

THE DECAY OF  $^{143}\text{Ce}$

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by

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The decay of 33 hour  $^{143}\text{Ce}$  to levels in  $^{143}\text{Pr}$  has been studied, using Ge(Li), NaI(Tl), and Si detectors. A number of new gamma ray transitions have been discovered. A decay scheme based on the experimental results is discussed and some spin assignments are suggested. This investigation was not exhaustive and suggestions are made as to further experiments which could be done.

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## CHAPTER I

### INTRODUCTION

One of the aims of nuclear spectroscopy is to learn as much as possible about the properties of the various energy levels in nuclei. With the tools and techniques available the experimentalist can deduce such properties as the energy, spin and parity of the levels.

It is the purpose of this chapter to briefly discuss some of these concepts before going on to describe in greater detail some of the models which can be applied to the nucleus.

#### 1.1 Spin and Parity

The angular momentum or spin of a particle consists of two parts. The orbital angular momentum which must have integral value (in units of  $\hbar = 1.05 \times 10^{-27}$  erg - sec.) which it possesses because in a classical sense, it is moving in a closed path about a centre, and the intrinsic angular momentum which it always possesses and which has no classical analogue. Protons and neutrons are both fermions, that is they obey Fermi Dirac statistics and each particle has an intrinsic angular momentum  $1/2 \hbar$ . The spin  $I\hbar$  of a nucleus is the vector sum of the spins of all the individual constituent nucleons. It is an experimental fact that nucleons with the same orbital angular momentum tend to pair off their spins giving a resultant spin of zero. Thus it is found that the ground state spins of odd A nuclei are half integral, those of even A nuclei are integral and those of even even nuclei are zero. Excited states are the result of differing configurations of nucleons and so have characteristic spins.

The parity of a level is a characteristic of the wave function describing the state under spatial reflection. The wave function has odd or even parity depending on whether the wave function does or does not, respectively change sign under spatial inversion. Thus for a static electric dipole a reflection at any instant through the origin of all three coordinates reverses the position of each charge, and so the system is said to have negative parity. If there is no change of sign, as for the magnetic dipole, the system is said to have positive parity.

## 1.2 Electromagnetic Transitions

After a nuclear interaction the nucleus can be left in an excited state. That is, an excited state is an unstable configuration which returns to the stable ground state by the emission of energy. This energy may be carried away either by particle emission or electromagnetic radiation, i.e., gamma rays.

In terms of classical electrodynamics, a source of electromagnetic radiation is formed by a localized system of charges and currents which vary in time in some periodic manner. The multipole order of an electric field depends on the physical arrangement of the charges and so electromagnetic radiation can be classified by its multipole order "L" according to the angular momentum "L" which the radiation field removes from the nucleus. This includes electric dipole (E1) radiation, magnetic dipole (M1) radiation, electric quadrupole, etc. Observation of certain physical characteristics of the radiation such as its spatial distribution as a function of angle of emission or the numerical ratio of



internal conversion electrons to photons indicates the classification of the radiation.

As in other physical processes, transitions in which photons are emitted or absorbed must conserve angular momentum. The predominant multipole order emitted will be that consistent with the smallest transfer of angular momentum possible between the states. The selection rule for the possible  $L$  values is

$$I_i + I_f \geq L \geq |I_i - I_f|$$

In addition since each state has a definite parity, a second condition follows from parity conservation; the change in parity of the radiating system is given by  $(-1)^L$  for electric transitions  
 $(-1)^{L+1}$  for magnetic transitions.

Transitions for which  $I_i = I_f = 0$  are forbidden and no transitions occur with  $L = 0$ .

### 1.3 Internal Conversion

As an alternative mode to the emission of a gamma ray, an excited nucleus may transfer its energy to one of the orbital electrons, ejecting it from the atom. This internal conversion process can be classified according to multipolarities in the same way as for the competing electromagnetic transitions, and if the nucleus is treated as a point nucleus the transition probability will have the same nuclear matrix elements as for the gamma emission process. Thus, the ratio of the transition rates for the two processes is independent of the detailed motions of the nucleons and can be calculated quite accurately. Thus the internal conversion

coefficient,  $\alpha_i$ , for the ejection of an electron from the  $i^{\text{th}}$  shell is defined as the ratio  $T_e / T_\gamma$  for that shell. The total internal conversion coefficient  $\alpha = \sum \alpha_i$ , is related to the total decay probability of a transition by  $T_T = (1 + \alpha) T_\gamma$ .

#### 1.4 Beta Decay

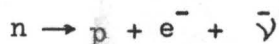
Along with electrons of definite energy, the nucleus is known to emit beta particles which have a continuous energy distribution. This presents a problem in that it appears as if energy is conserved only for the very few events where the beta particle is emitted with the maximum or end point energy. Furthermore, the law of conservation of angular momentum appears to break down, the number of nucleons does not change in beta decay so the spin of the nucleus remains either integral or half integral and yet the electron carries away a half integral value of spin. Pauli<sup>(1)</sup> in 1933 "saved" the two conservation laws by postulating the emission, along with the beta particle, of another particle, the neutrino, with the following properties; zero charge, one half spin and nearly zero or zero rest mass. Fermi<sup>(2)</sup> 1934 showed how the neutrino could fit into a beta decay theory using currents in the nucleus in analogy to electromagnetic gamma ray emission. Finally, Reines and Cowan<sup>(3)</sup> (1953, 1959) provided direct evidence for the neutrino using the inverse reaction:



If one works through Fermi's theory of the mechanism of beta decay one arrives at a result for the total decay rate:

$$\ln 2 / T_{1/2} = G^2 M^2 f(Z, E_0)$$

where  $c$  is a constant,  $M$  is the nuclear matrix element containing the neutron and proton states and the interaction Hamiltonian and  $f(Z, E_0)$  is a tabulated function of the atomic number of the beta emitter and the end point energy  $E_0$ . The expression  $f T_{1/2}$  is known as the comparative half life; from the above expression it can be seen that it is inversely proportional to the square of the matrix element, and hence depends on the initial and final wave functions and on the operator connecting them. The value of  $f T_{1/2}$  can thus be correlated with the type of transition. For instance the most fundamental beta decay is that of the neutron:



The matrix element for this reaction should be unity, as the wave function for a single proton ought to be the same as that of a single neutron. In other cases, such as transitions between mirror nuclei,  $M$  will be near unity and the transitions are called allowed and favoured transitions. Some examples are given below:

| Reaction                                 | $T$ (sec)          | $E_0$ | $fT_{1/2}$ |
|------------------------------------------|--------------------|-------|------------|
| $n \rightarrow H^1 + e^- + \bar{\nu}$    | 750                | 2.53  | 1200       |
| $H^3 \rightarrow He^3 + e^- + \bar{\nu}$ | $3.93 \times 10^8$ | 1.036 | 1020       |
| $O^{14} \rightarrow N^{14} + e^+ + \nu$  | 0.832              | 7.85  | 820        |

The product  $fT_{1/2}$ , although not precisely a constant, is strikingly near to it, considering that the half life varies by a factor about  $10^9$  over the range of known beta emitters.

Thus transitions can be classified according to certain selection rules and the classifications recognized by their

characteristic  $\log fT_{1/2}$  values.

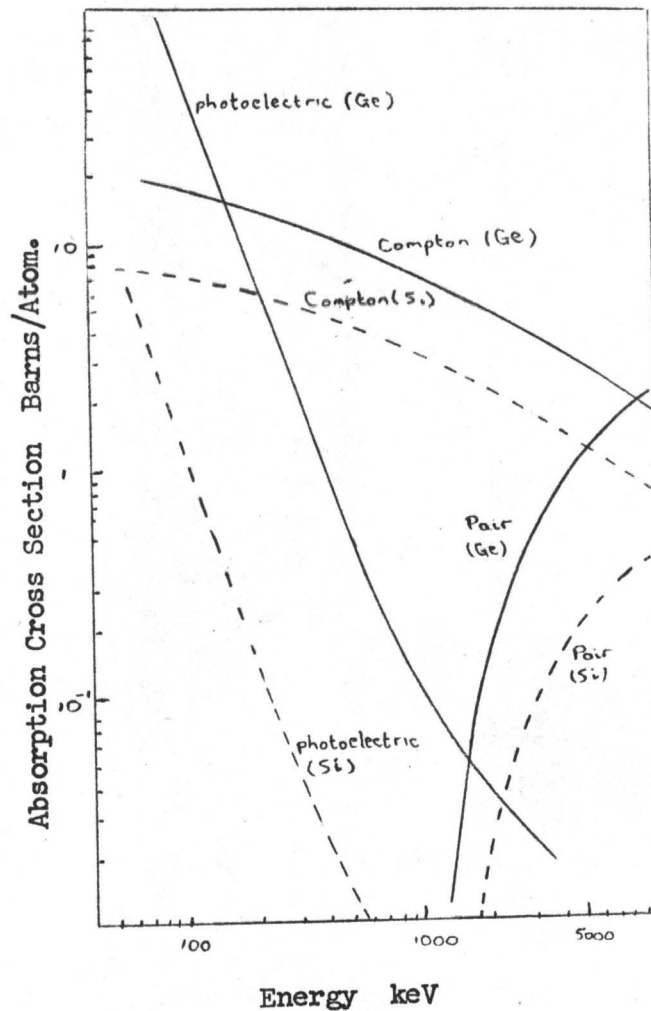
| Type                              | $\Delta I$         | $\Delta \pi$ | $\log fT_{1/2}$ |               |
|-----------------------------------|--------------------|--------------|-----------------|---------------|
| allowed {                         | super allowed      | 0            | no              | $3.5 \pm 0.5$ |
|                                   | allowed unhindered | 0 or 1       | no              | $5 \pm 1$     |
|                                   | hindered           | 0 or 1       | no              | $6 \pm 1$     |
| first forbidden<br>( non unique ) | 0 or 1             | yes          | $6.5 \pm 3$     |               |
| first forbidden<br>( unique )     | 2                  | yes          | $9.5 \pm 2$     |               |
| second forbidden                  | 2 or 3             | no           | 13              |               |
| third forbidden                   | 3 or 4             | yes          | 18              |               |

#### 1.5 The Interaction of Photons with Matter

There are three main processes by which energy, in the form of gamma radiation, may be absorbed by matter. These are the photoelectric effect, the Compton effect, and the pair production effect.

In the photoelectric effect the photon energy is wholly absorbed by the atom and an atomic electron is ejected with an energy  $E - B_j$  where  $B_j$  is the binding energy of the shell in which the electron was located. The vacancy created by the ejection of this electron is filled by electrons from outer shells "falling" into it, and this process may be accompanied by the emission of fluorescent radiation. Alternatively it is possible that no fluorescent radiation is emitted but that an electron from an outer shell is ejected. For instance a vacancy in the K shell may be filled by an electron from one of the L shells and another L electron emitted with energy  $E_K - 2E_L$ . An emitted electron of

this sort is called an Auger electron. Because a third body (the nucleus) is necessary for conservation of energy and momentum the most tightly bound electrons, that is the K electrons, are responsible for the largest part of the photoelectric cross section.



The Compton effect is the name given to the process in which a photon is inelastically scattered from a free electron.

In pair production a photon is transformed into an electron positron pair, the excess energy  $h\nu - 2mc^2$  appears in the form of kinetic energy of the two electrons. This process will be accompanied by gamma radiation when the positron annihilates.

The variation with energy of the photoelectric, Compton and pair production cross sections in silicon and germanium are shown in figure 1.

## CHAPTER II

### NUCLEAR MODELS

#### 2.1 Introduction

The necessity for models of the nucleus arises because the nuclear force, that is the interaction between nucleons, is not yet known and in any case, in all but the very lightest nuclei there are far too many nucleons to allow for an exact mathematical treatment, and too few to allow a proper statistical treatment.

There are a number of different models. They fall, roughly, into two groups; those which are most useful in describing the low energy excitation of nuclei, and those which have found application in treating the higher excitation arising in nuclear reactions. The shell model and the collective model are examples of the first kind, while the optical model and statistical model are examples of the second.

The models which will be considered in this chapter are those of the first group.

#### 2.2 The Shell Model

The shell model attempts to describe the effective force on each particle by means of a central potential  $V(r)$ . In analogy with atomic electrons one might then expect to find a shell structure in nuclei. Indeed it was the slight extra stability associated with certain values of the neutron and/or proton number, i.e. the magic numbers, which first caused Mayer<sup>(4)</sup> (1949) and Haxel, Jensen and Suess<sup>(5)</sup> (1949) to suggest



nuclear shell structure. In the case of the atom, the nucleus acts as a strong centre of attraction which makes it possible to treat the weaker electron-electron interactions by perturbation methods. No such force centre exists in the nucleus. Indeed scattering experiments show that the nuclear force is short range and acts nearly always between pairs of nucleons. It is therefore surprising that the structure of the nucleus can be treated with any simple potential at all. The model assumes that each nucleon moves in a static central potential created by the averaging of the interactions between all the other nucleons in the nucleus. The assumed potential may be a square well, a harmonic oscillator or some more complicated potential. Each nucleon is described by its own set of quantum numbers which determine its nuclear level. In general the Pauli exclusion principle and the level energy determine the order of filling of the levels. Nucleons must then move independently of one another as long as the system remains at minimum energy.

The square well and harmonic oscillator potential both give especially stable nuclei at the neutron and proton numbers 2, 8 and 20, and the addition of a strong spin orbit interaction, which couples the intrinsic spin and orbital angular momentum of each nucleon, to a central potential gives magic numbers at 2, 8, 20, 50, 82 and 126 nucleons. These are just the magic numbers observed experimentally.

In the extreme single particle model, the filled shells are regarded as an inert core and particles in the unfilled shells

react only with the potential due to this core. If there is only one particle outside of a closed shell the only contribution to the total spin is from that particle. Similarly, nuclei which are one particle short of a closed shell have a spin contribution from the single hole. If there are two particles or holes in a shell, these will couple to form degenerate levels with  $j = j_1 + j_2$  where  $j$  takes all the values permitted by the laws of combination of angular momentum and the Pauli principle.

In the single particle model the interactions of the "loose" particles outside closed shells is taken into account. The assumption is made that the loose particles are not perturbed appreciably from the singleparticle orbits described by the quantum numbers  $(n, l, j)$ , but however the degeneracy between  $k$  particles with the same  $(n, l, j)$  is removed.

There is a "pairing force" which makes it possible to predict the ground state spin value. Thus, in an even  $Z$ , odd  $N$  nucleus, the protons couple to  $J = 0$ , and the neutrons to  $J = j_n$ , the  $j$  value of the neutron shell which is filling. Then the resultant angular momentum of the whole nucleus is  $J = j_n$ . Similarly in an odd  $Z$  even  $N$  nucleus,  $J = j_p$ , and in an even even nucleus  $J$  remains zero. By making some assumptions about the type of force involved one can also arrive at rules which predict the ground state spins of odd-odd nuclei. These are Nordheim's<sup>(6)</sup> (1950) rules (as modified by Bernstein and Breman<sup>(7)</sup> (1960). The so called strong rule predicts that the spin of an odd odd nucleus is  $|j_p - j_n|$  if one of the combining nucleons has  $\underline{l}$  and  $\underline{s}$  parallel

and the other has  $\underline{l}$  and  $\underline{s}$  antiparallel. The weak rule states that  $j = |j_p - j_n|$  or  $j_p + j_n$  in the case where both the combining nucleons have  $\underline{l}$  and  $\underline{s}$  parallel or both have  $\underline{l}$  and  $\underline{s}$  antiparallel.

Excited states can also be described in terms of the energy levels predicted by this model although, not always with complete success.

There are other nuclear properties, however, for which the shell model does not work so well. It cannot give reliable results for wave function dependent quantities such as transition probabilities and quadrupole magnetic moments. This is true, especially for nuclei removed from closed shells. Here the very large quadrupole moments which are observed indicate that perhaps a spherical potential is no longer realistic. Thus the need is felt for a description in terms of the deformation of the nuclear surface which ignores the action of the individual particles, but incorporates the collective effects of many particles.

### 2.3 The Collective Model

Nuclei with closed shells appear on both experimental and theoretical grounds to be spherical. If a nucleon is added, it will tend to distort and polarize such a nucleus. Although a second particle couples with the first to give a zero spin, it can still add its polarizing effect to that of the first. The stability of the core against deformation is gradually broken down as extra nucleons are added until a nucleus with a deformed shape has a lower energy than one with a spherical shape. At this point the ground state of the nucleus will correspond to a non-spherical nuclear shape. About the most clear cut evidence for this is the

spacing of the lowest excited energy levels in even even nuclei. It is observed that either the energy of the second excited level is twice that of the first excited level, or, that the energy of the second excited level is about 3.33 times that of the first level. This leads to an analogy with excited energy states in a diatomic molecule. Vibrational states in the first case, rotational in the second.

Because of this analogy Bohr and Mottelson<sup>(8)</sup> (1953) were able to develop the collective theory from Rayleigh's<sup>(9)</sup> (1877) calculations for surface oscillations of a continuous liquid drop. In this description the radius vector of the nuclear surface is described by the function

$$R(\theta, \phi) = R_0 \left[ 1 + \sum_{\lambda=0}^{\infty} \sum_{u=\lambda}^{\lambda} \alpha_{\lambda u} Y_{\lambda}^u(\theta, \phi) \right]$$

where  $R_0$  is the radius of the equilibrium spherical shape. Collective motion is expressed by allowing the coefficients  $\alpha_{\lambda u}$  to be functions of time.

It can then be shown that the collective states of a nuclear drop have excitation energies  $\sum n_{\lambda} \hbar \omega_{\lambda}$ . The integer  $n_{\lambda}$  is the number of phonons of order  $\lambda$  in the excited state. For irrotational motion a phonon of type  $\lambda u$  carries angular - momentum quantum number  $\lambda$  with z component  $u$  and parity  $(-)^{\lambda}$ . Since the frequency  $\omega_{\lambda}$  is a rapidly increasing function of  $\lambda$ , one need only consider small values of  $\lambda$ .

The lowest order for which deformation of the nuclear surface is apparent is the  $l = 2$  state. The name quadrupole is applied to

this type of deformation because it produces features in the electric field at a distance from the charge distribution that are characteristic of a static quadrupole. For a nucleus, we assume in this case that the deformation is in the form of a surface vibration.

According to the collective model, the electromagnetic radiation field produced during a transition from the first excited vibrational state to the ground state results from a rearrangement of the nuclear charge from a quadrupole to a spherical distribution. The emitted electromagnetic radiations has the characteristics of an  $E2$  ( electric quadrupole) transition.

The addition of several nucleons outside a closed shell causes the nucleus to be permanently deformed. In this situation, the excitation of a nucleus can be divided into that associated with changes in intrinsic shape (vibrations) and that associated with changes in its orientation (rotations). In principle the moments of inertia of nuclei can be determined from the energies of their rotational states. If a nucleus is axially symmetric in shape, its rotational energy levels can be described by the three constants of motion:  $I$ , the total angular momentum of the system;  $M$ , the projection of  $I$  on a space fixed axis; and  $K$ , the projection of  $I$  on the nuclear symmetry axis.

The rotational effect can be either rigid, in which case particles actually move in circles around the axis of rotation, or wave like, in which case particles perform oscillator motions and only the geometrical shape of the drop changes. Wave like rotations can be observed only in deformed nuclei because the apparent

rotational motion must then be solely a surface phenomenon. Experiments corroborate such a requirement, for all known nuclides having clearly defined rotational states display relatively large static quadrupole moments.

If the rotation is wave like there can be no rotation about the symmetry axis. The quantum number  $K$  is therefore a constant for each set, or band, of rotational levels and represents an intrinsic angular momentum for that band. The quantum number  $R$  represents the contribution of the rotational motion and is of such magnitude as to make  $I = K, K + 1, K + 2$ , etc.

With this model the energies of the levels in any single band are found to be given by the formula:

$$W_I = W_0 + \frac{h^2}{2\mathcal{I}} \left[ I(I+1) + a(-1)^{I+1/2} (I+1/2) \delta_{K,1/2} \right]$$

which is based on the energy relationship of rotational levels in a linear molecule. In this formula  $W_0$  is an energy, dependent on the intrinsic structure of the system.  $\mathcal{I}$  is the effective moment of inertia of the nucleus. Its value is found empirically to lie somewhere between that expected for a rigid nucleus and that for a liquid in irrotational motion. The second term, occurs because the rotational motion is strongly coupled only to total angular momentum states for which the orbital angular momentum is different from zero; there is a decoupling of the rotational motion of  $K = 1/2$ , for then, only the spin angular momentum is not zero. The magnitude of this decoupling is given by the parameter  $a$ .



