THE APPLICATION OF TWO PARAMETER ANALYSIS IN THE SUM-COINCIDENCE SCINTILLATION SPECTROMETER

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BY

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Winnipeg, Canada.

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THE APPLICATION OF TWO PARAMETER ANALYSIS IN THE SUM-COINCIDENCE SCINTILLATION SPECTROMETER

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The existence of the 475 Kev. gamma ray transition in Ba^{134} is confirmed, and its relative intensity is measured to be $1.6\pm0.3\%$.

The 255 Kev. and 648 Kev. transitions in In^{113} are confirmed with measured relative transition intensities of $2.5\pm0.2\%$ and $0.02\pm0.01\%$ respectively. The end-point energy of the inner bremsstrahlung radiation following the decay of Sn^{113} is found to be 1.00 ± 0.15 Mev.

A previously unconfirmed cascade in Cs^{131} is established. The intensity of the 670 Kev. transition is estimated to be 0.2 to 0.3%.

TABLE OF CONTENTS

FIGURES

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ABSTRACT

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Chapter 1

INTRODUCTION

Since the discovery of radioactivity by Becquerel in 1896, systematic studies of the particles and radiations emitted by disintegrating atoms have enabled physicists to learn much about the properties and structure of the nucleus. Several models of the nucleus based largely on the results of these studies have been proposed and have met with considerable success. However, no single model yet proposed is consistent with all the information which has been collected.

Present models of the nucleus treat it as a concentrated collection of nucleons. These nucleons tend to arrange themselves, subject to certain rules, so that a minimum in the energy of the nucleus may be attained. This minimum energy state is called the "ground state".

Nuclei, in excited states, may result from the disintegration of other nuclei. It has been found both from experimental data and from theoretical calculations that only certain definite energy levels are possible. The nucleus, once excited to some particular level, seeks to return to the ground state either directly with the emission of a gamma ray, whose energy corresponds to the energy difference between the levels, or via several lower levels with the emission of several gamma rays.

The present thesis is concerned with the measurement of gamma radiation emitted from de-exciting nuclei.

In the study of the emitted gamma radiation several areas of investigation are of interest. In order to construct an energy level and gamma transition scheme for a nucleus, we require some information as to the relative intensities and energies of the emitted gamma rays. It is also desirable to do time and angular correlation studies of the gamma rays which occur in cascade when an excited nucleus returns to its ground state via some intermediate levels.

Early methods of study employed mainly the Geiger tube and the proportional counter. The Geiger tube could offer no direct energy resolution and its efficiency was very low. The proportional counter could provide good energy resolution, but was also inefficient as a detector.

The development and improvement of the scintillation spectrometer in 1949 was the beginning of a new era. This detector was able to provide a much more efficient means of counting than the previous forms of counters and could employ much higher counting rates of the order of several thousand per second. It could, furthermore, provide an accurate measure of the energy of the gamma rays. Further improvements in experimental technique and the advances made in electronic instrumentation have greatly improved the experimental situation.

The recent development of the solid-state detector, and in particular for the case of gamma rays, the lithium-drifted germanium diode, promises to be an advance comparable in importance to the introduction of the scintillation counter.

SCINTILLATION AND COINCIDENCE SPECTROMETRY

The primary importance and subsequent wide use of the scintillation counter was due largely to the fact that it was a radiation detector that could provide good energy measurement and at the same time also provide good counting efficiency. High counting rates which led to shorter counting periods could be readily employed.

In the scintillation process light is produced by the interaction of radiation with the scintillation crystal. The interaction processes are:-

i photoelectric process

ii Compton process

iii pair production process.

The light is converted to an electrical signal through the use of a photomultiplier which is, essentially, a noiseless amplifier. The magnitude of the electrical signal is subject to statistical fluctuations because of the statistical nature of the conversion processes involved. The most significant step in the conversion is the photon-to-photoelectron conversion at the cathode of the photomultiplier and this is responsible for most of the broadening of the amplitude distribution obtained. A typical peak-width of 7 to 8% is obtained for the photoelectric line with a 661 Kev. gamma ray. Each of the interaction processes leads to a characteristic portion of the scintillation spectrum.

Efficiency as a function of energy and crystal size may be found in the literature for the various separate or combined processes (Mott and Sutton 1958). Of primary importance is the photoelectric process in which the entire gamma ray is absorbed. The probability of multiple Compton collisions increases with crystal size, so that, in large crystals, complete absorption of the gamma ray energy is enhanced.

As previously mentioned a nucleus may decay to its ground state with the emission of several gamma rays in cascade. The improved speed of the scintillation spectrometer makes possible a study of these cascades. Several methods of spectroscopy utilizing two or more detectors, and requiring electronically that only those events which correspond to detection in the detectors within certain short periods of time of the order of 10^{-8}-10^{-9} seconds, be analyzed, have been employed (Siegbahn 1955). Very much information concerning the positions and in some cases the order of the excited levels may be gained using these methods.

A new method of coincidence spectroscopy was developed by Hoogenboom (Hoogenboom 1958). It is referred to as the sum-coincidence spectrometer, and in it the outputs of two matched-output detectors are fed into a linear adding circuit and a restriction that the two outputs add up to a certain energy equivalence is imposed before analysis is undertaken. Further discussion of the details of the spectrometer will follow in a later section.

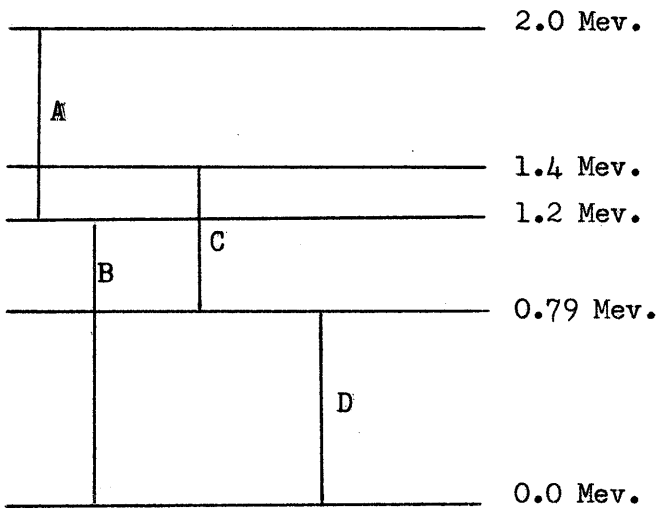
The main features of this method are that for a given cascade of gamma rays with energies A and B and a sum requirement of A+B the sum spectrum shows;

i two peaks corresponding to the full energy of the gamma rays and corresponding to the photopeaks of an ordinary scintillation spectrum

ii an absence of the Compton distributions normally found.

If in addition, the sum width be chosen to be fairly narrow, it is found that the resultant peaks are much narrower than the corresponding singles photopeaks. This feature may be of some importance when it is required to resolve gamma rays which have similar energies. An excellent summary and discussion of the characteristics of the sum-coincidence may be found in a thesis by Naqvi (Naqvi 1961).

To see the value of this method consider a simple hypothetical decay scheme as below:



We have here two cascades; gamma rays A and B, also gamma rays C and D.

An ordinary scintillation spectrum could not, without great difficulty, tell us that two gamma rays, one of energy 0.80 Mev. and the other of 0.79 Mev., exist. A simple coincidence spectrum would indicate the existence of two cascades, but would not clearly indicate which of the rays were in cascade. In the sum-coincidence method it is required that, in addition to the gamma rays being in coincidence, they add up to some particular value.

Suppose we choose the value of the sum requirement to be 2.0 Mev. We would get peaks at 1.2 Mev. and at 0.8 Mev. Neglecting for the moment, the possibility of a Compton event in one crystal appearing at some energy that might make it appear as a genuine photoelectric event, we now shift the sum requirement to 1.4 Mev. Peaks now appear at energies of 0.61 Mev. and 0.79 Mev. In this manner it is possible to determine both the energy of the gamma rays in each cascade, and the combination of rays in the two cascades.

The above discussion has, in fact, oversimplified the problem since substantial Compton-photopeak adding may occur at an energy of 1.4 Mev. We may get a Compton interaction involving gamma ray A whereby it deposits an amount of energy 0.2 Mev. in one crystal and is in coincidence with gamma B, or conversely, gamma ray B may, by a Compton

interaction, deposit 0.6 Mev. in one crystal and be in coincidence with gamma A in the other crystal. There is also the obvious extension to two Compton interactions in coincidence. Methods of recognition and removal of these interfering fictitious cascades have been developed (Schriber and Hogg 1964, Brown and Roulston 1965).

In the above method only one particular sum value has been used at one time. We have developed a method by which, with the use of a two-parameter analyzer, we may use the entire range of sum values.

ANGULAR CORRELATION

Since the introduction of the scintillation counter much effort has been spent on the study of the angular correlation between cascading gamma rays in a nuclear de-excitation. Theoretical calculations show that in the decay of a nucleus, if the first of two gamma rays in a cascade be in a particular direction, then the second may have a preferred direction of emission, i.e. the directions of emission are not randomly orientated. The calculations show further, that this directional correlation is dependent on the spins of the levels between which the particular transitions occur. Knowing then from experimental work the correlations between the cascading gamma rays, some information about the spins of the excited states from which the emissions occur may be learned.

The first substantial theory on angular correlation was published by Hamilton in 1940 (Hamilton 1940). Several years later a somewhat more generalized theory was presented by Goertzel (Goertzel 1946). In the period between these two presentations experimental work on angular correlation was not in too satisfactory a state. The counting rates that could be obtained by using Geiger counters and the lack of electronics which might provide short resolving times greatly hindered experimental progress and lengthened the counting times required.

The first experimental evidence of the existence of an angular correlation between coincident cascading gamma rays was produced by Berringer (Berringer 1943) in his work on Chlorine-38. Several other materials that he investigated did not seem to agree with the results that were to be expected from the existing theories. Deutsch and Brady confirmed in 1947 in their work with Geiger tubes the predicted values for several radioactive nuclei including Cobalt-60 and Sodium-24. They repeated their work in 1948 with the then newly introduced scintillation counters and found the results of the two studies to agree (Deutsch and Brady 1947, 1948).

