

The Gamma Rays of the Decay of
Neptunium-237 Populating
States in Protactinium-233

A Thesis

Submitted to

the

Faculty of Graduate Studies

In Partial Fulfilment

of the Requirement for the Degree

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by

Mark Skalsey

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A dissertation submitted to the Faculty of Graduate Studies of
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To my wife,

Peg

Acknowledgements

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Abstract

The decay of Np-237 was examined using Ge(Li) gamma ray spectrometers. Chemical separation of the contaminating daughter product was effected through anion exchange. Forty-two gamma rays were identified in the singles spectrum of which five are reported here for the first time: 115.5, 140.6, 153.5, 194.7, and 222.5 keV. A new level is proposed at 303.8 keV. Gamma-gamma coincidence measurements were used to study the level diagram of the excited states of Pa-233. The single particle half-life of the 86.51 keV level was calculated and compared to the experimental value. The intensity ratio of two "K-forbidden" transitions was calculated and compared to the experimental value.

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CHAPTER I

Equipment and Methods

1. Ge(Li) Gamma Ray Detector:

The germanium, lithium-drifted, Ge(Li), gamma ray detector is the currently accepted method of obtaining the energies and intensities of gamma rays (see table 1 for the detectors used in this study.) Surpassing the now obsolete sodium iodide, thallium-doped NaI(Tl) detector, Ge(Li) detectors, having very much superior resolution, are able to resolve two gamma rays very close in energy into separate components.¹ A gamma ray impinging on a germanium crystal detector deposits its energy in the crystal in the form of excitation and ionization events at a rate of approximately 2.85 eV per electron-hole pair.¹ These pairs are then collected at the anode and cathode under the influence of a d-c bias across the crystal. The signal is then sent through a charge sensitive preamplifier giving a fast rising voltage pulse. The pulse height is proportional to the number of charges collected and hence the energy

Table 1
Detectors

Ge(Li) DETECTORS USED IN THIS STUDY

Detector	Size	Resolution	Bias
Ortec X-ray #1	80 mm	250 eV @ 5.9 keV 525 eV @ 122 keV	-1000v
Ortec X-ray #2	80 mm	219 eV @ 5.9 keV 590 eV @ 122 keV	-900v
Nuclear Diodes 30cc	30cc	2.14 keV @ 1.33 MeV	+2500v
Ortec 35cc	35cc	3.0 keV @ 1.33 MeV	+1800v
Nuclear Diodes 50cc	50cc	1.95 keV @ 1.33 MeV	+2800v

of the gamma ray.

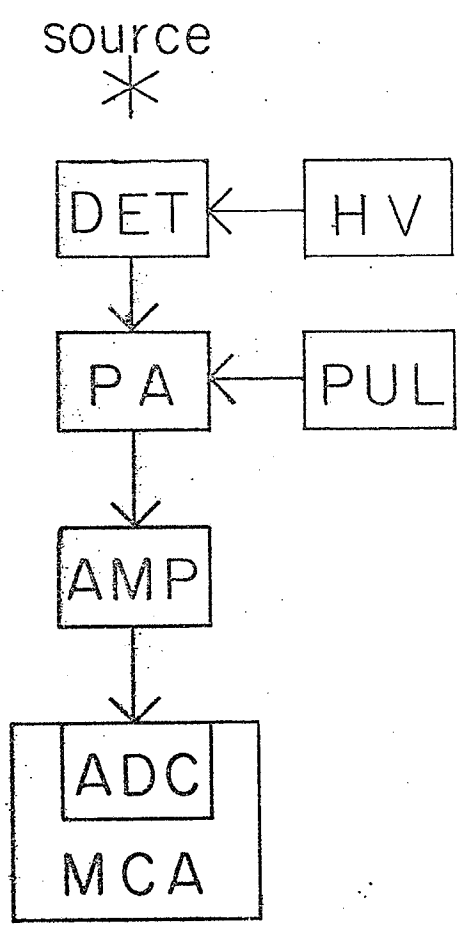
The excitation and ionization mechanisms in the crystal involve various well-known processes viz. 1) the production of electron-positron pairs which occurs at high energies with a threshold of 1.022 MeV 2) compton scattering in the intermediate energy range 3) photo-electric absorption, which predominates below 150 keV.¹ The detector operates as a diode with the germanium crystal lightly doped with gallium to produce a p-type region. Lithium is deposited on one face very heavily giving a strong n-type region. Due to impurities in the germanium the conduction of the crystal is too great to permit the application of a d-c bias across the crystal.² The lithium is drifted into the crystal from the face. Lodging between atoms in the crystalline structure, the lithium reduces the conductivity making the region "intrinsic." The intrinsic region is separate from the p and n-type regions, forming a p-i-n junction diode. At room temperature the lithium atoms have a high mobility in the germanium crystalline structure. To keep the lithium atoms in the crystal, it is maintained at liquid nitrogen temperature (77°K).⁴ For this reason the crystal is mounted on a cold finger immersed in liquid nitrogen and maintained continuously at low

temperature. To reduce the noise of the system the first stage of the charge-sensitive preamplifier is also mounted on the cold finger and maintained at liquid nitrogen temperature.

2. Singles Measurements

To record the gamma ray spectrum of a radioactive source, a Ge(Li) detector is used for what is called "a singles measurement"(see figure 1.) To accomplish this a high voltage d-c bias (H. V.) is established across the detector crystal (DET.) The charge is collected when a gamma ray enters the crystal and is amplified by the charge-sensitive preamplifier (P.A.) The pulser (PUL.) is a device which simulates the detector output. The pulser output is fed into the preamplifier primarily for the purpose of testing the operation of the system. The output of the preamplifier is fed into the main amplifier for further amplification and pulse shaping. At all points throughout this circuit the pulse height is proportional to the incident gamma ray energy. After leaving the main amplifier the pulses are analyzed according to their heights and this information is stored in a computer-type memory, the function of the multi-channel analyzer (MCA.) The analysis is performed by a part of the multi-channel

Figure 1
Singles Measurements



SINGLES MEASUREMENT

analyzer called the analogue to digital converter (ADC). When a pulse of a particular height enters the analogue to digital converter, one count is added to a memory channel, say N. If the pulse would have had a slightly larger (or smaller) height, a count would have been registered in memory channel (N+1) or (N-1). This mode of operation is referred to as "pulse height analysis." The multi-channel analyzer in use in this laboratory is a Nuclear Data Model ND-2200, Nuclear Data, Inc., Palatine, Illinois. This analyzer, used with three analogue to digital converters, has a total memory of 4096 channels with the possibility of storing up to one million counts in each channel. The memory may be used as one 4096 channel unit or two 2048 channel units or three 1024 channel units. The analyzer also is responsible for controlling the various output and display devices.

The resultant two-dimensional array, channel number versus counts per channel, is referred to as a spectrum. To first approximation the channel numbers are taken to be linearly related to the energy, viz.:

$$E = FX + B$$

where E is the energy, X is the channel number and F and B are constants usually called the conversion gain and the digital zero shift, respectively. For a more

accurate determination of energies, the well-known mixed source technique is used to determine the non-linearity of the system. The source under investigation is placed in front of the detector along with several standard calibration sources of known energies (see table 2). A spectrum is then accumulated and analyzed. Two known energy calibration photo peaks are chosen and F and B are calculated from these. All the other known calibration photo peaks positions are combined with the values of F and B to give a set of calculated energies E_{cal} . The difference between the calculated energy and the known energy is the energy deviation from linearity. The energy deviation is plotted against channel number yielding a "non-linearity curve." Taking the positions of the photo peaks of unknown energy and combining with F, B and the energy deviation, an accurate determination of the energies of photo peaks may be obtained.

The intensity of a gamma ray is found by obtaining the area under the photo peak and subtracting the contribution due to background radiation. Of course, the Ge(Li) detectors are not 100% efficient in detecting gamma rays. Assuming the direction of emission of gamma rays is isotropic, the first loss in efficiency is that not all the gamma rays are emitted

Table 2
Energy-Intensity Standards

Calibration Standards

Source	Energy (keV)	Intensity	T $\frac{1}{2}$
Am-241	26.345 $\pm$ 0.001	7.0 $\pm$ 0.6	433 y.
	59.537 $\pm$ 0.001	(100)	
Ba-133	80.998 $\pm$ 0.008	52.6 $\pm$ 1.0	7.2 y.
	276.397 $\pm$ 0.012	11.4 $\pm$ 0.3	
	302.851 $\pm$ 0.015	30.2 $\pm$ 0.6	
	356.005 $\pm$ 0.017	(100)	
	383.851 $\pm$ 0.020	14.4 $\pm$ 0.3	
Co-57	122.061 $\pm$ 0.010	(100)	270 d.
	136.473 $\pm$ 0.015	13.4 $\pm$ 1.7	
Cs-137	661.64 $\pm$ 0.08	(100)	30 y.
Eu-155	45.299 $\pm$ 0.002		1.81 y.
Mn-54	834.81 $\pm$ 0.04	(100)	313 d.
Ta-182	67.750 $\pm$ 0.001	295 $\pm$ 10	115 d.
	100.107 $\pm$ 0.002	(100)	
	152.430 $\pm$ 0.003	49 $\pm$ 3	
	179.390 $\pm$ 0.004	24 $\pm$ 2	
	222.104 $\pm$ 0.004	52 $\pm$ 2	
	229.316 $\pm$ 0.005	24 $\pm$ 3	
	264.070 $\pm$ 0.005	23 $\pm$ 3	

in the direction of the detector. The solid angle subtended by the detector, taking the source as a point at the center of a sphere, is the first factor in determining the efficiency. The number of gamma rays actually detected in the photo peak divided by the number of gamma rays emitted in all directions in space is called the "absolute efficiency" of the detector. The absolute efficiency is obviously dependent on the distance between the source and the detector, but it is also a function of the energy. To see this, consider a source at a fixed distance from the detector emitting gamma rays of both high and low energies. For gamma rays emitted off the axis determined by the centers of the source and the detector the probability is greater for gamma rays of lower energy to deposit all their energy in the crystal. Because of the increase in photo electron and Compton cross sections with decreasing energy the probability of energy escaping from the edge is lower for lower energy gamma rays. Effectively, for the lower energy gamma rays the cross section for absorption in the crystal is increased. However, the lower the energy, the more likely is the absorption of gamma rays in the front of the crystal in a dead region that has a high concentration of lithium. Likewise, the efficiency falls off at high energies as

the mean free path of the gamma rays increases with energy and the probability of absorption decreases.

To compensate for the energy dependence of the absolute efficiency it is necessary to calibrate each detector: the process is called "efficiency calibration." Again standard sources are used whose energies and intensities are well-known. To do an efficiency calibration on an absolute scale for a given source-to-detector distance it is also necessary to know the exact source strength at the time calibration is performed. Knowing the source strength and the intensities of the standard gamma rays gives the total number of gamma rays emitted. The number of counts under the photo peaks gives the number of gamma rays detected, the quotient then is the efficiency which can be plotted against the energy of the gamma rays.

3. The Gamma-Gamma Coincidence Measurements

Gamma-gamma coincidence is a method for investigating systematics in the energy level diagram of a nucleus.³ With this method it is possible to observe cascades of gamma rays as they fall through the various excited states to the ground state. It is assumed that the lifetime of any particular excited state is very short, generally of the order of

picooseconds. The time difference that the gamma-gamma coincidence circuit can resolve will be discussed later.

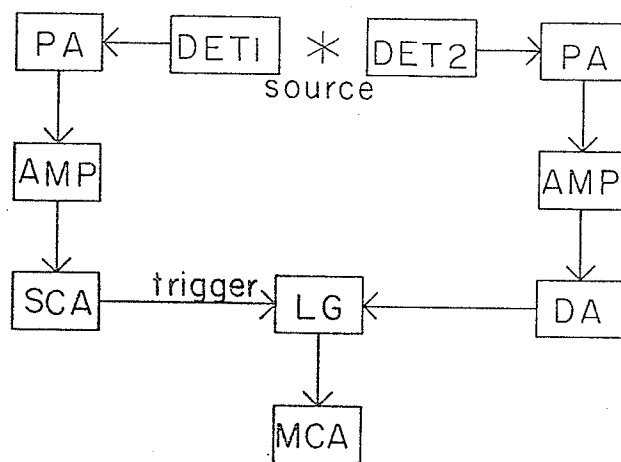
The simplest type of gamma-gamma coincidence circuit is one placing only an energy requirement on a regular singles measurement: see figure 2a. Gamma rays enter detector #1 and the signal is amplified, then fed into a single channel analyzer (SCA). The output of the single channel analyzer is logic pulses; if the voltage of a pulse falls between V and $V + \Delta V$ one logic pulse is generated, if a pulse does not fall in this range no pulse is generated. The value ΔV is called the window width, V is the lower discrimination level and $V + \Delta V$ is the upper discrimination level.

The logic pulse from the single channel analyzer triggers coincidence circuits in the linear gate (LG). The gate is then opened and the amplified results of the scanning detector #2 are recorded in the multi-channel analyzer. The major fault in this system is that whatever is seen with detector #2 when a gamma ray of the proper energy enters detector #1 is recorded by the multi-channel analyzer. There is no time requirement on the spectrum of gamma rays recorded.

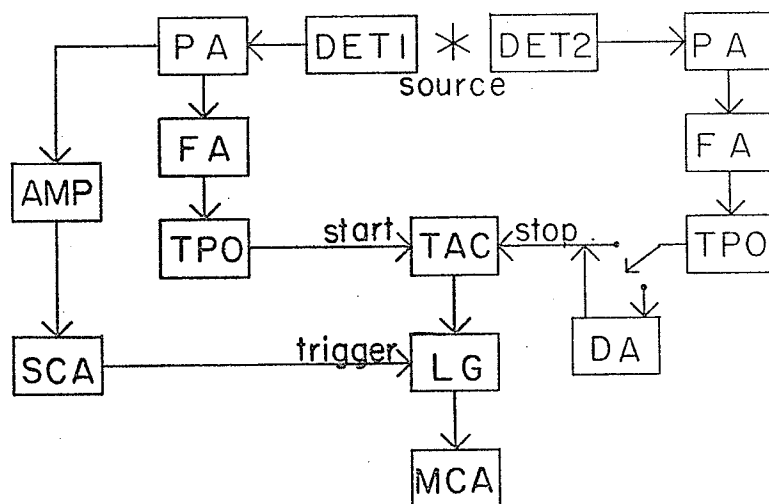
To put a time requirement into the system a leading edge timing circuit is constructed: see figure

Figure 2a
Slow Coincidence

Figure 2b
Time Resolution Measurement



SLOW COINCIDENCE



TIME RESOLUTION MEASUREMENT

2b. The fast amplifiers (FA) are present to increase the preamplifier output to a large enough amplitude to fire the time pick-off units (TPO.) When the time pick-off unit detects a sharp increase in voltage, the leading edge of a pulse, a logic pulse is generated and hence the name "leading edge timing." The outputs of the time pick-off units are fed into a time to analogue converter (TAC). The output of the time to analogue converter is a pulse whose height is proportional to the time difference of logic pulses entering the "start" and then the "stop" inputs. The time to analogue converter output is recorded on the multi-channel analyzer with an energy gate set by the single channel analyzer. The particular source used in setting up this circuit was selenium-75 whose decay scheme is shown in figure 3.⁴ Setting the single channel analyzer gate on the 135.99 keV line and using the Ortec Ge(Li) X-ray detector #1 as the gating detector and the Ortec 35cc Ge(Li) detector as the scanning detector, a time spectrum was acquired as shown in figure 4. The full width at half maximum of this time peak is the resolving time of the system. The delay amplifier is used to set the time scale. Introducing a known delay of several microseconds into the "stop" channel, each time to analogue converter