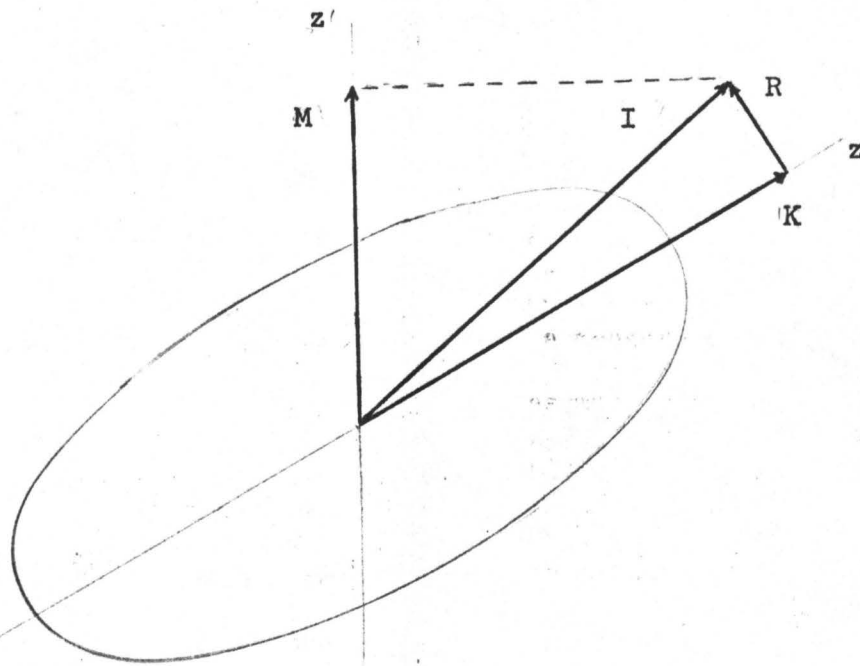


FIGURE 2: Coupling Scheme for Deformed Nuclei.

However, rotation is merely the simplest collective motion of deformed nuclei. The nuclear deformation is usually described in terms of two parameters, beta and gamma; beta is a measure of the total deformation, and gamma is a measure of the asymmetry from a spheroidal shape. Fig. 3. shows some possible spin values for the rotational bands built on the different types of collective motion.



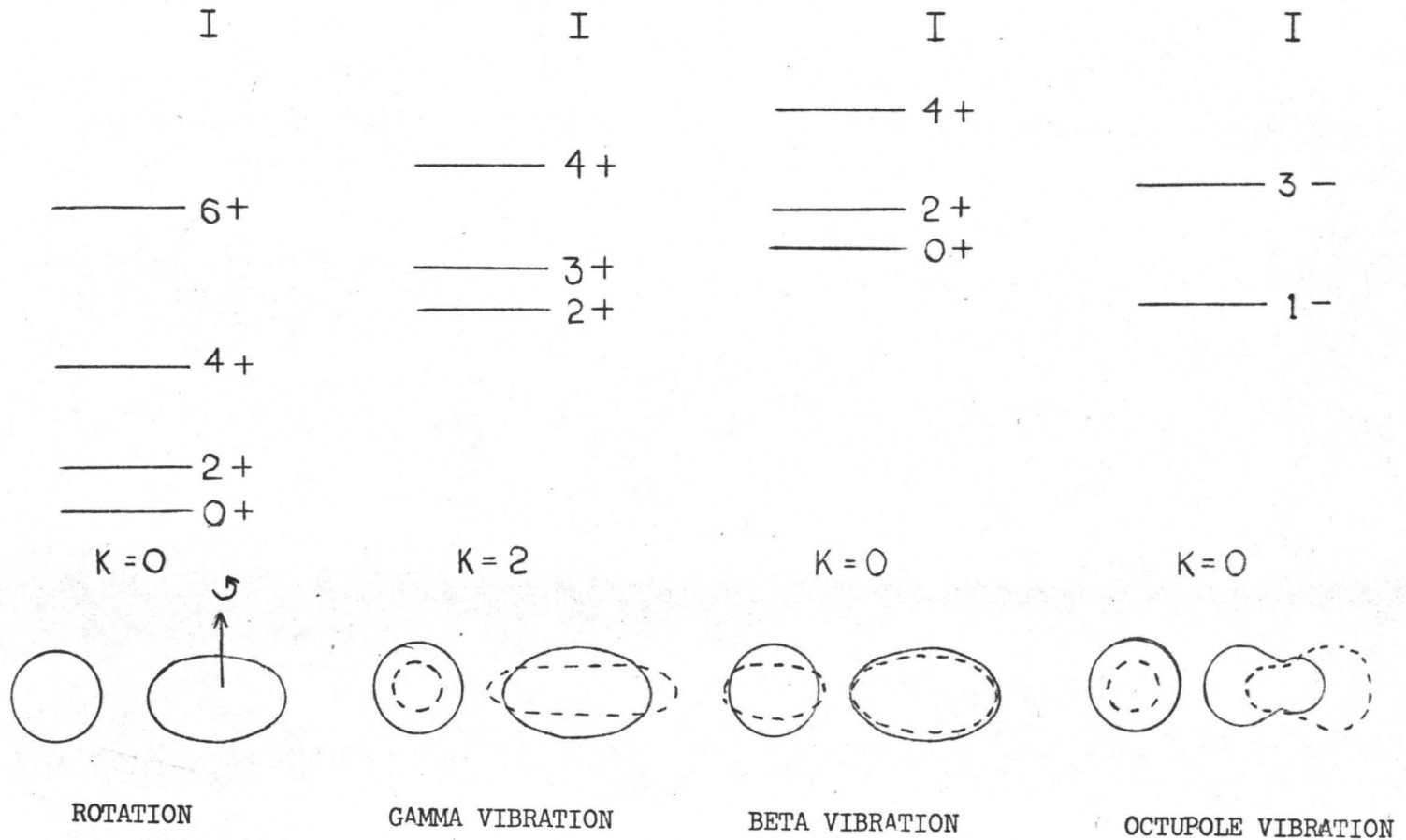


FIGURE 3: Some possible spin values for the rotational bands built on the different types of collective motion, cross sectional views perpendicular and parallel to the symmetry axis are shown. The solid and dotted lines indicate the extremities of the nuclear shape reached in each type of vibration.

## 2.4 The Unified Model

The sort of deformations mentioned above are obviously going to have an effect on the individual particle states. Nilsson<sup>(10)</sup> in 1955 extended the shell model by calculating the energies of the single particle levels in an anisotropic harmonic oscillator potential. To the oscillator potential with the usual  $l \cdot s$  term included Nilsson added a term proportional to  $l^2$  and a deformation parameter  $\beta$  in such a way that the shell model states would still be retained for zero deformation. Using Nilsson's notation, a wave function may be expressed as

$$\psi = \sum a_{N \Lambda \Sigma} | N \Lambda \Sigma \rangle$$

Here  $N$  is the total number of oscillator quanta,  $\Lambda$  is the particle's angular momentum and  $\Sigma$  is its component along the symmetry axis.  $\Sigma$  is the component of spin along the symmetry axis and thus  $\Omega = \Lambda + \Sigma$ . As the deformation increases,  $l$  and  $j$  cease to be good quantum numbers and states of the same  $\Omega$  are distinguished by the asymptotic quantum numbers  $[N, n_3, \Lambda]$  where  $n_3$  is the number of oscillator quanta along the symmetry axis and  $\Lambda$  is the projection of the orbital angular momentum on this axis.

In order to see which state this model predicts for any nuclide, one simply counts up the orbitals at the appropriate deformation placing two particles in each. Spins predicted in this manner agree very well with those observed for a large number of nuclides in the three well known regions of deformed nuclei  $A \sim 25$ ,

$150 < A < 180$  and  $220 < A$ . It is, of course, also possible to build rotational and vibrational bands on each particle state, as discussed in the last section.

The equilibrium shape of a nucleus of given  $A$  can be obtained by adding up the energies for all the filled single particle states for a series of  $\beta$  values and then plotting the total energy as a function of  $\beta$ . The equilibrium distortion corresponds to the minimum in this total energy versus  $\beta$  curve. There seems to be a factor of  $\sim 3$  between deformation parameters obtained by this method and deformations predicted from experimental electrical quadrupole moments.

## 2.5 Theory of Intermediate Coupling in the Unified Model

The coupled system consists of the core and the extra nucleon; the Hamiltonian for this system consists of three parts:

$$H = H_s + H_p + H_{int}$$

Where  $H_s$  is the Hamiltonian associated with the quadrupole vibrations of the surface of the core, it is equivalent to that of a system of harmonic oscillators.  $H_p$  is the Hamiltonian for the odd nucleon in an effective average potential, and  $H_{int}$  is the Hamiltonian for the particle surface interaction.

As we have seen above the coupled system possesses simple solutions in the limit of weak and strong coupling. In the former case, the particle and the collective types of motion are approximately independent and the effect of coupling can be treated as a small perturbation. In the latter case, the system bears

analogies to molecular structures; the nucleus acquires a large deformation and the stationary states can be characterized by the motion of the particles with respect to the deformed nucleus and the vibration and rotation of the structure as a whole.

In many nuclei, however, neither the weak nor the strong coupling solutions are adequate. A method for dealing with this kind of situation has been developed by Bohr and Mottelson<sup>(11)</sup> (1953) and a short description of the method as developed by Choudhury and Kujawski<sup>(12)</sup> 1966 (follows).

In order to study the predictions of this coupled system, eigenvectors of the uncoupled system are chosen as a basis of the space. These basis eigenvectors are denoted by  $j; N R; I M$  and they satisfy the following equation:

$$(H_s + H_p) |\alpha j; N R; I M\rangle = [\hbar\omega (N + 1/2) + E_j] |\alpha j; N R; I M\rangle$$

Here  $j$  is the single particle angular momentum and  $\alpha$  represents the radial quantum numbers of the particle such as  $n$  and  $l$ ;  $N$  is the number of phonons of surface oscillations, each having an angular momentum of two units;  $R$  is the total angular momentum of the surface;  $I = j + R$  is the total angular momentum of the system,  $M$  its  $z$  components; and  $E_j$  is the energy of the single particle in the quantum state of angular momentum  $j$ .

The matrix elements of  $H_{int}$  can be evaluated, and hence the total Hamiltonian arrived at. Diagonalization then yields the wave functions that describe the energy levels of the model. These wave functions can be used to calculate the electric and magnetic transitions and comparison with experiment can then be made.

### CHAPTER III

#### SURVEY OF PREVIOUS WORK

The 33 hour beta emitter in Cerium was first observed by Pool and Kurbatov<sup>(13)</sup> 1943, they correctly assigned the activity to  $\text{Ce}^{143}$ . Since then the decay of this isotope has been investigated by many workers, including Martin et al<sup>(14)</sup> (1955) Rao and Hans<sup>(15)</sup> (1962) Gopinathan et al<sup>(16)</sup> (1964) Mancuso et al<sup>(17)</sup> (1965) all of whom used scintillation coincidence spectrometers.

The prominent energy levels were fairly well established by Martin et al who studied the gamma ray spectrum with a ten channel scintillation coincidence spectrometer and cubical crystals of  $\text{NaI(Tl)}$  about 2 1/4 inches on a side. The internal conversion electron spectrum was observed with photographic magnetic spectrographs, the beta ray spectrum was analyzed with a double focusing magnetic spectrometer, and a beta-gamma coincidence experiment was done using a anthracene crystal as a beta detector. Levels were established at 0, 57, 232, 351, 724, 918, and 1160 keV. Beta rays were observed going to all these states except the ground state, their energies being 300 (6%), 540 (12%), 730 (5%), 1110 (40%), and 1400 (37%) keV.

Gopinathan et al carried out gamma-gamma coincidence with  $\text{NaI(Tl)}$  crystals of size 1.5 inches by 1.5 inches and 2 inches by 2 inches and a 512 channel analyzer. Their decay scheme proposed levels at 0, 57, 351, 493, 725, 942, 1160, and 1395 keV. Mancuso

used 2 in. by 2 in. NaI(Tl) crystals and a 256 channel analyzer and confirmed the existence of levels at 57, 350, 945, and 1167 keV. In addition they established a level at 493 keV. As a result of angular correlation experiments the following spin assignments were made 57 ( $5/2$ ), 350 ( $7/2$ ), 493 ( $5/2$  or  $7/2$ ), 725 ( $5/2$ ). The decay schemes of Mancuso and Gopinathan are presented in Fig 4. By measuring the L subshell ratios with a 100 cm. radius iron free beta spectrometer Gelletly<sup>(18)</sup> et al (1967) determined the 57 keV transition to be predominantly M1, with less than 0.2% E2 admixture. Similarly the 293 keV M1-E2 transition de-exciting the 350.7 keV level was found to have a  $37 \pm 4\%$  E2 admixture. The half life of the 57 keV state was measured by Graham<sup>(19)</sup> et al as  $4.17 \pm 0.09$  n- sec. and that of the 350 keV state. 0.3 n- sec. The ground state spin of  $^{143}\text{Ce}$  was measured by Maleh<sup>(20)</sup> and that of  $^{143}\text{Pr}$  by Burdick<sup>(21)</sup> et al. As well as these experimental papers Choudhury and Kujawski<sup>(22)</sup> published in 1965 a paper which gave the calculated results for the low lying levels of  $\text{Ce}^{143}$ . Their calculations were based on the intermediate coupling model and will be discussed in the last chapter.

As can be seen from Fig. 4 there are discrepancies in the spin assignments and in the energy measurements of the previously suggested decay schemes. In view of this and in view of the availability of the recently developed high resolution lithium drifted germanium detectors and a large (4096 channels) multichannel analyzer it was felt that another investigation could profitably be made. This investigation included gamma singles and gamma-gamma coincident measurement and will be discussed in chapter 5.



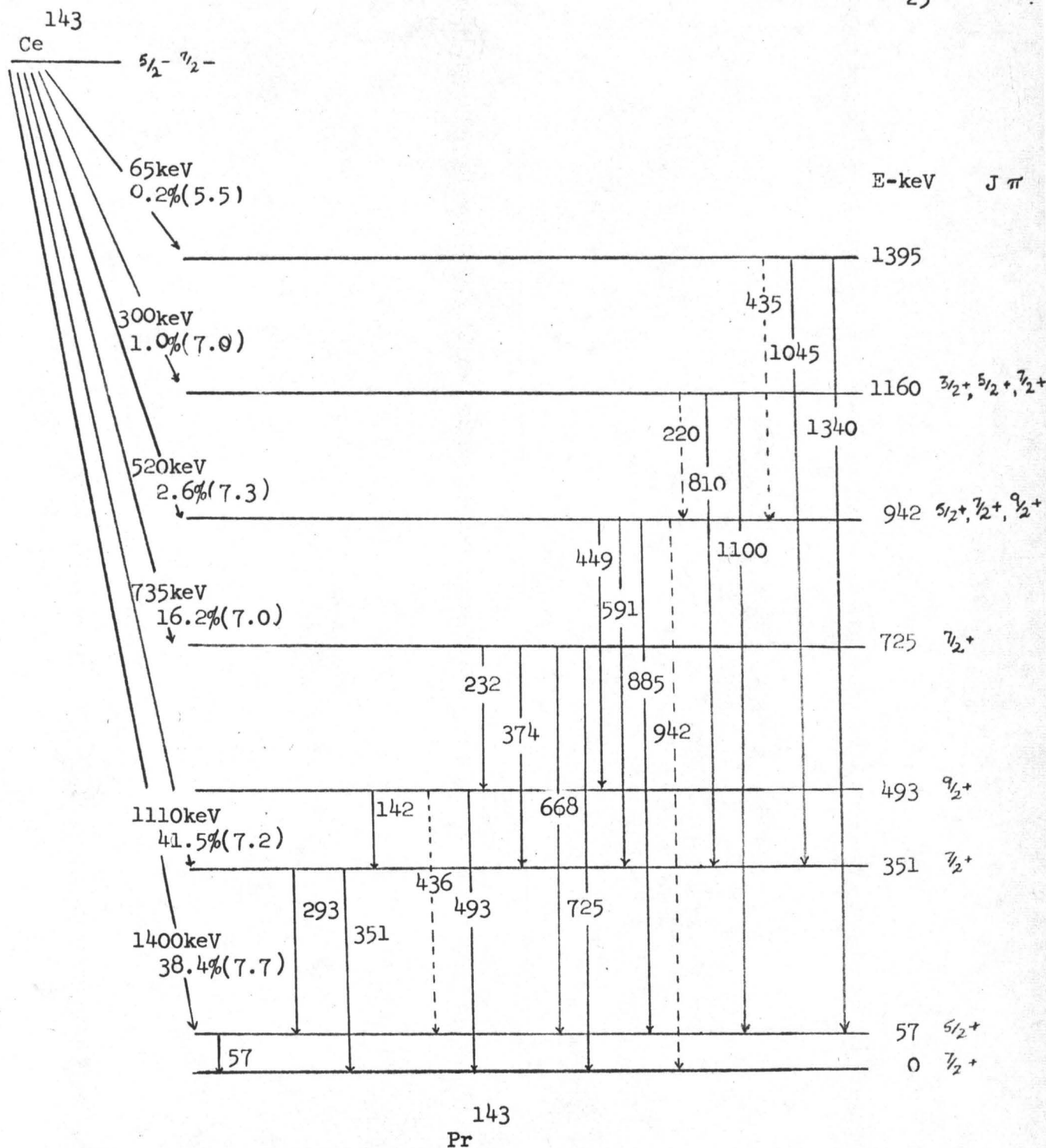
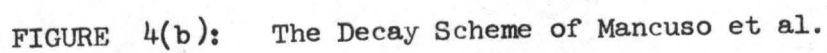


FIGURE 4(a) ; The Decay Scheme of Gopinathan et al.





## CHAPTER IV

### THE APPARATUS

#### 4.1 Solid State Detectors

The semi-conductor detector is similar in operation to the gas ionization chamber. Both consist essentially of an active region in which positive-negative charge pairs are produced by the passage of the ionizing radiation, and across which an electric field exists in order to remove these charges.

The material from which a detector is made must approach the following requirements:

- (i) The average energy required to produce an electron-hole pair should be as small as possible.
- (ii) The material should contain few free carriers at the operating temperature.
- (iii) Recombination rates of holes and electrons during the charge collection must be very small.
- (iv) The material should contain elements of high atomic number as this improves the gamma absorption properties of the detector.

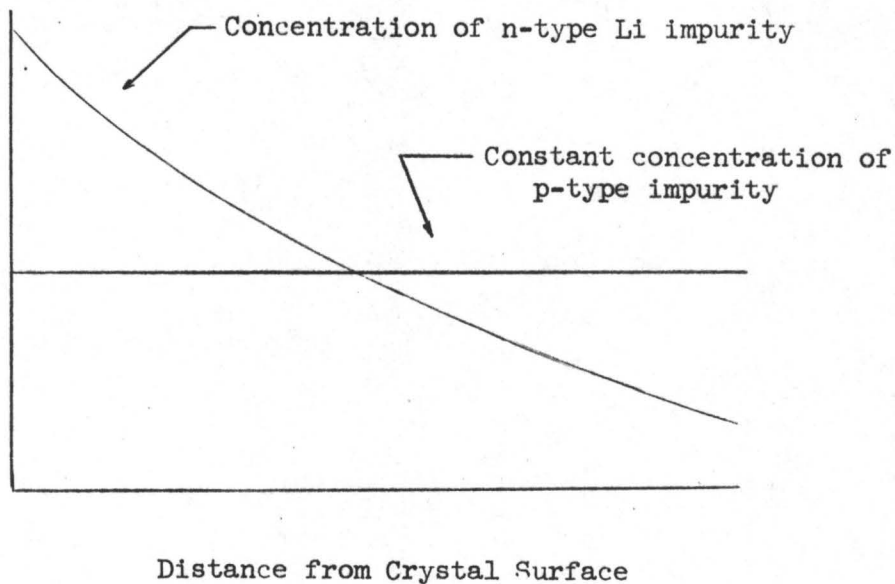
Only silicon and germanium are known to approach these properties.