About this time, largely through the work of Racah and Lloyd, the theoretical aspects of angular correlation were unified with group theory. Biedenharn and Rose published a lengthy paper in 1953 which formulated the mathematical aspects of the theory (Biedenharn and Rose 1953).

Some question as to the value of the theory arose due to a lack of agreement between the theoretical values predicted for the correlation functions and the experimental values obtained. Feingold and Frankel proposed that the lack of agreement was due to an inaccurate evaluation of the experimental data (Feingold and Frankel 1955). They were able to show that certain corrections to the experimental data need to be made to account for the finite sizes of both

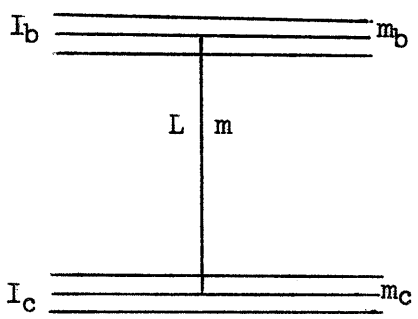
the source and the detectors used. McGowan applied their correction factors to the well known cascade which occurs in Nickel-60 following the decay of Cobalt-60, and found very good agreement between the experimental values and the theoretically predicted values.

The theory of angular correlation varies in complexity from the first theory of Hamilton to the highly mathematical theory formulated by the later workers such as Racah. We will present here a somewhat simplified, but still sufficient, treatment of the theory.

Let us consider a single gamma transition of angular momentum $\underline{L}$ between two nuclear levels B and C with respective spins I_b and I_c. For conservation of angular momentum we require $\underline{I}_b = \underline{I}_c + \underline{L}$. Let L_z be the projection of $\underline{L}$ on an arbitrary Z axis. The emitted gamma ray is said to be of multipolarity 2^L and has a magnetic quantum number m. The states B and C may be characterized by the quantum numbers I_b, m_b, and I_c, m_c respectively. Note that having chosen an axis in space we require, in addition,

$$m_b = m_c + m.$$

Consider the figure below:-



Each component $m_b - m_c$ of the transition between given magnetic states has a particular directional distribution, $F_L^m(\emptyset)$ which is independent of I_b and I_c and where $\emptyset$ denotes the angle between the direction of emission and our chosen Z axis. This distribution can be calculated directly from the Poynting vector for multipole radiation (Preston 1962).

In parallel to atomic spectroscopy, it might be expected that a Zeeman-like effect should occur in these nuclear transitions. Even in very strong magnetic fields, however, for nuclear spectroscopy the splitting is of the order of 10^{-8}ev., and as a result cannot be detected.

In order to calculate the directional distribution $F_L^m(\emptyset)$ of the unresolved line B - C, we need to know, in addition, the relative populations $P(m_b)$ for each sublevel and the relative transition probabilities $G(m_b, m_c)$ for each of the sublevel transitions. The directional distribution is then given by:

$$F_L(\emptyset) \sim \sum_{m_b m_c} P(m_b) G(m_b, m_c) F_L^m(\emptyset)$$

Let the absolute transition probability be considered as the product of a nuclear and a geometric factor, the nuclear dependent on physical properties, and the geometric which, due to our choice of Z axis, will contain the quantum numbers m_b and m_c. The nuclear factor will clearly enter as a constant.

The geometric factor on the other hand is not a constant but is dependent on our choice of the Z axis, and yields a transition probability factor of form

$$G(m_b, m_c) = (I_c L\ m_c m \mid I_b m_b)^2$$

where the right hand side of the equation corresponds to the Clebsch-Gordan coefficient for the vector addition of $\underline{I}_c$ and $\underline{L}$, and the scalar addition of m_c and m.

We need now consider the relative populations. They depend on the energies of the different magnetic states and on the manner in which level B was created. The dependence on the method of creation arises from the fact that if the nuclei are randomly orientated, then $F_L(\emptyset)$ must be independent of angle.

Let us now consider a particular cascade consisting of two gamma rays emitted in the de-excitation from level A to level B, and then to level C. Let the multipolarities of the successive gamma rays be L_1 and L_2 respectively. We seek now to determine an angular or directional correlation function $W(\emptyset)$ between the two gamma rays. Let us choose our axis of quantization to be along the direction of emmission of the first gamma ray. $W(\emptyset)$, the directional function between the two gamma rays, then becomes the same as the function $F_L(\emptyset)$ of the second gamma with respect to the emission axis. Once the relative populations are known, we can then determine the correlation function. The population

of the magnetic state, m_b, is given by the sum of all the transitions, $m_a - m_b$, leading to the state m_b. Assuming now that all the m_a states are equally populated and if

$$m_1 = m_a - m_b, \text{ then } P(m_b) = \sum_{m_a} G(m_a, m_b)\ F_{L_1}^{m_1}(\emptyset=0)$$

This choice of Z axis introduces a simplification in that a photon propogated in a definite direction can carry in that direction of motion only the angular momentum $\hbar$.

Combining now our above equations and letting $m_2 = m_b - m_c$ we have

$$W(\emptyset) \sim \sum_{m_a m_b m_c} (I_b, L_1, m_b \pm 1 \mid I_a, m_a)^2 F_{L_1}^{\pm 1}(0)\,(L_c L_2 m_c m_2 \mid I_b m_b)^2 F_{L_2}^{m_2}(\emptyset)$$

This equation was first derived by Hamilton and was worked out assuming dipole and quadrupole radiations. Calculation of the various functions required in his formulation was found to be very tedious and complicated for all but the dipole and quadrupole radiations. Later work by Racah, Lloyd and Alder led to an expression of the function by the equation:

$$W(\emptyset) = 1 + A_2 P_2(\cos \emptyset) + \ldots\ldots\ldots\ldots A_k P_k(\cos \emptyset)$$

where P_n are the associated Legendre Polynomials in cosine $\emptyset$, and k is the maximum value which is determined by the rule

$$k = \text{Min. } (2I_b, 2L_1, 2L_2)$$

The coefficients A_n are given by the expressions

$$A_n = F_n(L_1 I_a I_b) F_n(L_2 I_c I_b)$$

which can readily be found evaluated in the literature (Siegbahn 1955).

Chapter 2

APPARATUS

System

The basic operation of the sum-coincidence spectrometer was earlier indicated. This brief description will be expanded here.

The schematic block diagram of a normal sum-coincidence spectrometer is shown in Figure 1a. Pulses from two scintillation detectors after having been equally amplified are fed into a linear adding circuit. The adder circuit puts out a pulse proportional in size to the energy sum of the detected gamma rays. This pulse is then fed into a single channel analyzer which is set to accept some particular sum energy. If the required sum value is attained the single channel analyzer gates a pulse height analyzer, and analysis of the pulse from one of the detectors takes place.

In order to distinguish between possible events where two gamma rays are detected in one detector within its resolving time and events whereby one gamma ray is detected in each crystal, a fast coincidence circuit may be introduced. In this case analysis takes place only when two conditions are satisfied. Firstly, the above single channel analyzer sum condition must be met, and secondly, the fast coincidence circuit must receive a pulse from each detector within a time period of the order of 40 nanoseconds. Gating of the pulse