Figure 3
Se-75 Decay

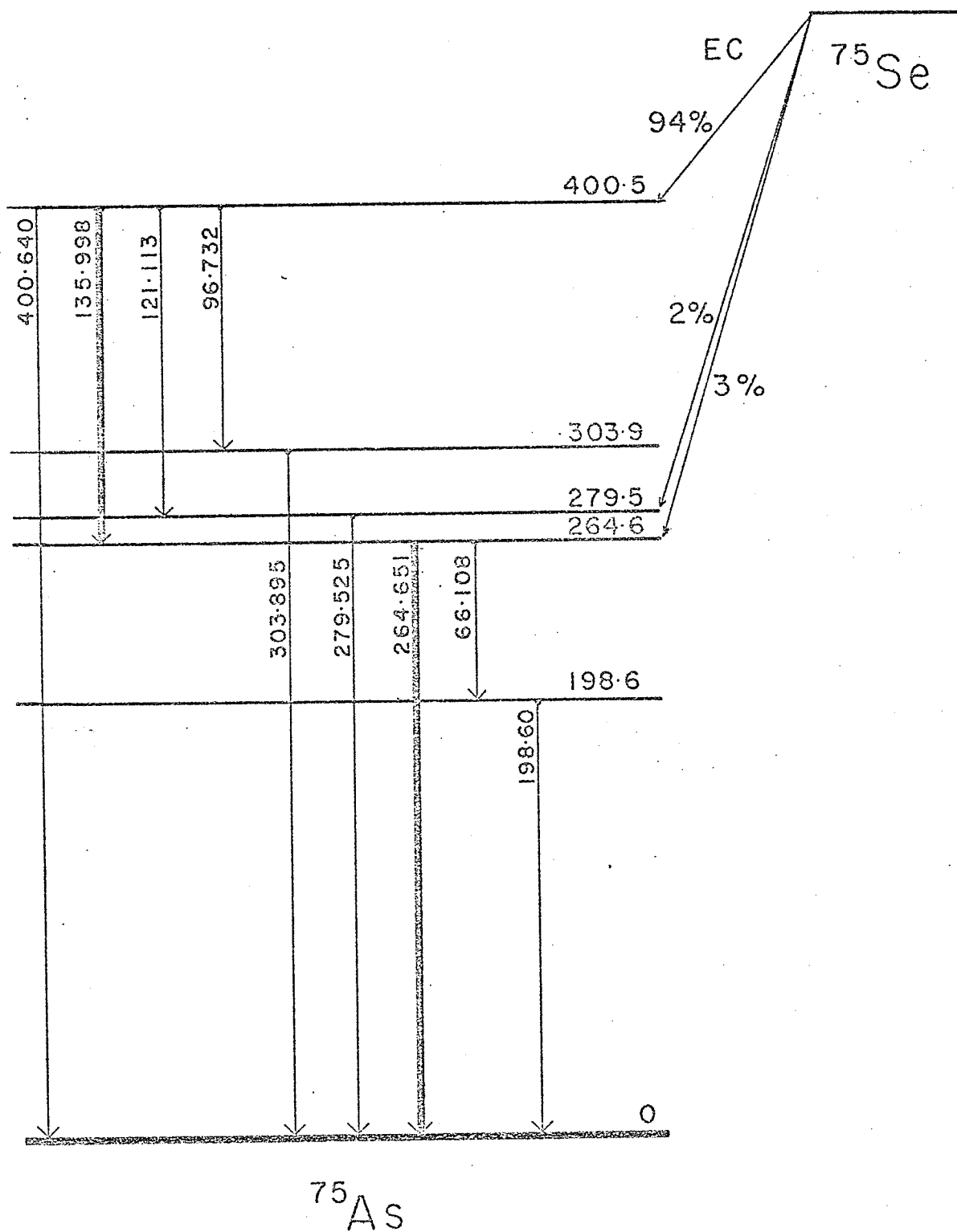
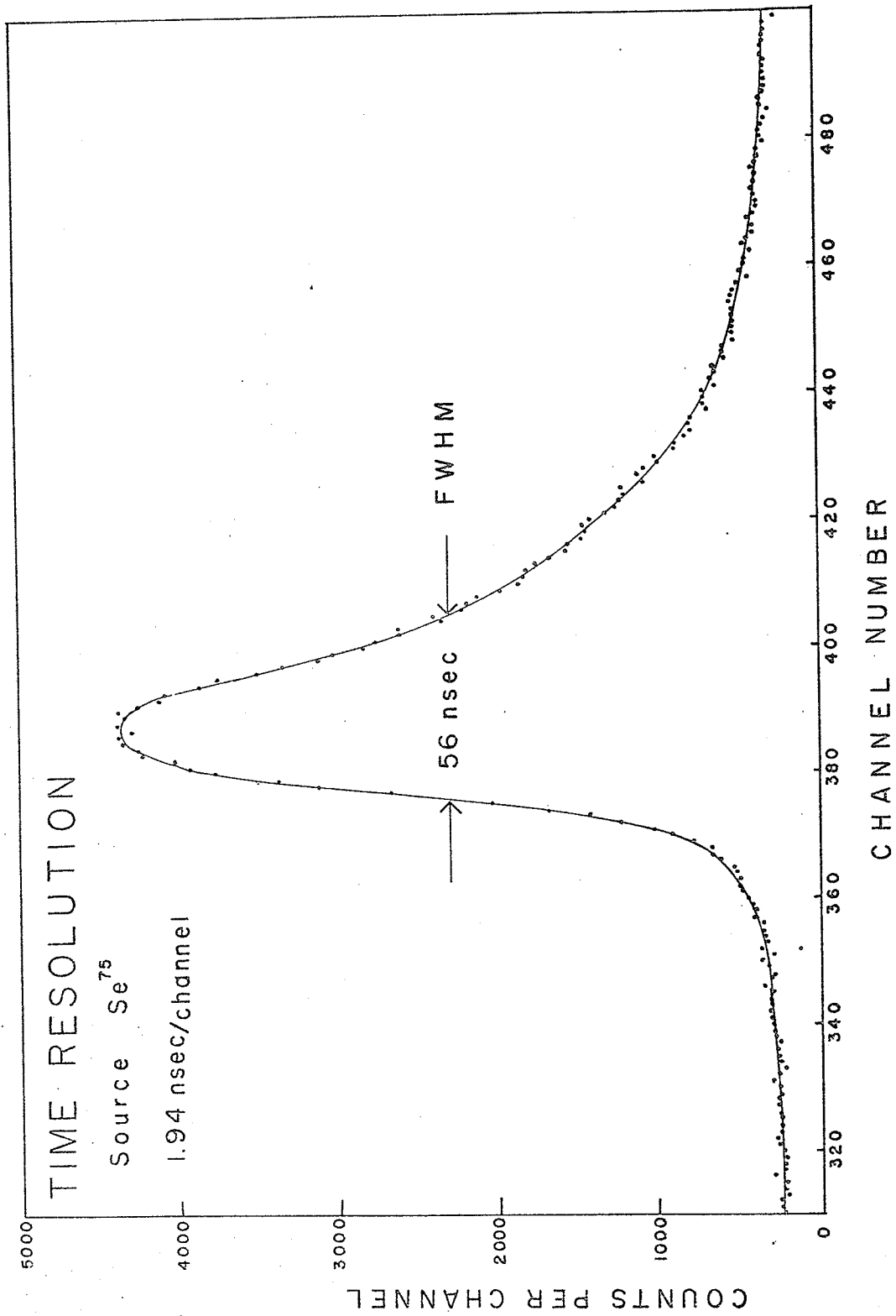


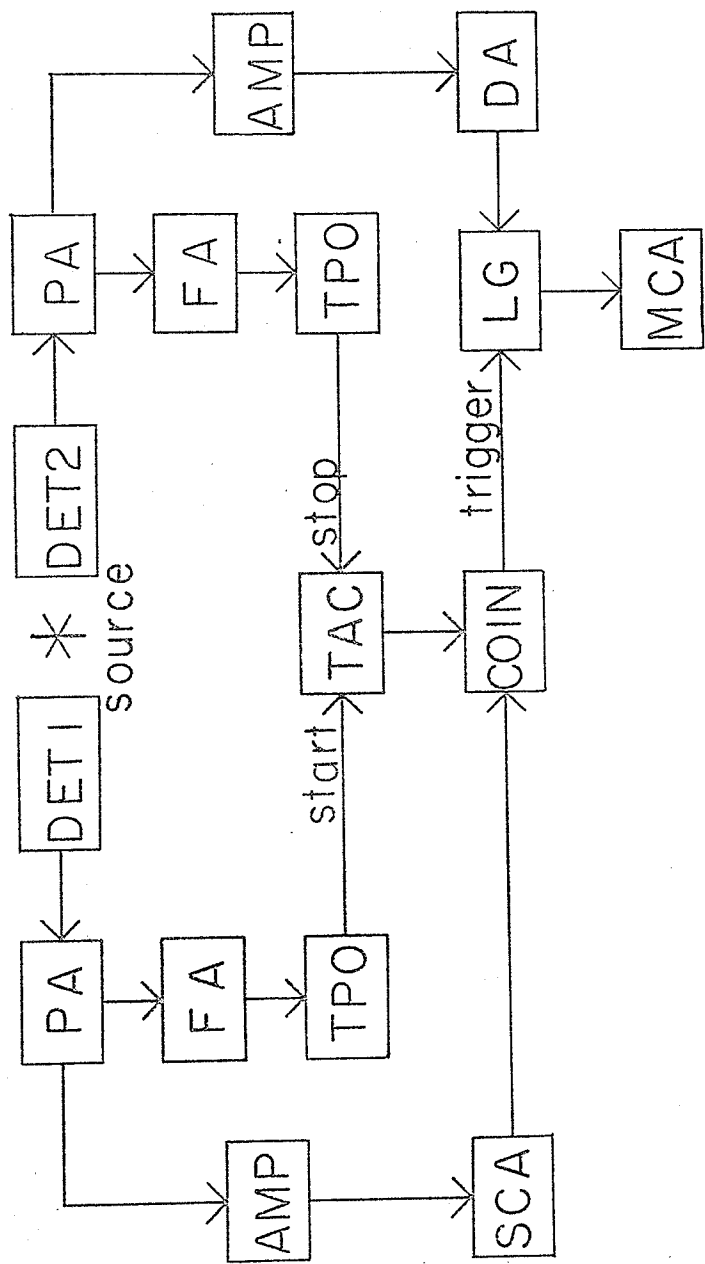
Figure 4
Resolving Time



output pulse is heightened by a constant quantity. This moves the time peak into the higher channel range of the spectrum. Locating centroids of the original and delayed peaks, the number of channels between divided by the delay time gives the scale. The resolving time was measured to be 56 nanoseconds.

Contained in the time to analogue converter is a single channel analyzer. Setting the window of this single channel analyzer on the time peak, a logic pulse is generated if the time pulse is of a proper height. Figure 5 shows how using this mode of operation for the time to analogue converter as "fast" coincidence and using a single channel analyzer as "slow" coincidence the total coincidence system with time and energy requirements is constructed. The names "fast" and "slow" come ^{from} the relative time duration of the logic pulses from the time to analogue converter and the single channel analyzer. The system was tested using again selenium-75 and also barium-133 whose decay scheme is shown in figure 6.⁵ For selenium-75 the single channel analyzer was set to gate on the 135.99 keV line. Figure 7a shows the singles spectrum and figure 7b shows the coincidence spectrum. Clearly three lines are in coincidence with the 135.99 keV line: 66.05 keV; 198.6 keV; 264.6 keV. Referring to

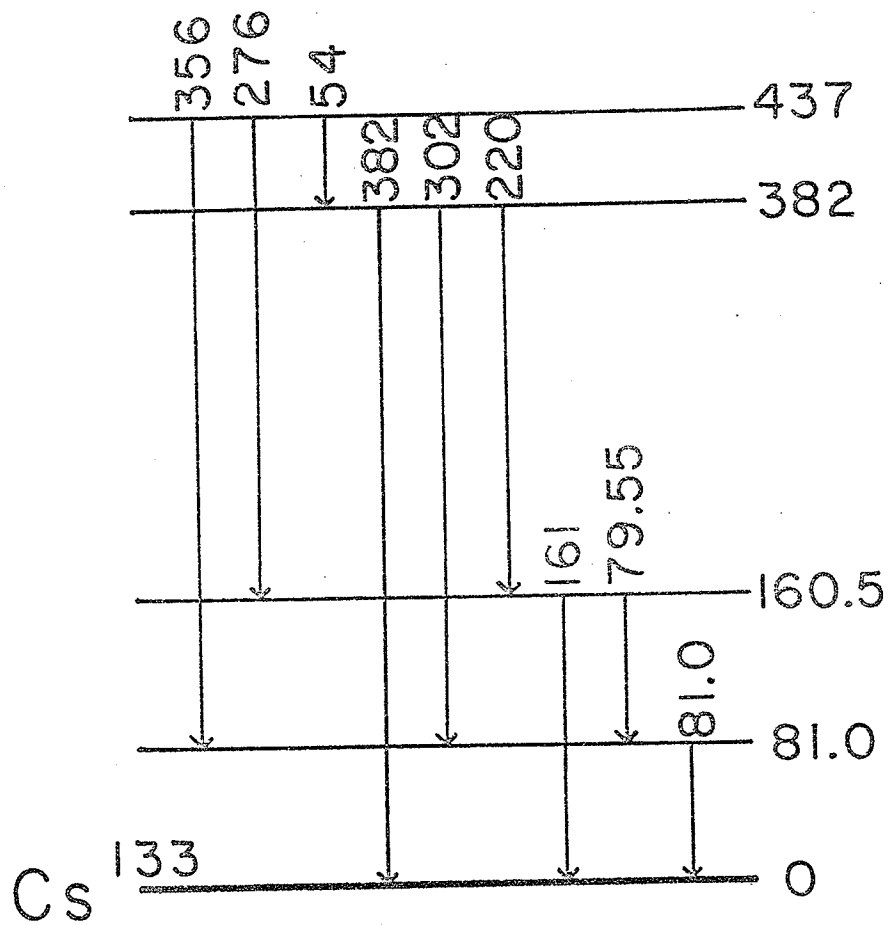
Figure 5
Coincidence System



GAMMA - GAMMA COINCIDENCE

Figure 6
Ba-133 Decay

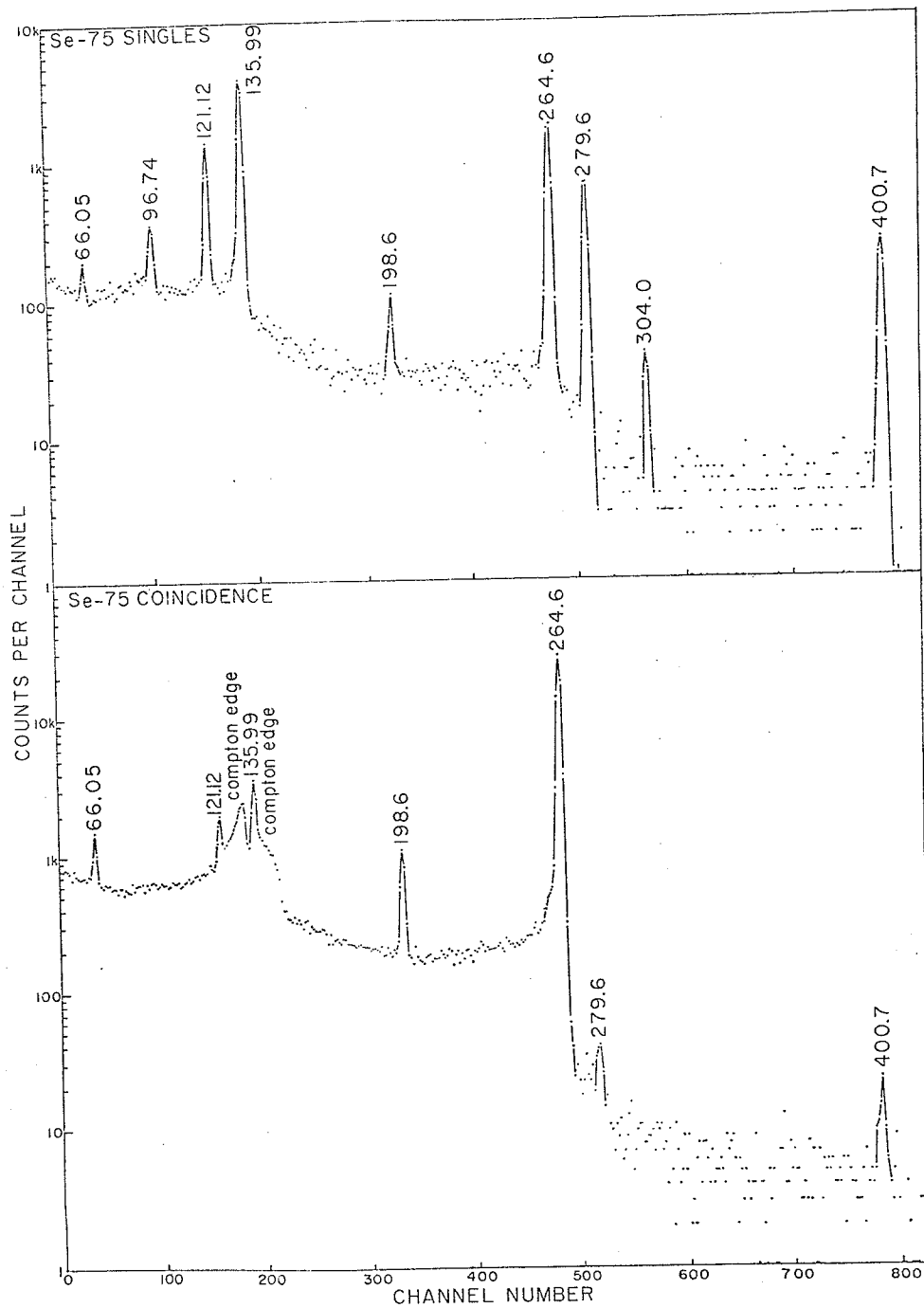
E.C. $\swarrow$ Ba^{133}
7.2 y



THE DECAY OF BA-133

Figure 7a
Se-75 Singles

Figure 7b
Se-75 Coincidence



the decay scheme these three lines and only these are in cascade with the 135.99 keV gamma ray. Notice also the structure just to the left and right of the 135.99 keV line in the coincidence spectrum. These are Compton edges at approximately 130 keV and 140 keV. They arise from the 264.6 keV and the 279.6 keV gamma rays. When setting the single channel analyzer gate at 135.99 keV not only will the 135.99 keV gamma rays open the gate but the background under the peak will open it, too. Imagine a gamma ray entering the scanning detector and Compton scattering off an electron. The new energy of the gamma ray re-emitted is the difference of the original energy and the amount of energy deposited in the detector, and is given by:

$$E'_\gamma = \frac{E^\circ_\gamma}{1 + \frac{E^\circ_\gamma}{m_e c^2} (1 - \cos \theta)}$$

where m_e is the mass of the electron, E°_γ is the original energy of the gamma ray and θ is the angle between the original and new trajectories of the gamma rays. If θ is near 180° we have back scattering and the chances are very good the back scattered gamma ray will leave the detector without interacting again. But the trajectory will take it now into the gating detector where it will be registered in coincidence

with the energy loss in the scanning detector. If the energy of the scattered gamma ray falls within the boundaries of the gate, the Compton edge will be counted as being in coincidence. Also the opposite situation may occur with the initial gamma ray first entering the gating detector and then back scattering into the scanning detector. If the energy loss in the gating detector, the difference of the initial gamma ray energy and the energy of the back scattered gamma ray, falls within the boundaries of the gate, the backscattered gamma ray will be registered in coincidence.

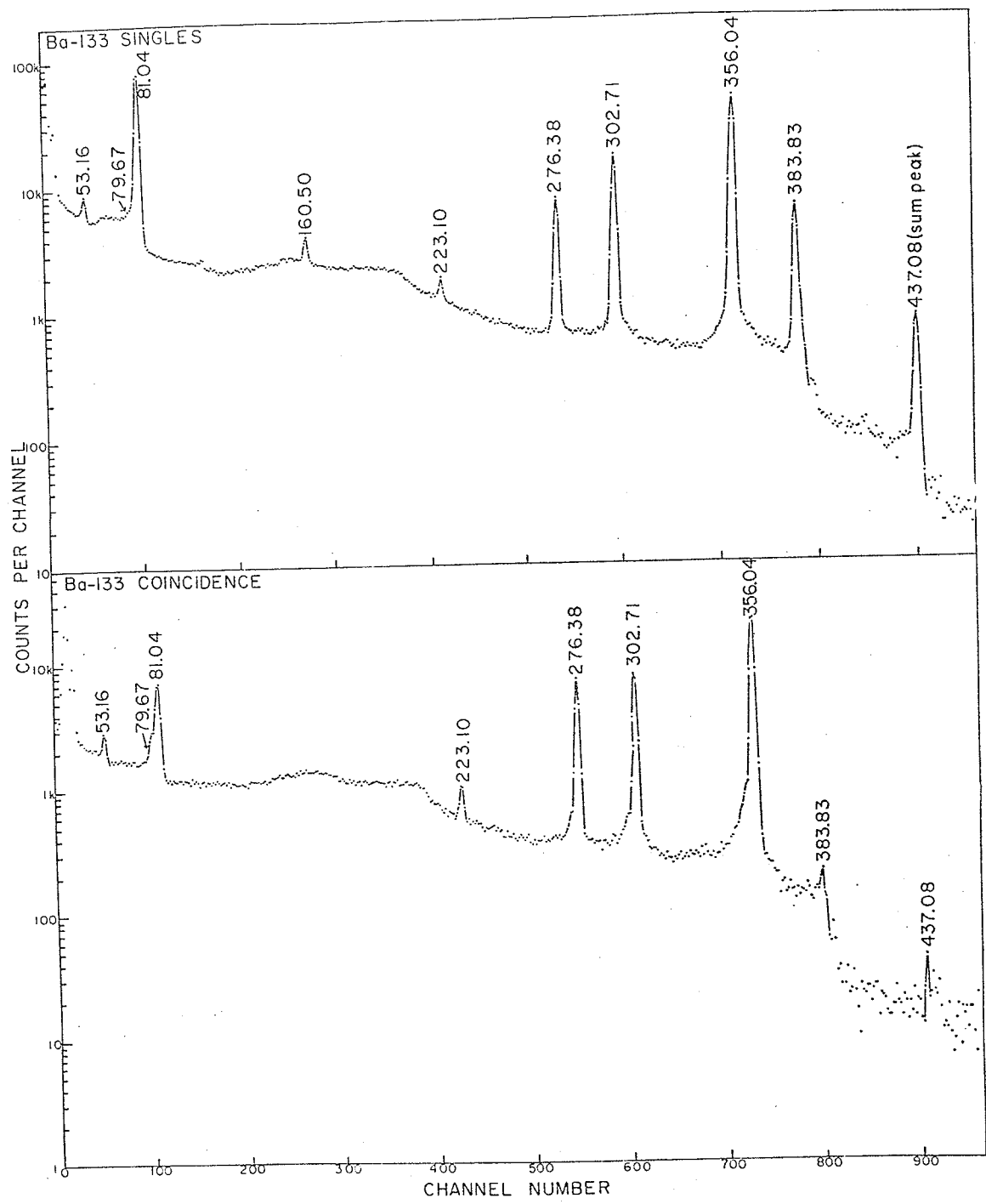
For barium-133 the single channel analyzer was set to gate on the 81.04 keV line. Because of the proximity of the 79.67 keV line, gamma rays in cascade with this line will also show up in the coincidence spectrum. Figure 8a shows the singles spectrum for barium-133 and figure 8b shows the coincidence spectrum. Here we see seven lines in coincidence: 53.16 keV; 79.67 keV; 81.04 keV; 223.1 keV; 276.38 keV; 302.71 keV; 356.04 keV. Again referring to the decay scheme we see that each of the seven is in cascade with either the 81.04 keV or the 79.67 keV gamma rays.