In the fabrication process of a germanium detector a high concentration of lithium is deposited on the surface of a richly p-type doped section of a single germanium crystal. Raising the temperature of the crystal to 400°C causes the lithium to diffuse

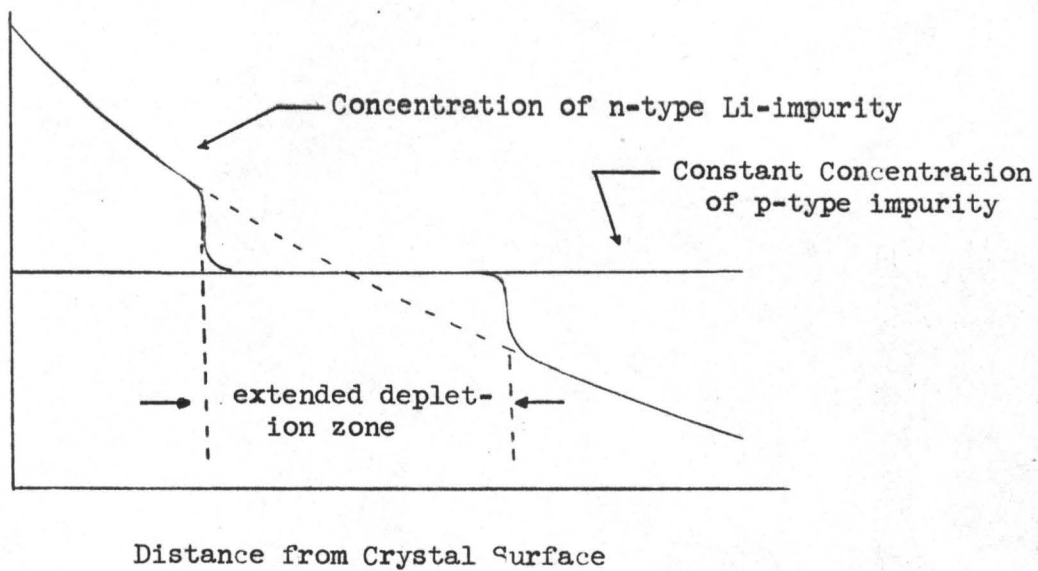
inward from the surface to give the situation shown in figure 5a. With a reverse bias of several hundred volts applied to the detector and with the temperature at about  $50^{\circ}\text{C}$ , the lithium ions are drawn across the junction from the n-side to the p-side where they combine with holes to form intrinsic semi-conductor material. The result of this process is shown in figure 5b. Until recently all detectors were made in the "planar" fashion, shown in figure 6a. The lithium was painted on one side of a relatively thin slice of germanium and then dried inward. These devices are limited in size by the cross sectional area of the initial germanium crystal and the depth of the depletion zone.

At the present time larger detectors are being made in the "wrap around" fashion, where the lithium is drifted simultaneously from all sides but one of the crystal to give a detector as shown in figure 6b. Detectors of this sort have the advantages of large active volumes (up to 40 ccs), and small variations in the depletion zone depth.

For many duties solid state devices are replacing NaI(Tl) detectors. They have one big advantage and this is their greatly improved resolution (typically a factor of 20 better). This improved resolution is a result of the more direct detection process (electron-hole production and collection) as opposed to the multi-step scintillation process, (excitation luminescence, light conduction, photo emission, etc.) each with its own resolution-destroying statistical variations. The disadvantage which solid state detectors have, that of inferior efficiency due



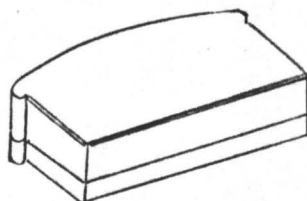
(a) Impurity Concentrations after Diffusion but before Drifting.



(a) Impurity Concentration after Drifting.

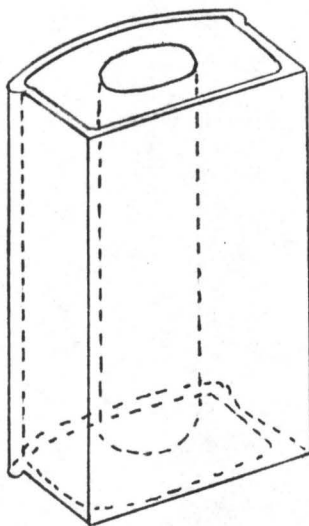
FIGURE 5 : THE LITHIUM DRIFT PROCESS

FIGURE 6



Li rich layer  
depletion zone  
p-type crystal

(a) Planar Type



p-type crystal  
Li rich layer  
depletion zone

(b) Wrap Around Type

The Two Different Types of Detectors after

; Comparable Drifting Times.

to smaller size, is largely being overcome by improved fabrication techniques.

#### 4.2 Amplification

Amplification is achieved in two stages, preamplification and main amplification with pulse shaping. A Tennelec model T.C. 130 preamplifier was used in the present case for the first stage. It is an instrument specifically designed for use with cooled detectors and incorporates a very low noise, cooled field-effect transistor. It is connected to the detector by as short a lead as possible in order to preserve the low noise characteristics. A Tennelec Model TC 200 double delay line linear amplifier was used for the second stage. For coincidence spectra where a cross-over point was required, it was used in the D. D. mode. However, for singles spectra it was found that the R.C. shaping was able to yield a substantially improved resolution.

#### 4.3 The Multichannel Analyzer

Technically, the aim of pulse spectroscopy is to obtain a statistically significant sample of the pulse height distribution associated with the detector response to the radiation characterizing a particular excitation or de-excitation process. To achieve this aim, it is necessary to sort the amplified pulses according to height and record the relative frequency of occurrence of a given incremental pulse height range. The multichannel analyzer is a specialized computer designed to do this so that a measure of the pulse height distribution as a whole is obtained in one counting period. The early multichannel pulse height analyzers used a

voltage sensitive discriminator and a scalar in each channel. This was very clumsy and expensive and the introduction of analog to digital conversion (A.D.C.) by Wilkinson in 1950 was a major advance. The principle is quite simple. A capacitor is charged to a voltage proportional to the peak voltage of the input pulse. During the linear discharge of the capacitor, pulses from a periodic pulse generator are counted by a scalar, the state of the scalar indicates in digital form the magnitude of the input pulse.

The response characteristics of the analyzer are determined mainly by the performance of the A.D.C. The integral linearity, a measure of the linearity of the pulse height channel-number relation, is determined by the quality of the ramp circuit. The stability of the analyzer depends upon the stability of the ramp, the oscillator and the associated gating circuits.

Since radioactive decay is of a random nature the pulses from detectors are distributed randomly in time. In order that only one pulse occurs in the converter circuit during analysis, there is a linear gate which closes during the analysis time required for the pulse already in the converter and opens when the analysis is completed.

Since the time required for analysis depends upon the address assigned to the pulse, the overall dead time for a particular counting experiment depends upon the nature of the spectrum involved. The actual live time of the analyzer during a given counting period is measured by scaling a clock oscillator.



The analyzer used was a Nuclear Data 4096 channel analyzer, each channel or location being able to contain an 18 bit word, corresponding to a maximum count of 262,144.

#### 4.4 Coincidence Circuit

The gamma-gamma coincidence measurements were carried out with the above two dimensional analyzer. A block diagram of the circuit used is shown in figure 7. In this diagram D.D. 2 represents the Tennelec double delay line amplifiers. The bipolar pulses from each were fed to, on the one side, a zero strobe (Camberra model 1420) and, on the other, a single channel analyzer (Camberra model 1435). These units were both used as zero strobes, that is, they detected the zero crossing point of the double delay line shaped pulses, and generated a timing signal when the input pulse crossed the zero voltage baseline. These timing pulses were then routed to a fast coincidence unit (Camberra Model 1440) which generated a rectangular logic pulse whenever the two pulses arrived within the adjustable resolving time of each other. A resolving time of about 100 n sec. was employed in the present experiments. The logic pulse was then fed to the analyzer and enable it to accept the two coincident linear pulses from the detectors. This information was then stored in the form of a matrix. Each event being placed in matrix position (i, j), where channel i was proportional to the pulse height from crystal 1 and channel j was proportional to the pulse height received from crystal 2.

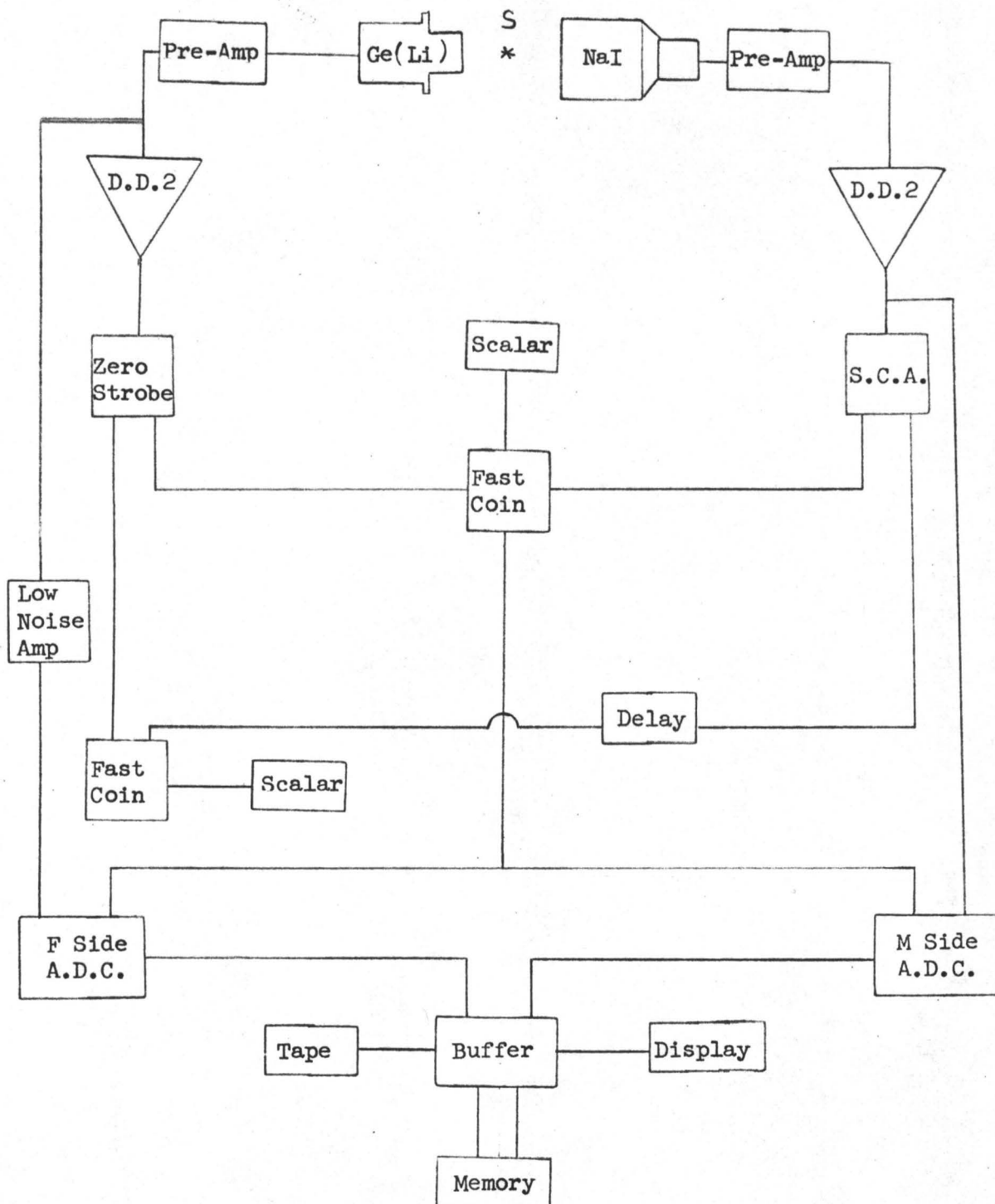


FIGURE 7

TWO DIMENSIONAL COINCIDENCE SPECTROMETER.

When being used with a coincidence configuration the memory of the analyzer is divided into two, half for oscilloscope display and half for storage. For Ge-NaI coincidence experiments it is usual to have a  $1024$  by  $256$  configuration. Thus when a coincident event is detected the information from the Ge(Li) detector is written in a 10 bit word and the information from the NaI(Tl) detector on an 8 bit word, these combine to fill one of the 18 bit locations in the memory. Each event is stored sequentially. When the storage half of the memory is full, that is when it contains  $2048$  coincident events it dumps, automatically, onto to magnetic tape and then continues with further counting. The tape is seven bits wide, the seventh bit is a parity instruction, so each word is split into three.

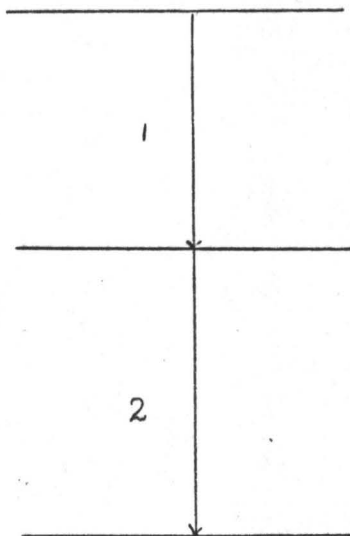
Four tapes containing a total of about  $7.1 \times 10^6$  events were obtained, these being fed to an I.B.M. 7040 computer for sorting.

The computer cannot sort all this information into matrix form in one operation. The size of the matrix was  $256$  by  $1024$  which is too big for the computer memory. So the memory forms a  $256$  by  $128$  matrix, then it checks the tape and places appropriate events into the proper place in the sub matrix. When it has gone through the tape it then dumps the contents of the memory onto a second tape and continues to place events into the second sub matrix and so on until the eight sub matrices have been covered. This is done for each tape then appropriate tapes are added.

The coincidence experiment was performed in a  $180^\circ$  geometry with the source mounted in an anti-compton shield to decrease the probability that either detector "sees" radiation scattered from the other. However, the  $180^\circ$  backscatterings are now enhanced with respect to other scattering angles with the result that backscattering peaks and Compton edges become "sharper".

As well as true events a number of chance events, which are due to the finite resolving time of the detector, are recorded. It can be shown that the ratio of true to chance events is given by  $1/2\tau N_0$ , where  $\tau$  is the resolving time.

Consider a simple cascade:



The counting rate in one counter will be given by  $N_1 = N_0 \omega_1 \epsilon_1$ , and that in the other

$$N_2 = N_0 \omega_2 \epsilon_2$$

Thus the true gamma-gamma coincidence rate will be given by

$$N_{12} = N_0 \omega_1 \epsilon_1 \omega_2 \epsilon_2$$

and the chance rate by

$$N_{ch} = 2\tau N_1 N_2 \text{ if each}$$

detector accepts counts for a time  $\tau$  ..

The ratio of true to chance coincidences will therefore be:

$$\frac{N_{12}}{N_{ch}} = \frac{1}{2\tau N_0}$$

Thus for a given  $\tau$ , determined by the apparatus,  $N_0$  must be kept as small as possible to make the true to chance ratio as large as possible. Further the example given above is the most favourable case. It can be shown in a similar way that if there is a cross-over transition the ratio will be given by:

$$\frac{N_{12}}{N_{ch}} = \frac{\delta_1}{2 \tau N_0}$$

if  $\delta_1$  is very small it becomes very difficult to keep the true to chance rates at an acceptable level. In practice, however, the situation was too complicated for this kind of analysis to be of any use. The chance spectrum has the same shape as the singles. In these experiments the source used was sufficiently weak that the ratio of total chance counts to total singles count was less than 2%, this being measured by the ratio of counts on the two scalars shown in figure 7.

## CHAPTER V

### THE EXPERIMENTS

#### 5.1 The Ge(Li) Gamma Ray Singles Measurements

The gamma singles experiment was done with a planar type Ge(Li) detector from Nuclear Diodes. Its area was  $1.5 \text{ cm}^2$  and its depletion depth 0.4 cms. giving an active volume of 0.6 ccs. The resolution of the whole system was 2.5 keV (full width of half maximum) at 293 keV. Two, almost exactly, reproducible geometries were used when counting. In one, the "distant geometry" the source was placed 4.2 cms. from the detector and no absorber was used. For the other, the "normal geometry", the source was held  $\sim 0.95$  cms from the detector in a plastic holder which was constructed to also hold an absorber. This consisted of two thin ( $\sim 1\text{mm}$ ) pieces of lead and a thin ( $\sim 1\text{mm}$ ) piece of cadmium, the cadmium being on the detector side of the lead. The lead served the purpose of absorbing the low energy (less than 80 keV) x-rays and background and thus helping to reduce the analyzer dead time to an acceptable level ( $\sim 20\%$ ). The cadmium absorbed the lead x-rays.

The cerium was obtained in powder form being 93%  $^{142}\text{Ce}$ . The main impurities being  $^{140}\text{Ce}$  (7%),  $\text{Eu}^{151}$  and  $\text{La}^{139}$ . Irradiations were made in the McMaster reactor with about 0.1 mg sealed in a quartz capsule and irradiated for 24 hours in  $\sim 10^{13} \text{ n/cm}^2$  flux. The run in the normal geometry was made for 39 hrs. 34 min. and the run in the distant geometry for 46 hrs. 19 min. During these

runs the accumulated data was dumped onto tape every few hours so that a comparison of half lives could be obtained. In addition runs with an aged source and with  $\text{La}^{140}$  were made to aid in the identification of impurities and with a number of standard sources to enable energy calibrations to be made.

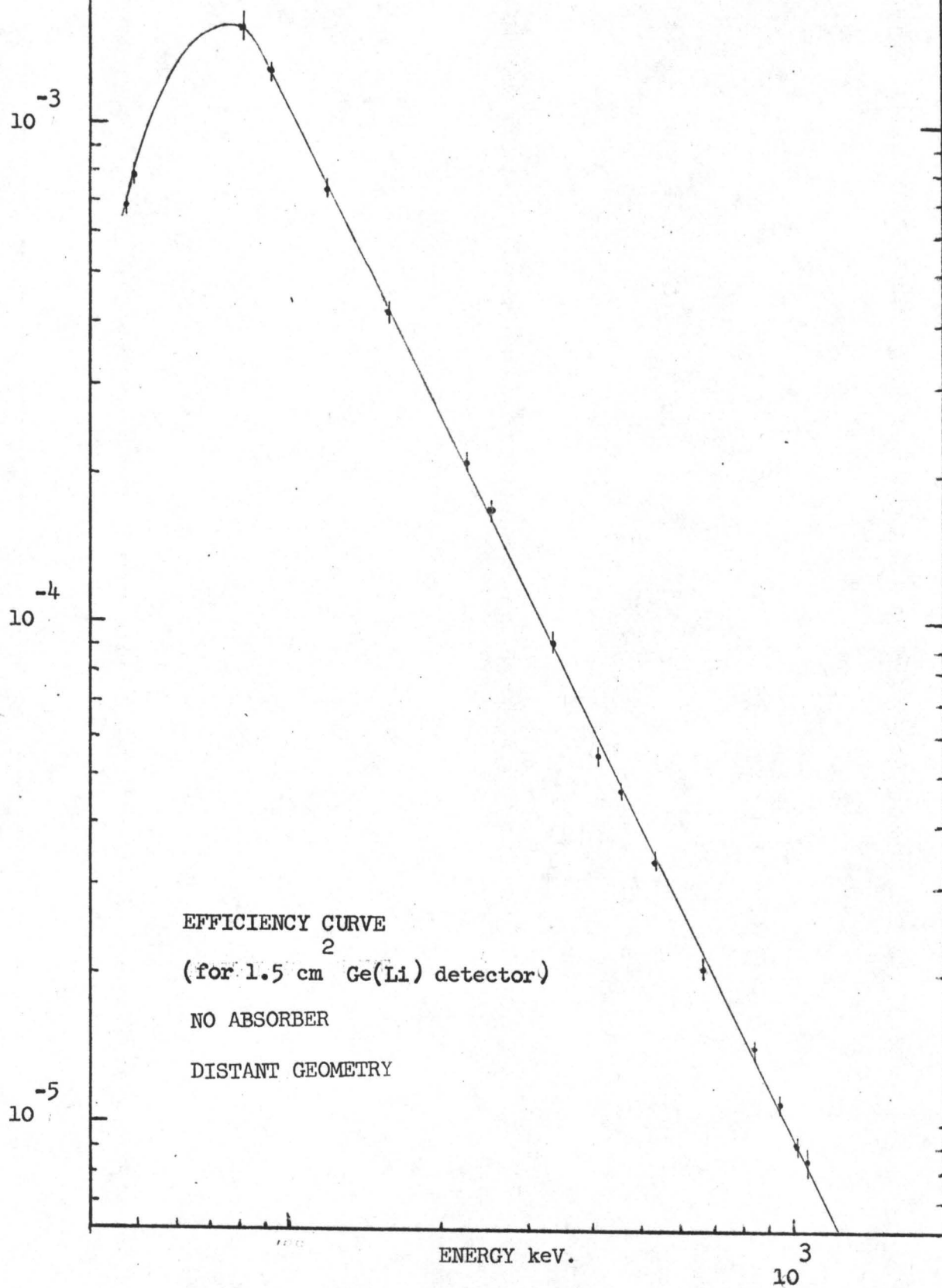
## 5.2 Efficiency and Energy Determination