Figure 1

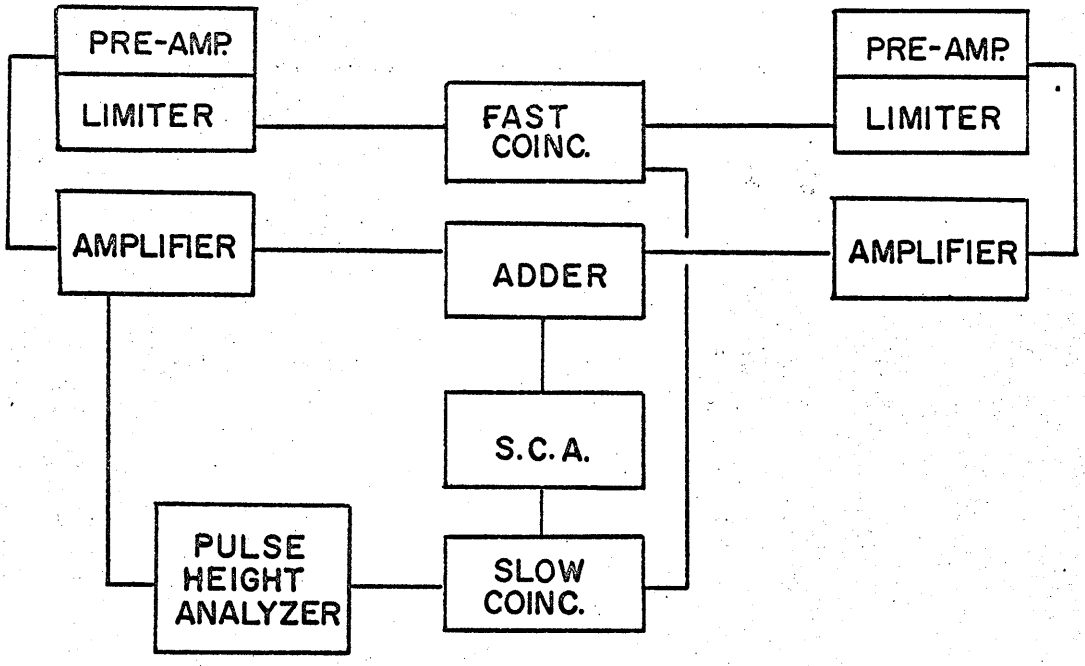
Schematic Block Diagram

a - Single Parameter

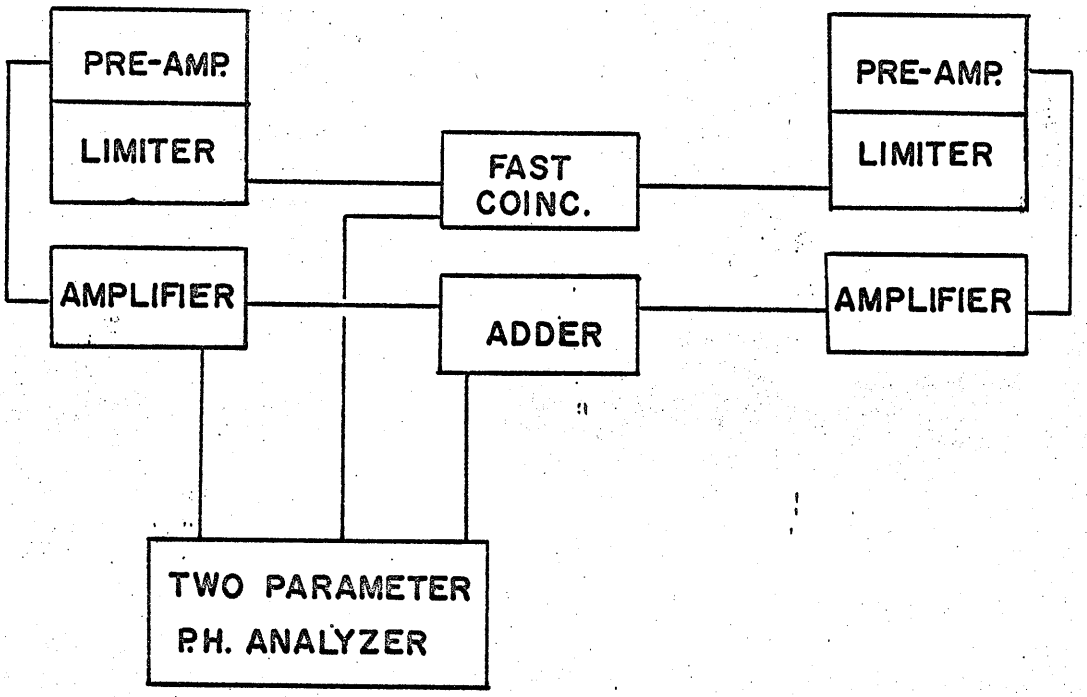
b - Two Parameter

SUM COINCIDENCE METHOD

SINGLE PARAMETER OPERATION



TWO PARAMETER OPERATION



height analyzer is achieved by means of a slow coincidence circuit.

Spectrometers of this type have been used widely since their introduction in 1958 by Hoogenboom. An extension to three detectors can easily be seen for triple coincidence studies. It may be noted that at any one time only one particular sum value may be employed. In the decay of nuclei with several levels and corresponding cascades, this may be a serious restriction if the cascades are weak in intensity, or if the life of the isotope is short.

The recent development of multi-parameter analyzers has allowed the introduction of an arrangement whereby analysis may be performed for all sum values at one time. Figure 1b shows in block form such an arrangement. Pulses from one of the detectors are fed into one parameter as in the previous system, and those from the adder circuit are fed into the second parameter. Gating of the analyzer is achieved by use of the fast coincidence circuit. In this method all gamma rays which are in fast coincidence are analyzed. This permits much more rapid acquisition of experimental data.

Electronic Circuitry

A detailed description of the electronic circuitry may be found elsewhere (Brown 1964) and will hence not be repeated here. The detectors employed are Harshaw (NaI(Tl)) integral line detectors. One of the detectors has a

$1\frac{3}{4}$" diameter by 2" crystal coupled optically to a 2" R.C.A. photomultiplier tube type 8053 while the other has a 3" by 3" crystal coupled to a 3" photomultiplier type 8054. The use of two different sized crystals is very convenient in distinguishing between Compton peaks and genuine photopeaks in a sum-coincidence spectrum and an enlargement on this property may be found in the literature (Brown and Roulston 1965). The power supply is a Hamner unit with slight changes made to improve stability. The pulses obtained from the photomultipliers are positive and are fed into a charge-sensitive pre-amplifier which was designed in this laboratory. The pre-amplifier and a timing pulse shaper of design of Birk et al (Birk et al 1961) are contained in a header unit built as a base for the photomultiplier. This unit, as well as the amplifiers and logic circuits, are all transistorized.

The amplifiers used are of a design by Chase and Svelto of Brookhaven National Laboratory (Chase and Svelto 1961). Some slight alterations have, however, been introduced to make their use more flexible. By simply changing a D. C. balance on two of the amplifier cards and shorting out a third card, the units may be used as linear double delay line amplifiers capable of pulses up to 10 volts, or as wide band low gain amplifiers. The remaining units are of a design by Goulding and McNaught of Chalk River Nuclear Laboratories (Goulding and McNaught 1960).

Particular care needs to be taken in signal matching for the sum-coincidence method. The pulses which the adder circuit receives from the two amplifiers must be accurately matched, or distortion of the spectrum display results. To achieve this balance all signal cables are suitably terminated, and pulses from the separate detectors, as seen by the adder circuit, are matched to within a small fraction of an analysis channel. Care needs to be taken so that balance is achieved in the exact experimental configuration since the amplification of the photomultipliers may be influenced by stray magnetic fields. A source is brought near one of the detectors, and proper shielding is used to prevent the radiation from also entering the second detector. The position of one particular gamma ray photopeak on a pulse height analysis of the adder output is noted. The roles of the two detectors are then reversed and proper adjustment of the amplification of the second detector is then made to allow the position of the gamma ray peak to appear in the same position, as for the first detector.

Shielding

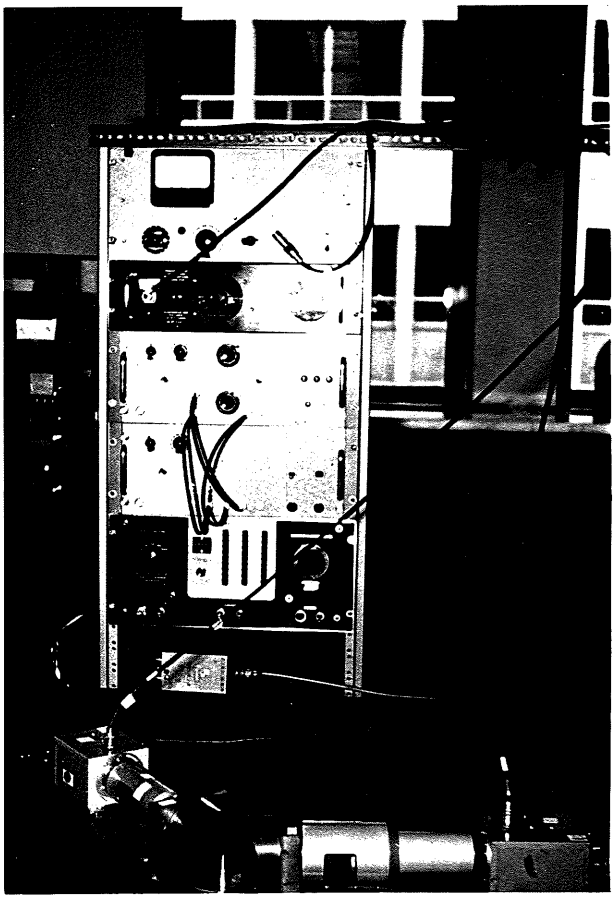
A prominent part of a scintillation spectrum which increases in size with increasing energy is the broad Compton distribution. It arises due to events where a gamma ray suffers a Compton collision in the crystal and then escapes thereby losing only part of its initial energy in the crystal.

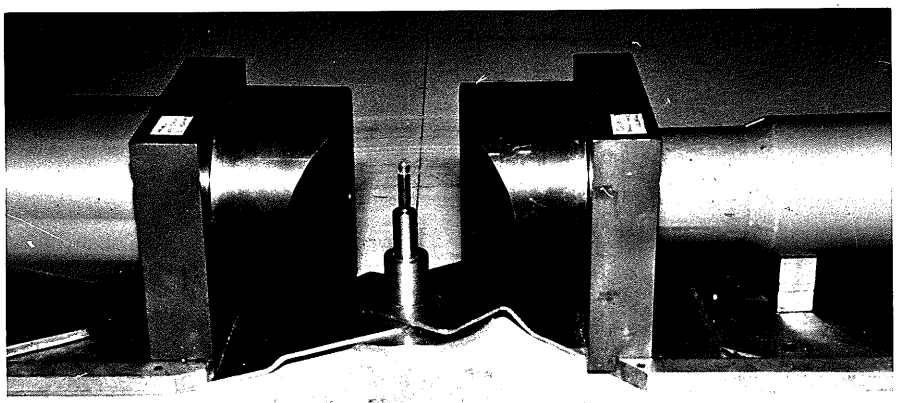
Figure 2

Apparatus

a - Control Panel and Detectors

b - Shields





Such events are most probable for gamma rays which strike a scintillation crystal near its edges. Those gamma rays which strike the crystal near its centre frequently undergo further collisions and may lose their entire energy thereby giving rise to pulses in the photopeak. Since the Compton part of the spectrum is of little use, and is, in fact, a hindrance, steps are taken whenever possible to reduce it.

Lead shields, three inches in thickness, have been constructed for this purpose. (Figure 2). They have a tapered hole bored in them which is focused for a source distance of 10 cms. and exposes only a $1^{1}/3$ inch diameter circular portion at the centre of the crystal face to the radiation.

