the ^{141}Pm decay described in Section 3.2, whereas the study of the Sm isotopes ^{141g}Sm and ^{141m}Sm required the use of a wider variety of methods to determine the decay schemes and their characteristics. The study of ^{141}Sm involved isotopes with similar enough half-lives, 11.3m in the case of ^{141g}Sm and 22.1m in the case of ^{141m}Sm , that the counting methods had to be modified. As an added feature, the decay of ^{141m}Sm involved the population of an $11/2^-$ isomeric level in ^{141}Pm which was observed to be a delayed state. The measurement of this half-life is discussed in Section 3.1.3.D.

By using two Ge(Li) detectors placed in 180° geometry we were able to perform several useful prompt-coincidence experiments as described below in Section 2.2.2.A. Other prompt and delayed coincidence experiments performed with Ge(Li) detectors inside a split-ring NaI(Tl) annulus are outlined in Section 2.2.2.B.

2.2.2.A. Multiparameter Ge(Li)-Ge(Li) Spectrometer

With the availability of larger volume and more efficient Ge(Li) detectors it has become practical to use them in obtaining 2 parameter coincidence information.

The goal of such an experiment is to simultaneously provide coincidence information for any and all peaks or regions of the spectrum in a single run.

Despite the relative inefficiency of Ge(Li) detectors, as opposed to NaI(Tl) detectors, there are decided advantages associated with their use in a multiparameter experiment. The most important features are the excellent energy resolution and the greater peak-to-Compton background ratio.

As a result, satisfactory coincidence information can be obtained with fewer counts per channel using a Ge(Li)-Ge(Li) system than are normally required when using Ge(Li)-NaI(Tl). The electronics arrangement used in this lab is a relatively standard technique and is discussed in more detail in several theses and journals (Epp 70a). A diagram of the arrangement can be seen in Fig. 1.

With minor modifications, it is possible to obtain more information than described above. This information refers to a third parameter, namely the time between the two coincident events. It is with this in mind that I describe an alternate Ge(Li)-Ge(Li) coincidence spectrometer as seen in Fig. 2.

This is still a double coincidence experiment where we wish to record the linear signal from each of the detectors. The added feature in this configuration is the presence of a time to pulse height convertor (TPHC) in place of the fast coincidence unit. This provides us with