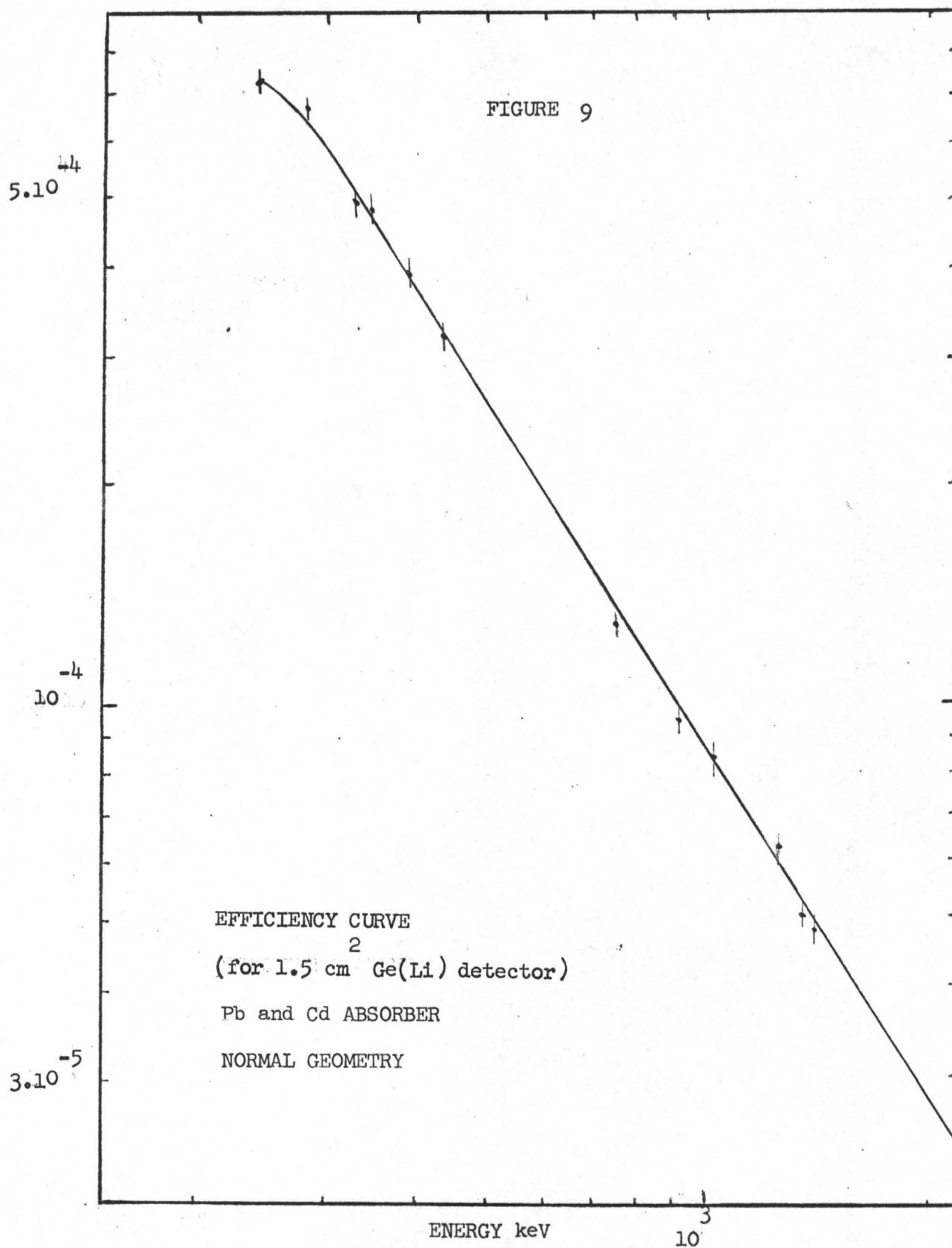
The photopeak detection efficiency of the crystal as a function of energy for the two geometries used was measured using a number of standard sources. The strengths were measured using a NaI detector and the tabulated efficiencies and photo-fractions of Heath<sup>(23)</sup> et al. The sources used were  $^{241}\text{Am}$ ,  $^{203}\text{Hg}$ ,  $^{139}\text{Ce}$ ,  $^{203}\text{Hg}$ ,  $^{51}\text{Cr}$ ,  $^{198}\text{Au}$ ,  $^{22}\text{Na}$ ,  $^{207}\text{Bi}$ ,  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ . This method gave a good result for the distant geometry as is shown in figure 8. But for the normal or close up geometry small variations in the positioning of the sources made the method unsatisfactory. The experiment was repeated by another member of the group using a  $^{152}\text{Eu}$  source with relative intensities of gamma lines as given by Dzhelepov et al<sup>(24)</sup>, the result is shown in figure 9.

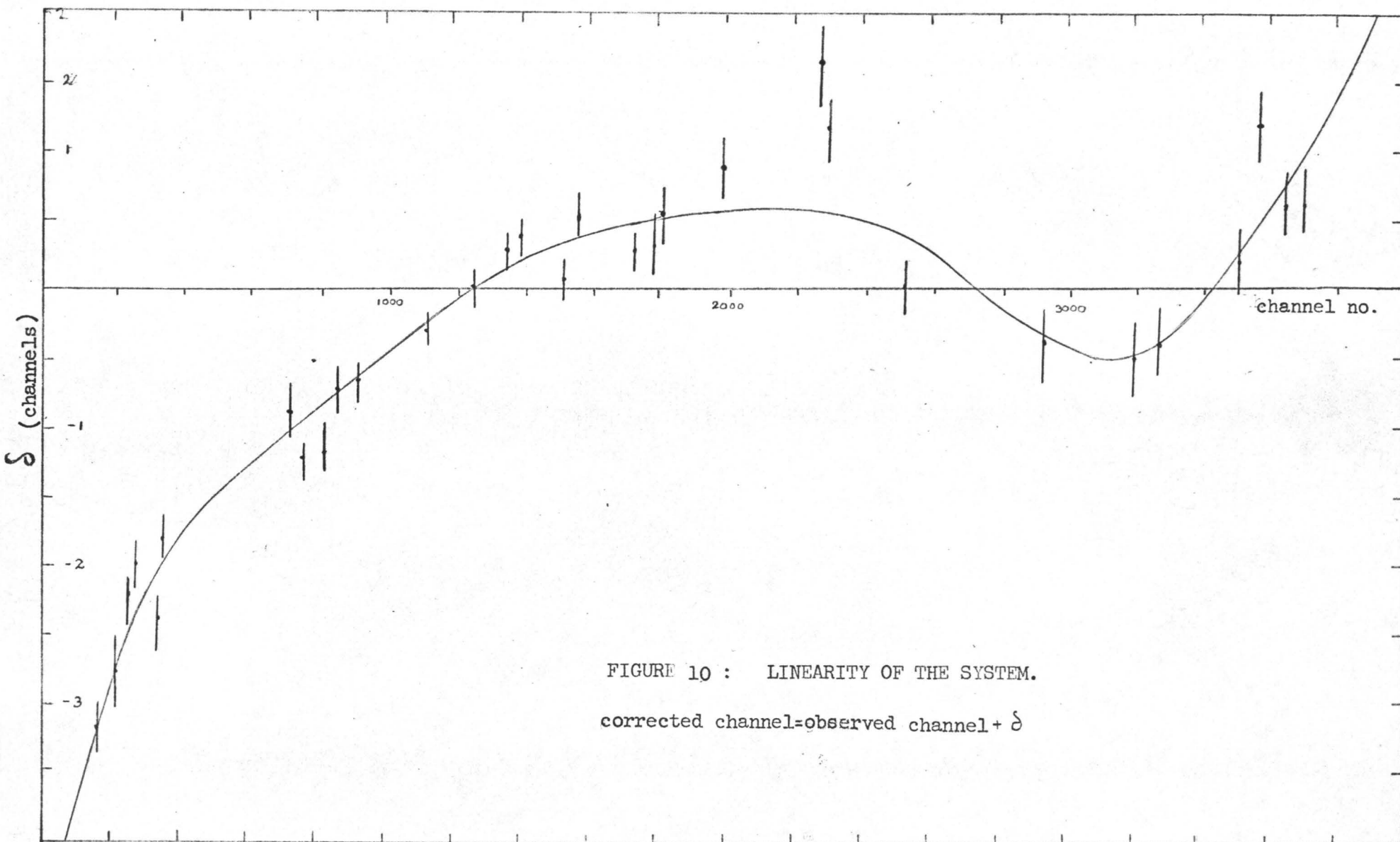
The above sources were also used for the linearity calibration which was obtained as follows: From the spectrum obtained with all the calibration sources in front of the detector the channel numbers of the centers of the peaks were obtained as accurately as possible ( $\pm 0.2$  channels). Two of these peak positions were arbitrarily chosen and assumed to have a correct peak position



FIGURE 8





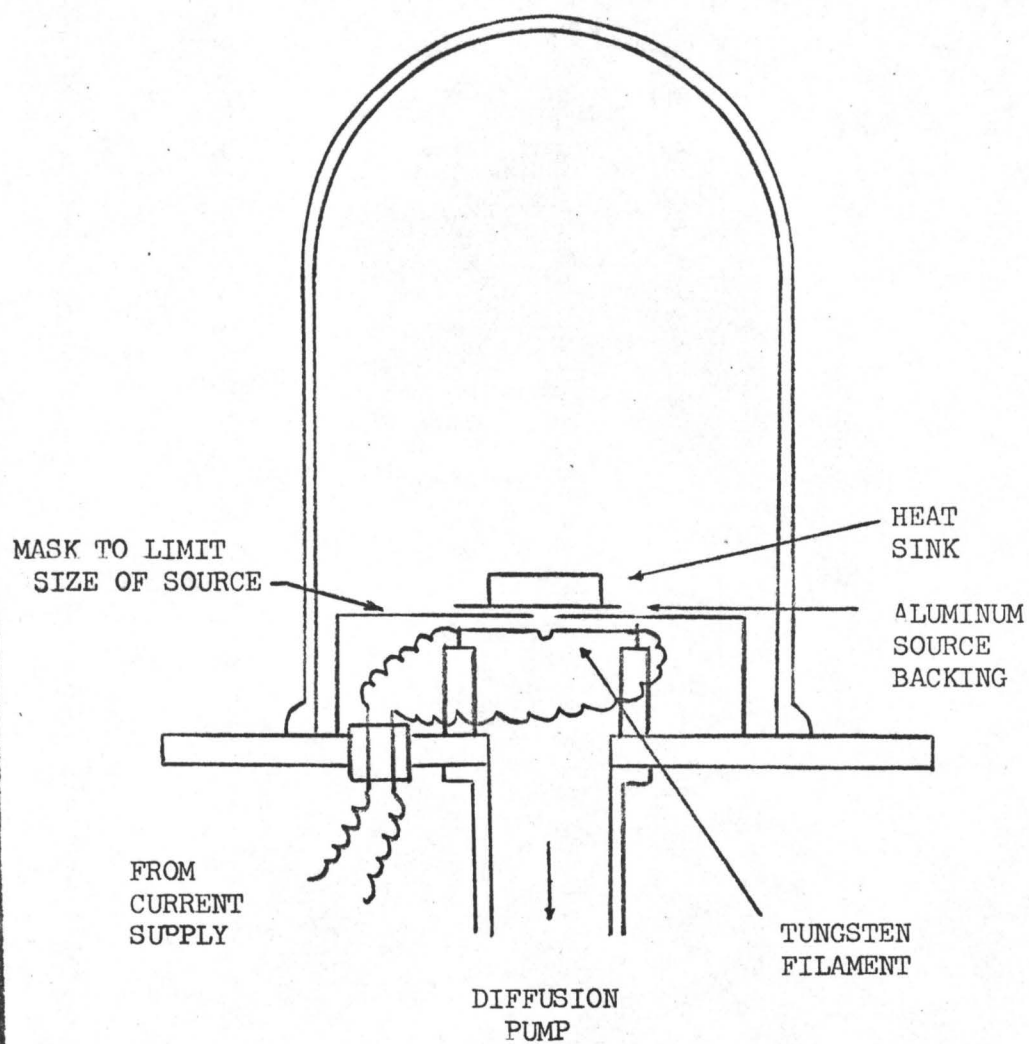


corresponding to their particular energy. Because of the slight non linearity of the detector, amplifiers and analogue to digital converter circuits, the other peak positions then stood in need of correction. The correct values were determined from the two fixed positions and the difference between the observed values and the calculated values was plotted as a function of channel number. The resulting graph was then used to give the correction for all observed peak positions. This is shown in figure 10.

### 5.3 The Si(Li) Beta Ray Measurements

The beta ray spectrum was measured using a Simtec Si(Li) detector, which had a 1mm depletion depth and  $1 \text{ cm}^2$  surface area. The resolution was 3.9 keV (full width at half maximum) at 251 keV. The detector was kept in a vacuum at liquid nitrogen temperatures. These sources were prepared by using a vacuum sublimation technique. After removal from the reactor, the quartz capsule containing the sample was broken and the contents were mixed with a very small ( $\sim 1 \text{ mg}$ ) amount of potassium ferrocyanide. A small quantity of concentrated hydrochloric acid was then added and the resulting solution was evaporated to near dryness. The small drop remaining was then transferred to an indentation in a tungsten filament, figure 11, and dried under a heat lamp. The vacuum system was pumped down to a pressure of about  $10^{-3}$  mm of Hg. The source was first outgassed by heating the filament to  $\sim 800^\circ\text{C}$ . Then with a  $5 \text{ mg/cm}^2$  aluminum source backing and a 0.4 cm diameter collimator in position the filament was heated to  $\sim 1400^\circ\text{C}$ . The voltage being pulsed (5 seconds off

FIGURE 11



APPARATUS FOR PREPARATION OF SOURCES BY VACUUM  
SUBLIMATION.

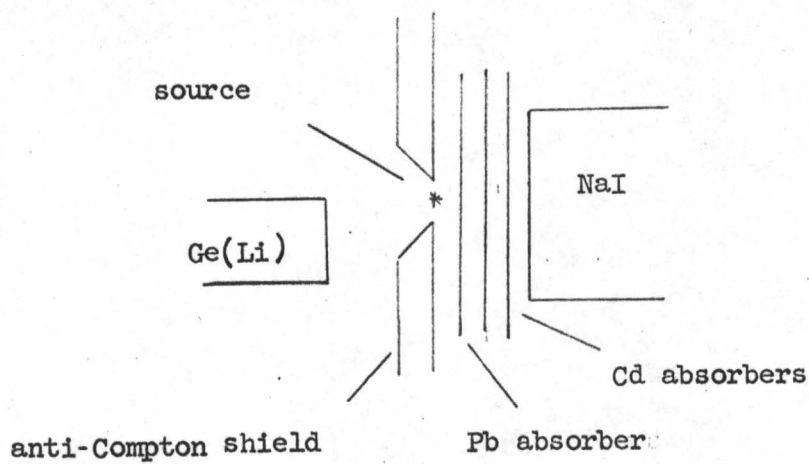
10 seconds on) to avoid melting the aluminum.

#### 5.4 The Ge(Li) - NaI(Tl) Coincidence Experiments

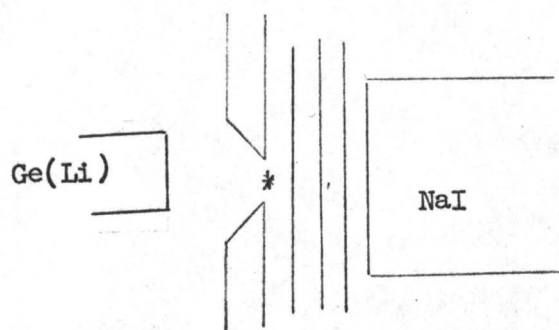
The gamma-gamma coincidence experiments were carried out with the 0.6 cc Ge(Li) detector which gave a resolution of 3.5 keV at 293 keV and a 3 in. x 3 in. NaI(Tl) detector, which had a resolution of 12% (f.w.h.m.) at 293 keV. A total of four tapes of data were acquired. About  $4 \times 10^7$  events were recorded with a geometry as shown in figure 12a. Another  $1.5 \times 10^7$  events with a geometry as in figure 12b. Finally, a low energy experiment was done, no absorber was used, the voltage was increased on the NaI detector and the gain increased on the amplifier on the Ge(Li) side. The geometry was as shown in figure 12c. A further  $1.5 \times 10^7$  events were recorded.

FIGURE 12

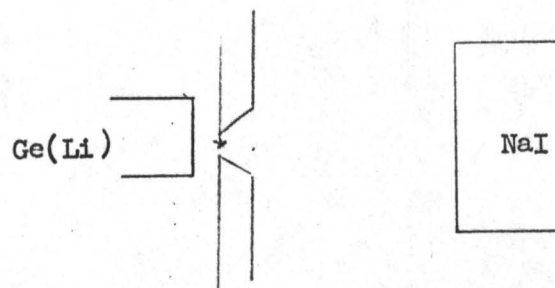
## Coincidence Experiment Geometries.



12(a)



12(b)



12(c)



## CHAPTER VI

### RESULTS

#### 6.1 The Ge(Li) Gamma Ray Measurements

Two runs were made, one for each of the two geometries which are described in chapter 5. In each case data was accumulated in the buffer storage and dumped onto magnetic tape at recorded intervals during the run. The energies and intensities were determined from dumps taken with ~60%, and 100% of the final number of counts recorded, although for some of the weaker peaks the statistics at the 60% point were not good enough to make a determination possible. From the relative intensities of the lines in successive dumps it was possible to make an estimation of the half life of each peak and on the basis of this, some peaks were assigned to impurities.

The intensities, relative to the intensity of the strong 293.3 keV transition, were found by determining the peak areas and using the appropriate efficiency curves shown in chapter 5. The energies were determined from the peak positions, using the non linearity correction curve and the procedure which is also discussed in chapter 5.

The final spectra for the runs in both geometries are presented in figures 13 and 14. All the lines which are observed are presented in table 1. Where a line is observed in both runs the figures given are the weighted means of the measurements. The transition energy of

FIGURE 13.

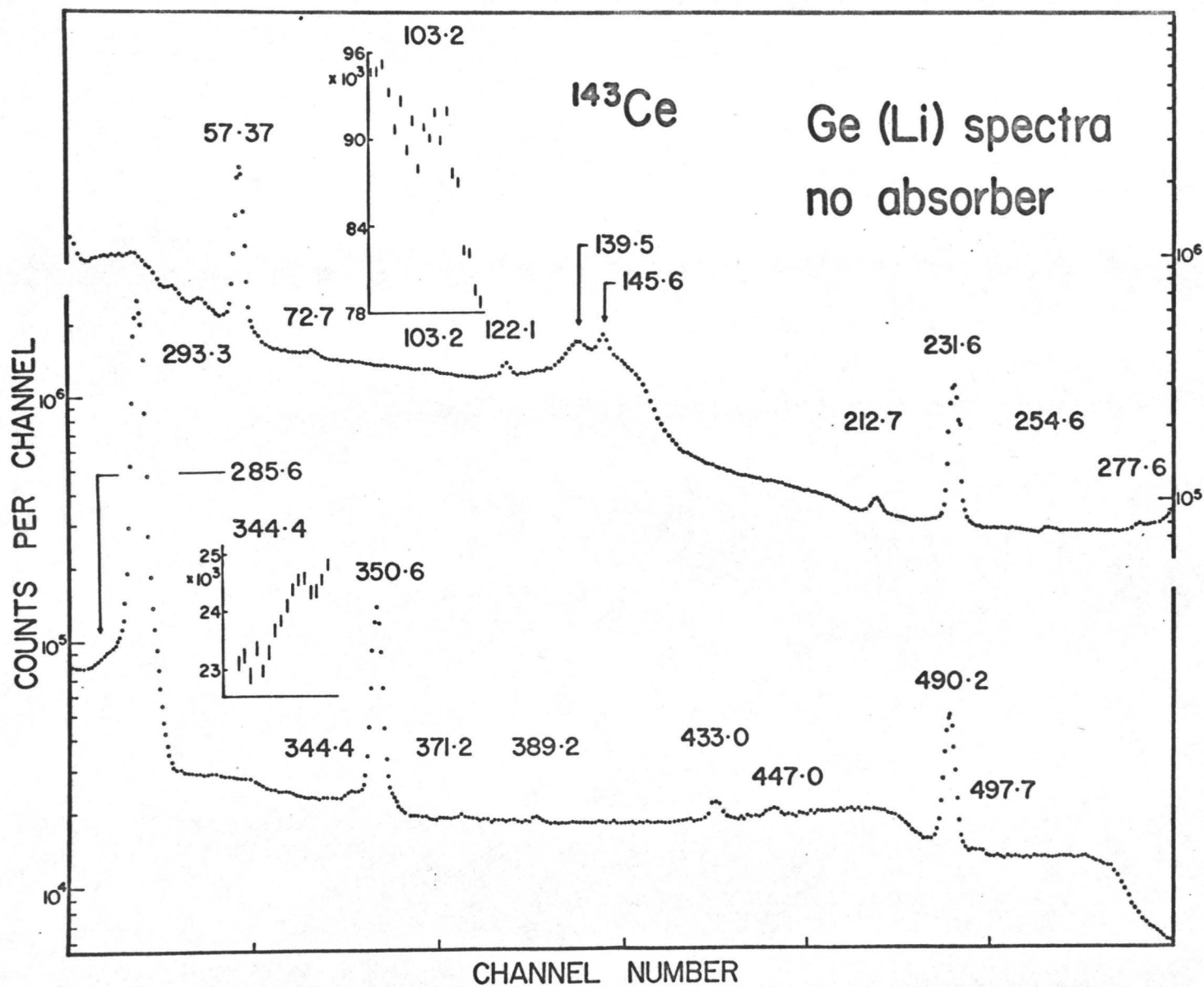


FIGURE 14.