The effect of this shielding on a spectrum obtained from a 3" by 3" scintillation crystal may be seen in Figure 3. The familiar gamma ray spectrum of Na^{22} has been taken both with and without the shield at a distance of 10cms. and normalized for peak height of the 1.28 Mev. photopeak. The decrease in the Compton distribution is very noticeable, it being of the order of 20% for the 1.28 Mev. gamma ray and about 60% for the 0.511 Mev. gamma ray. The improvement in the relative photopeak efficiency of the higher energy gamma ray as compared to the lower energy one is very clear. Taking account of the fact that there are two 0.511 Mev. gamma rays for each 1.28 Mev. gamma ray, the ratios of 1.28 Mev. to 0.511 Mev. photopeak areas with and without the lead shield are

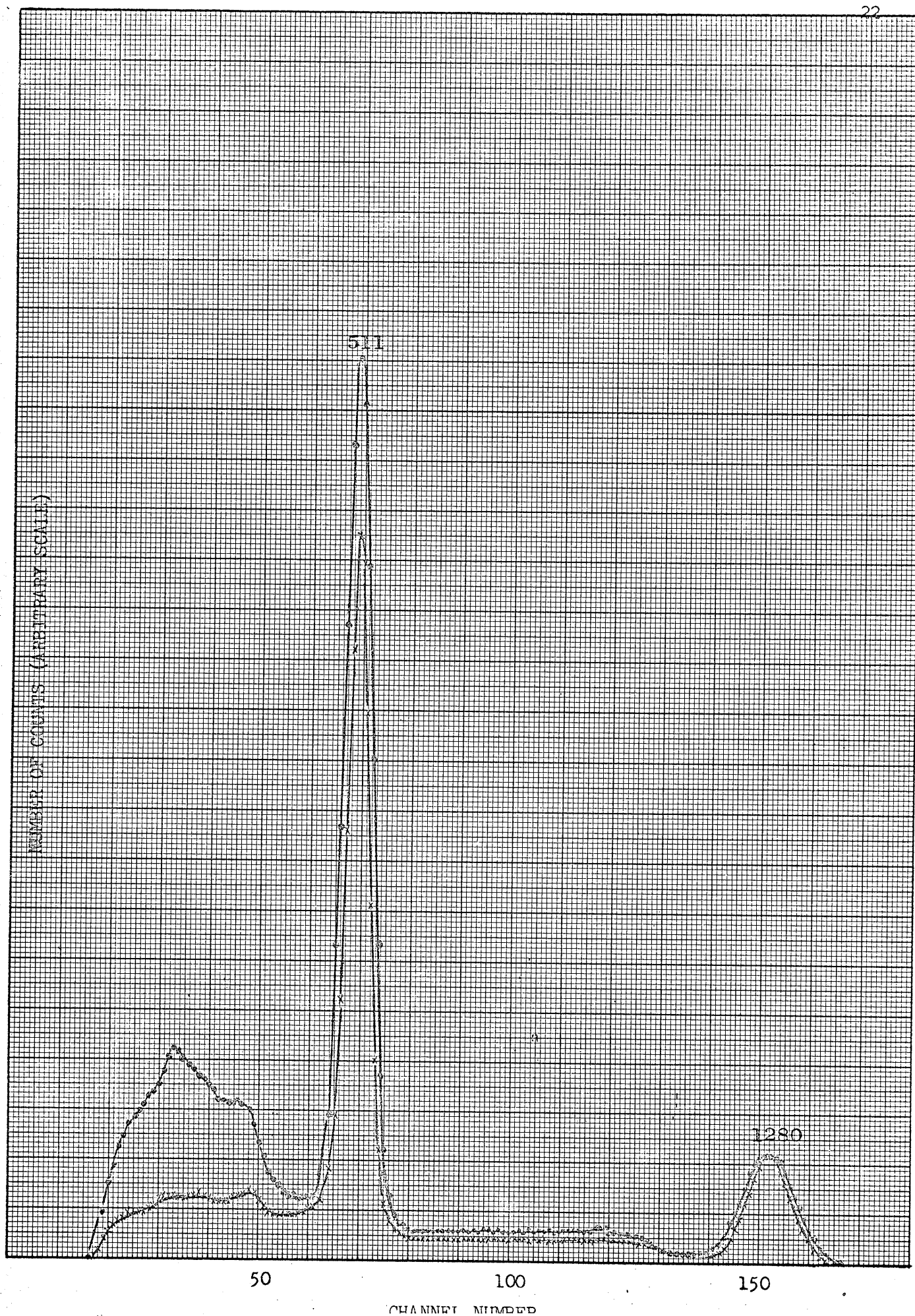
Figure 3

Shielding Effect on Compton Reduction

Energies in Kev.

•———• Without Shields

x———x With Shields


511
1280
NUMBER OF COUNTS (ARBITRARY SCALE)
50
100
150
CHANNEL NUMBER

respectively 0.41 and 0.29. This reduction of the Compton contribution improves the prominence of the photopeaks in a sum-coincidence spectrum.

Two Parameter Analysis

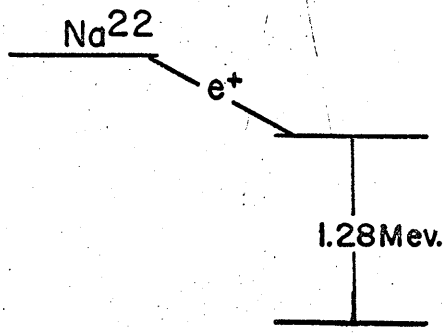
The two parameter analyzer used is a Nuclear Data ND-160 4096 Analyzer System manufactured by Nuclear Data Inc. Illinois. Inputs of various sizes from external amplifiers, or an internal amplification system, may be employed. External gating is readily available. For this project low level pulses from a wide band low level amplifier were used along with the internal amplifiers of the analyzer. For two parameter operation the 4096 channels may be programmed in several forms, with the 64 channel by 64 channel arrangement being found most convenient for our sum-coincidence studies. Display of the contents of the memory on a cathode ray screen may also be of several forms with the contour and isometric displays being of most value for two parameter studies.

In order to see what shape of spectra we might expect, consider Figure 4 which shows three one parameter sum-coincidence spectra of Na^{22}. Na^{22} is a positron emitter which decays into the first excited state of Ne^{22}. This decay is followed almost instantaneously by the decay of Ne^{22} to its ground state with the emission of a 1.28 Mev. gamma ray.

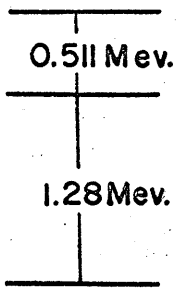
Figure 4

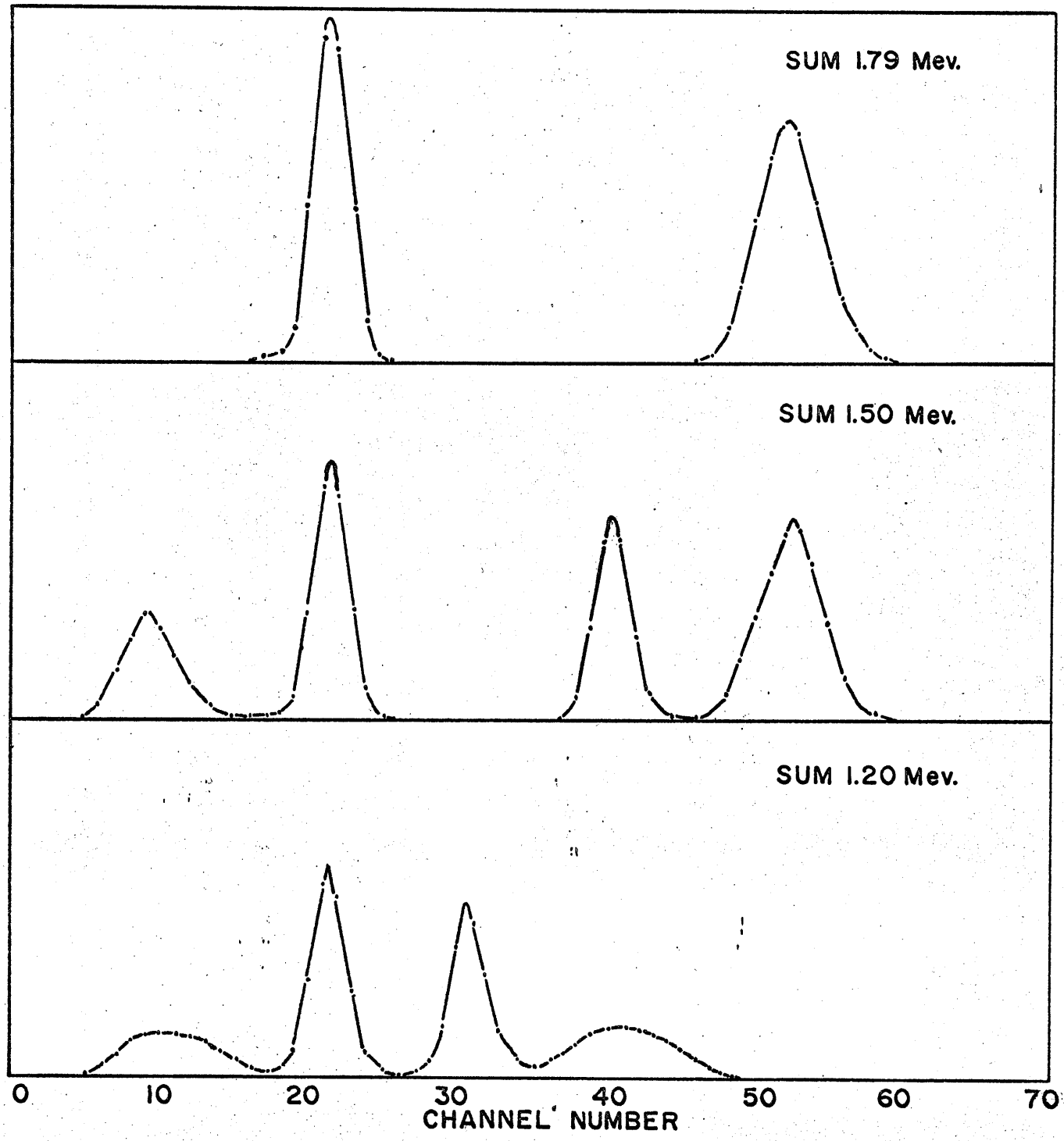
Single Parameter Sum-Coincidence Analysis of Na^{22}

SUM COINCIDENCE SPECTRA of Na^{22}



EQUIVALENT DECAY





An equivalent decay is as shown. Suppose a sum-coincidence spectrum is taken with a sum value of 1.79 Mev. Peaks at 1.28 Mev. and 0.511 Mev. corresponding to the photopeaks of the two gamma rays are to be expected, and are found. Let the sum value be reduced now to 1.50 Mev. and a spectrum taken. Photopeaks at 0.511 Mev. and 1.28 Mev. are again found, along now with their corresponding Compton summing compliments of 0.99 Mev. and 0.22 Mev. Finally let a sum value of 1.20 Mev. be chosen. Since this is too low an energy value for the 1.28 Mev. photopeak, it no longer appears. The 0.511 Mev. photopeak is still allowed, however, and appears with its sum compliment of 0.69 Mev. Some Compton-Compton addition may also be noted. The expected peak area asymmetry for two different sized scintillation crystals is also quite clear.

The 0.511 Mev. photopeak can be seen to retain its channel position in all three of the spectra. The 1.28 Mev. peak does so as well until it disappears for the 1.20 Mev. sum case. The Compton sum compliments, as seen most clearly in the second and third spectra for the 0.511 Mev. compliments, move in position to the left. In the two parameter study where many sum values are used a similar behaviour for photopeaks and Compton contributions is to be expected.