The theoretical true to random ratio is given by the expression:

$$\frac{T}{R} = \frac{1}{2 \tau_r \frac{dN}{dt}}$$

Figure 8a
Ba-133 Singles

Figure 8b
Ba-133 Coincidence



where τ_r is the time resolution of the system and $\frac{dN}{dt}$ is the source strength. This assumes that there is no background under the line being gated on and that the single channel analyzer has been set perfectly. With a source strength between five and ten microcuries and a time resolution of approximately 50 to 60 nanoseconds the true to random ratio will be between 40 and 20. The selenium-75 spectra were analyzed to find the experimental true to random ratio. Using the 135.99 keV and the 264.6 keV lines the true to random ratio was found to be 27.

4. Data Analysis

Once a spectrum has been acquired with the multi-channel analyzer, the data is in magnetic core storage. There are several output devices that can be attached to the multi-channel analyzer to retrieve the data. An IBM typewriter can be used to print channel by channel the contents of the memory. A plotter may also be used to produce a graph of the spectrum. The primary data acquisition unit, however, is a paper tape punch. After punching, the paper tape is then read into an IBM-1620 computer which will punch computer cards containing the data. The cards may then be read into the IBM-360 computer and the data stored on

direct-access devices. The IBM-360 computer is incapable of handling paper tape directly.

With the data stored in the IBM-360 system analysis may be performed by various computer programs developed in this laboratory and run from a terminal in the laboratory. These programs are in a library called Cutipie. Plotting, listing, peak fitting and data smoothing are some of the functions that can be performed. The peak fitting program, Cutie,⁷ is the heart of the program library. Analysis with this program gives the position, area, FWHM and associated errors of photo peaks in gamma ray spectra. Cutie will automatically search a spectrum and locate the approximate peak positions. A least squares fit is made to each peak, and there are two choices for the shape of the peaks. The first and simplest is a true gaussian which is placed on a straight line background, but the best peak shape is a gaussian on the high energy side with a decaying exponential on the low energy side. The background used with this shape is a polynomial of second order. The only input that is required to start automatic analysis is the region to be analyzed and the approximate FWHM in this region. Manual operation is also possible by giving the program the approximate peak positions. Up to six peaks may be

fitted in any one region and this makes the program ideal for simple X-ray groups.

Coincidence spectra tend to show a great deal of scatter channel by channel at the higher energy ends. Convolution of each point using least squares method smooths the data into a more nearly continuous curve. The position and area of photo peaks remain unchanged after application of this program (see Appendix.)

5. Source Preparation

Neptunium-237 is an alpha particle emitter with a half-life of 2.14×10^6 y.⁸ The source was purchased from Amersham/Searle Corporation, The Radiochemical Centre, Amersham, Buckinghamshire, England. Approximately 100 milligrams were obtained in 2 ml of nitric acid. The daughter product in the decay of Np-237 is protactinium-233, a beta emitter with a half-life of 27 d.⁹ This presents a disturbing problem immediately. In all gamma spectra of Np-237 the gamma spectrum of Pa-233 will be present as a contaminant. One milligram of Np-237 will have a strength of 0.71 microcuries. If this quantity is originally free from all Pa-233, in one week the strength of the Pa-233 will be 0.11 microcuries, in two weeks 0.21 microcuries and in a month 0.36 microcuries. Fortunately the daughter

of Pa-233, uranium-233, is long lived, $1.62 \times 10^{5.10}$ y., and hence will not contaminate the spectra.

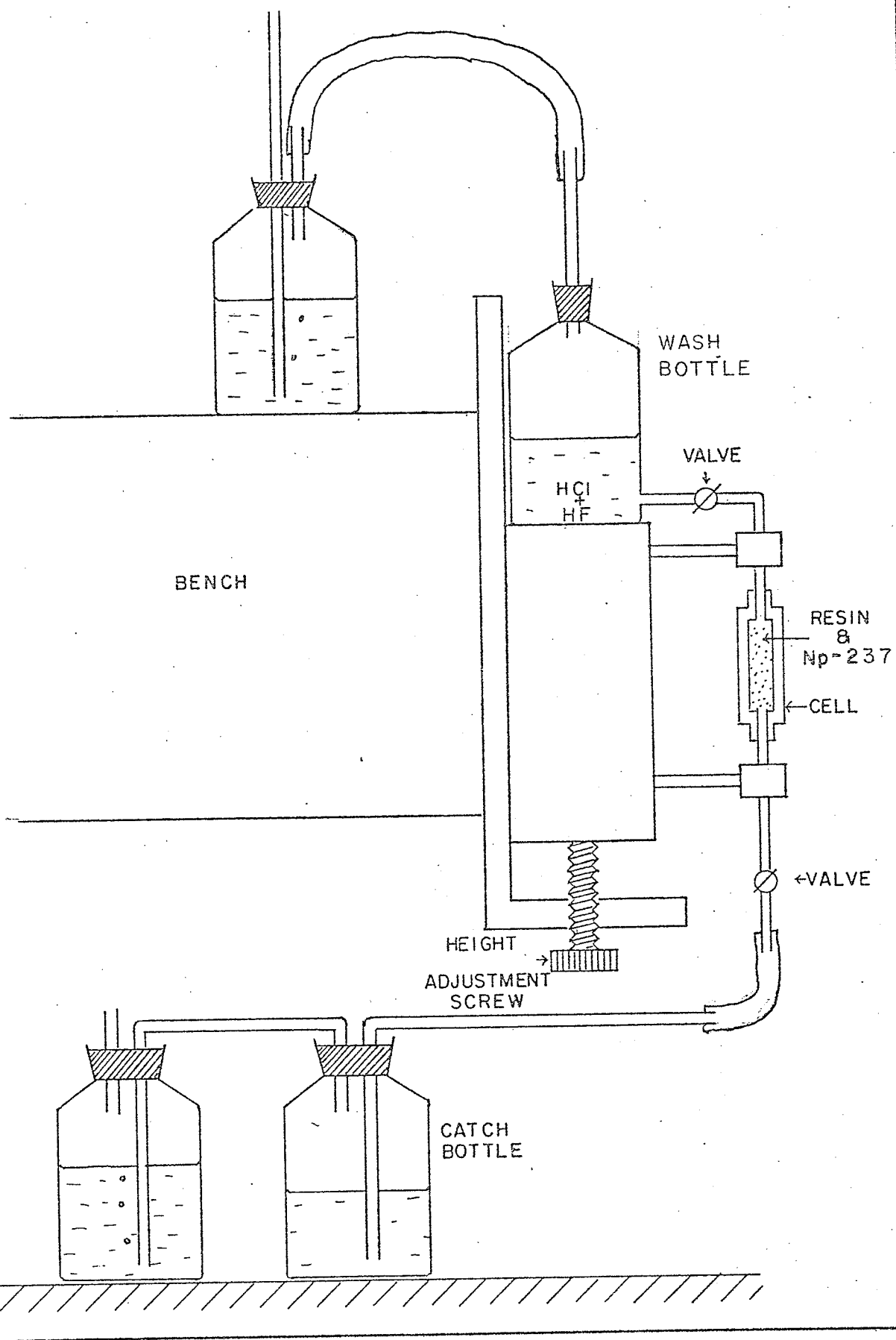
To remove the disturbing influence of Pa-233 as much as possible, chemical separation through anion exchange was effected. For singles measurements an ion exchange column was used as described below. For the coincidence measurements an initial separation was performed and then the protactinium was allowed to grow freely into the source. A micro column was filled with the ion exchange resin Bio-Rad AG 1x8. The neptunium is in a +6 valence state and is retained in the column at a molarity of 3. ^{HCl} Passing through the column at this molarity is protactinium and americium in valence states of +5 and +3, respectively. The neptunium can then be released by changing the molarity of the wash to 0.1. This manner of purification was repeated once a month to produce a series of sources pure enough to be useful. The sources still contained Pa-233 in amounts that increased with time, but a previous investigation in this laboratory of the spectrum of Pa-233 gave the energies and intensities of the gamma rays. With this knowledge identification of the effects of the contaminant became simplified and routine.

Sources were prepared on special plastic

microscope slides. A nine-sixteenths inch diameter hole is near one end of each slide. A square of mylar 150 millionths of an inch thick was glued over the hole. A drop of the recently purified neptunium solution was placed on the mylar. This operation was performed in a glove box to prevent contamination from this long lived neptunium isotope. The drop is evaporated to dryness and another drop is deposited, then dried and so on until a source of sufficient strength was obtained. Another layer of mylar was then placed over the source thereby minimizing the possibility of contamination of the laboratory.

While the slide sources were sufficient to do coincidence experiments, something better was needed for the singles measurements. A lucite cell was constructed to hold the resin and the source. The cell was then inserted into the continuous purification system depicted in figure 9. Working as above with the same resin and using 3N HCl as the wash the concentration of protactinium was maintained at a level lower by a factor of two or three over that obtainable with slide sources. However, the resin acted as a reducing agent for the neptunium, slowly changing the neptunium valence state and causing the source to be washed out of the cell. An alternate procedure,

Figure 9
Continuous Purification System



proposed by Browne,¹¹ was to use the same resin but a different valence state of neptunium, the +4 state, and a different washing solution, 10N HCl + 0.3N HF. To oxidize the resin before use, it was treated with this mixture of hot acid. The hydrofluoric acid was used in the wash because the solubility of protactinium is very high in HF solutions. This procedure worked extremely well, reducing the protactinium concentration approximately fifteen times over the previous used continuous purification procedure. However, the hydrofluoric acid had a tendency to attack the cell and the tubing, limiting the amount of time the new continuous purification system could be used.

It was found that the gamma ray spectra taken in the presence of the resin still contained considerable contributions from the gamma rays of protactinium. The reason for this was that the Pa was impacting in the resin crystals. When a five MeV alpha particle departs from a neptunium nucleus the resultant protactinium nucleus receives a recoil energy of approximately 100 keV. If the recoil of the protactinium is directed into the resin crystal, the protactinium will be lodged in the crystal and the washing solution will be unable to remove the contaminant. The best results were obtained by washing the neptunium for a long period and

then releasing the Np from the column. The gamma ray spectrum is then recorded for a short period and then the neptunium is repurified on the column and the process is repeated. With this method the concentration of protactinium was reduced to 5×10^{-10} parts relative to the neptunium. This concentration gave a specific activity of 0.06% protactinium relative to neptunium.

CHAPTER II

Energies and Intensities

1. Selective Absorption Across the K Edge of a Material

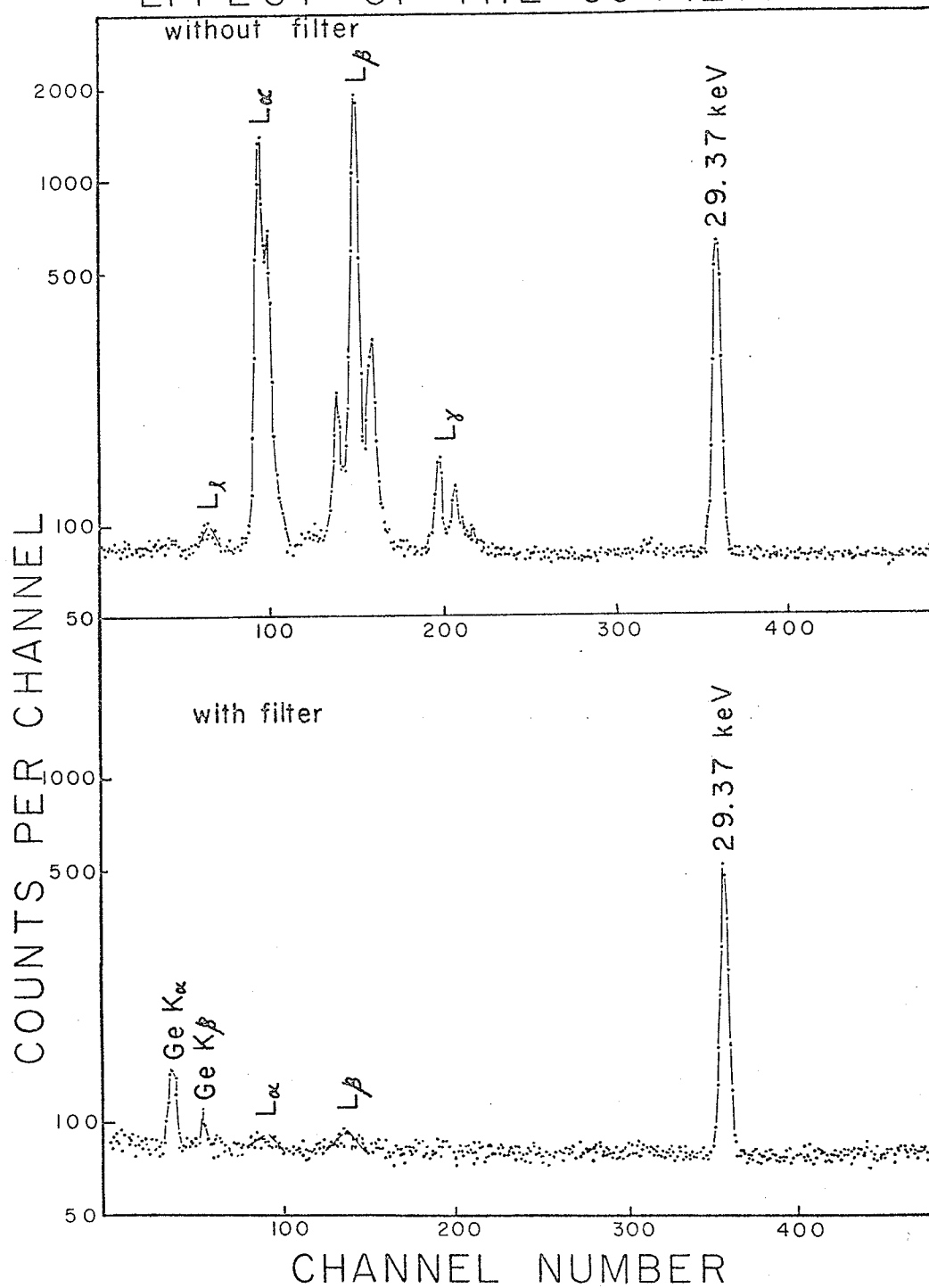
Consider recording a gamma ray spectrum consisting of two photo peaks of different energies. Say this spectrum is recorded with a strong source placed near the detector. Two peaks will appear corresponding to the two gamma rays. Also a third peak will appear much less intense and slightly wider: the sum peak. The energy of this new peak is the sum of the energies of the two gamma rays. Two gamma rays entering the detector within the resolving time of the detector and its preamplifier will look like one gamma ray to the detector. The sum peak is then due to the small time interval for which the detector will fail to resolve two signals and will treat the composite signal as single. The increase in width arises from the slight difference in time of entry of the two gamma rays. In general, this situation is not desired; there are enough peaks in a spectrum without generating even

more. A simple solution is to reduce the counting rate (time consuming but effective.) A better way which does not work well much above 100 keV but is very effective at low energies is to use a filter. Gamma rays passing through matter are absorbed but the absorption is a function of the energy. For each element the energies of the K and L absorption edges¹² are tabulated. A possible problem in this work is the strength of the L X-rays of heavy elements, with energies ranging from about 11 to 20 keV. These X-rays are plentiful and they sum with themselves and with gamma rays of high intensities. To find an effective filter and get it the right thickness would greatly simplify spectra taken in the low energy region. The K absorption edge of germanium is at 11.104 keV, making it an ideal material for such a filter. Obtaining an old detector crystal that had been drifted through, we sliced a thin wafer 0.25 millimeters in thickness or 0.130 g/cm². Placing the filter between the source and detector effectively eliminated from the spectrum the L X-rays from heavy elements. As an example figure 10a shows the L X-rays and the 29.37 keV gamma ray from the decay of neptunium-237, and figure 10b shows the effect when the germanium filter is interposed between detector and source. The L X-rays are very much

Figure 10

Ge filter

EFFECT OF THE Ge FILTER



reduced compared to the 29.37 keV line, but germanium K X-rays are now present through fluorescence. In practice, the counting rate was kept low and the filter used to make sure that no summing was occurring.