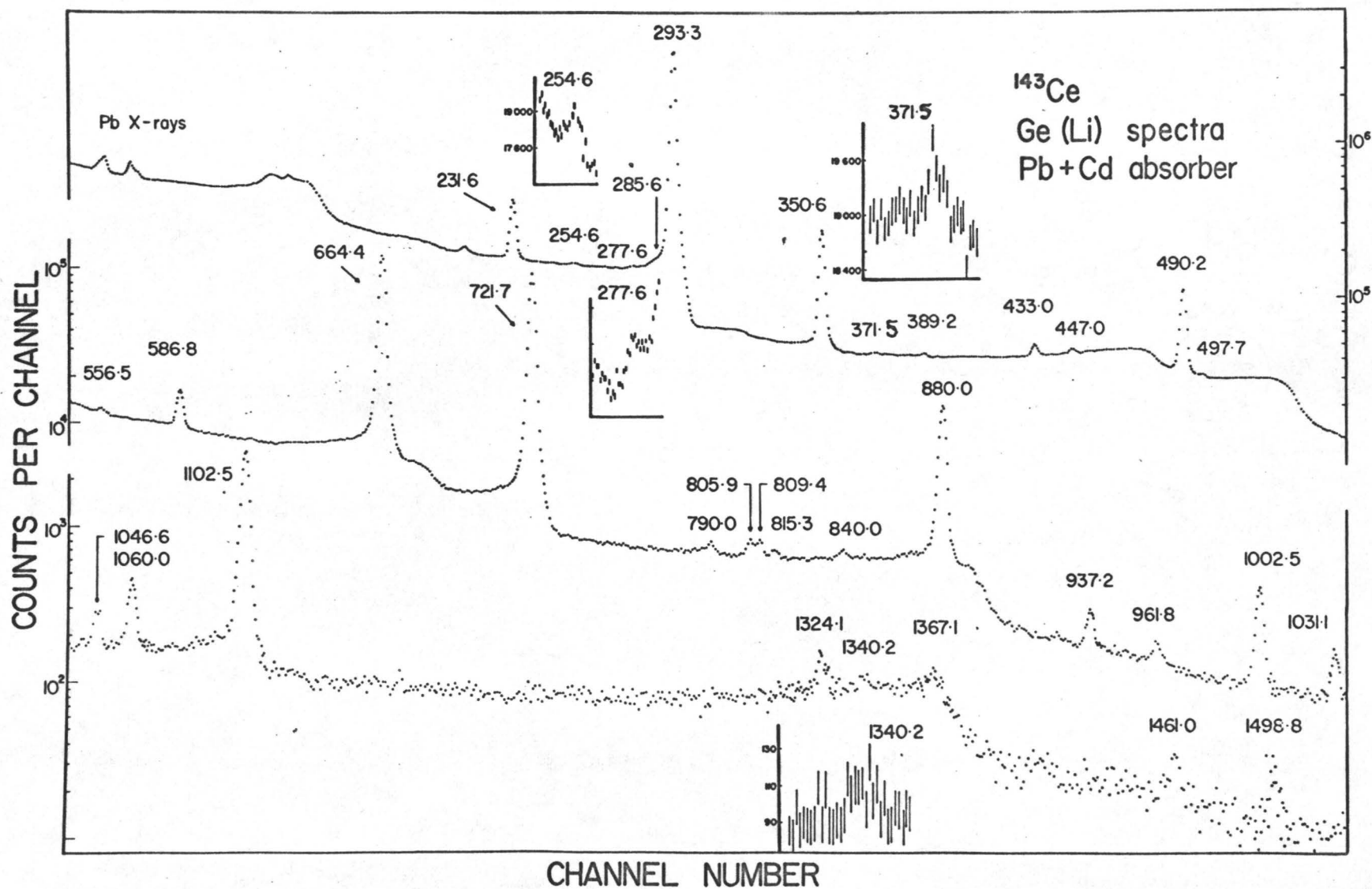


TABLE I

Energies and Intensities of Lines Observed in the Singles Spectra.

| Energy<br>keV | Estimated<br>(E) | Intensity        | Comments                                      |
|---------------|------------------|------------------|-----------------------------------------------|
| 57.37         | 0.05             | 20.4 $\pm$ 4.0   |                                               |
| 72.7          | 0.4              | 0.1 $\pm$ 0.05   |                                               |
| 74.7          | 0.4              | 0.1 $\pm$ 0.05   | lead x-ray                                    |
| 84.8          | 0.7              | 0.05 $\pm$ 0.02  | lead x-ray                                    |
| 103.2         | 0.6              | 0.06 $\pm$ 0.02  |                                               |
| 122.1         | 0.5              | 0.3 $\pm$ 0.05   | 0.8 of this<br>is due to<br>$^{152}\text{Eu}$ |
| 139.5         | 0.5              | 0.2 $\pm$ 0.1    |                                               |
| 145.6         | 0.5              | 1.4 $\pm$ 0.2    | $^{141}\text{Ce}$                             |
| 212.7         | 0.4              | 0.36 $\pm$ 0.08  |                                               |
| 231.6         | 0.3              | 5.2 $\pm$ 0.5    |                                               |
| 254.6         | 0.7              | 0.02 $\pm$ 0.005 |                                               |
| 277.6         | 0.7              | 0.02 $\pm$ 0.005 |                                               |
| 285.6         | 0.7              | 0.04 $\pm$ 0.02  | at limit of<br>detection                      |
| 293.3         | 0.3              | 100 $\pm$ 10     |                                               |
| 344.4         | 0.7              | 0.03 $\pm$ 0.01  | $^{152}\text{Eu}$                             |
| 350.6         | 0.3              | 6.9 $\pm$ 0.6    |                                               |
| 371.5         | 0.7              | 0.03 $\pm$ 0.01  |                                               |
| 389.3         | 0.5              | 0.06 $\pm$ 0.02  |                                               |

TABLE I. (cont'd.)

| Energy<br>keV | Estimated<br>(E) | Intensity        | Comments          |
|---------------|------------------|------------------|-------------------|
| 433.0         | 0.4              | 0.3 $\pm$ 0.04   |                   |
| 447.0         | 0.4              | 0.15 $\pm$ 0.05  |                   |
| 490.2         | 0.3              | 4.3 $\pm$ 0.3    |                   |
| 497.7         | 0.5              | 0.2 $\pm$ 0.05   |                   |
| 556.5         | 0.5              | 0.07 $\pm$ 0.02  |                   |
| 586.9         | 0.4              | 1.0 $\pm$ 0.2    |                   |
| 664.4         | 0.3              | 12.2 $\pm$ 1.0   |                   |
| 721.8         | 0.4              | 10.1 $\pm$ 1.0   |                   |
| 790.1         | 0.5              | 0.04 $\pm$ 0.01  |                   |
| 805.9         | 0.5              | 0.08 $\pm$ 0.03  |                   |
| 809.4         | 0.5              | 0.06 $\pm$ 0.03  |                   |
| 815.3         | 0.4              | 0.02 $\pm$ 0.005 |                   |
| 840.0         | 0.4              | 0.4 $\pm$ 0.1    | $^{152}\text{Eu}$ |
| 880.0         | 0.5              | 2.1 $\pm$ 0.2    |                   |
| 937.2         | 0.4              | 0.05 $\pm$ 0.01  |                   |
| 961.8         | 0.5              | 0.3 $\pm$ 0.07   | $^{152}\text{Eu}$ |
| 1002.5        | 0.4              | 0.1 $\pm$ 0.01   |                   |
| 1031.2        | 0.5              | 0.03 $\pm$ 0.004 |                   |
| 1046.6        | 0.6              | 0.02 $\pm$ 0.005 |                   |
| 1060.0        | 0.5              | 0.06 $\pm$ 0.008 |                   |
| 1102.9        | 0.4              | 0.4 $\pm$ 0.05   |                   |
| 1324.1        | 0.6              | 0.06 $\pm$ 0.01  |                   |

TABLE I. (cont'd.)

| Energy<br>keV | Estimated<br>(E) | Intensity         | Comments                                  |
|---------------|------------------|-------------------|-------------------------------------------|
| 1340.2        | 0.7              | $0.02 \pm 0.006$  | at limit of<br>detection                  |
| 1367.1        | 0.8              | $0.02 \pm 0.006$  | perhaps<br>entirely<br>Compton of<br>1595 |
| 1404.8        | 0.8              | $0.004 \pm 0.002$ | short lived                               |
| 1461.0        | 0.8              | $0.06 \pm 0.02$   |                                           |
| 1498.8        | 0.7              | $0.07 \pm 0.02$   |                                           |
| 1595.3        | 0.7              | $0.9 \pm 0.2$     | $^{140}\text{La}$                         |
| 1731.5        | 0.7              | $0.2 \pm 0.5$     | short lived                               |

57.37 keV was measured by Graham<sup>(19)</sup> et al (1963).

The peaks at 122.1, 840.0 and 961.8 keV are due to  $^{152}\text{Eu}$ , although there is coincidence evidence which shows that there might be a weak 122 keV transition in  $^{143}\text{Ce}$ . The 145.6 keV line is due to  $^{141}\text{Ce}$ , and the 1595.3 keV peak (not shown) is due to  $^{140}\text{La}$ . This latter radiation is believed to be responsible for the Compton edge at 1367 keV, although there may be a weak  $^{143}\text{Ce}$  line also at this energy. Peaks observed at 1404 and 1731 keV decayed with too short a half life to be associated with the  $^{143}\text{Ce}$  decay.

A considerable number of these lines have not been previously reported. Some of these have been located in the decay scheme but discussion will be delayed until after the results of the coincidence experiments have been described.

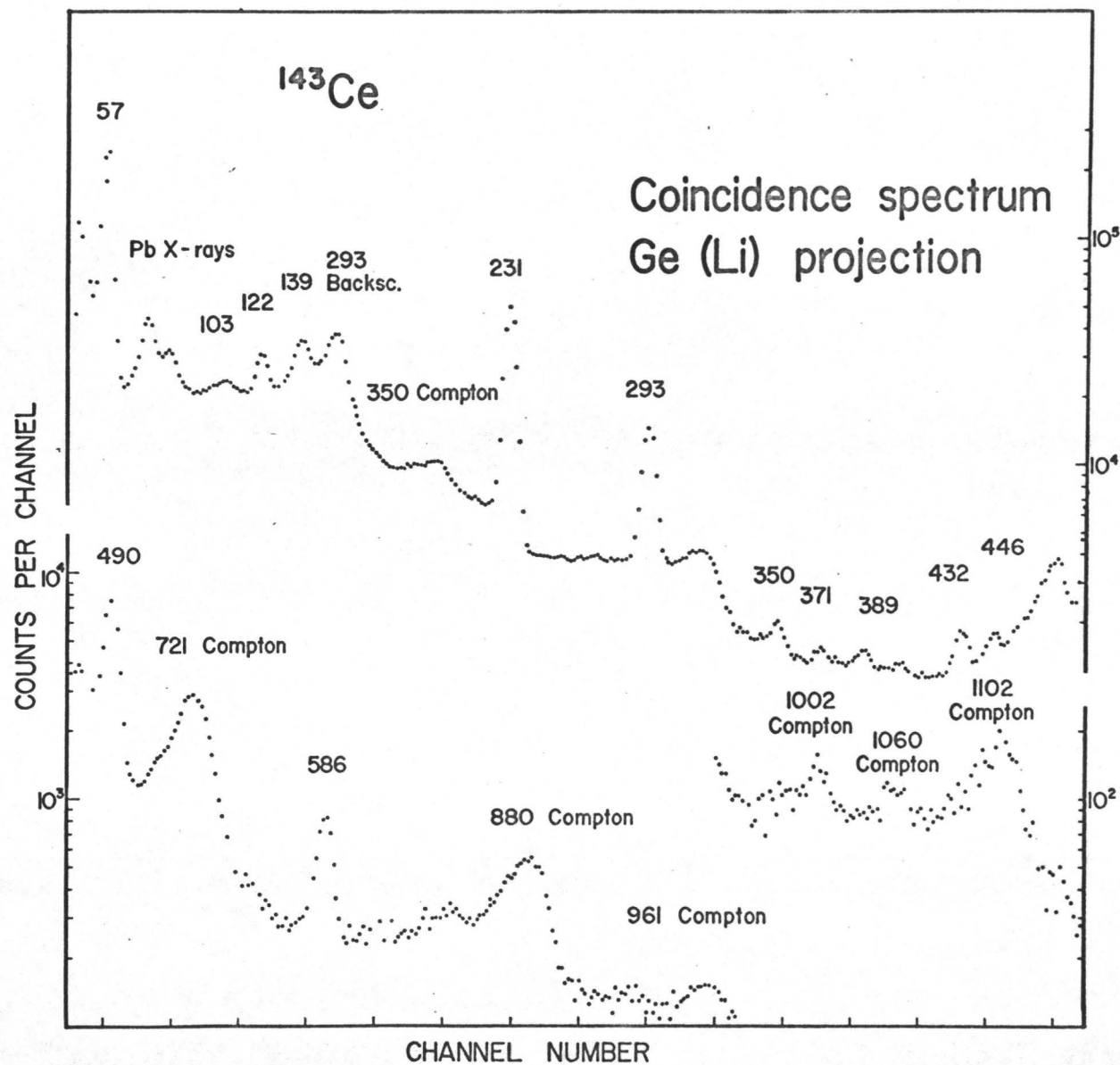
## 6.2 Gamma-Gamma Coincidence Results

The coincidence data was sorted into matrix form in the manner described in chapter IV. Projections onto both axes of the matrix were plotted and are presented in figures 15 and 16. These represent the data on the first three tapes. That is, the two tapes acquired with the geometry as shown in figure 12 and the tape acquired with the geometry as shown in figure 12b, are all summed. (The "low energy" experiment did not produce satisfactory results and the data was discarded).

Computer programmes were used which printed out the spectrum resulting from summing the spectra in coincidence with any specified



FIGURE 15.



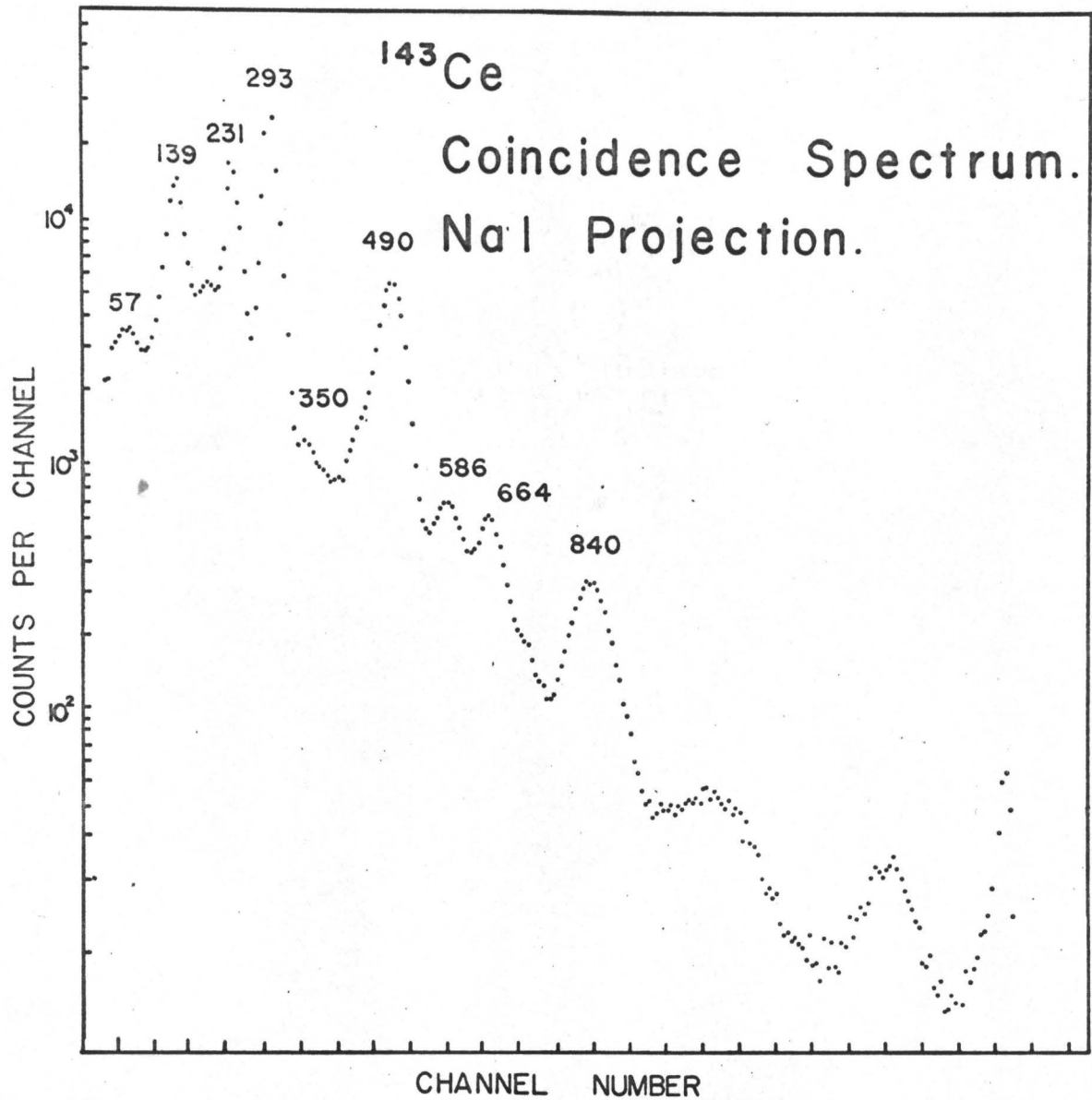
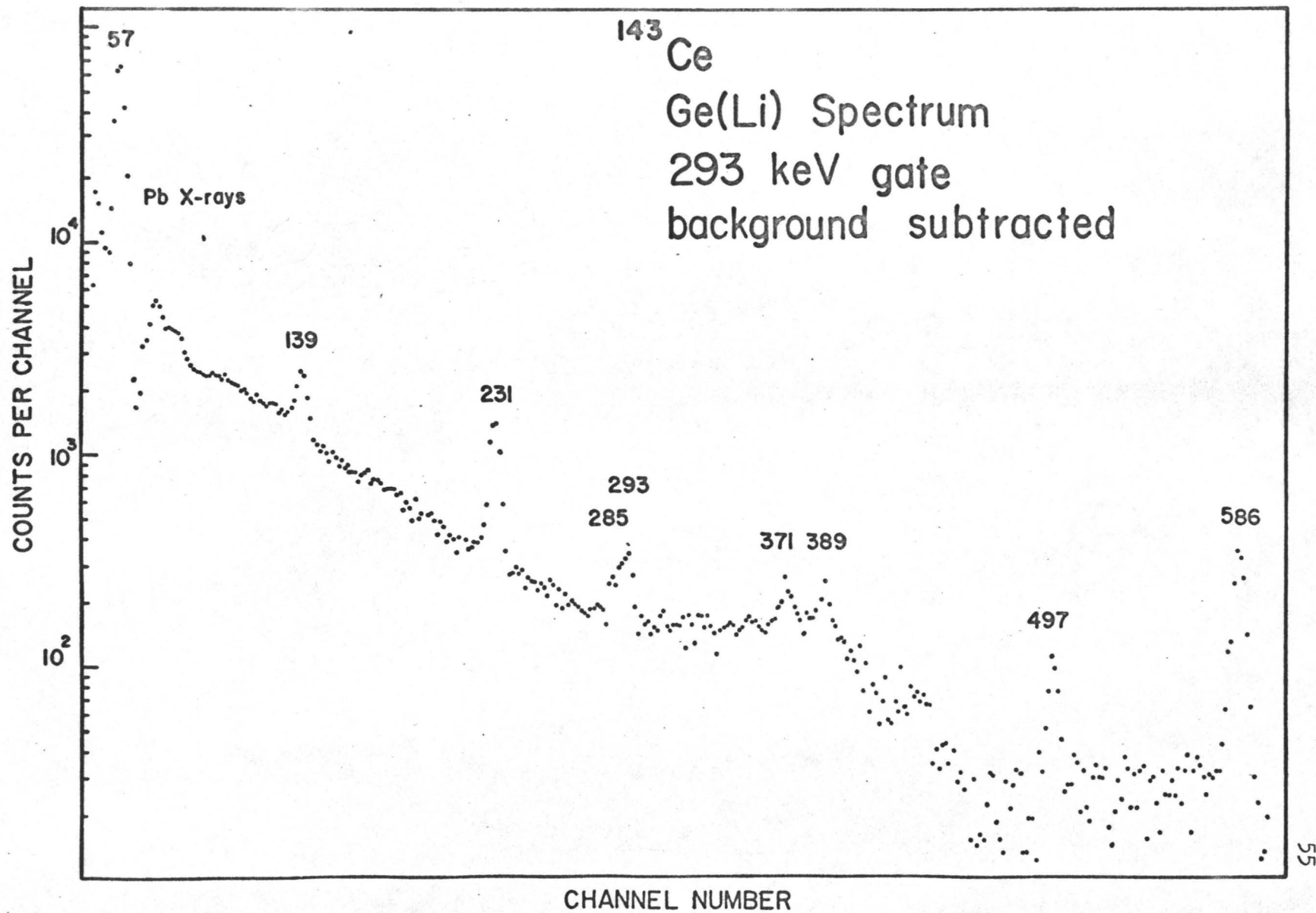


FIGURE 16.

channels along either axis. In this way, "gates" at any energy of any width could be chosen. The spectra shown are all labeled with the median energy of the gate. For example, in the case of the Ge(Li) spectrum in coincidence with 293 keV, shown in figure 17, the window was 30 keV wide and the background was subtracted. That is, spectra in coincidence with channels adjacent to the peak are summed, and then subtracted. In many cases, it proved very difficult to define a background in this way, the result was very often that in parts of the resulting spectrum too much was subtracted. In these cases several spectra with adjacent windows were plotted and the changes noted.