Chapter 3

SCINTILLATION SPECTROMETER STUDIES

SODIUM 22

The decay of Na^{22} has been very completely investigated by various experimenters. Because of the relative simplicity of its gamma ray spectrum it lends itself very well as a calibration and test source. Its use in the present case is as a test source for the two-parameter method of sum-coincidence analysis. The equivalent decay of Na^{22} and the forms of sum spectra expected were discussed in the previous section in conjunction with Figure 4.

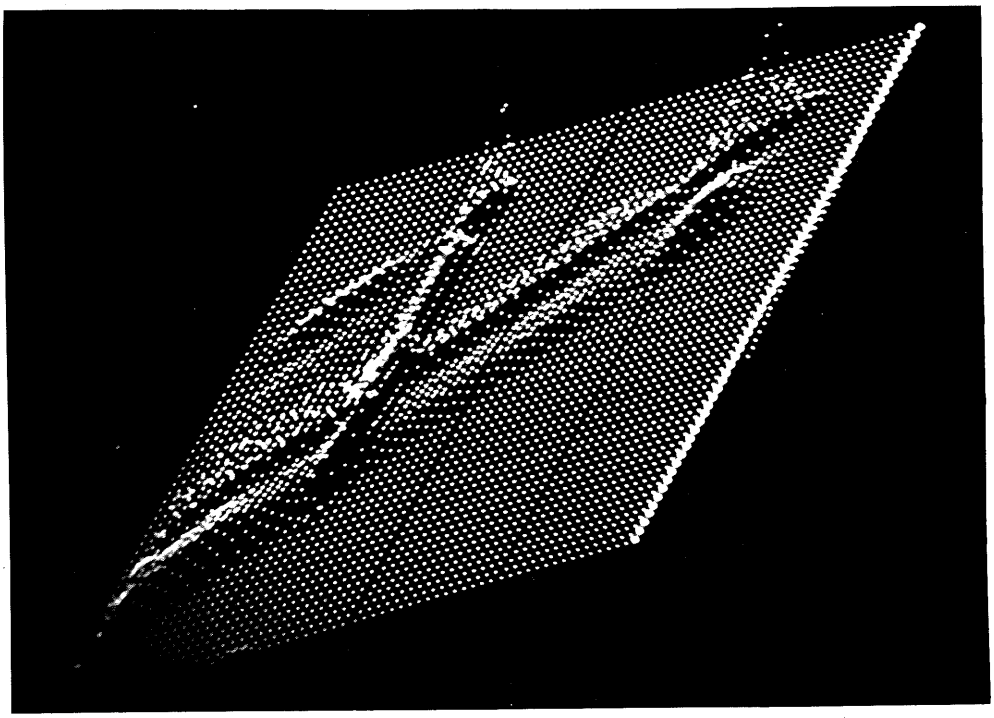
The two-parameter sum spectra as displayed on the cathode ray tube screen of the analyzer are shown in Figure 5. To discuss the displays here and in further spectra, let us define a three dimensional coordinate system. Let the lower left-hand corner of the spectrum be the origin with the X axis along the bottom of the spectrum and the Y axis running up the display. Counts are then in the Z direction. Under this definition of coordinates, the sum direction is the Y axis while the X direction defines the spectrum of pulses taken from one of the detectors.

Figure 5

Two Parameter Na^{22} Sum-Coincidence Spectral Display

a - Isometric Display

b - Contour Display



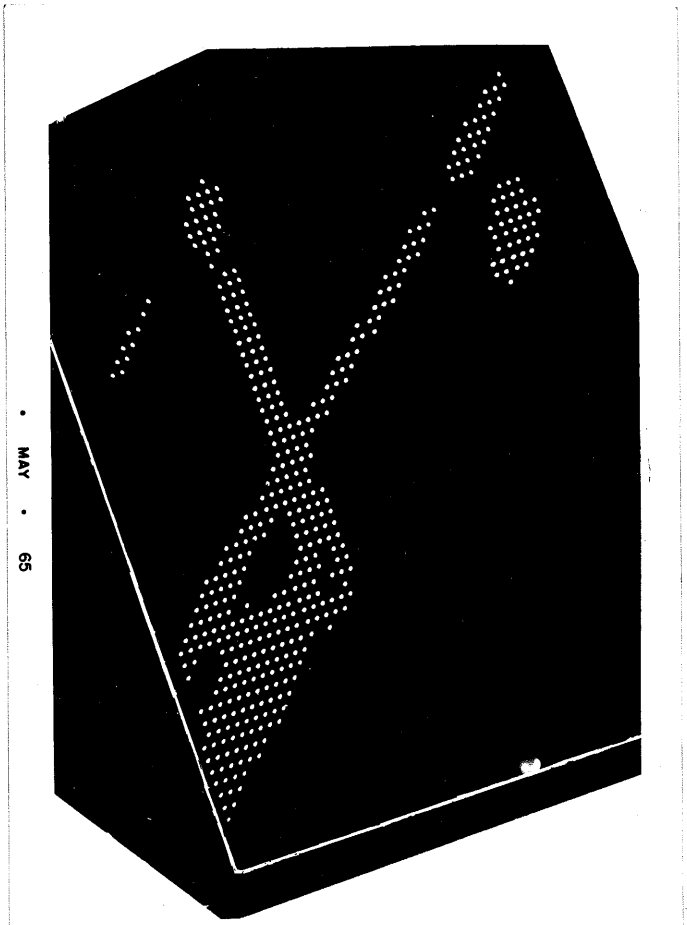

• MAY • 65

Figure 5a shows the isometric display of the Na^{22} sum-coincidence spectrum. Directions of increasing X and Y correspond to increasing singles and sum energies respectively. As previously indicated, the photopeaks of the gamma rays are expected to occupy the same X direction channel number for consecutive sum lines, while the Compton sum compliments are to move in the direction of decreasing X with decreasing sum energy. These features are very clearly visible on the display. The photopeaks appear as ridges parallel to the Y axis while the Compton portions are angled ridges. The sum values of 1.79 Mev. and 1.28 Mev. corresponding to the maximum sum value and the expected disappearance of the 1.28 Mev. photoridge are easily distinguished. Similarly a sum value of 1.02 Mev. corresponding to the crossing of the .511 Mev. photoridge with its compliment Compton ridge may be easily located. The lower sum values display the Compton-Compton addition.

Figure 5b is the contour display of the same sum spectrum. As in the isometric display, the photo and Compton ridges may be very easily identified. Particular sum values are again clearly shown.

The value of the two-parameter extension is now very clear. The entire range of sum values is at one time analyzed and displayed. Where in a single parameter spectrum

there may exist some difficulty in determining whether some particular peak is a photopeak or a fictitious Compton section, in the two-parameter by observing the position of the peak in several adjacent sum lines this problem is immediately resolved.

CESIUM 134

The beta decay of 2.2 year half-life Cs^{134} and the following gamma ray transitions in its daughter nucleus Ba^{134} has been subjected to much study in recent years. The presently accepted decay scheme of Ba^{134} is shown in Figure 6.

Several groups (Van Wijngaarden and Connor 1964, Segaert et al 1963) have reported the existence of a 475 Kev. gamma ray arising from a transition between the 1643 Kev. and 1168 Kev. levels. Various estimates of the intensity of this transition have been made.

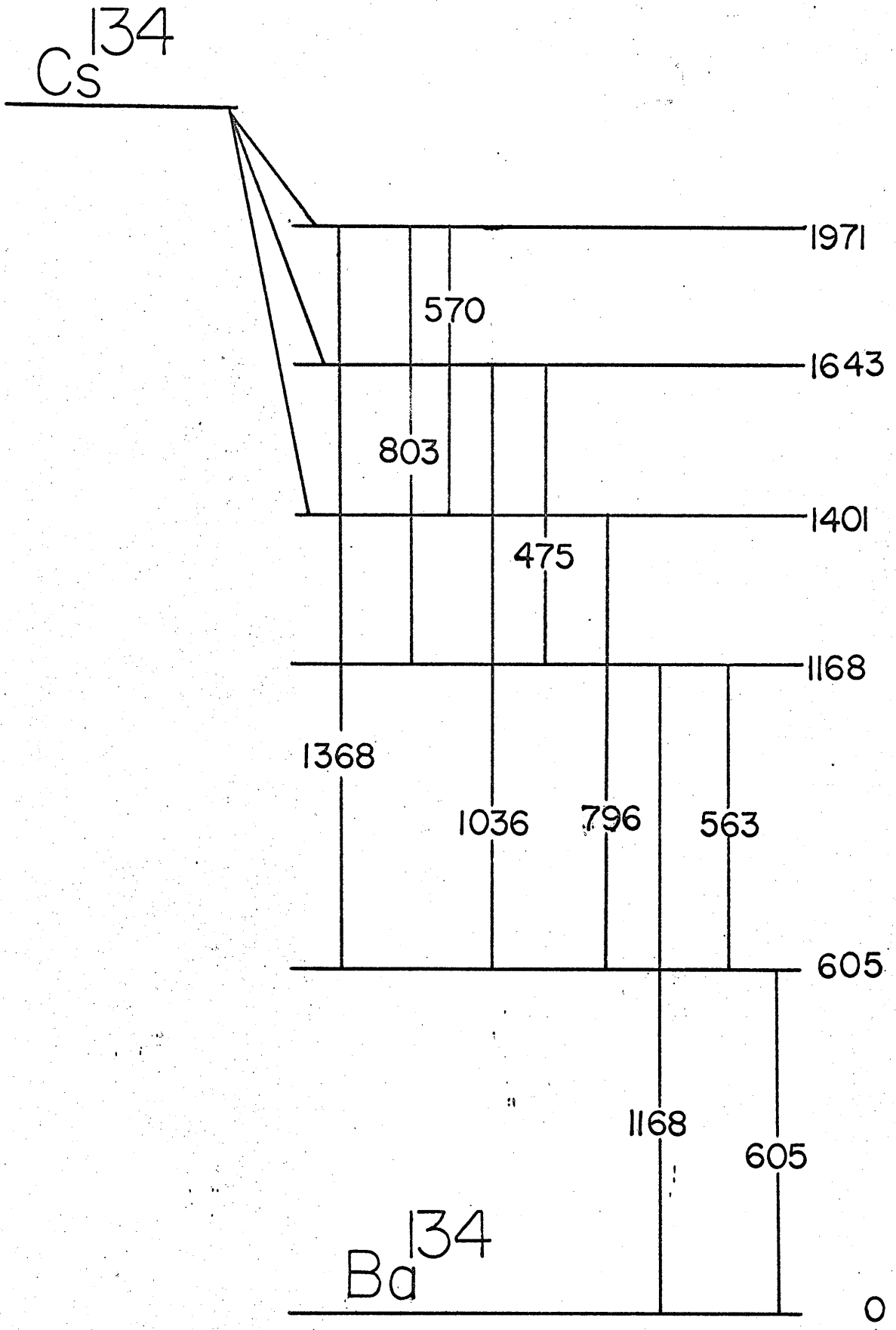
The purpose of the present study of the gamma decay of Ba^{134} is to attempt to confirm the existence of the 475 Kev. gamma ray and to determine its intensity.