2. Energy calibration of 29.37 keV and 46.53 keV gamma rays

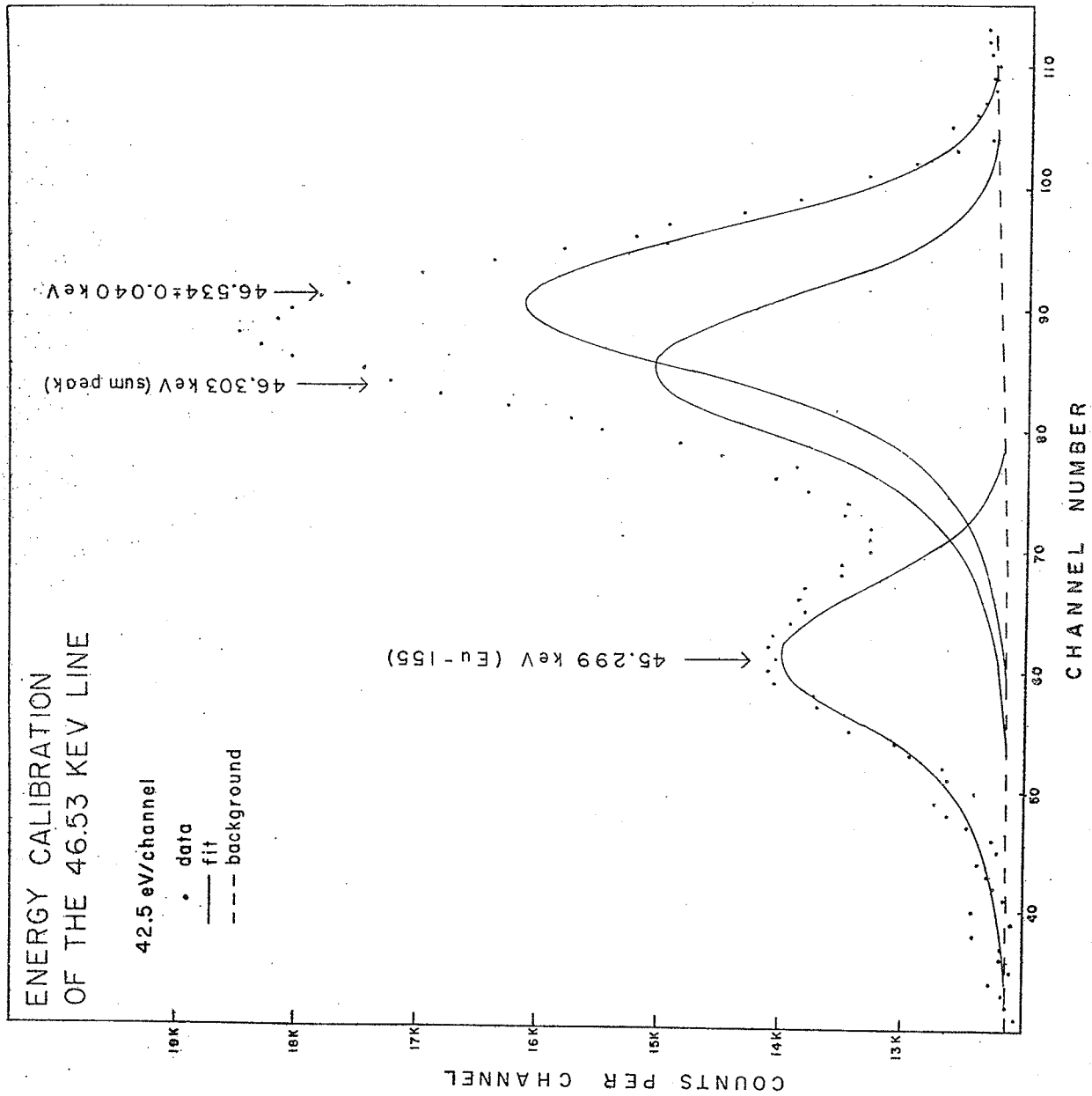
To calibrate the energy of gamma rays the mixed source technique is invaluable. In the low energy regions the K X-rays are very helpful tools. The energies of K X-rays are well known¹² and provide us with numerous useful calibration lines. It was known before starting this project that the lowest energy gamma ray in the decay of neptunium-237 has an energy of about 29.3 keV. Cline¹³ quotes 29.375 ± 0.020 keV, Browne¹⁴ however has the energy as 29.29 ± 0.10 keV. Calibration sources were chosen to place lines a few keV above and below this line. Americium-241 decays with the emission of a gamma ray at 26.345 keV. For the other source cesium-137 was chosen. The only gamma ray emitted in its decay is one at 662 keV and is therefore barely detected in a X-ray detector but the barium $K\alpha$ X-rays are at 31.817 and 32.194 keV. The germanium filter was used to absorb the L X-rays from the neptunium decay and also the L X-rays from the americium decay. The energy value of the neptunium

gamma ray was then found to be 29.373 ± 0.005 keV assuming perfect linearity between the 32.2 and 26.3 keV lines. (This is verified by calculating the energy of the Ba $K\alpha_2$ X-ray and finding 31.817 keV; the tabulated value is 31.817 keV.) Previous work by the present author on non-linearity in this region has shown that the assignment of five eV error in the curve itself is quite conservative for the region used (ie. the intrinsic error in energy from the linearity curve is 0.0 ± 5.0 eV,) hence the final value of 29.373 ± 0.010 keV is given for this gamma ray.

To calibrate the 46.5 keV gamma ray of neptunium the mixed source technique was again used. A neptunium source along with a europium-155 source were used. Europium-155 emits a 45.299 keV 14 gamma ray and gadolinium X-rays. These lines were used to calibrate the neptunium gamma ray. The germanium filter was not used here to present a challenge to our peak fitting program. A sum peak located at 46.303 keV is expected; summing between the protactinium L_{β_3} X-ray (16.930 keV) and the neptunium gamma ray at 29.373 keV. Figure 11 shows how the peak fitting program handled this situation. The energy of the neptunium gamma ray was found to be 46.534 ± 0.040 keV and the energy of the sum peak was found to be 46.303 ± 0.050 keV. The value of

Figure 11

46.53 keV Gamma Ray Calibration



46.534 $\pm$ 0.040 keV can be compared to Cline's value of 46.5 $\pm$ 0.5 keV and Browne's value of 46.46 $\pm$ 0.10 keV.

It must be pointed out here that energy calibrations are only as accurate as the standards used. Gamma rays calibrated on bent crystal spectrometers make excellent calibration sources. X-rays also make excellent calibration sources be they observed after a nuclear decay or excited by fluorescence, for their energies are very well known.

3. Energies and Intensities

The singles gamma ray spectrum of the decay of neptunium-237 displayed a large number of photo peaks. A total of 42 gamma rays were observed in the neptunium decay. It also must be remembered that the products of the decay of Pa-233 were present to some extent in all spectra, and 11 gamma rays from the decay of Pa-233 were observed. Also present in the spectra were X-rays of protactinium and uranium from the nuclear decays of Np-237 and Pa-233, respectively, and X-rays of neptunium produced by fluorescence of the source material were also observed in some sources. All this amassed together produced an extremely complicated spectrum.

Energy calibration of the neptunium gamma rays was

performed using the mixed source technique. A Np-237 source was placed in front of a detector along with various calibration sources and a spectrum accumulated. A non-linearity curve was drawn and the most intense neptunium lines were calibrated (see figure 12 for an example of a non-linearity curve. Shown here is an extreme case with a large non-linearity. The use of a better amplifier gave the same shape for the curve but with much less non-linearity.) This process was repeated several times with different combinations of calibration sources. The weaker gamma rays were then calibrated using the previously measured strong neptunium gamma rays as calibration standards.

Figure 13 shows the gamma ray spectrum for neptunium-237, taken with the germanium X-ray detector #2 using the germanium filter. Cline¹³ reports a gamma ray at 43.5 ± 0.5 keV. It was found, however, that this is a sum peak; summing between the $L\alpha$ X-ray and the 29.37 keV gamma ray. An interesting feature in this region is the gamma ray at 94.66 ± 0.05 keV, directly underneath the uranium $K\alpha_2$ X-ray. The presence of this gamma ray is inferred by comparing the intensities of the uranium $K\alpha_2$ and $K\alpha_1$ X-rays. Scofield¹⁵ has calculated the ratio of the intensities of $K\alpha_2$ to $K\alpha_1$ as a function of the atomic number, Z. Computer analysis of

Figure 12
Non-linearity Curve

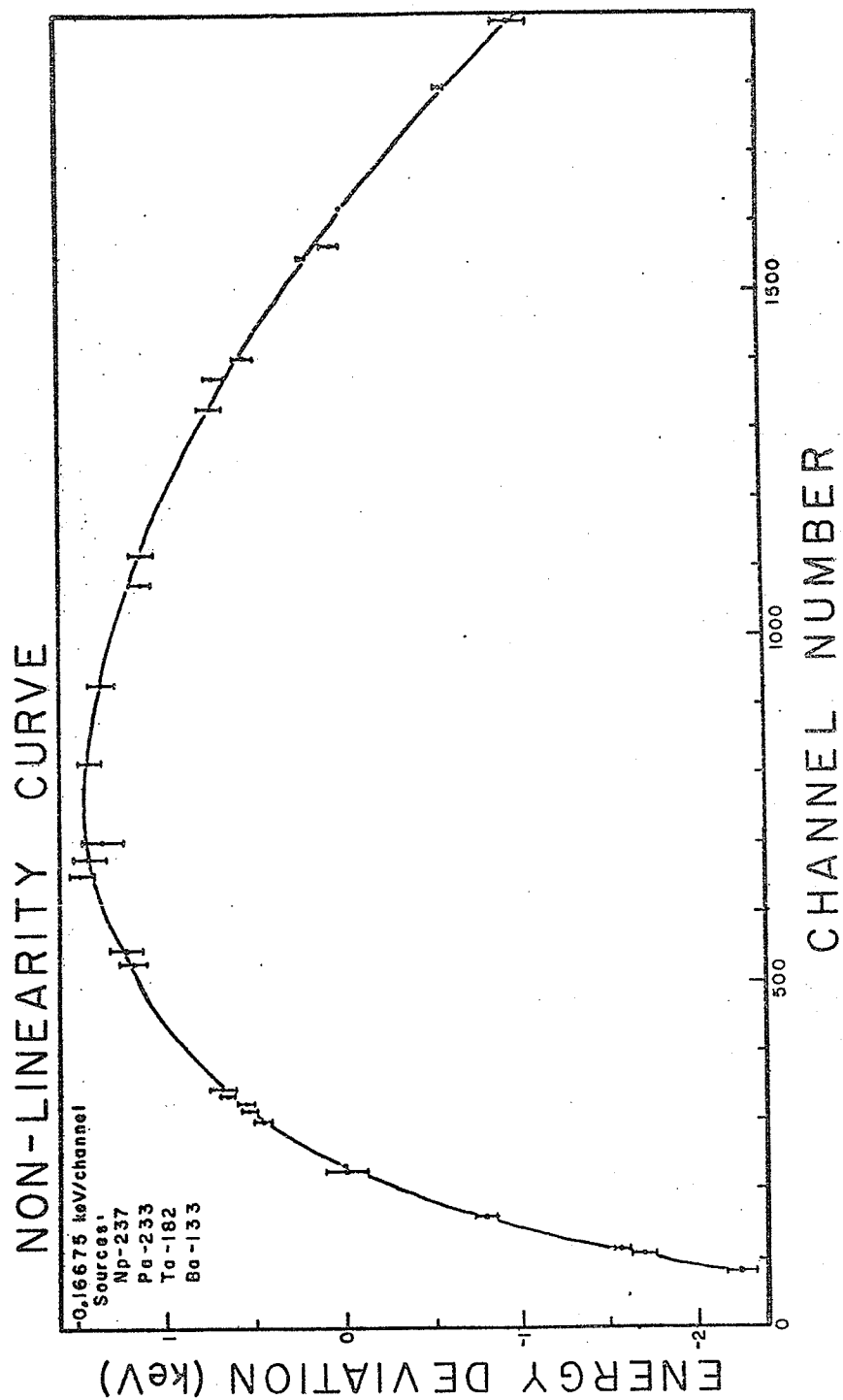
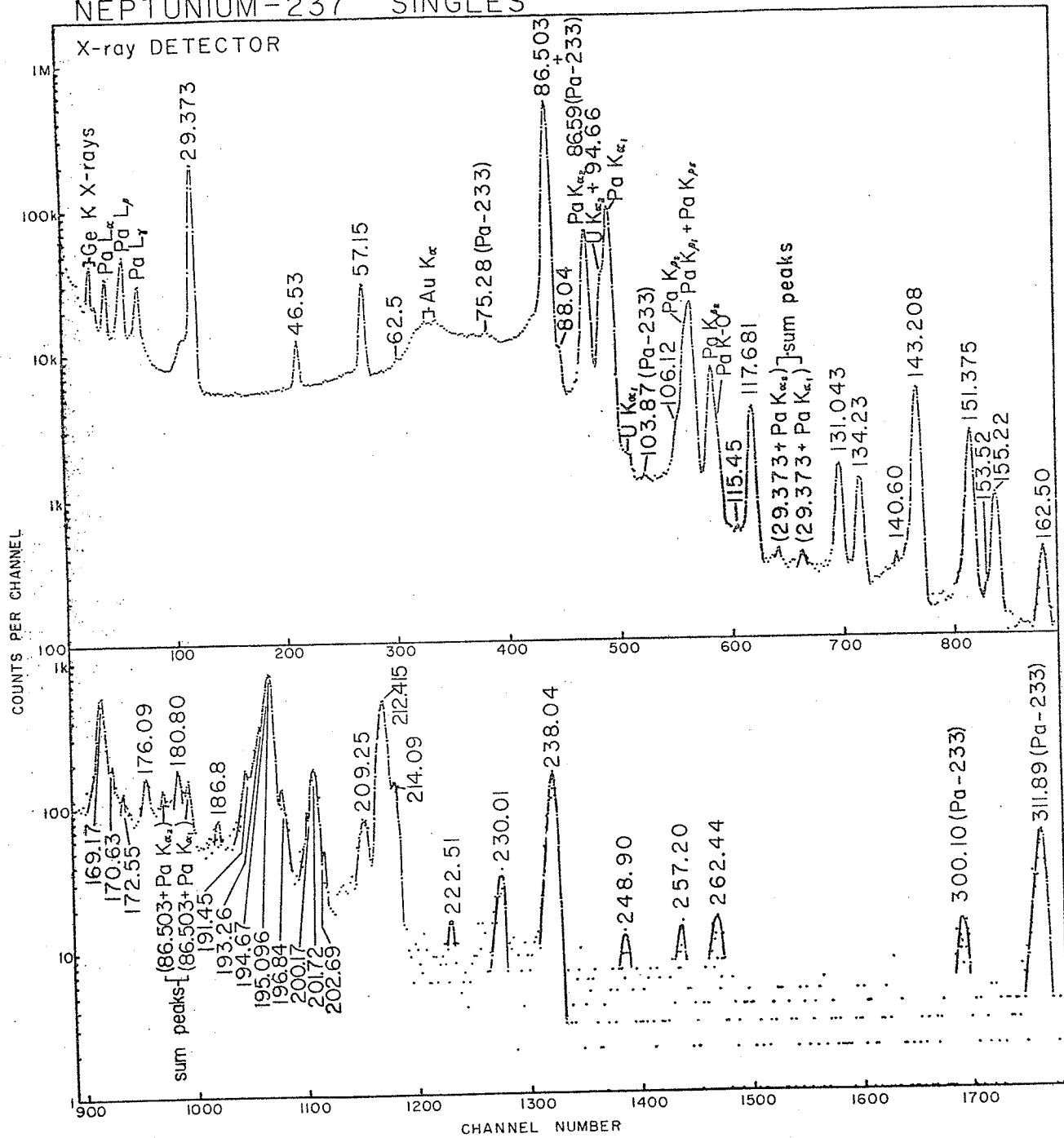


Figure 13
X-ray Detector Singles

NEPTUNIUM-237 SINGLES



the spectra gave the intensities for the four X-rays in the K_{α} group of protactinium and uranium. The K_{α_2} to K_{α_1} ratio for the protactinium X-rays agreed with Scofield's prediction of 0.622, but the same ratio for uranium produced a number far greater than Scofield's prediction of 0.625. The only possible solution for the disagreement of values was to propose a gamma ray of sufficient intensity directly beneath the uranium K_{α_2} X-ray. The peak fitting program was unable to separate the U K_{α_2} component from the gamma ray. Hence, the gamma ray must be directly under the X-ray and the energy of the gamma ray must equal the energy of the X-ray.

Another interesting region is centered on the 195.10 keV gamma ray. Apparently there are four photo peaks bunched together in this region. Analysis shows, however, that the fwhm of the four peaks is significantly greater than neighboring peaks. The conclusion drawn from this is that there is a fifth peak located somewhere in this group. Figure 14 shows how the peak fitting program handled this situation. A fifth peak was located close in energy to the most intense gamma ray in this group. With this fit the fwhm of the photo peaks became comparable to that of neighboring photo peaks.

Figure 14

Cutipie Analysis of 195.10 keV Group

CUTIPIE ANALYSIS OF THE 195 keV GROUP

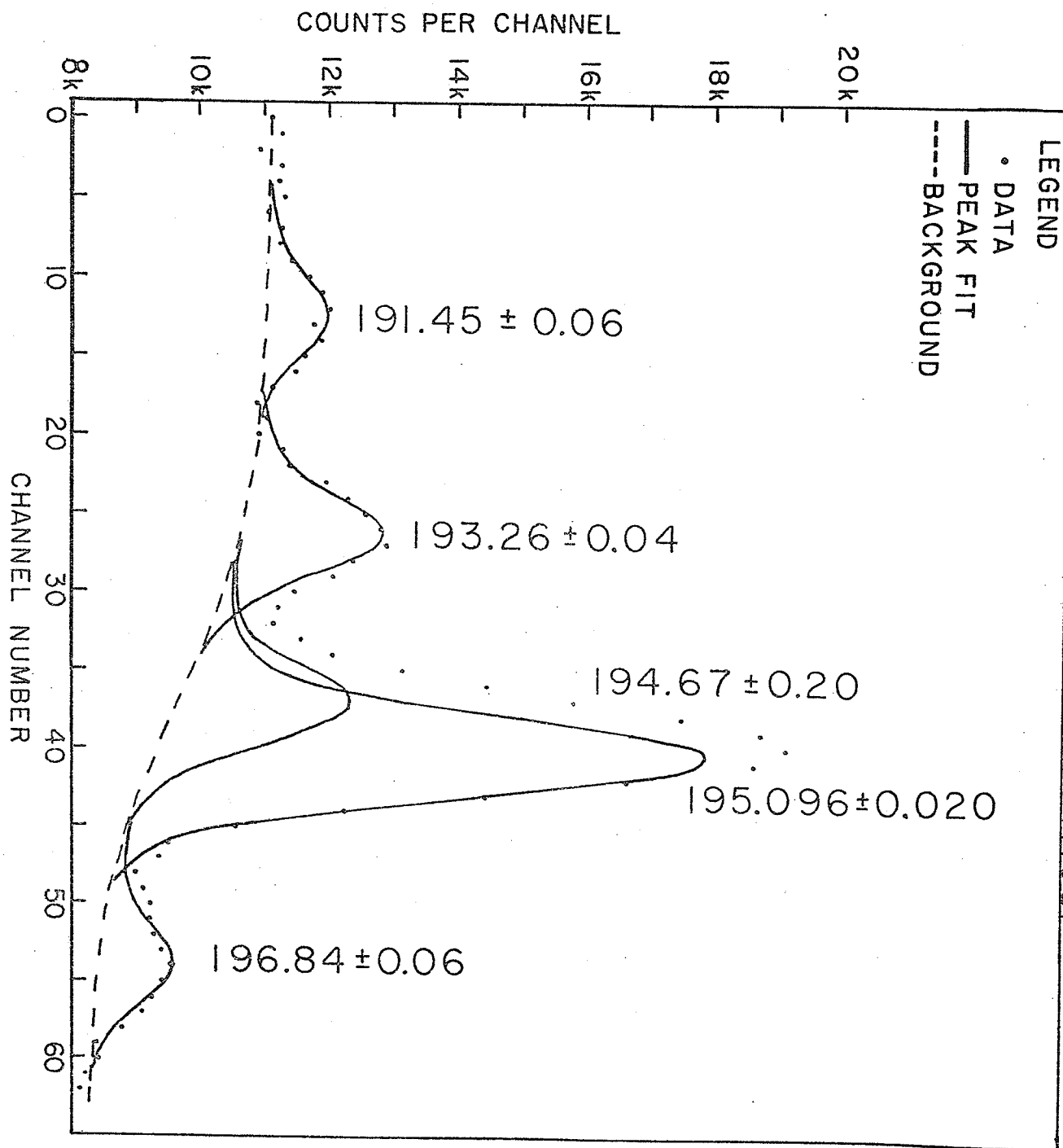


Figure 15 shows the gamma spectrum taken with the 30cc detector; ranging from 25 keV to 320 keV. The presence of the Pa-233 gamma ray photo peaks at the high energy end puts a small background under the neptunium gamma ray photo peaks. This background made the energy and intensity calibration slightly more difficult to achieve with great precision. The advantage of using the 30cc detector is the greater efficiency at higher energies compared to the X-ray detector. For example, compare the appearance of the 186.8 keV line in the two singles spectra (figure 13 and 15.) Obviously the 30cc detector is better suited to observe this peak. The main disadvantage of the 30cc detector is the resolution obtainable is considerably poorer than the X-ray detector resolution. This can be seen by comparing the group centered on the 195.10 keV line in the two spectra. The X-ray detector shows the structure of this group much more clearly than the 30cc detector.