In figure 17 we see peaks at energies 57, 139, 371, 497 and 586 keV with the correct intensities to be in direct coincidence with the 293 keV radiation. The 231 keV peak is due to a rather strong (5.2%) gamma ray, which is indirectly coupled to the 293 keV radiation via the 139 keV transition, and has the expected intensity for this mode of decay. In addition, there are peaks at 285 and 389 keV which have the correct intensities to represent direct feeds to the 350 keV level. It is, however, impossible to decide whether they each feed this level directly or via a cascade. A series of narrow windows in the 285 - 293 keV region on both the Ge(Li) and the NaI projections, indicated that the 389.3 keV transition is in coincidence only with the 293.3 keV radiation. This, and the fact that no cross-over transition is observed, leads to the conclusion that the 285.6 keV and 389.3 keV transitions are separately in coincidence with

FIGURE 17.



293.3 keV, giving new levels at 636.0 and 739.8 keV. No other observed transitions fit to these levels. In view of the weakness of these arguments these levels have not been included in the decay scheme.

Figure 18 shows the reverse experiment, a NaI spectrum with a gate at 293 keV, which gives essentially the same information as figure 17. In this case, the 285 keV peak and the 293 keV chance are not resolved nor are the 371-389 keV peaks. There can also be seen evidence for the 809.4-293.3 keV and the 1031.2-293.3 keV cascades.

The 490 keV NaI gate is shown in figure 19. This Ge(Li) spectrum shows the 231 and 446 keV peaks with the intensity expected if they are assumed to feed the 490 keV level. There is also a peak at 293 keV which arises because of 497 keV photons in the gate. If the gate is widened to include 9 channels (54 keV) on the 490 keV peak in the NaI projection spectrum of figure 16, then peaks can be seen which clearly define the 231-490, 446-490, 293-497, 350-497 and 490-446 keV cascades. This is shown in figure 20. There are also seen, in this spectrum, very weak and poorly defined peaks at 330, 398, 569 and 607 keV of intensities too low to be detected in the singles spectra (i.e. 0.02%). With the presently available data, however, nothing more can be said about these. The NaI spectrum in coincidence with the 490 keV gate in the germanium side (not shown) leads one to consistent conclusions.

The Ge(Li) spectrum with a gate at 231 keV which is 24 keV wide and has the background subtracted, is shown in figure 21. Clear

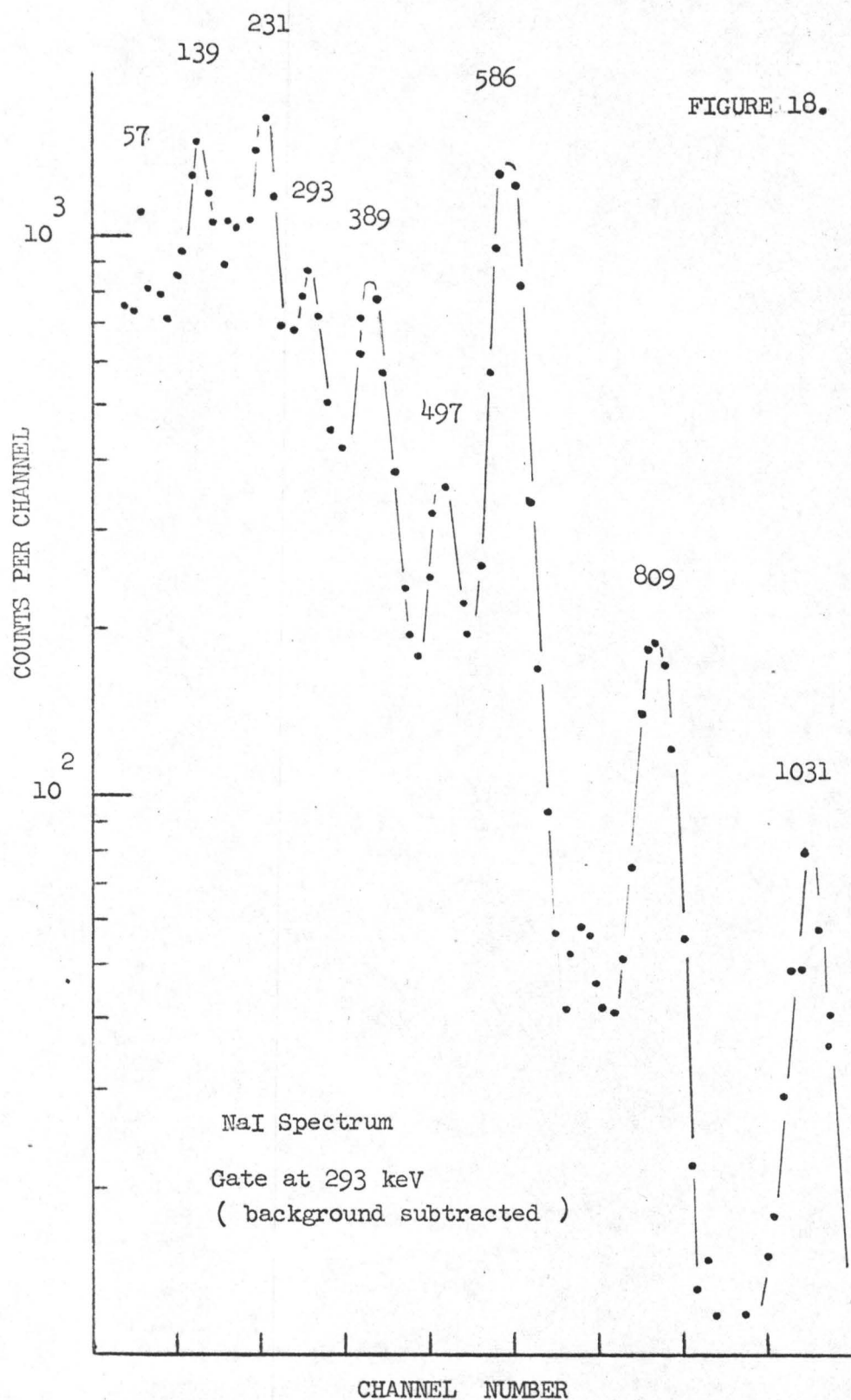
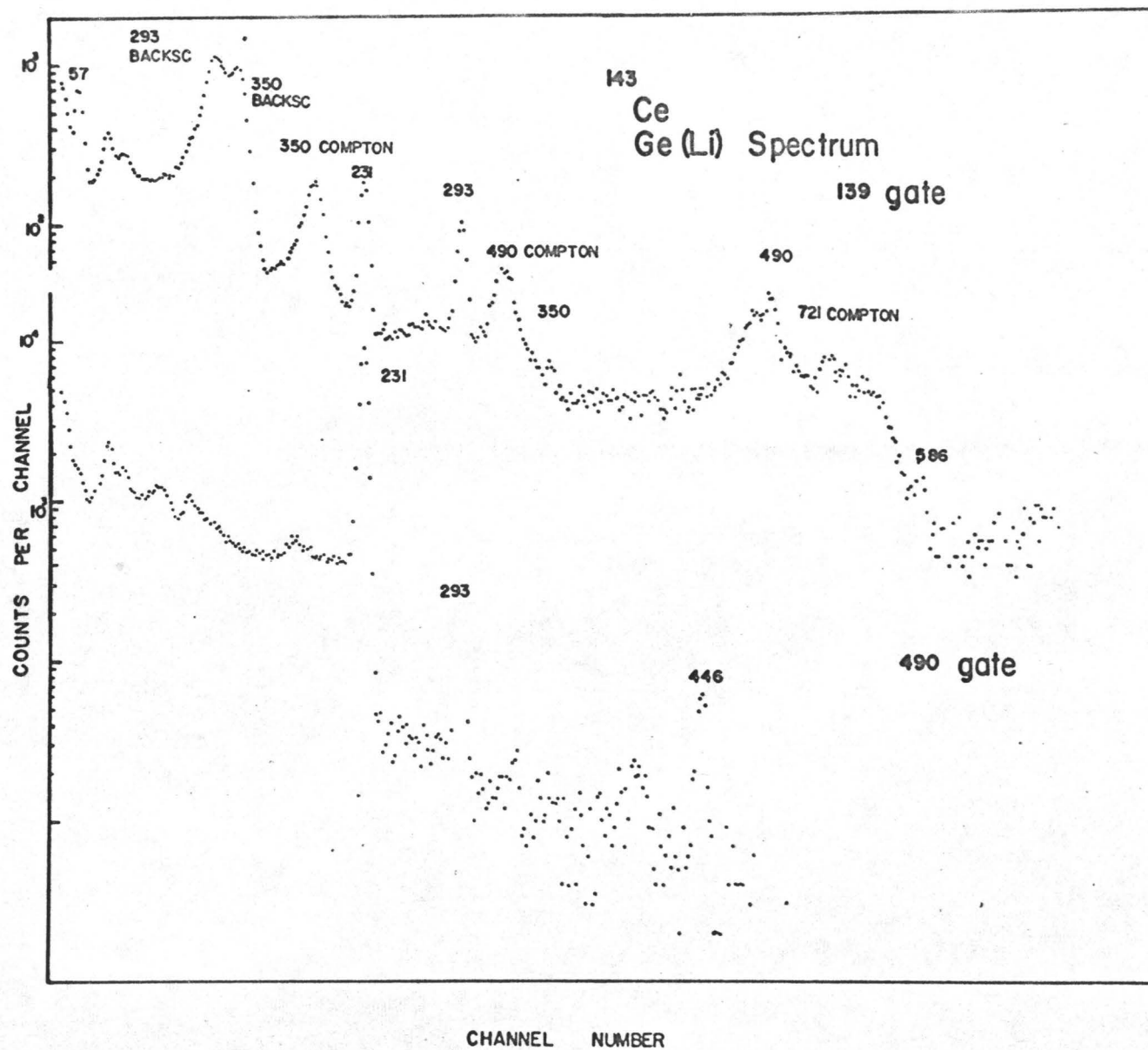


FIGURE 19.





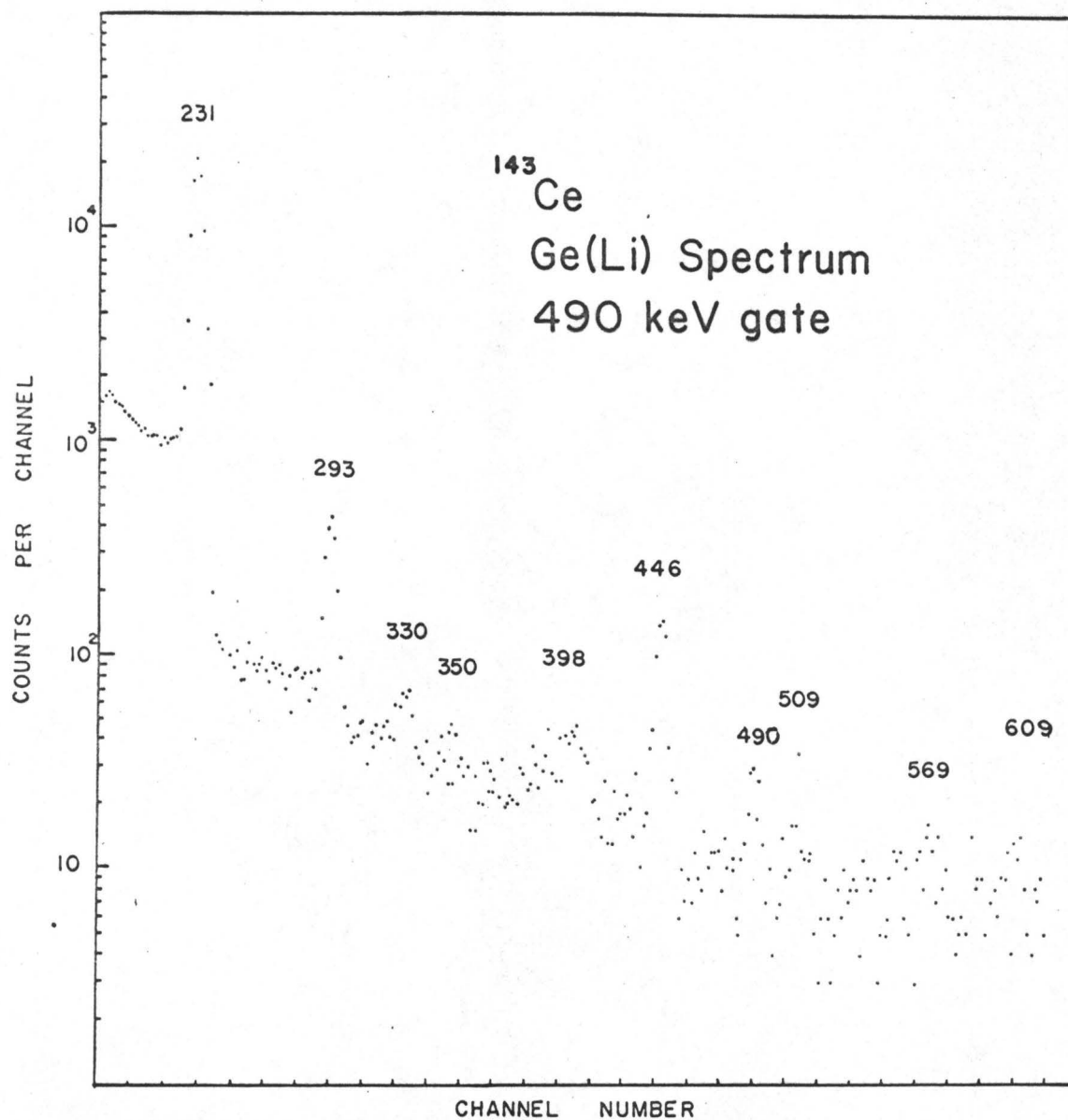
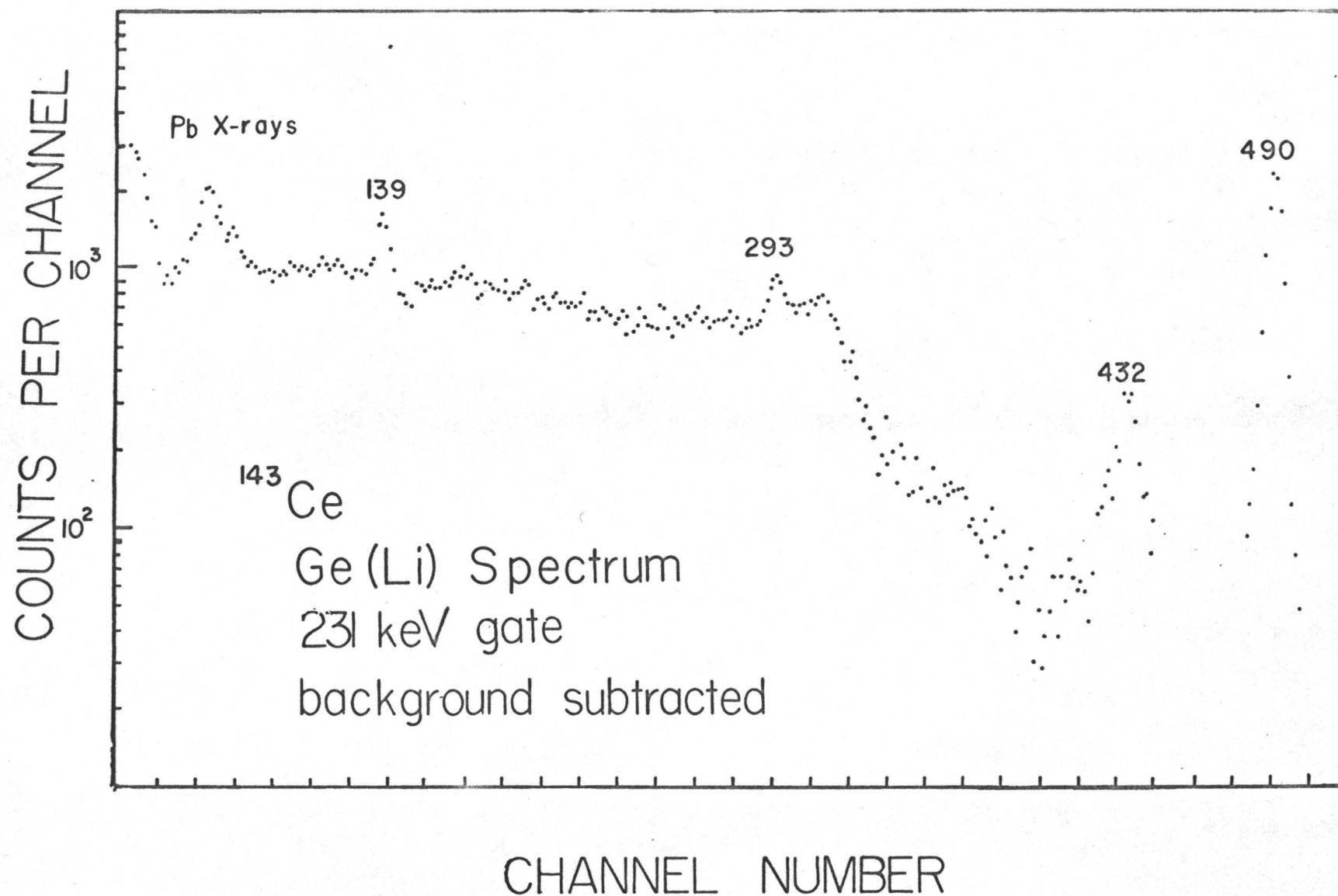


FIGURE 20.

FIGURE 21.