Two parameter sum-coincidence studies of the decay were undertaken. For weak intensity cascades the sum-coincidence method has its greatest value when the cascades arise from decays from the uppermost excited level to the ground state. In cases of cascades which begin at a lower level, there is often the possibility of interference in the form of fictitious Compton peaks being superimposed upon the genuine photopeaks. In the case of Ba^{134}, the 475 Kev. gamma ray comes from the decay of an intermediate level. Direct examination of the two parameter spectrum does not,

Figure 6

Decay Scheme of Cs^{134}

Energies in Kev.


Cs^{134}
1971
570
1643
803
1401
475
1168
1368
1036
796
563
605
1168
605
Ba^{134}
0

because of the heavy Compton background, indicate clearly the existence of an expected 475 Kev. photoridge. A detailed study of the individual sum lines is required.

Figure 7 shows one of the typical sum lines. A sum value of 1.64 Mev. is used. Examining the decay scheme in Figure 6, a sum-coincidence between the 475 Kev. and the 1168 Kev. gamma rays is expected. Peaks are indeed found at these energies. A large portion of the 475 Kev. peak, however, may be due to a Compton event in the crystal, which is being analyzed, involving the higher intensity 803 Kev. gamma ray where it deposits 475 Kev. of its energy in the crystal and then escapes. Since this 803 Kev. gamma ray is in coincidence with the 1168 Kev. gamma ray, the required sum value can thus be obtained. A study of peak shapes and the probability of such an event need be made. It is found that part, but not all, of this peak is due to such events. Immediately adjacent sum lines yield a similar result. A careful study of the probable portion of the peak due to 475 Kev.-1168 Kev. photopeak summation indicates a definite existence of the gamma ray with an intensity of 1 - 3% of the 605 Kev. transition intensity.

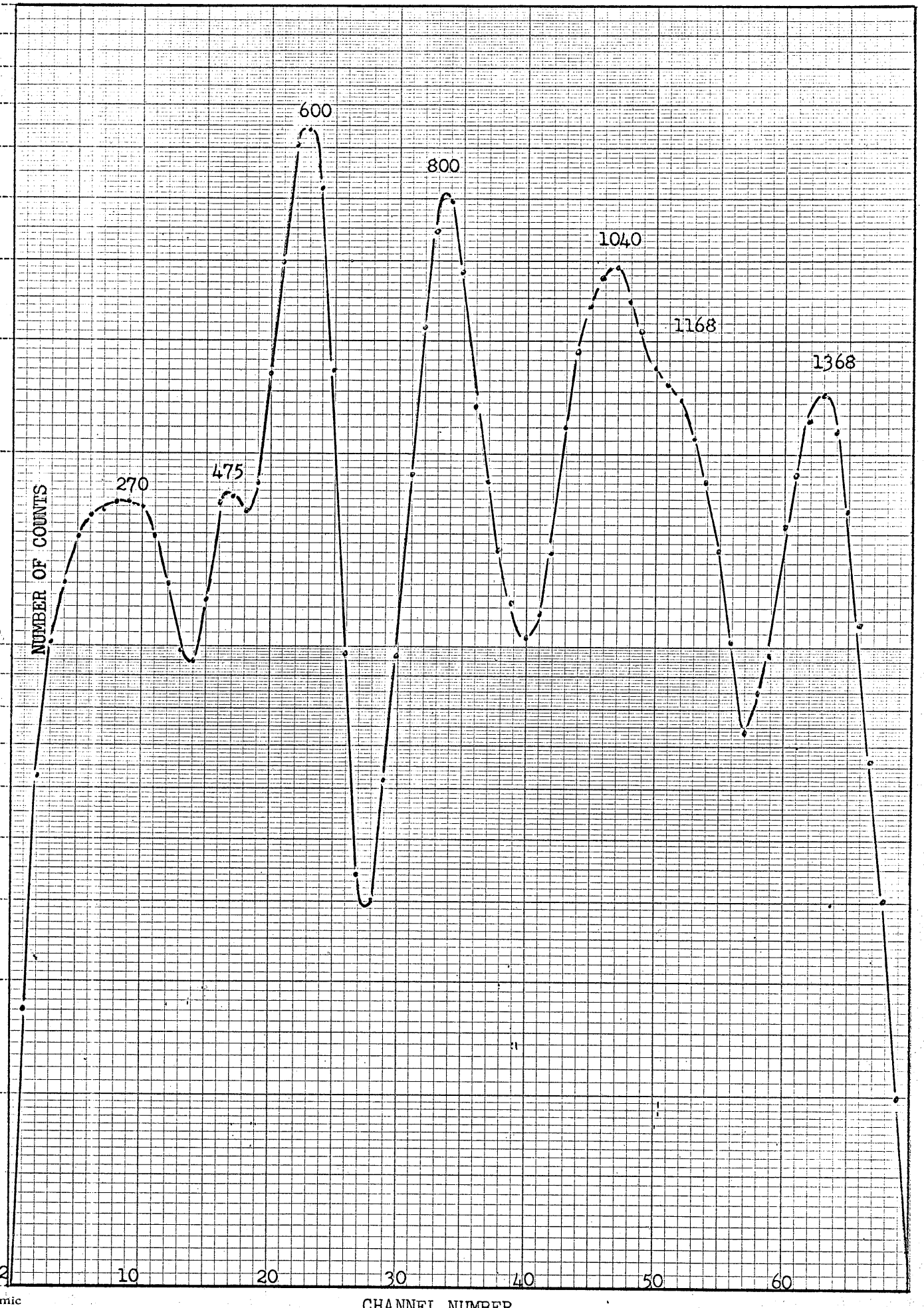
Attempts to find the intensity of the 475 Kev. transition from a normal scintillation spectrum are rather inaccurate because of the large Compton contributions of the adjacent high intensity peaks. In most cases very

Figure 7

Cs^{134} Sum-Coincidence Spectrum

Sum Window at 1640 Kev.

Energies in Kev.



i-Logarithmic
s x 10 to the inch

little, or no, direct indication of the existence of intensity of the gamma ray may be achieved until adjacent peaks are subtracted from the spectrum. Substantial errors may easily be introduced.

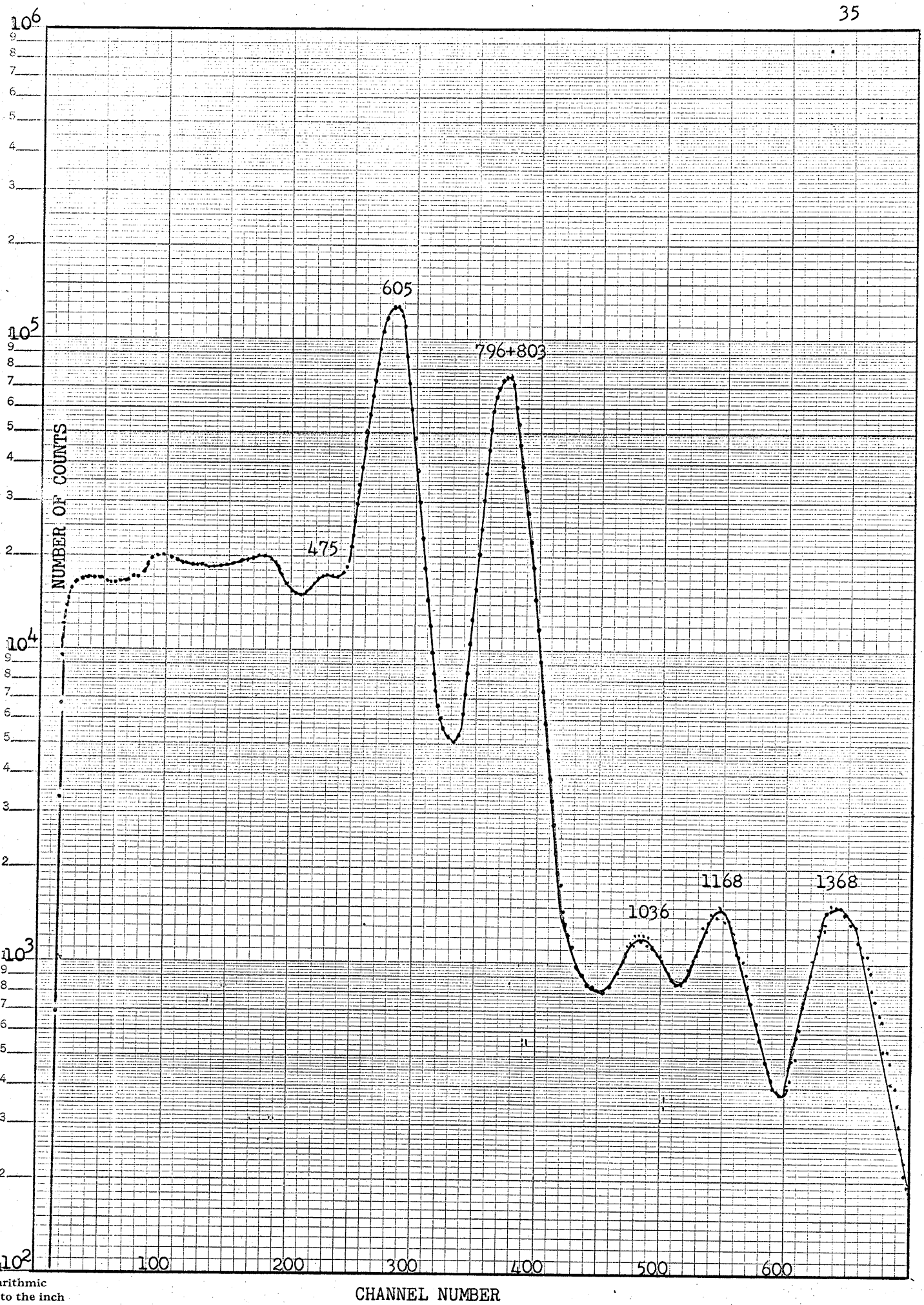
The use of a large scintillation crystal and careful shielding, as was earlier indicated, may quite considerably reduce the Compton background. Figure 8 shows the singles scintillation spectrum of the gamma radiation from Ba^{134} taken on a 3" by 3" detector at a source-to-crystal distance of 10 cms. and employing the previously mentioned lead shielding. The indication of a peak at 475 Kev. is much more definite here than in the case where smaller crystals and no shielding are used (Van Wijngaarden and Connor 1964, Sagaert et al 1963). Removing the contributions to the spectrum from the adjacent peaks, and correcting for detector efficiency as a function of energy, the intensity of the 475 Kev. transition is found to be $1.6 \pm 0.3\%$ of the 605 Kev. transition.