Table 3 gives the values obtained in this work for the energies and intensities of the gamma rays observed in the decay of neptunium-237. Several new gamma rays were discovered in this investigation: those of energy 115.5, 140.6, 153.5, 194.7, 222.5 keV and these can all be placed in the level diagram. All the gamma rays listed are accompanied by the values obtained by others and all

Figure 15
30cc Detector Singles

NEPTUNIUM-237 SINGLES

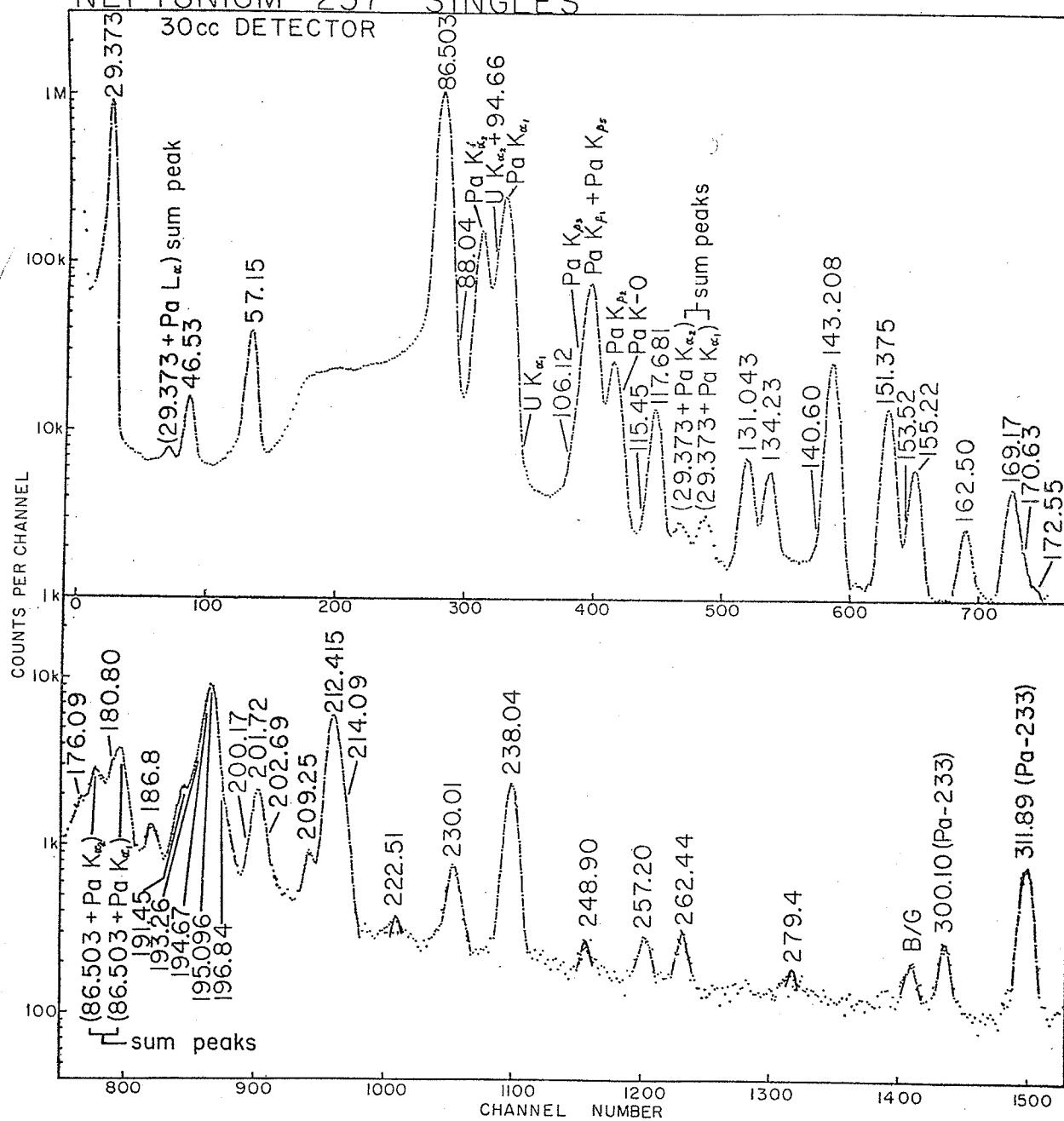


Table 3
Gamma Ray Energies and Intensities

Gamma Ray Energies and Intensities

E_{γ} (keV)				I_{γ} ($I_{86.5} = 100$)			
Vara and Gaeta (1969)†	Browne and Asaro	Cline	Present Work	Vara and Gaeta (1969)	Browne and Asaro	Cline	Present Work
29.6	29.29(10)	29.375(20)	29.373(10)	100	111 ± 16	75	132±7
46.7	46.46(10)	*	46.534(40)	0.77	1.11±0.16	-	0.95±0.2
57.1	57.15(10)	57.112(20)	57.15(4)	0.46	3.33±0.3	6.25	3.53±0.2
62.9	-	-	62.5(5)	1.5	-	-	~ 0.1
86.6	86.49(10)	86.486(10)	86.503(20)	100	100	100	100.0
-	88.07(10)	*	88.04(16)	-	1.27±0.16	-	1.1 ± 0.3
94.5	94.66(10)	*	94.66(5)	30.8	6.66±0.6	-	5.04±0.35
-	106.22(10)	106.300(200)	106.12(5)	-	0.47±0.05	0.80	0.36±0.07
-	-	*	115.45(20)	-	-	-	0.021±0.006
117.3	117.65(7)	117.720(20)	117.681(30)	-	1.35±0.16	1.40	1.47±0.10
-	131.11(7)	131.110(20)	131.043(30)	-	0.71±0.07	0.70	0.83±0.12
134.0	134.23(7)	134.280(20)	134.23(4)	0.77	0.56±0.06	0.60	0.66±0.13
-	-	*	140.60(10)	-	-	-	0.15±0.04
143.6	143.26(7)	143.250(10)	143.208(25)	3.08	3.33±0.32	3.61	3.76±0.23
-	151.31(7)	151.410(10)	151.375(35)	-	1.98±0.24	2.00	2.03±0.13
-	-	-	153.52(20)	-	-	-	0.058±0.013
-	155.20(7)	155.25(20)	155.22(4)	-	0.77±0.07	0.80	0.79±0.06
162.6	162.38(7)	162.520(30)	162.50(6)	0.38	0.31±0.03	0.30	0.33±0.06
-	169.09(7)	169.16(30)	169.17(5)	-	0.60±0.06	0.60	0.67±0.07
170.3	170.56(10)	170.640(50)	170.63(8)	0.077	0.159±0.024	0.150	0.133±0.02
172.6	-	172.600(200)	172.55(20)	-	-	0.0375	0.057±0.015
176.1	175.93(10)	176.060(50)	176.09(7)	-	0.175±0.024	0.13	0.139±0.028
179.9	180.66(10)	180.780(50)	180.80(8)	-	0.19±0.024	0.169	0.18±0.04
-	-	184.400(100)	-	-	-	0.069	-
185.9	186.86(30)	-	186.8(5)	-	0.056±0.112	-	0.026±0.026
-	-	187.4(1)	-	-	-	0.031	-
-	191.34(10)	191.42(30)	191.45(6)	0.77	0.230±0.032	0.237	0.140±0.024
193.0	193.05(10)	193.220(30)	193.26(4)	-	0.429±0.05	0.46	0.35±0.035
-	-	-	194.67(20)	-	-	-	0.41±0.12
-	194.91(7)	194.970(20)	195.096(20)	-	1.67±0.16	1.6	1.37±0.17
196.7	196.81(10)	196.800(100)	196.84(6)	0.77	0.214±0.024	0.20	0.186±0.027
-	-	199.900(100)	200.17(10)	-	-	0.038	0.029±0.011
202.7	201.68(8)	201.67(20)	201.72(5)	0.115	0.357±0.037	0.40	0.36±0.04
-	~203	-	202.69(25)	-	~ 0.03	-	0.039±0.015
-	209.07(8)	209.180(30)	209.25(5)	-	0.13±0.017	0.169	0.157±0.016
212.2	212.28(7)	212.33(20)	212.415(25)	0.77	1.26±0.16	1.40	1.35±0.09
214.5	213.92(10)	213.960(40)	214.09(5)	0.77	0.35±0.04	0.35	0.38±0.03
-	-	-	222.51(25)	-	-	-	0.014±0.014
-	229.84(10)	229.9(1)	230.01(10)	-	0.11±0.016	0.081	0.093±0.023
237.9	237.91(7)	237.91(20)	238.04(4)	0.15	0.54±0.05	0.550	0.61±0.07
-	248.60(40)	248.8(5)	248.90(10)	-	0.040±0.01	0.038	0.039±0.017
257	257.14(40)	257.15(50)	257.20(20)	-	0.056±0.014	0.05	0.053±0.028
-	262.48(40)	262.42(50)	262.44(15)	-	0.056±0.012	0.063	0.062±0.018
-	-	280.4(5)	279.4(4)	-	-	0.0188	0.014±0.014

† Vara, J.M. and Gaeta, R. 1969. Nucl. Phys. A130, 586.

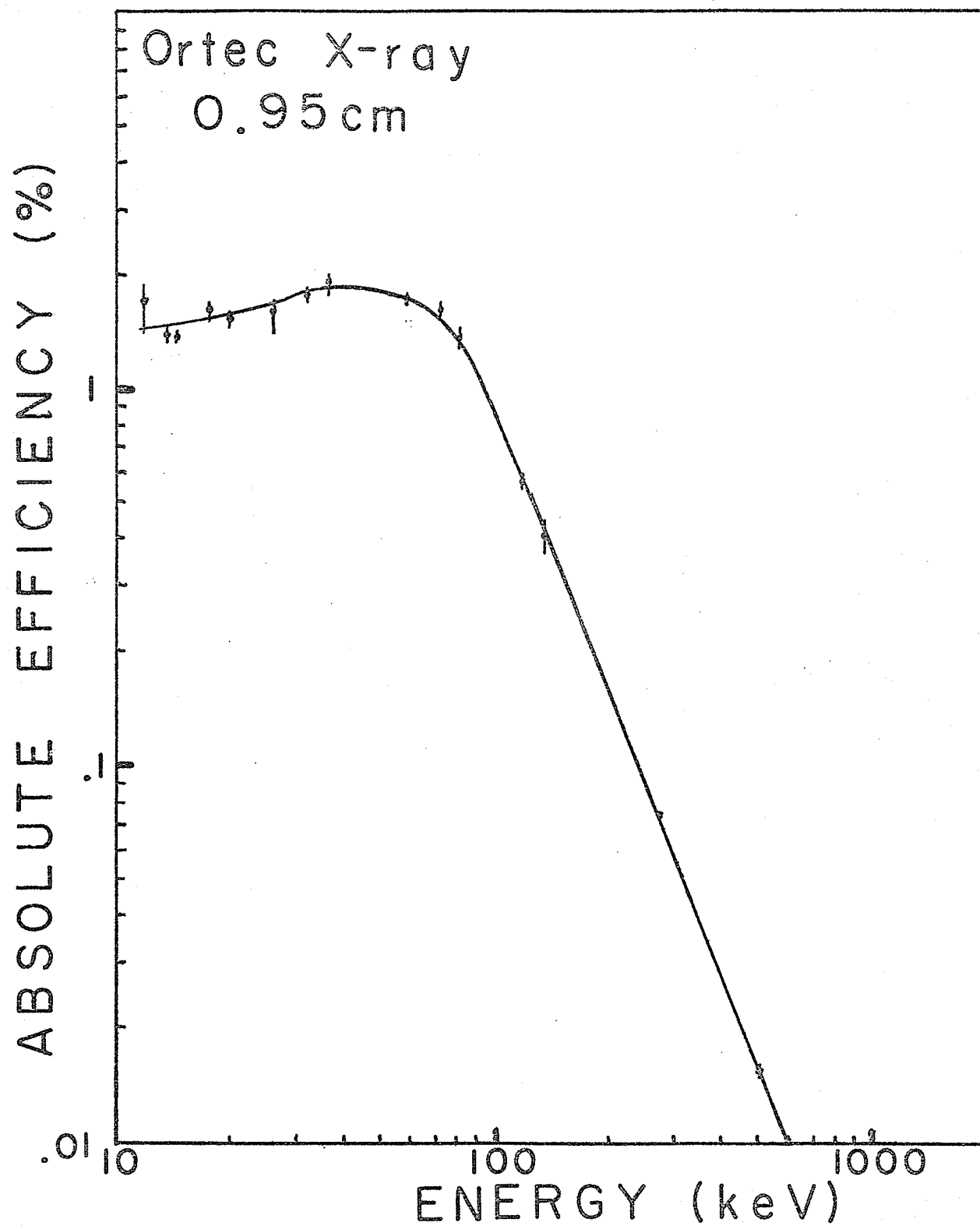
In addition 62 other γ -rays, unobserved by other workers, are listed.

* These five γ -rays together with transitions of energy 43.5 ± 0.5 , 70.5 ± 0.1 , and 99.8 ± 0.2 are marked on the spectrum but are not included in the list of ^{237}Np γ -rays

in Table III are placed in the level diagram except the 248.90 keV line. The intensities of these gamma rays were found as described in chapter 1 by first obtaining the area under the photo peaks and then compensating for the detector efficiency (see figure 16.) The area divided by the efficiency gave the intensity.

It can be seen the values of energy and intensity correspond closely between the works of Cline, Browne and Asaro and the present. The values of Vara and Gaeta are not in agreement with others.

Figure 16
X-ray Detector Efficiency



CHAPTER III

Coincidence Measurements

1. Coincidence Measurements

After determining the energies and intensities of the gamma rays it is necessary to construct a decay scheme. The spectrum of neptunium-237 shows a large number of alpha groups, indicating that several levels in protactinium-233 are populated directly. The levels above the ground state are called "excited states." The excited states of a nucleus are quantized in energy and angular momentum and the decay scheme depicts the excited states. The gamma rays are emitted when the nucleus undergoes a transition from one excited state to another or to the ground state. As can be seen from this discussion while this study is investigating the decay of neptunium-237, actually under investigation are the excited states of protactinium-233.

While it is possible to construct a decay scheme or level diagram from just the energies of the gamma rays, a much more reliable method involves the use of

coincidence measurements. In coincidence spectra we look for cascades of gamma rays i.e. one gamma ray feeding into an excited state and another depopulating the same state is a cascade. By setting the single channel analyzer to gate on one particular gamma ray we hope to see in the coincidence spectrum all the gamma rays in cascade with the gating line. Of course, since radioactive decay is a random process, coincidence spectra also contain random elements; this is the meaning of the true to random ratio. Consider just two gamma rays in a spectrum, one in cascade with the gating line and one not in cascade. Call the gamma ray in cascade the first and let it have a singles intensity I_1' , call the one not in cascade the second and let it have a singles intensity I_2' . Both peaks will appear in the coincidence spectrum, the second peak totally due to random coincidences with an intensity I_2 . The first peak, with intensity I_1 , will be a combination of random and genuine coincidences. To calculate the true to random ratio we assume the random components of both peaks are in the same ratio as the singles intensities. This is true provided there is nothing under the gating photopeak which might be in coincidence with either gamma ray. Separating I_1 into the random component I_{1R} and the genuine component

I_{IG} , the singles ratio gives:

$$I_{IR} = \frac{I_1'}{I_2'} I_2.$$

Having made this assumption it is now easy to calculate the true to random ratio:

$$\frac{T}{R} = \frac{I_1 - I_{IR}}{I_{IR}}$$

Once the coincidence system had been constructed and tested, a number of coincidence spectra were gathered. Altogether nine separate measurements were made, the average duration of a run being approximately two weeks. The nine major gamma rays studied in the coincidence measurements were 29.37, 57.15, 86.50, 117.68, 143.21, 151.38, 195.10, 212.42 and 238.04 keV. Of the nine gamma rays used for gating only three did not have other neptunium gamma rays in near proximity. Consequentially, six of the coincidence spectra had more than one gamma ray falling in the gate. Most of these six were strong peaks with a nearby weak peak. The worst case in this sense is the 195.10 keV gamma ray. The single channel analyzer set to gate on this peak also gated on four other gamma rays in the immediate neighborhood. Also opening the gate in all

coincidence measurements was the background under the peak. If back scattering from a strong gamma ray falls in the gate, the Compton edge will arise in the coincidence spectrum.

The statistics of the coincidence spectra were poor compared to those of the singles spectra, but this was to be expected. The problem of statistical scatter in the coincidence spectra was the impetus which led to the construction of the computerized smoothing program. With large background scatter weak lines were frequently obscured. Hence, a gamma ray that appears in cascade in the decay scheme will not necessarily stand revealed in the coincidence spectrum.

2. Coincidence Spectra

Coincidence spectra were acquired using the 35cc Ortec detector and the Ortec X-ray detector #2. For almost half of the coincidence runs the X-ray detector was used as the gating detector and the 35cc as the scanning detector. This arrangement worked well until the energies of the gating lines became too high. Above 150 keV it was necessary to use the 35cc detector as the gating detector and the X-ray detector as the scanning detector. The 86.50 keV gate was also run with the 35cc detector as the gating detector and the

X-ray detector was scanning.

29.37 keV gate:

The coincidence spectrum for the 29.37 keV line is shown in figure 17. Three lines are definitely enhanced above the singles intensities of the randoms: 57.15 keV, 143.21 keV and 151.38 keV. A possible fourth line that is probably enhanced is the 117.68 keV gamma ray. Referring to the decay scheme (figure 26) we see the 57.15 keV level is fed by the 29.37 keV gamma ray. The 151.38 keV gamma ray populates the 86.51 keV level which de-excites by the 29.37 keV gamma ray. But to accommodate the 143.21 keV and the 117.68 keV gamma rays it is necessary to insert a crossover transition from the 94.68 keV level to the 86.51 keV level. This transition has been proposed by E. Browne and F. Asaro¹⁶ previously through a similar line of reasoning. This coincidence spectrum lends additional support for the existence of this transition. The K X-rays shown in the figure are composites of the uranium and protactinium X-rays.