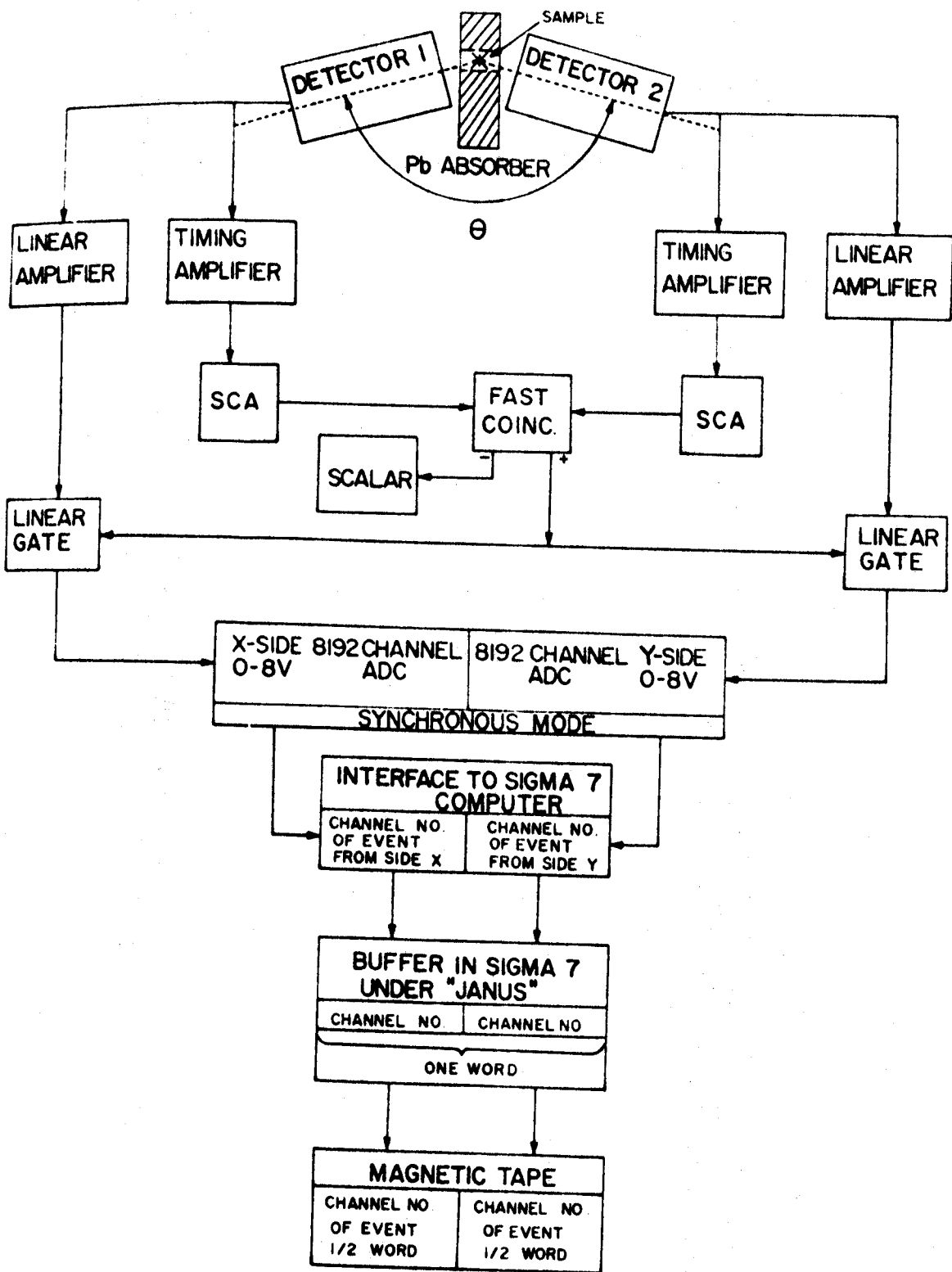


Figure 1. Block diagram of the apparatus used to collect two-dimensional "megachannel" γ - γ coincidence spectra, using the Sigma-7 computer.

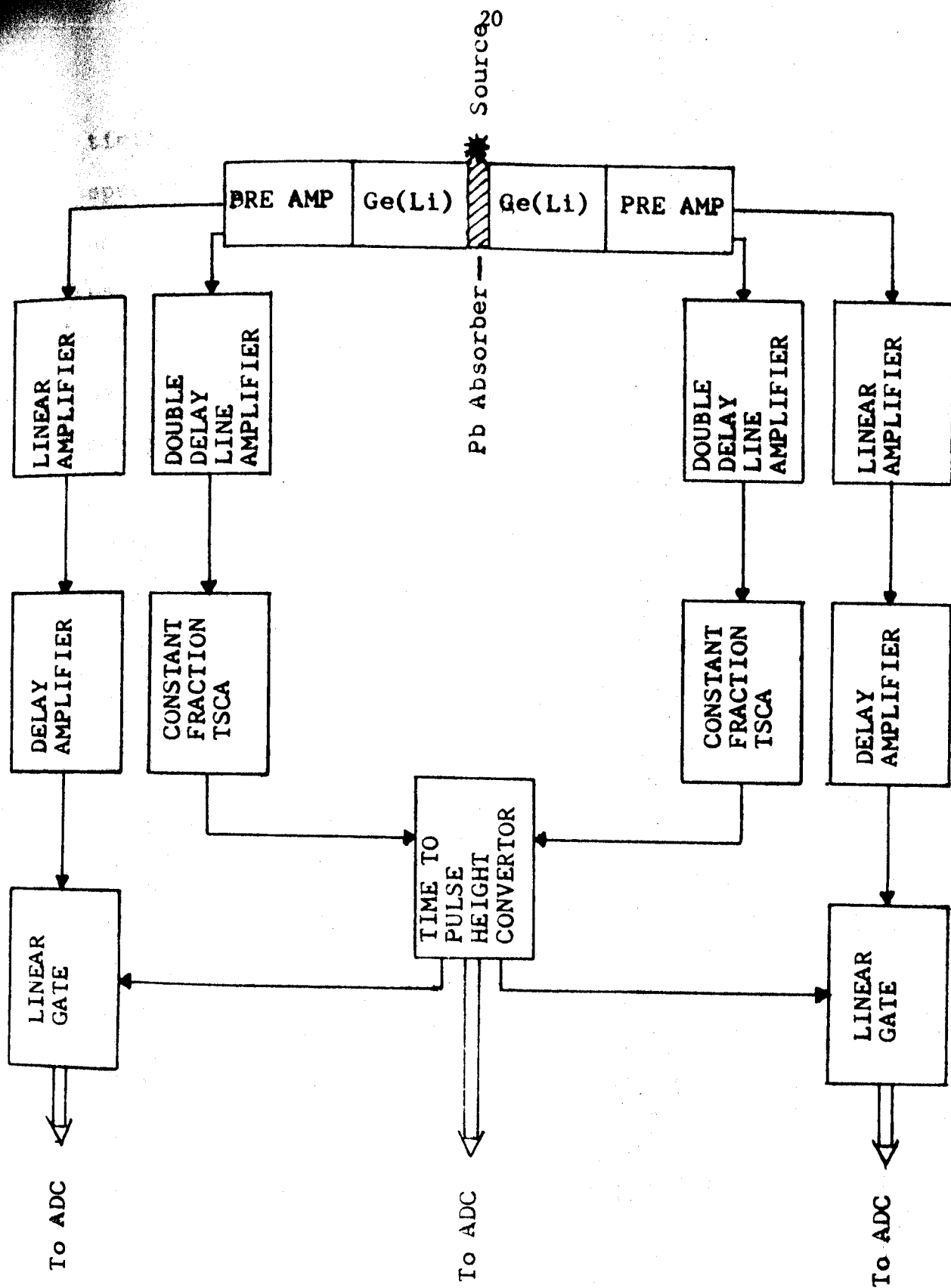


Figure 2. Schematic illustration of $\text{Ge(Li)}\text{-Ge(Li)}$ multiparameter coincidence apparatus with time-to-pulse-height convertor (TPHC).