Recent work on the gamma decay of Ba^{134} employing high resolution lithium-drifted germanium detectors has been done by Ewan and Tavendale (Ewan and Tavendale 1964) at Chalk River. They report an intensity, for the 475 Kev. transition, of $1.50 \pm 0.15\%$, with which our findings are seen to agree.

Figure 8

Singles Gamma Spectrum Following Decay of Cs^{134}

Energies in Kev.

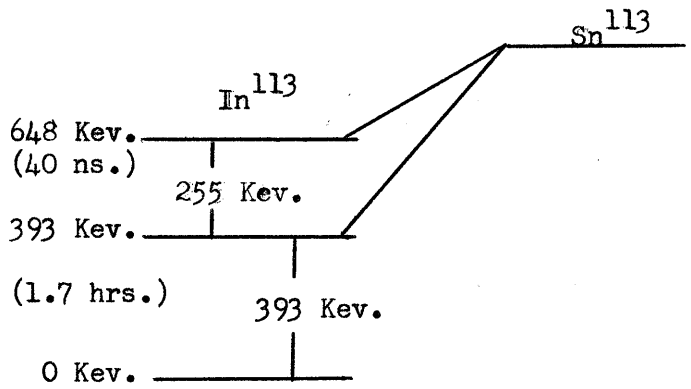


i-Logarithmic
s x 10 to the inch

TIN 113

The decay of 118 day half-life Sn^{113} to stable In^{113} by the process of orbital-electron capture has been quite widely studied. Some question as to the relative intensities of the resultant gamma ray transitions in In^{113} and as to the total decay energy between the ground states of the two nuclei, however, still exists.

The rather simple decay scheme of the Sn^{113} -In^{113} transition, as given in Nuclear Data Sheets, is shown below:



Both the 648 Kev. and the 393 Kev. levels are reported to be isomeric.

To check for possible weak cascades, a two parameter sum-coincidence spectrum was taken. No cascades, or coincidences involving gamma rays with energies above 50 Kev. were observed. Coincidences between the 24 Kev. X-ray and the 255 Kev. gamma ray have been reported (Burson et al 1959), but the triggering circuits used in the present apparatus do not allow coincidence studies below 50 Kev.

In Figure 9, the lower energy portion of the gamma ray spectrum taken on a 3" by 3" detector at a source-to-crystal distance of 10 cms. is shown. Prominent gamma ray photopeaks are found at energies of 393 Kev., 255 Kev., 24 Kev., 83 Kev. and 65 Kev. Two additional peaks which correspond to the backscattered peak and Compton edge of the intense 393 Kev. transition may also be noted. The first three peaks listed above are in agreement with the decay of Sn^{113}, with the 24 Kev. peak corresponding to the K X-ray of In^{113}, which follows the orbital electron capture. The 65 Kev. and 83 Kev. transitions do not, however, fit into the decay scheme. Only one other group has reported either of these gamma rays. Phillips and Hopkins (Phillips and Hopkins 1960) report the existence of an 83 Kev. peak in their spectra. They attribute this as due to impurity in the source used. This is assumed to be the case here for both the 83 Kev. transition and the 65 Kev. transition which could arise from an impurity of Sn^{119} in the source. The improvement in prominence of the 255 Kev. photopeak by using collimated shields is quite substantial.

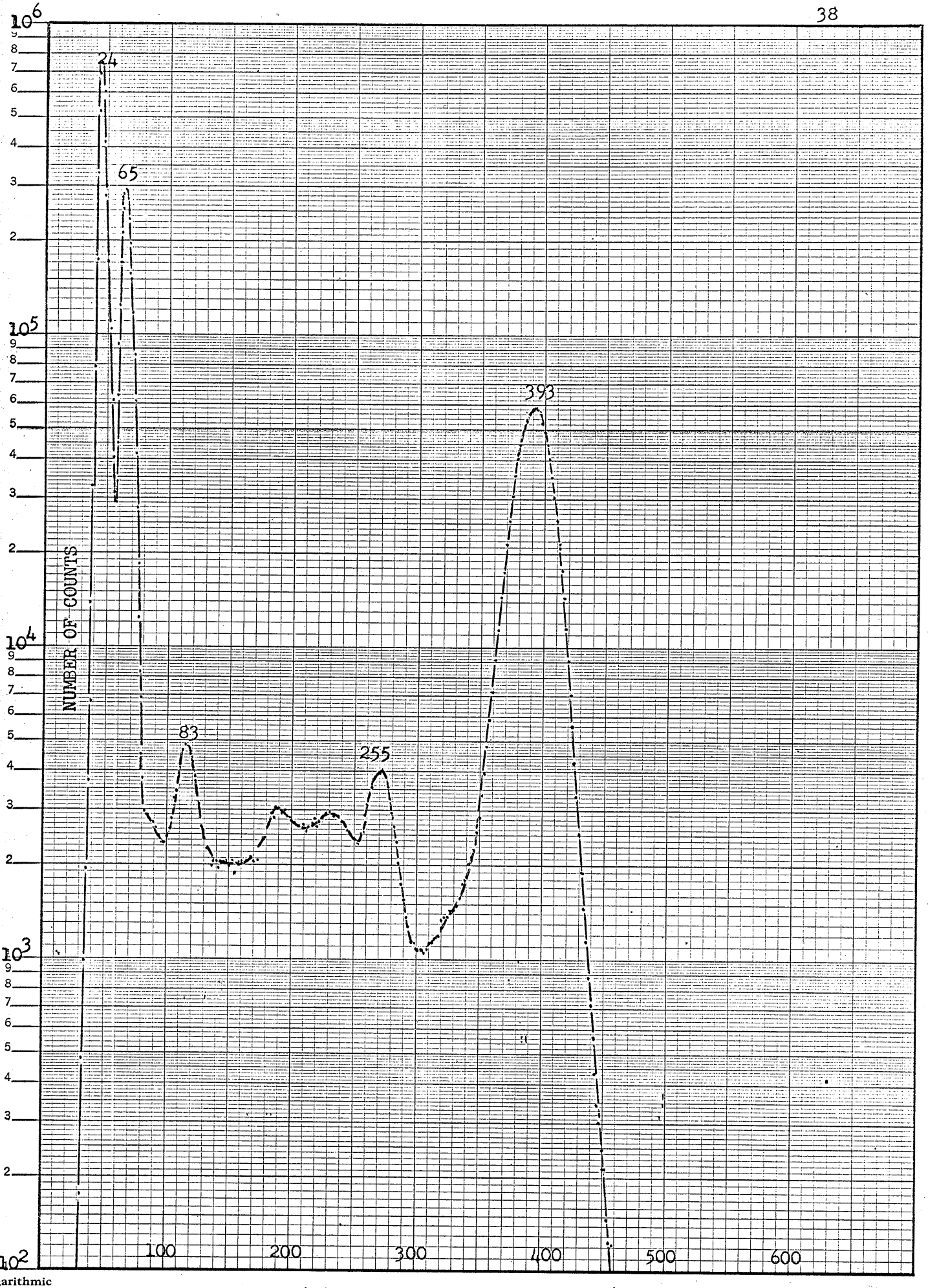
In order to investigate the higher energy region of the gamma ray decay, which is of low relative intensity, the 3" by 3" detector was shielded by at least 2 inches of lead in every direction except for the immediate direction of the photomultiplier. Background counts due to stray

Figure 9

Lower Energy Singles Gamma Spectrum

Following Decay of Sn^{113}

Energies in Kev.



Semi-Logarithmic
Cycles x 10 to the inch

radiation in the laboratory, or natural radioactivity which has been observed in the walls of the room, were reduced by at least an order of magnitude in this manner. The zero-channel position of the multi-channel analyzer was moved to a higher energy to reduce the analyzer dead time resulting from the low energy transitions.

The results of a 24 hour analyzing period with the source-to-crystal distance again being 10 cms. are shown in Figure 10. Following the above analysis, the source was removed, and a spectrum, with proper dead time corrections, was taken in order to determine the background.

Several new features are now clearly visible. To begin, there are very definitely counts beyond the 393 Kev. photopeak which are due to the source. The counts in this region, apart from background radiation, exhibit three parts, namely, two peaks, one at 648 Kev., the other at 786 Kev., and a decreasing continuum. A 648 Kev. transition in In^{113} has been previously reported by Phillips and Hopkins and is interpreted as a cross-over transition from the 648 Kev. state of In^{113} to its ground state. The 786 Kev. peak is very probably caused entirely by two 393 Kev. gamma rays being incident on the detector within its resolving time. Trials taken at different source-to-crystal distances support this. The fraction of the 648 Kev. peak that may be due to similar events involving the 255 Kev. gamma ray or a Compton event of the 393 Kev. and a 393 Kev. gamma ray does not exceed 5%.