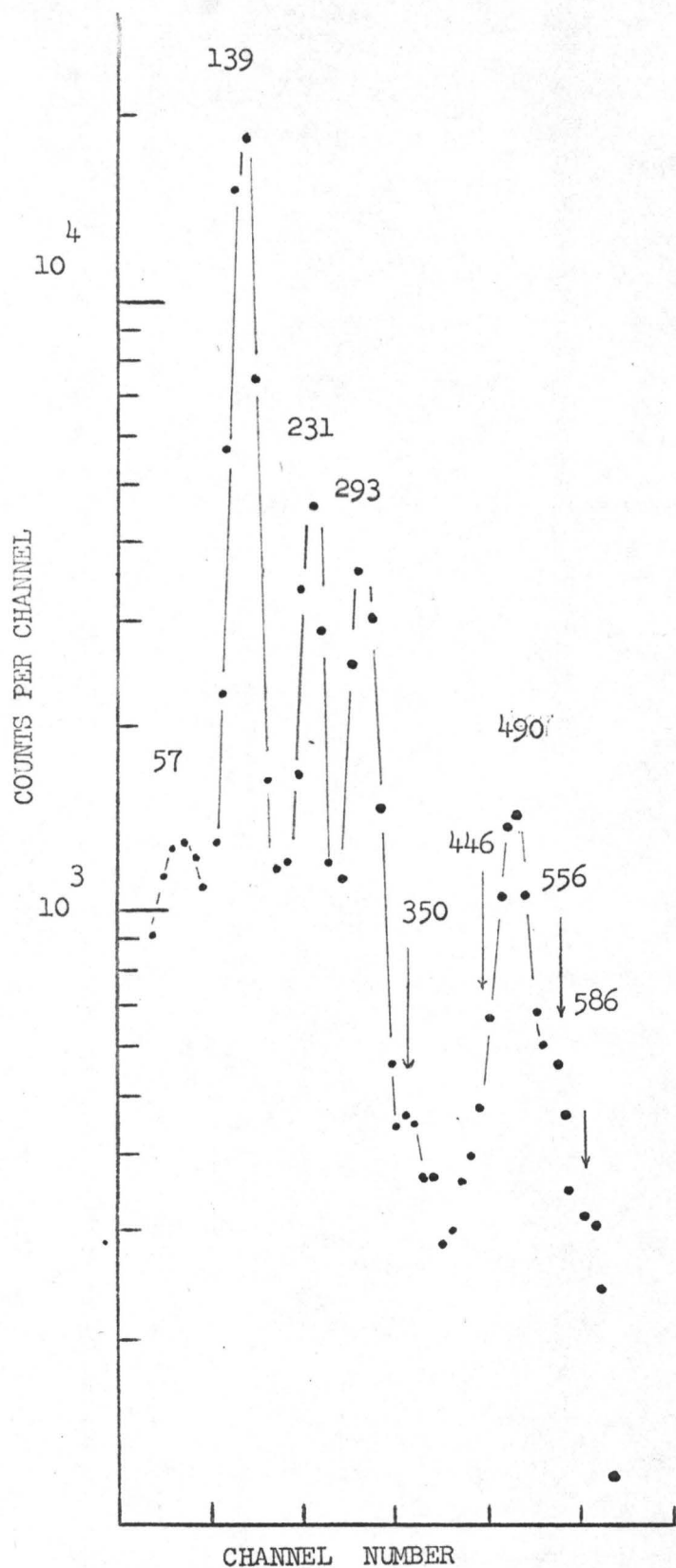
evidence is seen here of the 231-139, 231-432 and 231-490 keV cascades and the 231-139-293 triple cascade.

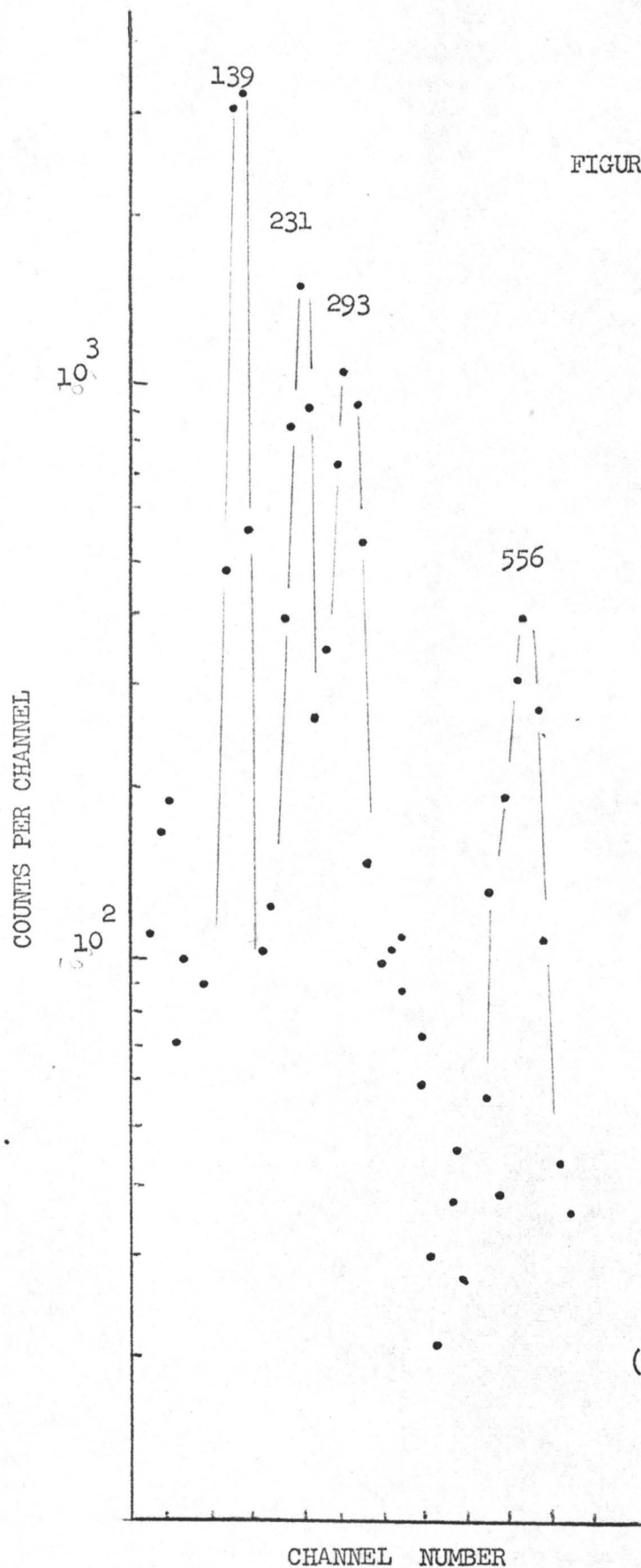
Figure 19 shows the Ge(Li) spectrum and figures 22 and 23, the NaI spectra, all with a gate at 139 keV. In figures 21 and 23 the background is unsubtracted and subtracted respectively. The 139 keV peak is complicated by the fact that the backscatter peaks of the 231, 293, 350, 490, 664 and 771 keV strong gamma transitions lie in this energy region, and most of the recorded events are self coincidences. In figure 19, the spectrum gated by NaI pulses between 120 and 160 keV, and there is a very strong complex at 140 keV due to coincidences between the 293 keV Compton and backscatter peaks, and prominent 350, 490, 664, and 720 keV Compton peaks. The only true peaks showing are those arising from 139-293, 231-139 and 293-139 keV cascades.

The same information is revealed in the reverse experiment with a gate set on the 139 keV Ge(Li) peak. The spectrum with background subtracted, which is shown in figure 23, seems to indicate a coincidence with a strong 556 keV gamma ray. A conclusion that there is a 139-556 keV cascade is contradicted by the absence of any evidence for a 490-556 keV cascade and in any case, the 556 keV transition is not nearly as strong as figure 23 indicates. Hence, this result is believed to be due to difficulties in background subtraction and is a good example of the sort of problem involved in dealing with true Ge(Li) peaks in the 130-160 keV region in the coincidence geometry used.

The NaI spectrum associated with a 57 keV Ge(Li) gate is shown in figure 24. This spectrum shows prominent peaks at 293, 664, 880, 1102

FIGURE 22.





NaI Spectrum  
(Gate at 139 keV  
(background subtracted))

and 1324 keV which are associated with the strong gamma rays feeding the 57.37 keV level. The relative peak heights, when corrected for the NaI detector efficiencies, are in rough agreement with the relative intensities as seen from the singles spectrum. There remains the question of the peaks at 122 and 490 keV which are observed in this spectrum. These present rather a problem since the underlying background in the Ge(Li) spectrum is only 10% of the peak, and if they were gamma rays directly feeding the 57 keV, they would have intensities of about 3%. When a background subtraction is made (not shown), the 122 keV peak is greatly reduced, and the 490 keV peak is found to be very much over subtracted. A closer examination reveals that the 490 keV transition is apparently in coincidence with something at about 30 keV. It is therefore suggested that the 103 keV transition, which is observed in the singles but not placed in the decay scheme, directly feeds the 490.2 keV level. Then the coincidences between the 490 keV transition and the Compton of the 103 keV radiation could be responsible for the 490 keV peak seen in figure 24. It proved impossible, however, to substantiate this. The 103.2 keV radiation is rather weak (0.06%) and lies in the region dominated by backscatter lines, especially that of the strong 231 keV radiation. In view of these difficulties the proposed new level at 593.4 keV is not included in the decay scheme.

Figure 25 shows the NaI spectrum in coincidence with a gate set on the 122 keV Ge(Li) photopeak, with no background subtracted. This spectrum has a strong 840 keV peak which corresponds to the 840 keV



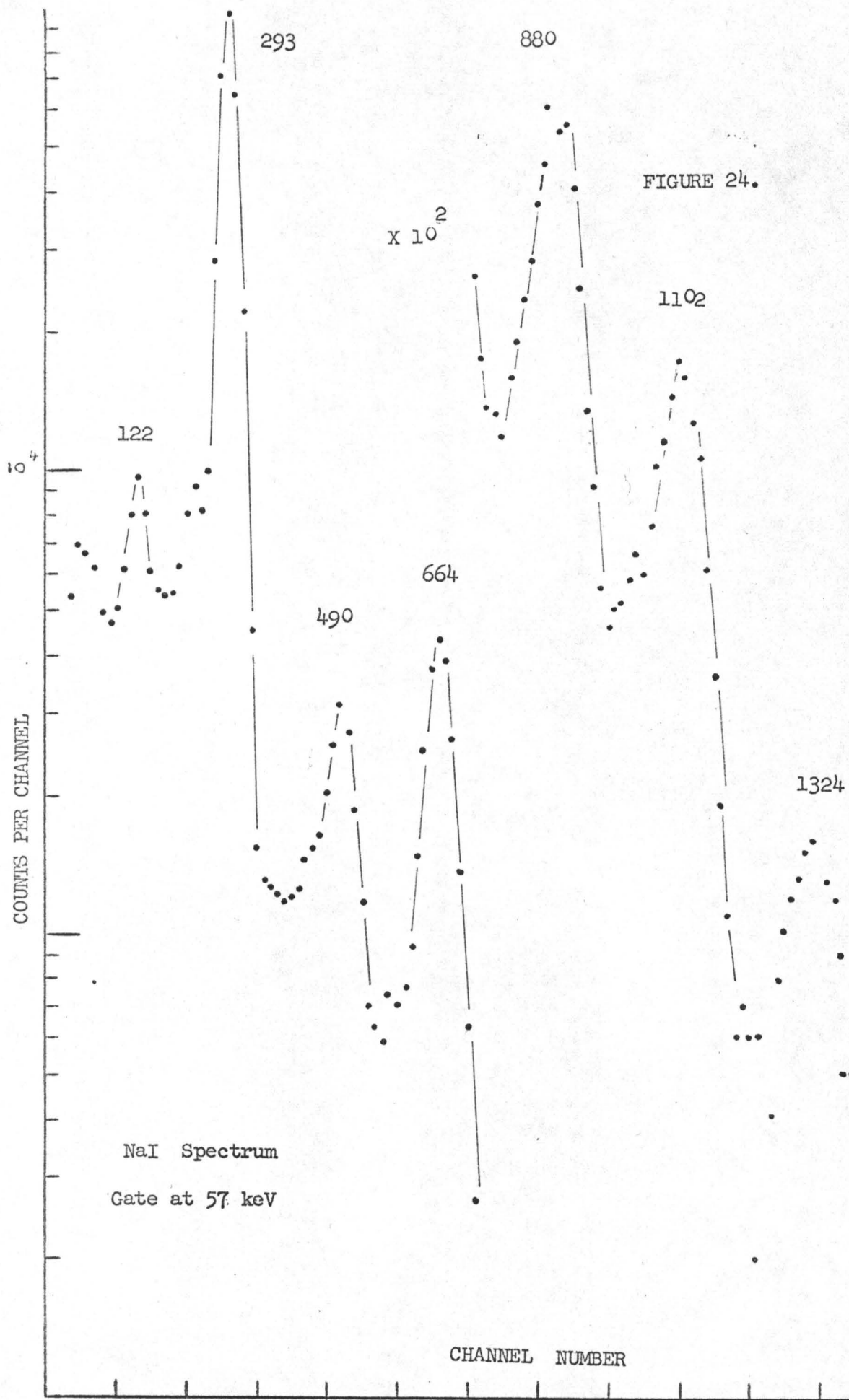
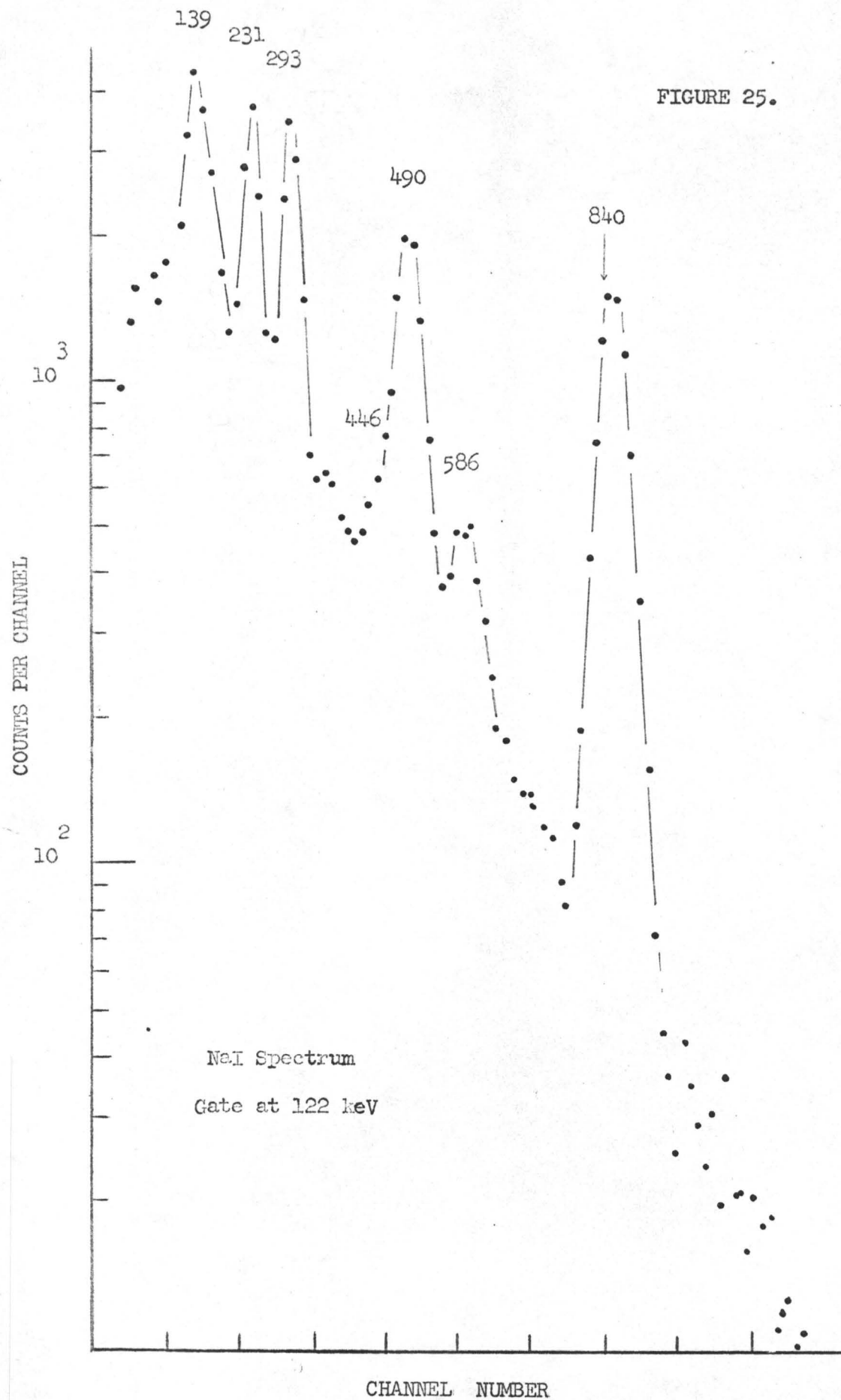
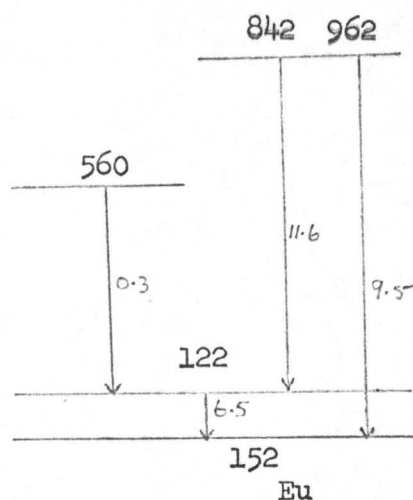




FIGURE 25.



gamma ray in  $^{152}\text{Eu}$ . The relevant portion of the decay scheme of the



9.3 hour component in the decay of  $^{152}\text{Eu}$  is shown in the inset. From the intensity of the 840 keV  $^{152}\text{Eu}$  impurity line, as measured in the singles run, and from the relative intensities given in the inset, the coincidence probabilities for the 842-122 keV and 560-122 keV cascades are found

to be 0.22 and 0.006% respectively. Thus the 840 keV peak in figure 25 corresponds in intensity to that expected for a 840-122 keV cascade of intensity 0.22%. The 560-122 keV cascade would yield a 560 keV photopeak of intensity about 3% of that at 840 keV (i.e. 50 counts). This peak would fall in the valley between the 586 and the 490 keV peaks. The 586 keV peak cannot be associated with the 122 keV transition in  $^{152}\text{Eu}$ , and spectra in coincidence with adjacent windows show that the 586 keV peak is to be associated with a gate at 122 keV. This is therefore believed to be evidence for a 586-122 keV cascade in  $^{147}\text{Ce}$ . The reverse experiment with a NaI gate set on the 586 keV peak shows that the relative intensities of the 293-586 keV and 122-586 keV cascades is 18:1, hence the intensity of the 586-122 keV cascade is about 0.05%. The intensity of the 122 keV cascade is therefore found to be approximately 0.01%. From a comparison of the intensities of the 122 keV

peaks seen in Ge(Li) spectra with gates at 840 and 880 keV the result is obtained that the 122 keV contribution due to  $^{143}\text{Ce}$  is 0.04%. The gates were 12 keV wide and therefore they should not overlap, and in this energy region the only other photons entering these gates will be just from the Compton of the 1102 which will not affect the 122 keV peak. Because of the difficulties associated with obtaining convincing evidence for a 122 keV transition in the  $^{143}\text{Ce}$  decay, it is shown as a dotted line in the decay scheme. The 0.04 indicates the maximum possible intensity.

A further examination of the coincidence data was made to look for transitions, defining new levels, in particular the Ge(Li) spectrum with a gate covering the 664 and 721 keV peaks was examined. No new coincidences were observed. The 212.7 radiation is sufficiently strong (0.3%) that it would have been seen if it fed any of the levels up to the 937.3 keV level, from intensity considerations it could not feed any of the levels above 937.3 keV. The conclusion is that it is a ground state transition. This is strengthened by the correct energy fit of a 277.6 - 212.7 keV cascade from the 490.2 keV level. A NaI gate at 277 keV indicated a peak at 212 keV of about the right intensity.

### 6.3 The Internal Conversion Measurement

The energy calibration of the system with the  $1\text{cm}^2$  Si(Li) detector was effected by means of a run with a  $^{207}\text{Bi}$  source. The spectrum of this electron capturing nucleus shows low energy auger lines and the K and L internal conversion lines of the 569.6 and 1063 keV transitions.

To get the energies of the observed peaks the values of the binding energies of  $\text{Pb}_{82}$  have to be subtracted. A  $^{143}\text{Ce}$  source yielded the spectrum shown in figure 26. The energies and intensities of the peaks observed are presented in table II. The K subshell conversion coefficient for the 231.6 keV transition was then obtained by normalizing to the value for the 293.3 keV transition. This value is obtained from the missing ratio as measured by Gellently<sup>(18)</sup> et. al., and from the calculated conversion coefficients of Sliv and Band.

| Conversion Coefficient   | Missing Ratio |
|--------------------------|---------------|
| M1 $5.75 \times 10^{-2}$ | $63 \pm 4\%$  |
| E2 $4.30 \times 10^{-2}$ | $37 \pm 4\%$  |

Then for the 293.3 keV transition

$$\alpha_K = \frac{5.75 \times 10^{-2} \times 63 + 4.30 \times 10^{-2} \times 37}{63 + 37} = 5.21 \times 10^{-2}$$

and hence the K subshell internal conversion coefficient for the 231 keV transition is given by

$$0.0521 = \frac{5.2}{10.1} \times \alpha_K$$

$$\alpha_K = 0.10 \pm 0.02$$

This then gives a multipolarity mixing of  $54 \pm 40\%$  M1.