57.15 keV gate:

The coincidence spectrum for the 57.15 keV line is shown in figure 18. There are no lines enhanced in

Figure 17
29.37 keV Coincidence

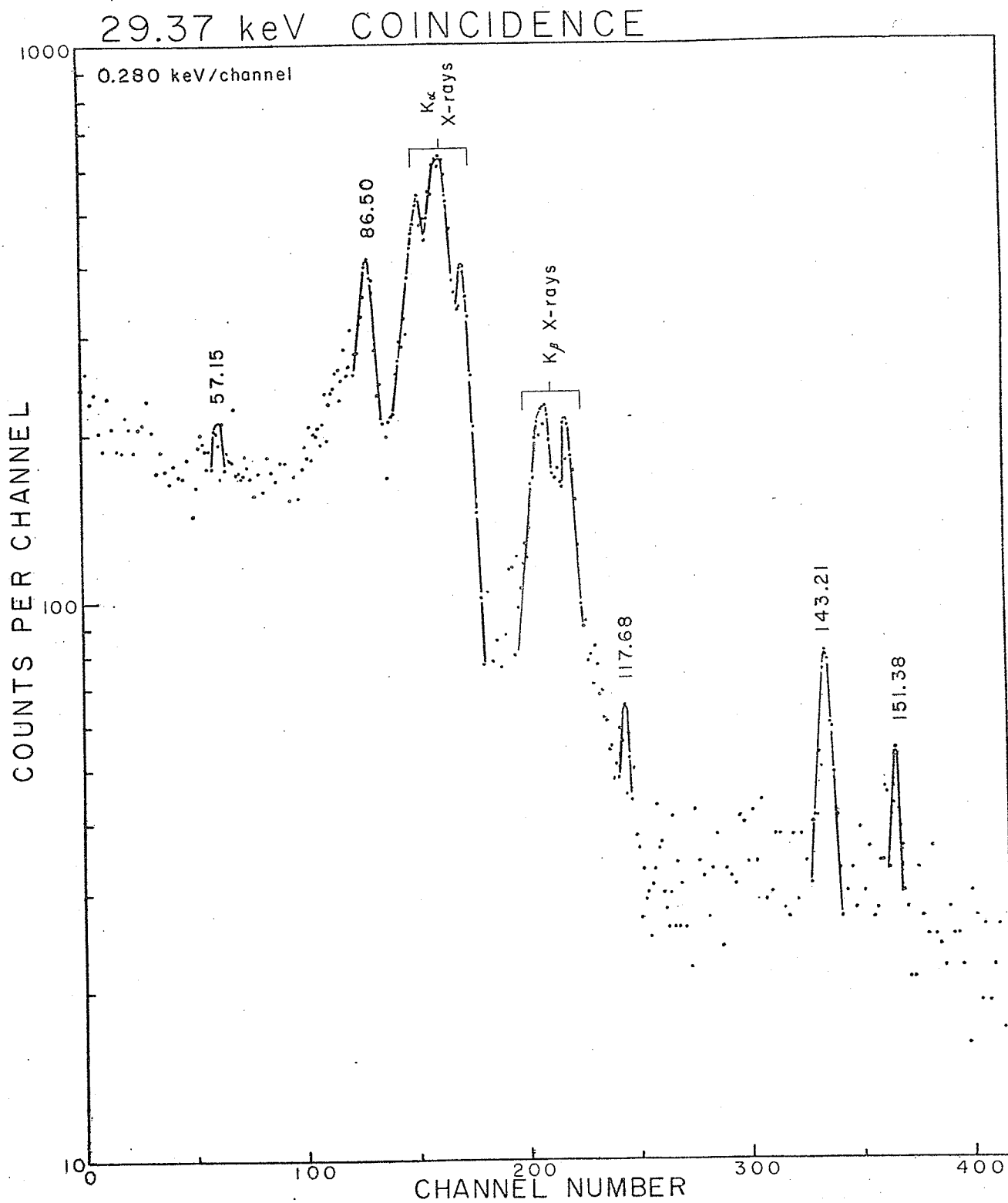
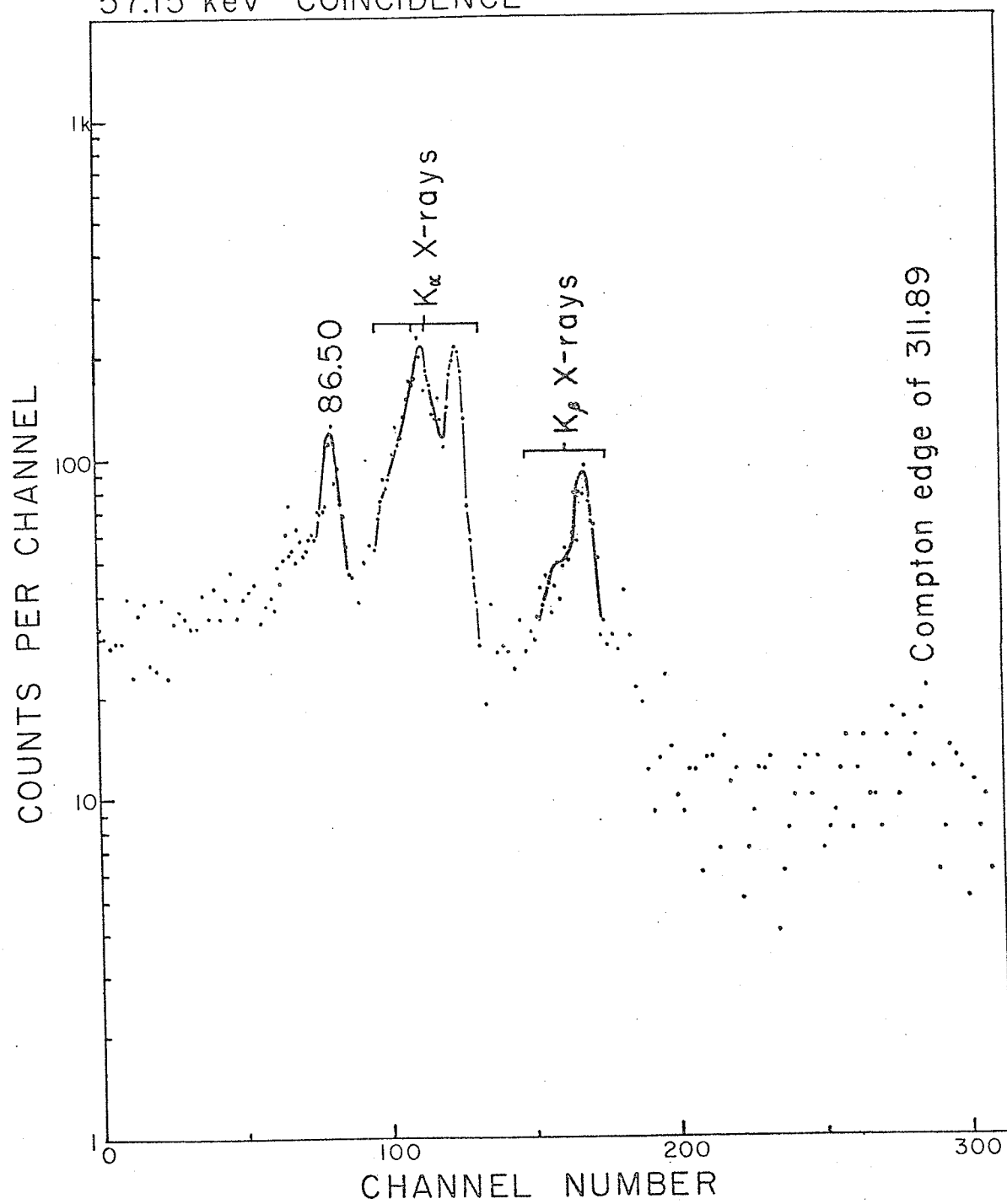


Figure 18
57.15 keV Coincidence

57.15 keV COINCIDENCE

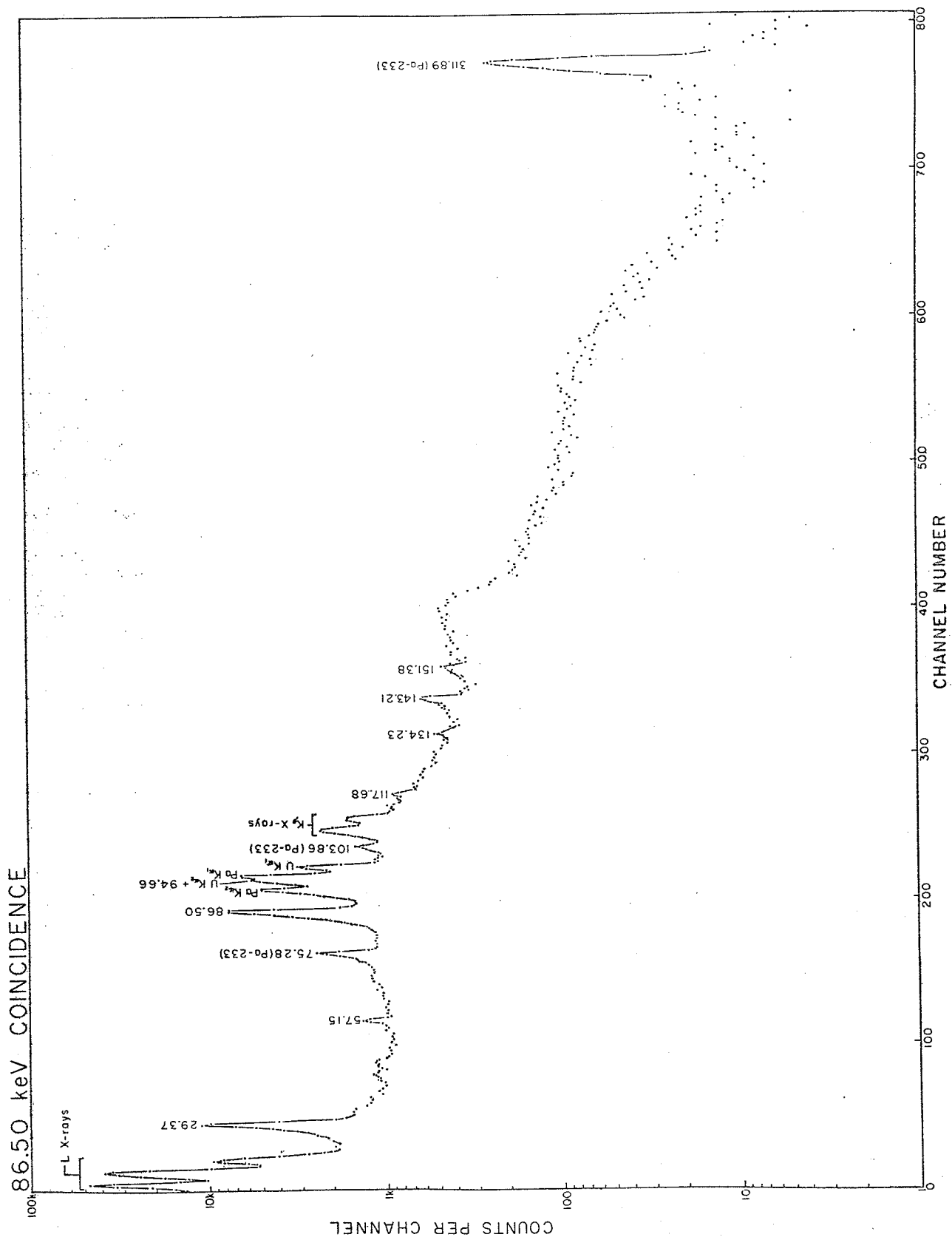


this spectrum although from the decay scheme numerous gamma rays are expected to be in coincidence. So here we have an example of a weak gating line with weak lines expected in coincidence but too weak to be seen against the background. The X-rays and the 86.50 keV gamma ray appear as random components.

86.50 keV gate:

The coincidence spectrum for the 86.50 keV line is shown in figure 19. Also falling in this gate was the 88.04 keV gamma ray from the decay of Np-237 and the 86.59 keV gamma ray from the decay of Pa-233. The 151.38 keV gamma ray feeds the 86.51 keV level and hence is expected and seen in coincidence. The 117.68 and the 143.21 keV gamma rays feed the 94.68 keV level and hence are expected to be in coincidence through the 8.17 keV transition. The singles intensity of the 131.04 keV line is greater than the 134.23 keV line. But the 131.04 keV line does not appear in the coincidence spectrum. So the 134.23 keV line is in coincidence with the 86.50 keV line. It is now necessary to introduce another crossover transition from the 103.69 keV level to the 94.68 keV level. This transition has been proposed again by Browne¹⁶ and this spectrum lends support to the hypothesis.

Figure 19
86.50 keV Coincidence



Also slightly enhanced is the 94.66 keV line. This enhancement is spurious, however. As can be seen from the decay scheme the 94.66 keV line is not in coincidence with the 86.50 keV line. When setting the gate on the 86.50 keV line it is possible that some of the $K\alpha$ X-rays will also open the gate. The 35cc detector used for gating has considerably poorer resolution than the X-ray detector. The enhancement of the 94.66 keV line is then explained as follows. Internal conversion of K electrons of the 143.21 and the 117.68 keV ^{M1} transitions will give rise to K X-rays in coincidence with the 94.66 keV gamma ray. If the K X-rays were falling into the gate the observed enhancement is expected.

By the same line of reasoning internal conversions of the 151.38 keV ^{M1} transition will populate the 86.51 keV level. If the gate opens from the K X-rays emitted after a transition populating the 86.51 keV level, the 86.50, 57.15 and 29.37 keV gamma rays are expected to be enhanced. The intensities given in the coincidence table (see below) for this gate assume the 86.50 keV line appearing in this spectrum is entirely due to randoms. But as seen here this is not completely correct, so the intensity values will be systematically too low. The 117.68 and 143.21 keV transitions will

also feed the 86.51 keV level through the 8.17 keV transition giving the same effect.

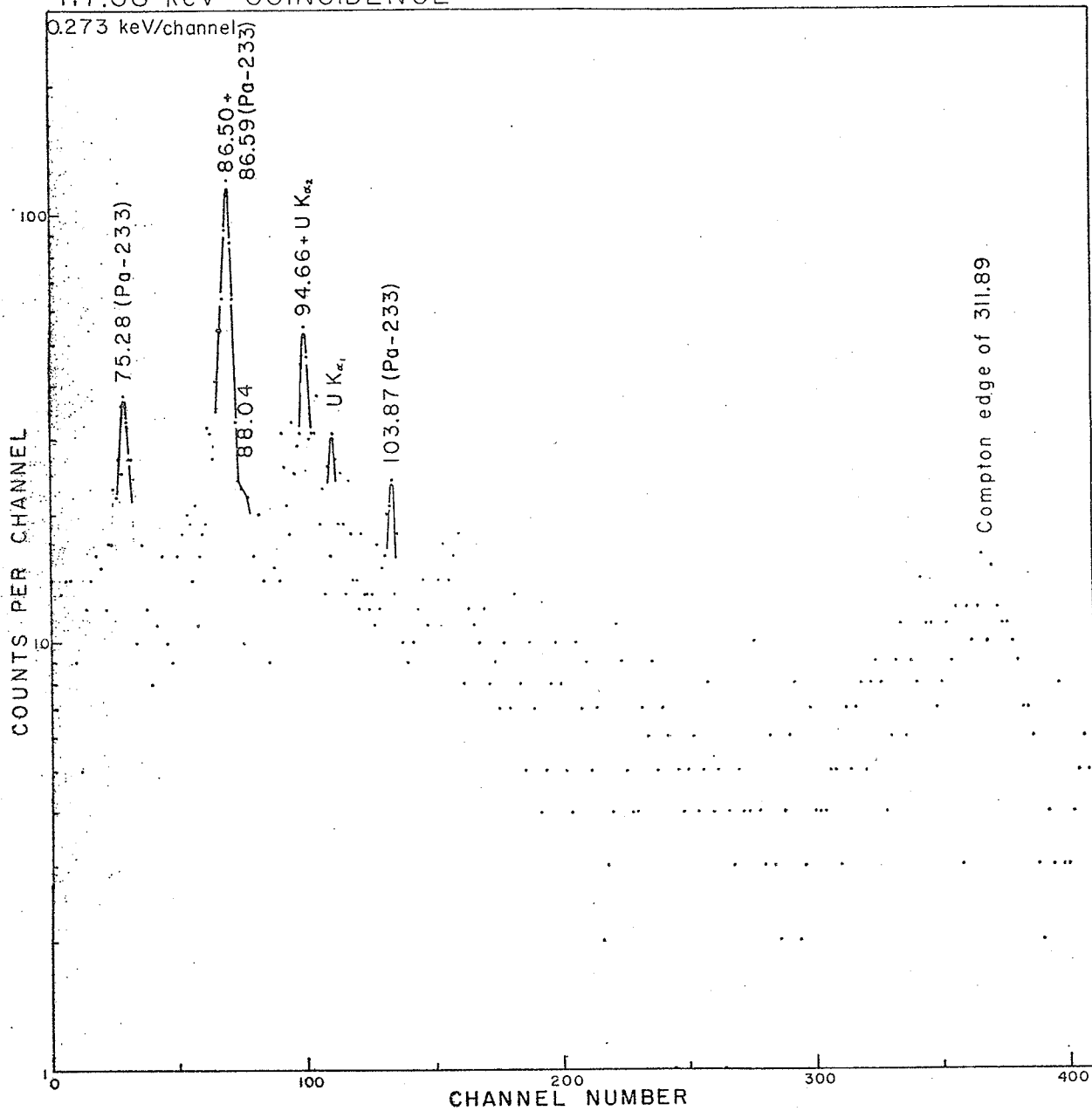
Finally the 86.59 keV gamma ray of protactinium-233 will also fall in this gate. We then expect to see and do see the 311.89 keV gamma ray of Pa-233 in coincidence, too.

117.68 keV gate:

The coincidence spectrum for the 117.68 keV gamma ray is shown in figure 20. The Compton edge for the 311.89 keV gamma ray is seen at approximately 160 keV. As explained in chapter 1 the Compton edge will arise due to the back scattering. Because Compton events of the 311.89 keV gamma ray in the gating crystal may fall into this gate, all gamma rays in cascade with the 311.89 keV gamma ray in the level scheme of uranium-233 will appear in the coincidence spectrum. Three gamma rays from the decay of protactinium-233 are seen then in this coincidence spectrum: 75.28, 86.59 and 103.87 keV. Referring to the coincidence spectrum we also see the 94.66 keV gamma ray clearly in coincidence. The 86.50 keV peak is considerably wider than in the singles spectrum. This is probably due to the 88.04 keV gamma ray being in coincidence with the 117.68 keV gamma ray. The crossover transition between the 94.68

Figure 20
117.68 keV Coincidence

117.68 keV COINCIDENCE



and the 86.51 keV levels requires that some enhancement of the 86.50 keV gamma ray be present. This is not inconsistency with the data as there is no entirely random gamma ray appearing.

143.21 keV gate:

The coincidence spectrum for the 143.21 keV gamma ray is shown in figure 21. The Compton edges of the 300.10 keV and the 311.89 keV gamma rays are present as well as the the other protactinium-233 gamma rays in cascade with them. The only neptunium gamma ray that shows a possible enhancement in this spectrum is the 88.04 keV gamma ray on the high energy tail of the 86.50 keV gamma ray. While any trace of the 94.66 keV gamma ray is obscured by the K X-rays.

151.38 keV gate:

The coincidence spectrum for the 151.38 keV gamma ray is shown in figure 22. For this measurement and all the following coincidence measurements the role of the X-ray detector and the 35cc detector was reversed. The 35cc detector was used as the gating detector and the X-ray detector was the scanning detector. The gamma rays in coincidence in this spectrum are the 29.37 keV, 57.15 keV and the 86.50 keV lines. Also

Figure 21
143.21 keV Coincidence

143.21 keV COINCIDENCE

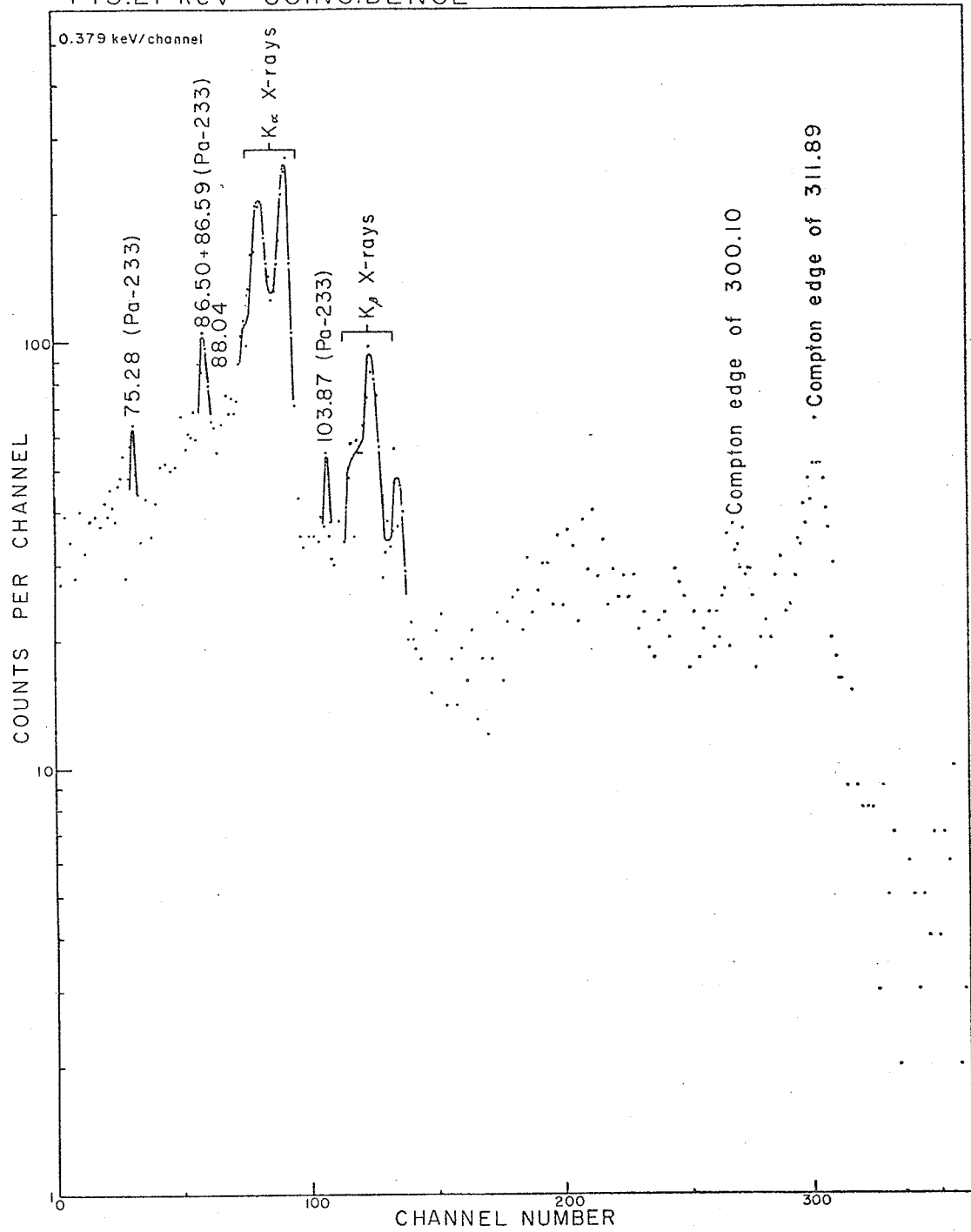
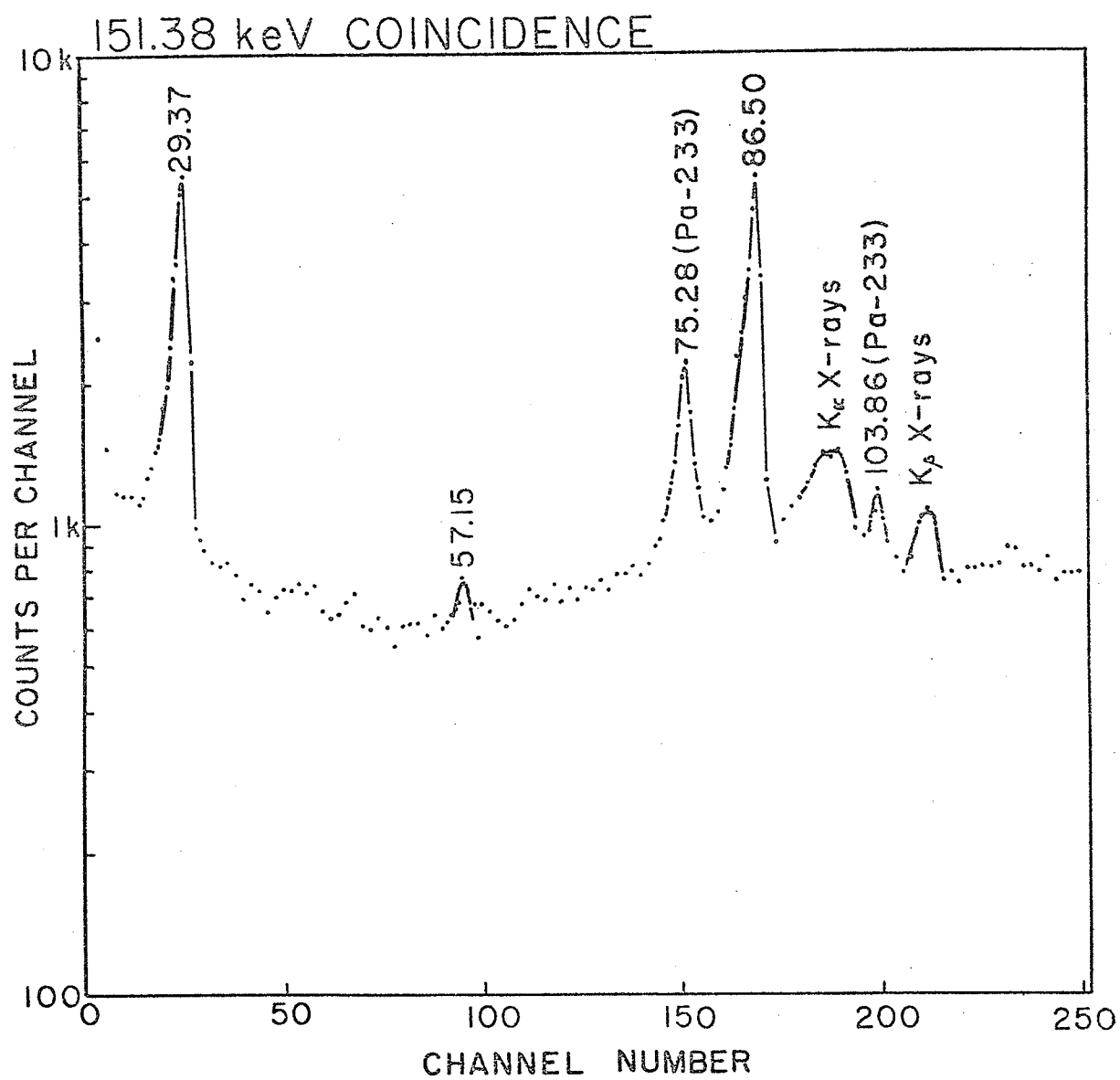


Figure 22
151.38 keV Coincidence



appearing in coincidence are the 75.28 keV and the 103.86 keV lines from Compton events in the gating detector of the 311.89 keV gamma ray in Pa-233. The 151.38 keV gamma ray feeds the 86.51 keV level giving the observed coincidences.

195.10 keV gate:

The coincidence spectrum for the 195.10 keV gamma ray is shown in figure 23. There are five gamma rays falling into this gate, from the 191.45 keV line to the 196.84 keV line. There is no enhancement of any gamma rays in this spectrum. The 29.37 and 86.50 keV lines appear as random components.

212.42 keV gate:

The coincidence spectrum for the 212.42 keV gamma ray is shown in figure 24. Also falling in this gate is the 214.09 keV gamma ray. The only peaks that show a definite enhancement are the 29.37 keV, 57.15 keV and 86.50 keV gamma rays. It also appears the 46.53 keV gamma ray may be slightly enhanced, however this may be due to summing between L_{β} X-rays and the 29.37 keV gamma ray. The enhancement of the 29.37 keV, 57.15 keV and 86.50 keV gamma rays may be explained by the 214.09 keV gamma ray which populates the 86.51 keV level. The

Figure 23
195.10 keV Coincidence

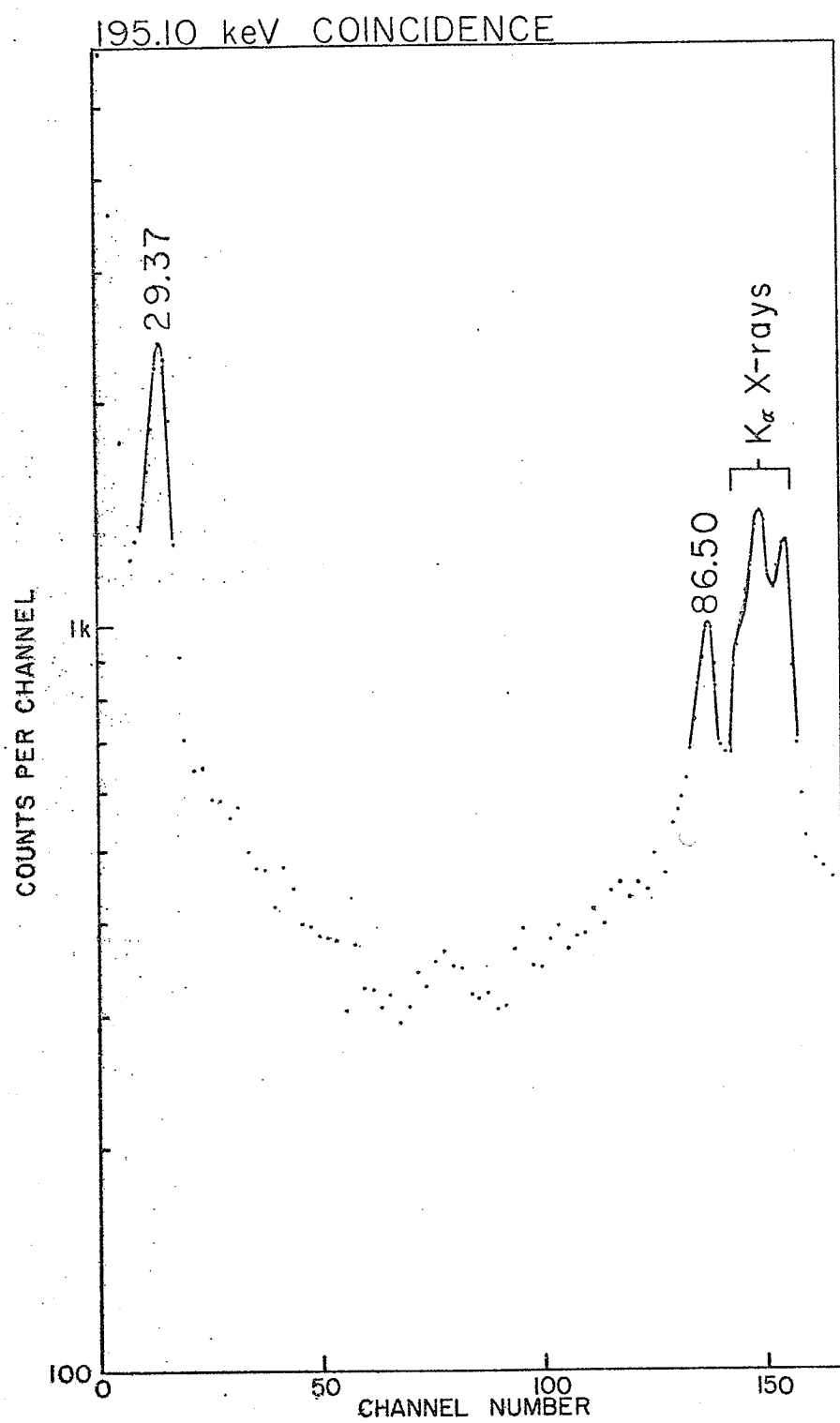
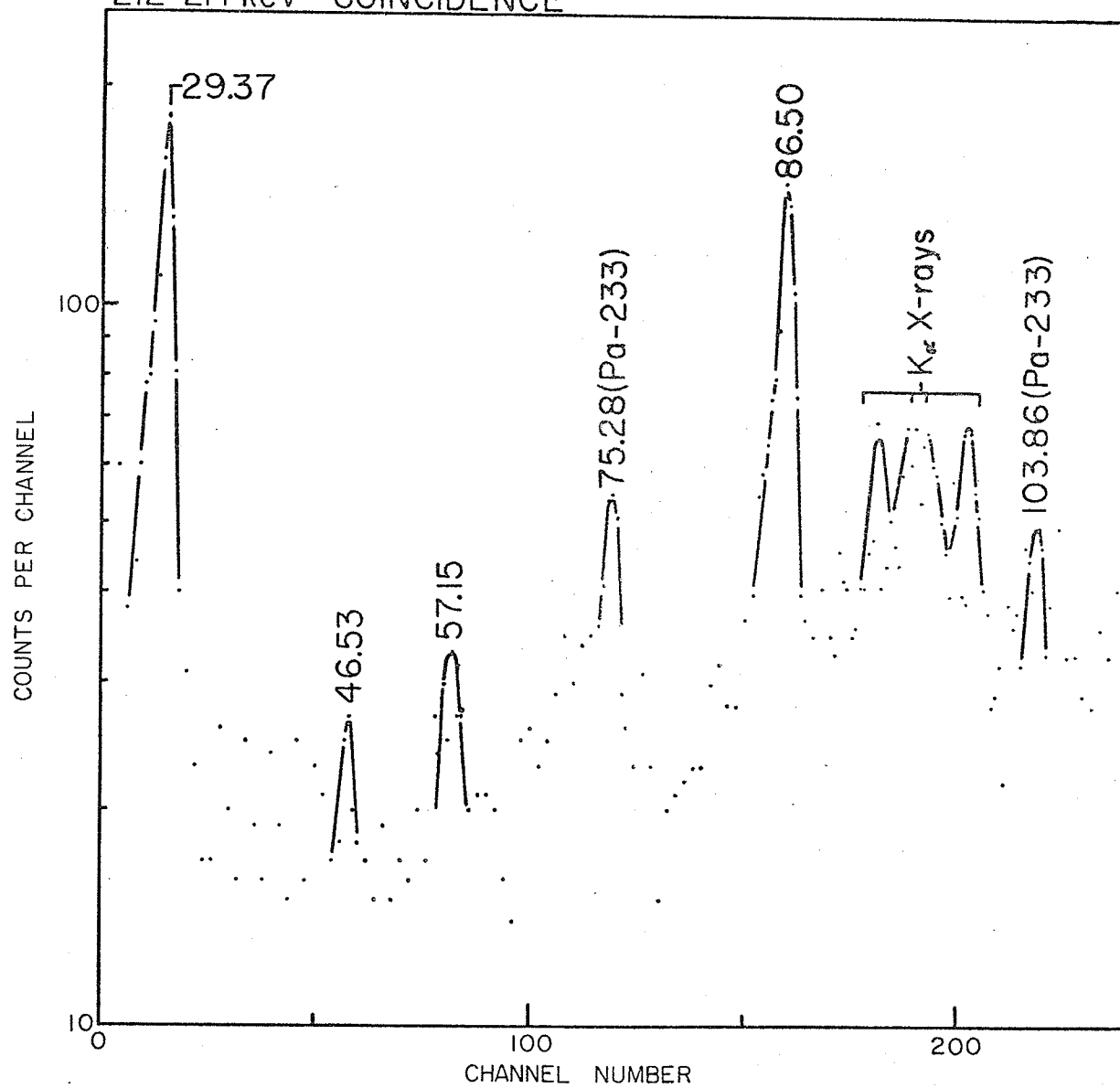


Figure 24
212.42 keV Coincidence

212-214 keV COINCIDENCE



212.42 keV gamma ray is a ground state decay and we do not observe the enhancement of the very weak 153.52 keV gamma ray feeding the 212.42 keV level.

238.04 keV gate:

The coincidence spectrum for the 238.04 keV gamma ray is shown in figure 25. Since the 238.04 keV gamma ray is a transition to the ground state, the only gamma rays expected in coincidence are those feeding the 238.0 keV level. The 62.5 keV gamma ray then is the only expected cascade. In the spectrum there does seem to be a slight enhancement at 62.5 keV but it is uncertain.

These nine coincidence spectra represent a gate on each of the most intense lines of the neptunium-237 decay. The results of the coincidence measurements are summarized in Table IV, the coincidence table. Horizontally are the various gating lines and vertically are the lines observed in cascade. The numbers in the table are the relative intensities for each coincidence spectrum with the strongest line normalized to 100. From this table and the energies of the observed gamma rays it is now possible to assemble a decay scheme for the excited states of

Figure 25
238.04 keV Coincidence

238.04 keV COINCIDENCE

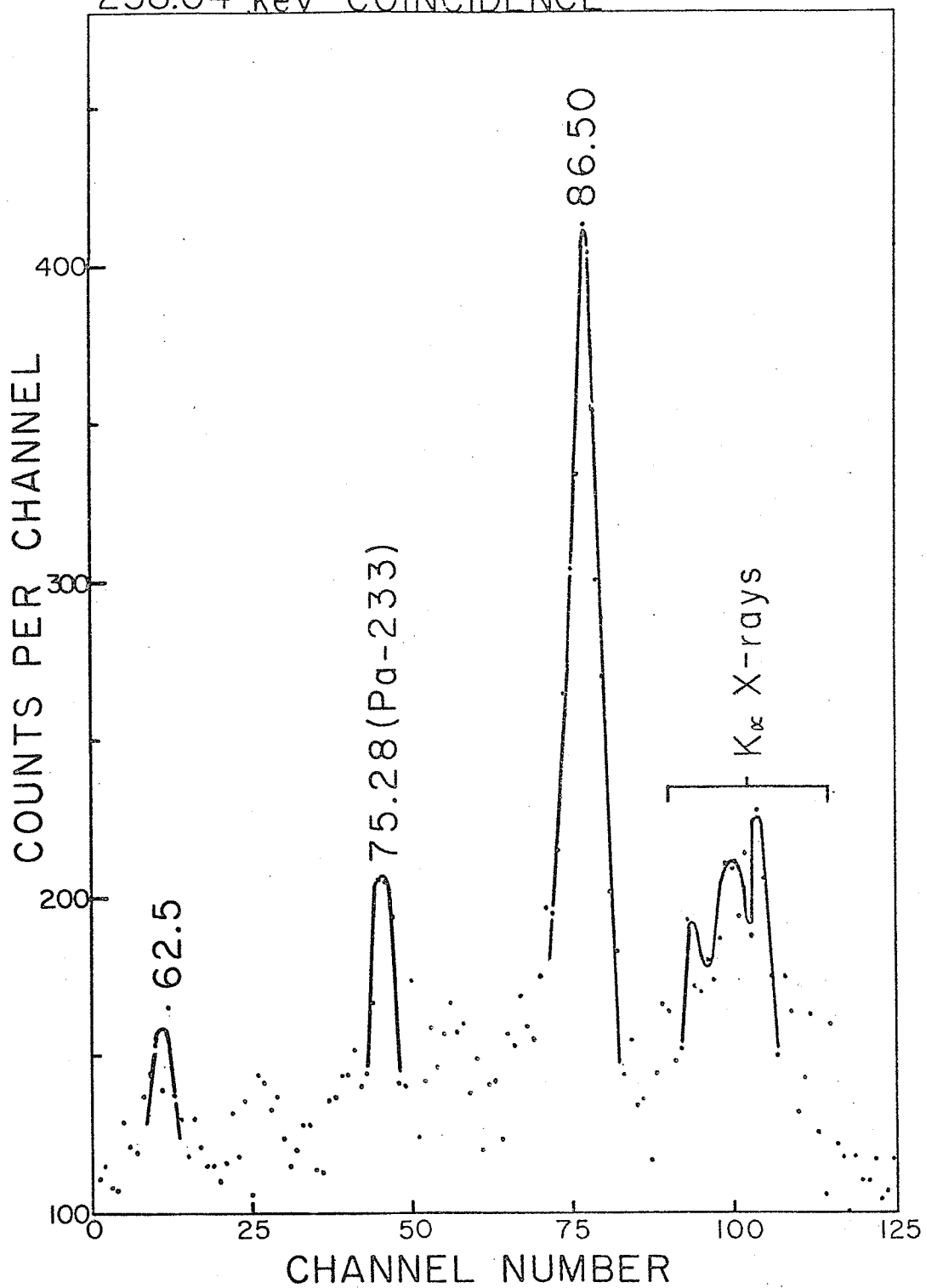


Table 4
Coincidence Results

COINCIDENCE TABLE

Cascades	Gates			
	29.37	86.50	151.38	214.09
29.37			94	100
46.53				??
57.15	95		2.4	2.7
86.50			100	79
117.68	18	16		
134.23		18		
143.21	100	100		
151.38	25	34		

Errors in this table are typically 50% or greater

protactinium-233.