#### 6.4 The Decay Scheme

The results discussed above are incorporated in the decay scheme shown in figure 27. The energy of each level in the decay scheme was

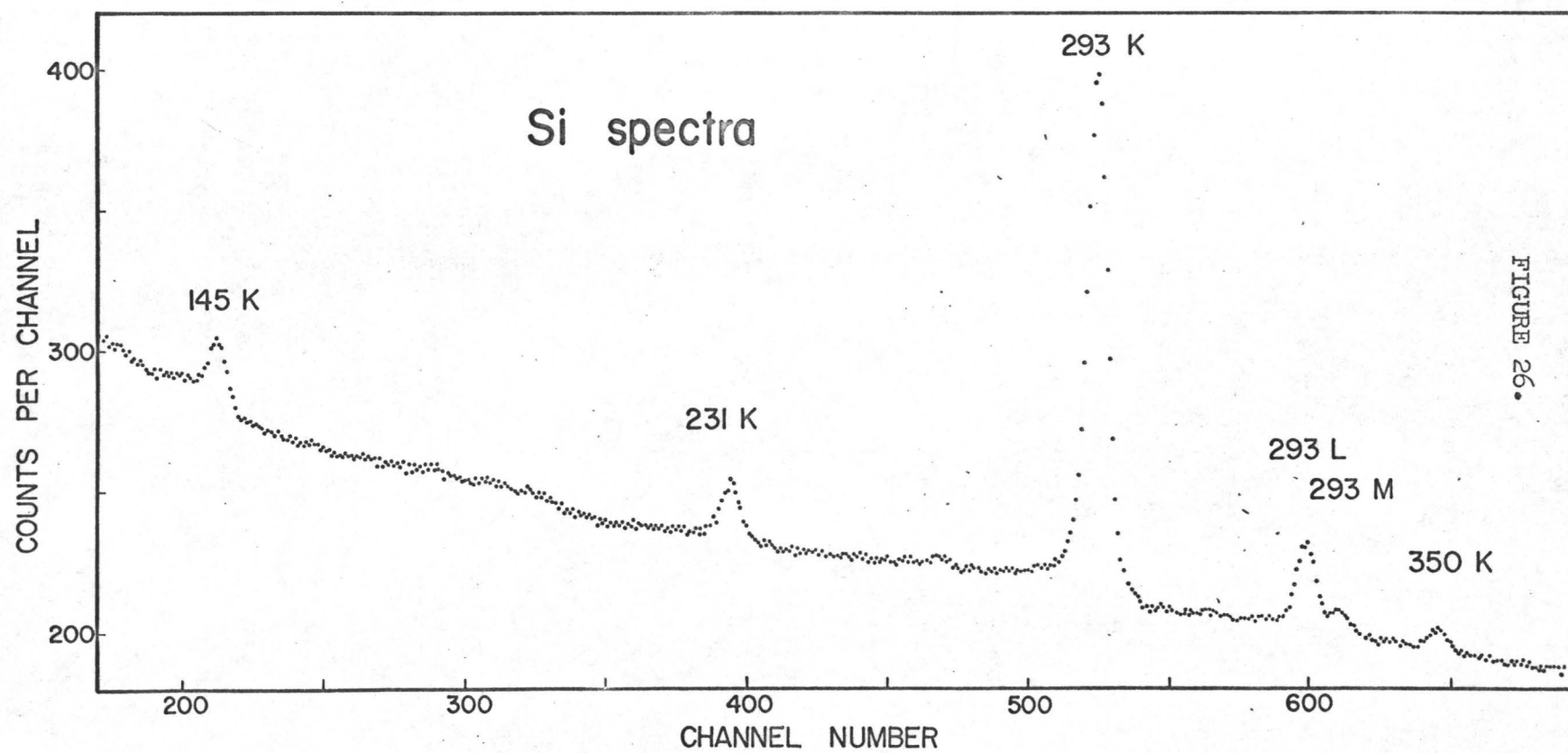


FIGURE 26.

TABLE II  
Lines in the Internal Conversion Spectrum

| Electron Energy<br>keV | Relative<br>Electron<br>Intensities | Origin of<br>Conversion<br>Line | Relative<br>photon<br>Intensities |
|------------------------|-------------------------------------|---------------------------------|-----------------------------------|
| 189.6                  | 10.1                                | K of 231 keV peak               | 5.2                               |
| 251.3                  | 100                                 | K of 293 keV peak               | 100                               |
| 285.0                  | 12.2                                | L of 293 keV peak               | 100                               |
| 308.3                  | 3.4                                 | K of 350 keV peak               | 6.9                               |



determined by a weighted average of the transition energies involved in the various paths of de-excitation to the ground state. The energies of the transition are given in keV and the intensities relative to 100 for the strong 293.3 keV transition. The location of many of the stronger lines in the decay scheme has been based on coincidence data; for transitions located in this fashion, a dot is placed on the lower level involved.

All the classified transitions are shown in table III which also compares the present work with the work of Gopinathan et. al. The first four columns present their results, the fifth and sixth give the present measurements, the seventh indicates the basis of classification, and the next shows the location of each line in the scheme. The last column shows the difference between the separation of the levels involved in the transition and the actual energy measurement of the transition.

The present energy measurements are considerably more precise than those of the earlier workers who were using NaI detectors and the coincidence conditions are more rigorously applied. In the main, the present results confirm the previous decay schemes. The 220 keV line listed by Gopinathan is not present; it could conceivably have arisen from pile up or back scatter in the NaI detectors. A number of new transitions, notably those of energy 122.1, 212.7, 277.6, 497.7, 790.1, 1002.5 and 1060.0 keV have been placed in the decay scheme. Three new levels at 212.6, 867.9 and 1059.8 keV have been established on energy fit involving 2, 2 and 3 lines respectively, and three other levels at



TABLE III  
<sup>143</sup>  
 GAMMA RAYS IN THE DECAY OF Ce

| Gopinathan et al Mancuso et al |           |                 |           | Present Work    |                 |           |                  |                             |
|--------------------------------|-----------|-----------------|-----------|-----------------|-----------------|-----------|------------------|-----------------------------|
| Energy<br>(keV)                | Intensity | Energy<br>(keV) | Intensity | Energy<br>(keV) | assign-<br>ment | Intensity | Classification   | E calculated-<br>E observed |
| 57                             | 25        | 57              | 23        | 57.37           |                 | 20.4      | 57.37 → 0        | 0                           |
| 142                            | 1.1       | 143             | 6         | 122.1           | γ-γ             | 0.08      | 1059.87 → 937.31 | +0.5                        |
|                                |           |                 |           | 139.5           | γ-γ             | 0.2       | 490.23 → 350.62  | +0.1                        |
|                                |           |                 |           | 212.7           |                 | 0.36      | 212.65 → 0       | 0                           |
| 220                            | 7         | ---             | ---       | ---             | ---             | ---       | ---              | ---                         |
| 232                            | 7         | 232             | 4         | 231.6           | γ-γ             | 5.2       | 721.78 → 490.23  | - 0.1                       |
|                                |           |                 |           | 277.6           | γ-γ             | 0.002     | 490.23 → 212.65  | 0                           |
| 293                            | 100       | 293             | 100       | 293             | γ-γ             | 100       | 350.62 → 57.37   | 0                           |
| 351                            | 9         | 350             | 9         | 350.5           | γ-γ             | 6.9       | 350.62 → 0       | +0.1                        |
| 374                            | 0.9       | 375             | 0.4       | 371.5           | γ-γ             | 0.03      | 721.78 → 350.62  | -0.3                        |
| 436                            |           |                 |           | 433.0           | γ-γ             | 0.3       | 490.23 → 57.37   | -0.1                        |
| 447                            |           |                 |           |                 |                 |           |                  |                             |
| 453                            | 0.8       | 452             |           | 447.0           | γ-γ             | 0.15      | 937.31 → 490.23  | -0.1                        |
| 493                            | 5.3       | 493             | 4         | 490.2           | γ-γ             | 4.3       | 490.23 → 0       | 0                           |
|                                |           |                 |           | 497.7           | γ-γ             | 0.2       | 847.86 → 350.62  | -0.4                        |

TABLE III (continued)

| Gopinathan et al. Mancuso et al. |           |        |           | Present Work |                     |           |                              |                             |
|----------------------------------|-----------|--------|-----------|--------------|---------------------|-----------|------------------------------|-----------------------------|
| Energy                           | Intensity | Energy | Intensity | Energy       | assignment          | Intensity | Classification               | E calculated-<br>E observed |
| 591                              | 3         | 595    | 2.2       | 586.9        | $\gamma$ - $\gamma$ | 1.0       | $937.31 \rightarrow 350.62$  | -0.1                        |
| 668                              | 15.1      | 668    | 13        | 664.4        | $\gamma$ - $\gamma$ | 12.2      | $721.78 \rightarrow 57.37$   | 0                           |
| 725                              | 17.1      | 725    | 12        | 721.8        | $\gamma$ - $\gamma$ | 10.1      | $721.78 \rightarrow 0$       | 0                           |
|                                  |           |        |           | 790.1        |                     | 0.036     | $847.86 \rightarrow 57.37$   | +0.4                        |
| 810                              | 0.7       | 817    | 0.7       | 809.4        | $\gamma$ - $\gamma$ | 0.06      | $1160.14 \rightarrow 350.62$ | +0.1                        |
| 885                              | 3         | 888    | 2         | 880.0        | $\gamma$ - $\gamma$ | 2.1       | $937.31 \rightarrow 57.37$   | -0.1                        |
| 942                              | 0.5       | 945    | 0.4       | 937.2        |                     | 0.05      | $937.31 \rightarrow 0$       | +0.2                        |
|                                  |           |        |           | 1002.5       |                     | 0.1       | $1059.84 \rightarrow 57.37$  | 0                           |
| 1042                             | 0.4       | 1045   | 0.4       | 1031.2       | $\gamma$ - $\gamma$ | 0.03      | $1381.61 \rightarrow 350.62$ | -0.2                        |
|                                  |           |        |           | 1060.0       | $\gamma$ - $\gamma$ | 0.06      | $1059.84 \rightarrow 0$      | -0.2                        |
| 1100                             | 1.4       | 1110   | 0.8       | 1102.9       | $\gamma$ - $\gamma$ | 0.5       | $1160.33 \rightarrow 57.37$  | 0                           |
| 1340                             | 0.1       | ---    | ---       | 1324.1       | $\gamma$ - $\gamma$ | 0.02      | $1381.61 \rightarrow 57.37$  | -0.1                        |

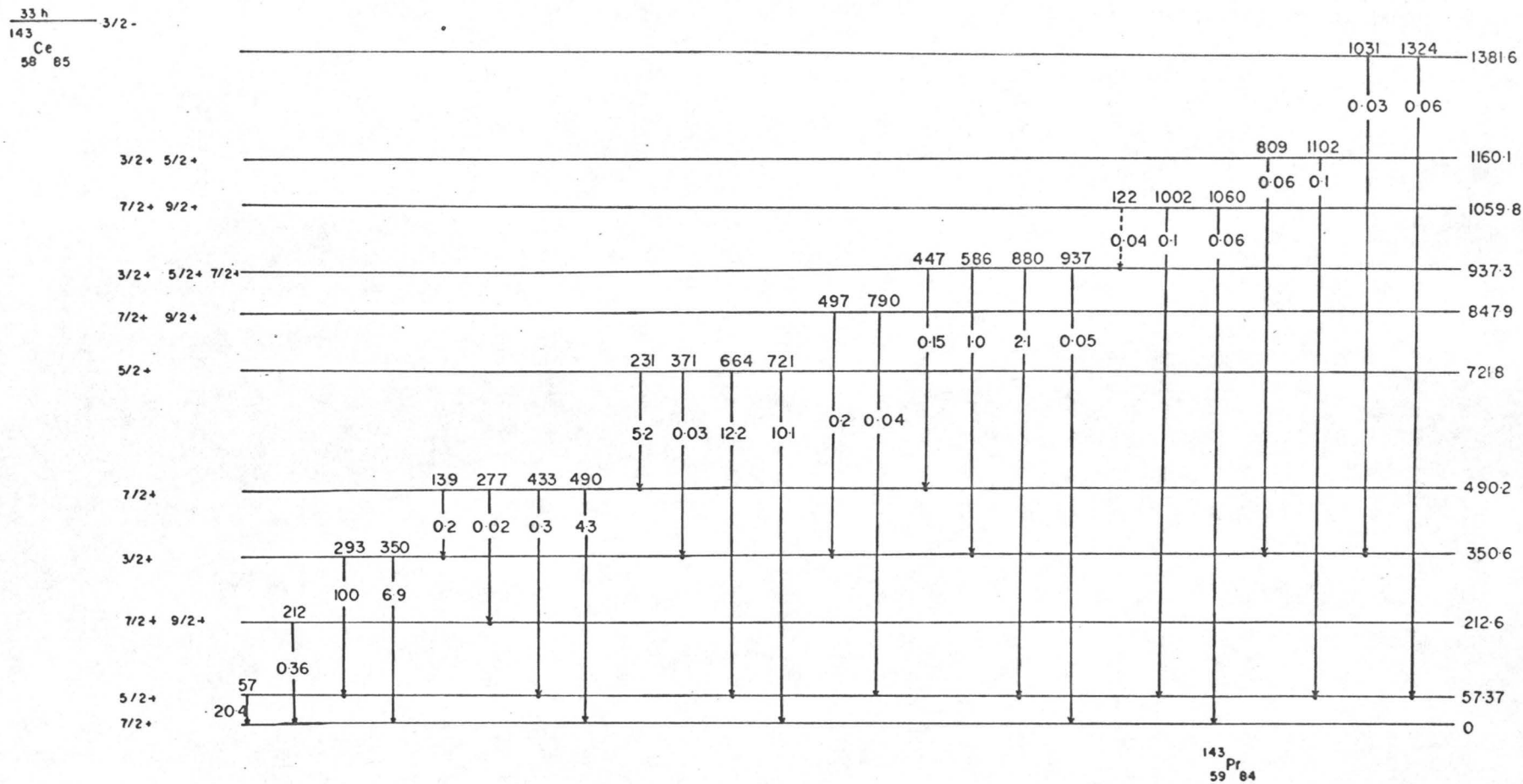


FIGURE 27: The Decay Scheme of  $^{143}_{58}\text{Ce}$ .

593.4, 636.2 and 739.9 keV are tentatively proposed on the basis of the coincidence data but are not included in the decay scheme on account of the weakness of the arguments.

There remain a large number of lines; 72.7, 254.6, 556.5, 809.9, 815.3, 1046.6, 1340.2, 1461.0, 1498.8 keV, which have not been placed. They all have intensity 0.08 or less, but nevertheless it should be possible to locate the stronger of them by further coincidence experiments with the existing equipment. The last two are probably due to impurities since the decay energy is only about 1400 keV. It is suggested that a coincidence experiment with a  $60^\circ$  geometry and a wedge shaped Compton shield, and a beta-gamma coincidence experiment would be valuable.

#### 6.5 The Intensity Balance

Table IV shows the result of the intensity balance, that is, it shows the difference between the transition intensity de-exciting the level and that feeding it. The number of transitions per 100 decays was found by normalizing to the total feed to the ground state. The beta feed to the ground state being assumed to be zero. It was thus possible to find the beta intensity feeding each level, from this could be found the partial half lifes of the beta transitions and hence, using the nomogram of Moszkowski<sup>(25)</sup> the approximate  $\log ft_{1/2}$  values for each beta transition could be determined.

#### 6.6 The Spins and Parities of Levels in $^{143}\text{Pr}$

The measurements of  $3/2$  for the ground state spin of  $^{143}\text{Ce}$  by Maleh,  $7/2$  for the ground state of  $^{143}\text{Pr}$  by Burdick et al and

TABLE IV

Intensity Balance for the Levels in  $^{143}\text{Ce}$ 

| Level Energy<br>keV | $\beta$ feed intensity<br>/100 decays | $\log ft_{1/2}$ |
|---------------------|---------------------------------------|-----------------|
| 1381.6              | 0.04 $\pm$ 0.03                       | -----           |
| 1160.1              | 0.25 $\pm$ 0.05                       | 7.6 $\pm$ 0.4   |
| 1059.8              | 0.19 $\pm$ 0.04                       | 9.0 $\pm$ 0.4   |
| 1046.7              | 0.05 $\pm$ 0.01                       | -----           |
| 937.3               | 1.7 $\pm$ 0.3                         | 7.9 $\pm$ 0.3   |
| 847.9               | 0.13 $\pm$ 0.03                       | 9.3 $\pm$ 0.4   |
| 721.9               | 15.5 $\pm$ 1.3                        | 7.8 $\pm$ 0.3   |
| 490.2               | -0.5 $\pm$ 0.5                        | -----           |
| 350.6               | 60.8 $\pm$ 5.6                        | 7.6 $\pm$ 0.3   |
| 57.37               | 22.74 $\pm$ 7.1                       | 8.6 $\pm$ 0.3   |
| TOTAL 100.9         |                                       |                 |



5/2 for the first excited state of  $^{143}\text{Pr}$  by Graham et al provide a firm basis for the further assignment of spins and parities. Gopnathan<sup>(26)</sup> assigns a spin of 3/2 to the 350 keV level and this assignment is strengthened by the  $\log ft_{1/2}$  value to this level. The  $\log ft$  values of beta transitions from  $^{143}\text{Ce}$  to most of the levels of  $^{143}\text{Pr}$  indicate that they are of the first forbidden type showing that these levels have even parity.

From the 664-57 keV directional correlation Mancuso et al assign a 5/2 spin to the 721 keV level and hence from the 231 - 490 keV directional correlation they give 5/2 or 7/2 for the spin of the 490 keV level. The absence of beta feed to this level implies that 7/2 is the correct spin assignment.

The absence of beta feed to the 213 keV level and the relative intensities of the transitions feeding it, lead to a tentative assignment of 7/2+ to this level.

The spin assignments to the higher levels were made on the basis of the  $\log ft_{1/2}$  values given in Table IV, and on the relative intensities of gamma transitions. The arguments are weak and it is here, especially, that the need is felt for further experiments. It is very difficult to make an assignment for the 1381.6 keV level, the  $\log ft_{1/2}$  value to this level is  $\sim 10$  which implies a spin of 9/2, 11/2 ; however, the two transitions de-exciting this level feed the 350 and the 57 keV levels which both have low spins, i.e. 3/2 and 5/2.

#### 6.7 The Interpretation of the Levels of $^{143}\text{Pr}$

No firm model-dependent predictions of the levels in this

decay have yet been successfully made. However, it is quite easy to make a general description of the nature of the lower lying energy levels. The shell model predicts a  $5/2_+$  ground state and a  $7/2_+$  first excited state as is observed in  $^{141}\text{Pr}$ . The experimental results show that these states have crossed in  $^{143}\text{Pr}$ , the cause being ascribed to the increasing quadrupole force.  $^{143}\text{Pr}$  has two neutrons outside the closed shell of 82 and nine protons outside the closed shell of 50 giving single particle states  $g_{7/2}$  and  $d_{5/2}$  available to the odd proton. It can then be expected that this odd proton will couple to the quadrupole vibrations of the even even  $^{142}\text{Ce}$  core. Hence the higher excited states may be interpreted as collective in nature with expected spins ranging from  $1/2$  to  $11/2$ . As was mentioned earlier Choudhury and Kujawski have applied the intermediate coupling approach to this nuclide. As they note in their paper one can assume that the odd proton having available both the  $1g_{7/2}$  and  $2d_{5/2}$  states is neither weakly nor strongly coupled to the collective surface vibrations of the even even  $^{142}\text{Ce}$  core. Their calculation predicts correctly the multipolarity and the half life of the 57 keV transition. However, they do not predict levels anywhere in the neighbourhood of 212, 350, 937, 1046, 1059, or 1160 keV. It can thus be said that the results of the present work do not improve the agreement with the theoretical results.



## SUMMARY

The gamma ray transitions emitted following the decay of  $^{143}\text{Ce}$  have been studied by singles and coincidence techniques, using Ge(Li) and NaI detectors. Thirty-eight gamma ray transitions have been associated with the decay of  $^{143}\text{Ce}$ , of which twenty-five have been classified in the proposed decay scheme. Ten excited levels have been established for  $^{143}\text{Pr}$ , of which seven had been postulated by earlier workers. In addition a classification of three further transitions defining new levels is tentatively proposed.

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