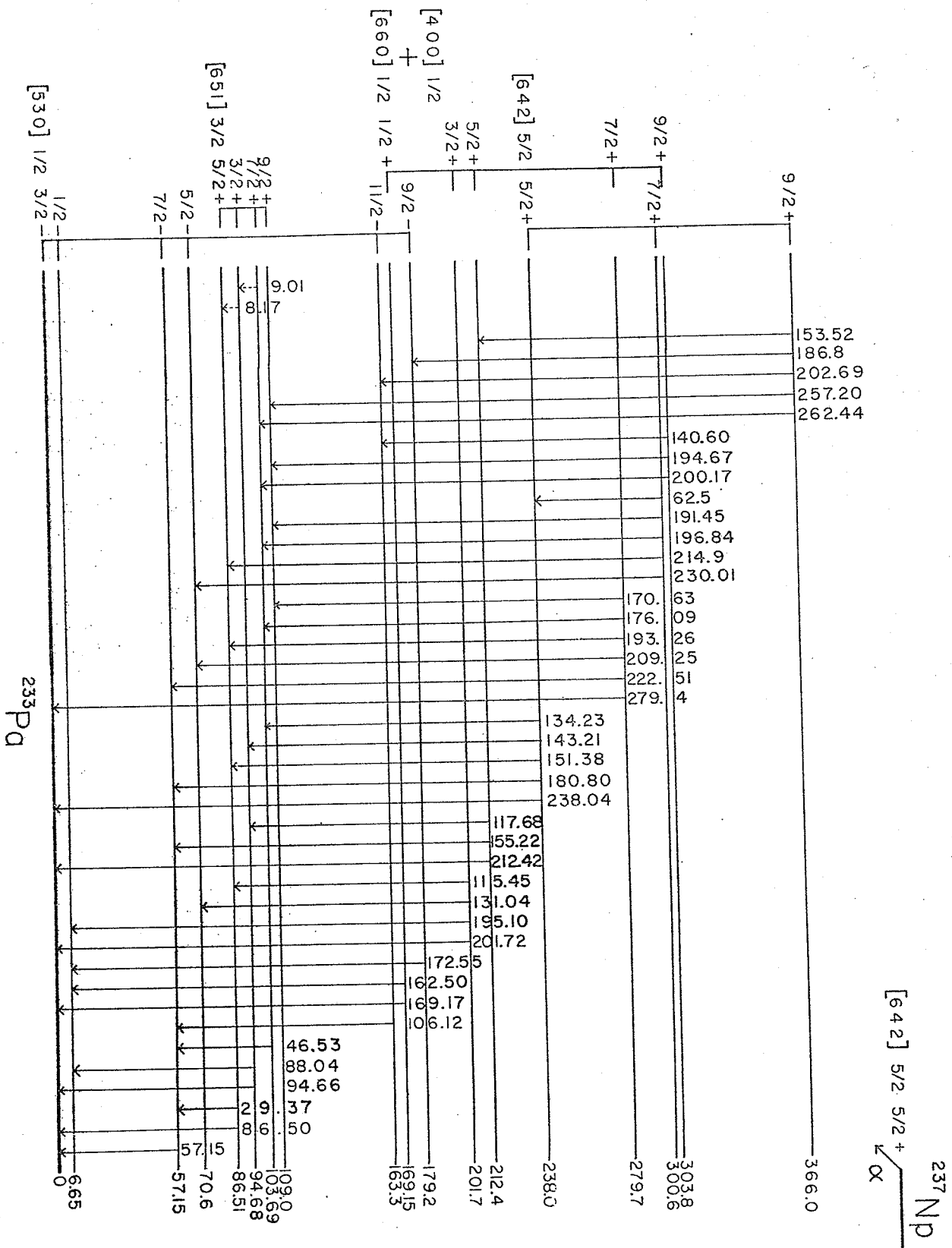
CHAPTER IV

Rotational Structure and the Decay Scheme

The success of the Nilsson model¹⁷ in describing the properties of heavy nuclei has been considerable. The heavier nuclei have a shape deformed away from the simple spherical case to an ellipsoidal shape. General interpretations of the rotations of deformed nuclei have made necessary modifications to the j-j coupling schemes of the shell model. The interpretations of these intermediate or rotational coupling schemes gives rise to the Nilsson rotational bands. The decay scheme for the decay of neptunium-237 is shown in figure 26. On the left side of the decay scheme are the Nilsson rotational band assignments for the energy levels from Browne.¹¹ The form of the Nilsson band assignments is $[Nn_z\Lambda] K I \pi$, where N is the total number of nodes in the wave function, n_z is the number of nodal planes perpendicular to the symmetry axis and Λ , the orbital angular momentum component along the symmetry axis, can take on values $N-n_z, N-n_z-2, \dots, 0$ or 1 . I is the spin of the level and K is the projection of I along the symmetry axis of the nucleus and π is the parity.

The energy levels within one rotational band are

Figure 26
Decay Scheme



given by the formula:¹⁸

$$E_I = E_0 + A[I(I+1) - K(K+1)]$$

where E_0 is the energy of the lowest member of the rotational band. This equation holds if the other modes of motion (i.e. vibrational) are negligible. In the case where the coupling of the vibrational and rotational motions is important an additional term is needed:¹⁹

$$E'_I = E_I + B[I(I+1) + C \delta_{K, 1/2} (-1)^{I+1/2} (I+1/2)]^2$$

Special consideration for the rotational bands with $K = 1/2$ is required because of the decoupling of angular momenta between bands (Coriolis decoupling.) For $K = 1/2$:²⁰

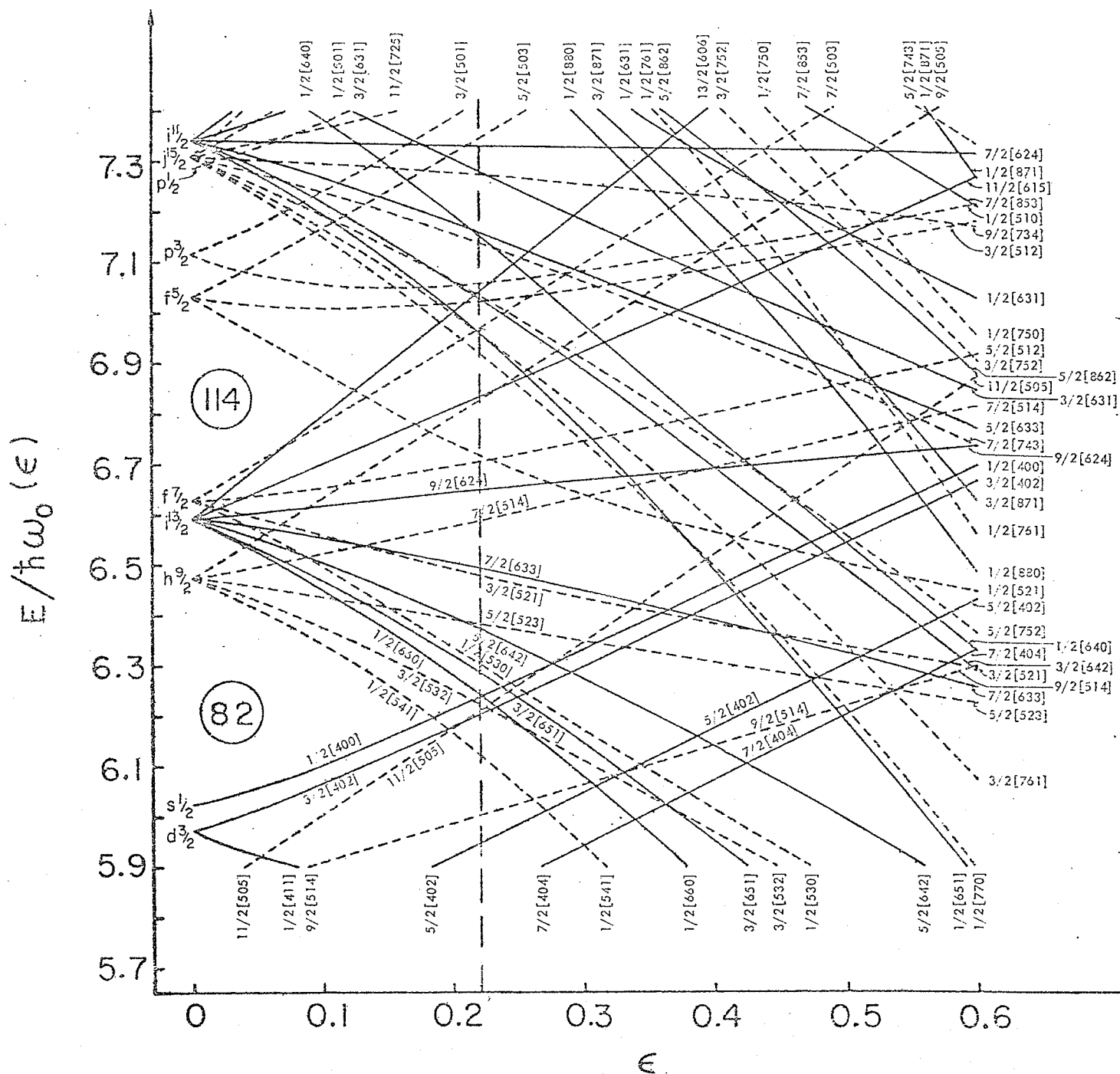
$$E_I = E_0 + A[I(I+1) - 1/2] + (-1)^{I+1/2} B(I+1/2).$$

Using these equations on the four rotational bands in the decay scheme, it was attempted to predict the energies of new excited states. At this time the level shown at 303.8 keV had not been observed. Calculating the expected energy of the fifth member of the mixed

rotational band $1/2^+ (4\ 0\ 0) + 1/2^+ (6\ 6\ 0)$ using the last equation, it was found that the $9/2^+$ member of this band should have an energy of 300.9 keV. The difference between calculated and experimental energies is less than 1% and can be considered to be in good agreement. Other possible evidence for this level comes from Baranov et. al.²¹ They observed an alpha particle group with an energy of 4.572 MeV in the decay of neptunium-237, this would correspond to a feed to a level of 305 keV.

Figure 27 shows the energy of Nilsson single particle orbitals as a function of the deformation. The particular diagram shown here is for odd proton levels in nuclei with atomic number greater than 82; this is the case for the Pa-233 nucleus with 91 protons. The amount of deformation in the Pa-233 nucleus is customarily taken to be $\epsilon = 0.22$.²² This is shown by a dashed vertical line in the figure. It can be seen that the locations of the lowest members of rotational bands in the figure does not correspond to the locations in the decay scheme. This is a consequence of configuration mixing. The last proton in Pa-233 is in the $1h_{9/2}$ shell level along with eight other protons. The Nilsson wave functions are single particle wave functions for particles outside the core.

Figure 27
Nilsson Diagram



This, however, is not the case for Pa-233, where many different mixtures of configurations can occur.

A comparison of the experimental and theoretical lifetimes of the 86.51 keV level was performed. The half-life of a level may be calculated, again assuming single particle orbitals, by studying the transitions de-exciting the level. For electric multipole transitions, the transition probability is given by:²³

$$T_{sp}^{(\lambda)} = \frac{4.4(\lambda+1)}{\lambda[(2\lambda+1)!!]^2} \left(\frac{3}{\lambda+3}\right)^2 \left(\frac{E_\gamma}{197 \text{ MeV}}\right)^{2\lambda+1} a^{2\lambda} S(I_i, \lambda, I_f)$$

where λ is the multipole order, a is the nuclear radius and S is the "statistical factor" given by:

$$S(I_i, \lambda, I_f) = (2I_f + 1) \left(\langle I_i \lambda I_f \frac{1}{2} | -\frac{1}{2} 0 \rangle \right)^2$$

where $\langle j_1 m_1 j_2 m_2 | j m \rangle$ is the Clebsch-Gordon coefficient. There are two transitions that de-excite the 86.51 keV level; the 29.373 keV and 86.503 keV gamma rays. The multipolarity of both these gamma rays is known to be E1.²⁴ The single particle Weisskopf half-life of the 86.51 keV level was calculated to be 1.5×10^{-12} sec, but the measured half-life is 3.6×10^{-8} sec.²⁵ The transition

is retarded by a factor of about 20,000 over the single particle value. This result is expected experimentally, E1 transitions in other nuclei exhibit retardations of similar magnitude, but this is not understood theoretically very well.²⁶

"K-forbidden" transitions were also studied in this investigation. A transition between bands is called K-forbidden if:

$$\Delta K = K_f - K_i > \lambda$$

where λ is the multipole order. The transition probability of a K-forbidden transition is:²⁷

$$T(\lambda) = \frac{8\pi(\lambda+1)}{\lambda[(2\lambda+1)!!]^2} \frac{1}{\hbar} \left(\frac{E_\gamma}{\hbar c} \right)^{2\lambda+1} B$$

where B is the reduced transition probability. All the information pertinent to the nuclear structure is contained in the reduced transition probability. For a K-forbidden transition the reduced transition probability is given by:²⁸

$$B(\lambda; I_1 K \rightarrow I_2 K + \Delta K) = M^2 \langle I_1 \lambda K + \Delta K - \lambda \mid I_2 K + \Delta K \rangle^2 \frac{(I_1 - K)!(I_1 + K + \Delta K - \lambda)!}{(I_1 + K)!(I_1 - K - \Delta K + \lambda)!}$$

where $\langle j_1 m_1 j_2 m_2 | j m \rangle$ is the Clebsch-Gordon coefficient and where M^2 is a constant that gives the mixing between the K 's of the initial and final states and states with $K_i + \lambda$ and $K_f - \lambda$. To eliminate the constant a ratio of transition probabilities is formed from two gamma rays depopulating the same level going to the same rotational band. The two transitions used were the 238.04 keV and the 180.80 keV gamma rays. The 238.04 keV transition was known to have a multipolarity of $E1$.¹⁶ The multipolarity of the 180.80 keV transition was assumed to be also $E1$ (spin change of one and parity change.) The ratio of the transition probabilities of the 238.04 keV to the 180.80 keV gamma rays was calculated to be 12.8. This value can be compared to the experimentally determined intensities of the two gamma rays. The experimental intensity ratio should equal the ratio of the transition probabilities. The experimental ratio was found to be 3.21 ± 0.80 . The discrepancy probably only indicates the 180.80 keV gamma ray is not a pure $E1$ transition as assumed. If we assume an $M2$ admixture the first observation is the $M2$ component is not K -forbidden. This would increase the theoretical intensity of the 180.80 keV gamma ray giving better agreement with the experimental result. Also Browne and Asaro have shown that there is

considerable Coriolis mixing in the rotational bands of this nucleus. Ignoring this mixing could also lead to the discrepancy of intensity ratios.

APPENDIX

Mathematical Smoothing of Gamma Ray Spectra

In acquiring coincidence spectra it is noted readily that there is a great deal of scatter in the contents of consecutive channels. This is especially noted in the higher energy range of a spectrum. This effect is easily explained by remembering that if a particular channel has N counts in it, the expected error is the Poisson error or the square root of N . If N is relatively small, this error is of a large magnitude compared to N . To overcome the difficulty in handling this the author has written a computer program called Smooth to do a mathematical smoothing of coincidence spectra.

Studying the literature on smoothing techniques²⁹ the author has picked a particular algorithm that is especially suited for this type of work. The smoothing is performed point by point across the spectrum, for each point a small neighborhood of points around the point to be smoothed is chosen. A least squares fit to a power function is performed in the neighborhood and a

new smoothed value for the one point is generated. This technique is called "convolution." Mathematically, the smoothed value S_i of the data D_i is found by:

$$S_i = N_m \sum_{j=-m}^m a_{mj} D_{i+j}$$

where N_m and a_{mj} are constants³⁰ depending on how large a neighborhood is selected: the neighborhood here has $2m+1$ points.

Of primary importance in this technique is the size of the neighborhood. It is found that for m less than two the procedure simply does not work. So the first possible value for m is two, this gives what is called a five-point smooth; m equal to three gives a seven-point smooth and so on. It is found that an n -point smooth works best on gamma ray photo peaks whose full width at half maximum lies between n and $n+2$ points. This criterion, the detector resolution, is then used to determine the size of the neighborhood to be used in the smoothing algorithm.

It is also noted that the resolution of Ge(Li) gamma ray detectors is a function of the energy of the photo peak. The functional relationship is that the FWHM increases as the square root of the energy. Using

this fact, smoothing of a gamma ray spectrum is accomplished by first dividing the spectrum into regions where the FWHM ranges from 5 to 7 points, 7 to 9 points, ect. and then smoothing these regions with a five-point smooth, seven-point smooth, ect. The maximum number of points that can be used in this program in a smooth is 25, making the maximum allowable FWHM 27 points. If this maximum is exceeded an error message is returned and the program terminates.

Comparing an original coincidence data set to the smoothed data set we find in all cases the intensities and positions of photo peaks remain the same within calculated experimental error. It is also generally found that the calculated experimental error for intensities and positions become smaller after the smoothing procedure is applied. Comparing an original data set with excellent statistics with the smoothed data set, several observations are made. First, having smoothed the data the peaks become wider but lower, conserving the area. The peak positions usually fall within experimental error but a χ^2 test between smoothed and original positions shows non-equivalence. The χ^2 test was used to examine the hypothesis that the smoothed peak positions were equal to the original peak positions. As was mentioned earlier, smoothing is done

by dividing the spectrum into regions. In the region where the FWHM varies between five and seven points a five point smooth is used. For peaks in this region that have a FWHM that is closer to seven than five it is more likely the smoothed peak positions will vary from the original positions. If the actual FWHM is close to the number of points used in the smooth the peak positions will be the same for smoothed and original peaks. Judging from these results the program should only be used on coincidence spectra where large errors are expected. Application of the smoothing program did level out the background into a more nearly continuous line, but did not reveal the presence of weak peaks previously unseen in the original spectrum.

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