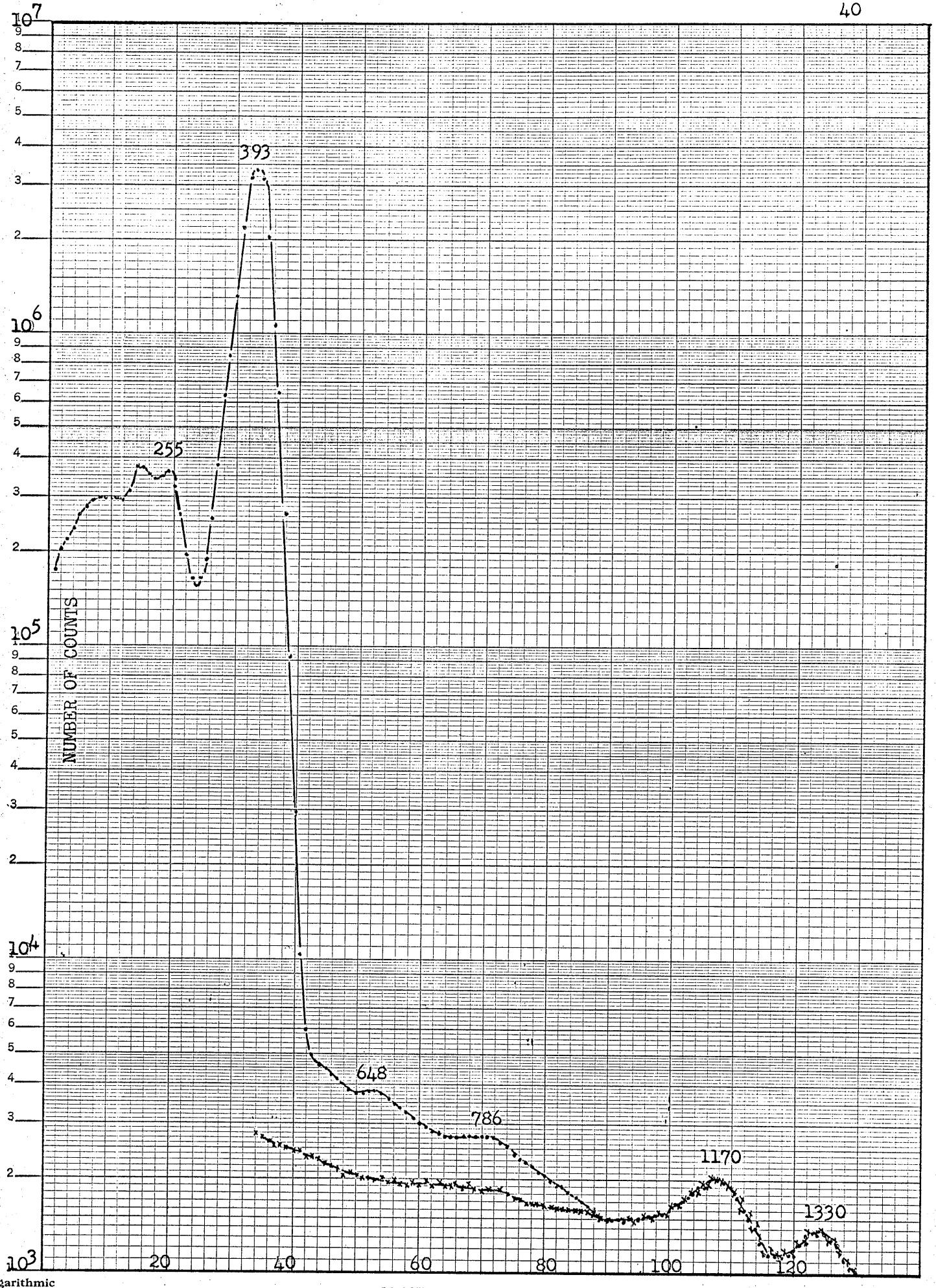
Figure 10

Higher Energy and Bremsstrahlung Spectrum of Sn^{113}

.____. - Singles spectrum

x——x - Background

Energies in Kev.



ni-Logarithmic
es x 10 to the inch

Finally, the continuum part of the spectrum remains. It is attributed to the inner bremsstrahlung radiation following the capture of the orbital electron. Peaks at 1.17 Mev. and 1.33 Mev. which are found in both the spectra are due to a slight contamination of Co^{60} on the lead used for the shielding.

Intensity studies of the 255 Kev. and 648 Kev. transitions have been made, taking into account efficiency as a function of energy. The values obtained are listed and compared to earlier reported results below:

Table I

Reference	Intensity of Gamma Rays		
	255 Kev.	393 Kev.	648 Kev.
Bhatki et al (1957)	5.0	100	-
Girgis and Von Lieshout (1958)	3.0 ± 0.3	100	<0.05
Burson et al (1959)	2.7 ± 0.2	100	<0.10
Phillips and Hopkins (1960)	2.5 ± 1.0	100	0.15 ± 0.10
Present work	2.5 ± 0.2	100	0.02 ± 0.01

The results for the 255 Kev. transition, except for the first, are all in general agreement. The present measure of the 648 Kev. transition agrees with the upper limits set by Girgis and Von Lieshout, and also by Burson et al.

Figure 11a shows an enlarged linear plot of the higher energy region discussed above. An estimated shape for the inner bremsstrahlung is made, and that portion of the spectrum that is attributed to the bremsstrahlung is shown. In Figure 11b, a Kurie-type plot of this contribution, with corrections made for scintillation efficiency is shown. Allowing also for the finite resolution of the scintillation spectrometer, the end-point energy of the inner bremsstrahlung is found to be 1.00 ± 0.15 Mev. This value is in general agreement with that of Phillips and Hopkins who report a value of 0.90 ± 0.30 Mev.

The interpretation of the bremsstrahlung end-point is a topic of some question. Phillips and Hopkins report it as being the end-point of the transition to the 393 Kev. level in In^{113}. Allowing for the 393 Kev. gamma ray transition, the total decay energy is then reported to be 1.3 ± 0.3 Mev. Morrison and Shiff (Morrison and Shiff 1940), from theoretical considerations, report that the ratio of the number of bremsstrahlung photons, N(p), to the number of K-electron captures, N(c), should be given by:

$$\frac{N(p)}{N(c)} = \frac{a\ E^2}{12\pi\ m^2c^2}$$

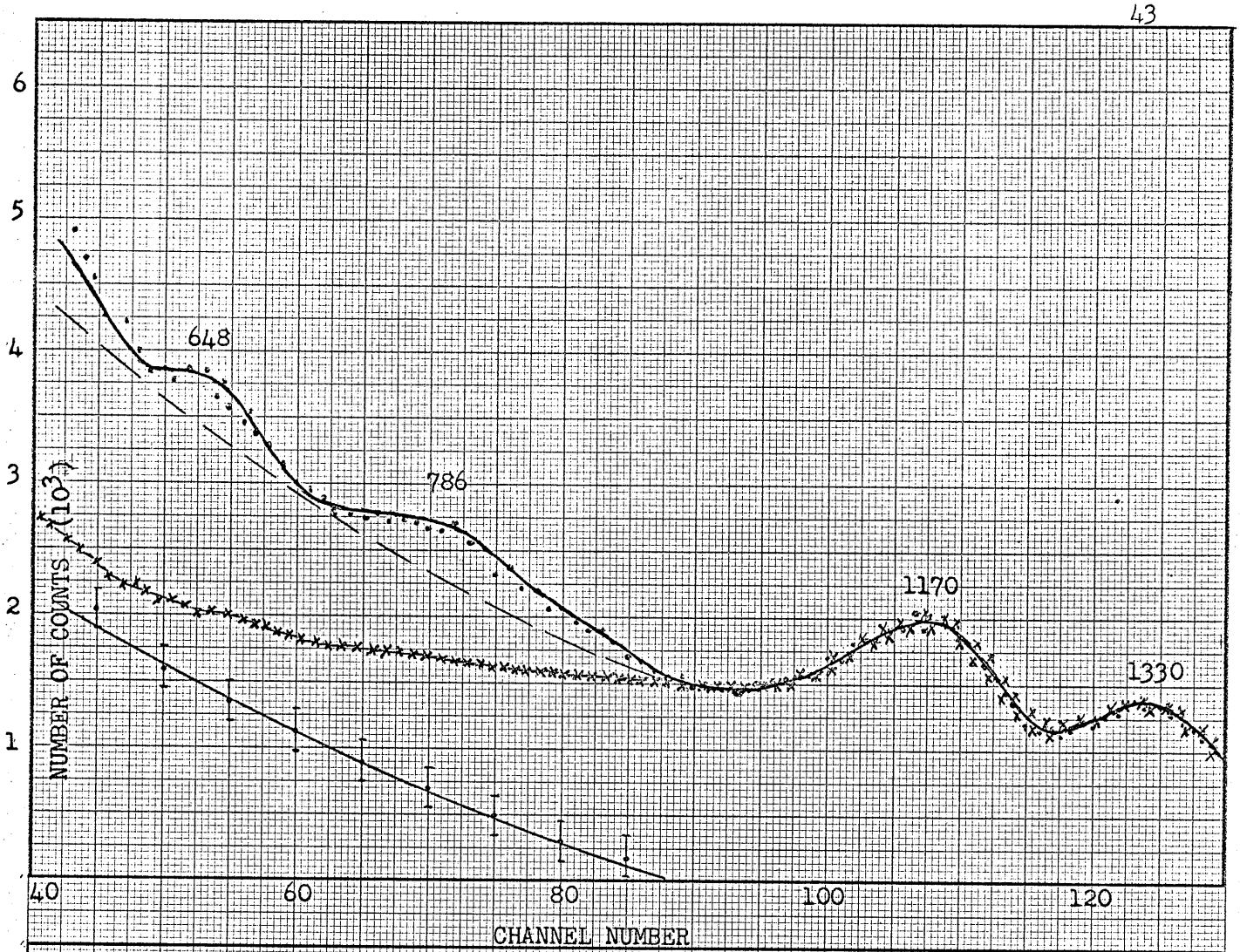
where a is the fine structure constant, E is the end-point energy, and m is the electron mass. Taking into account the

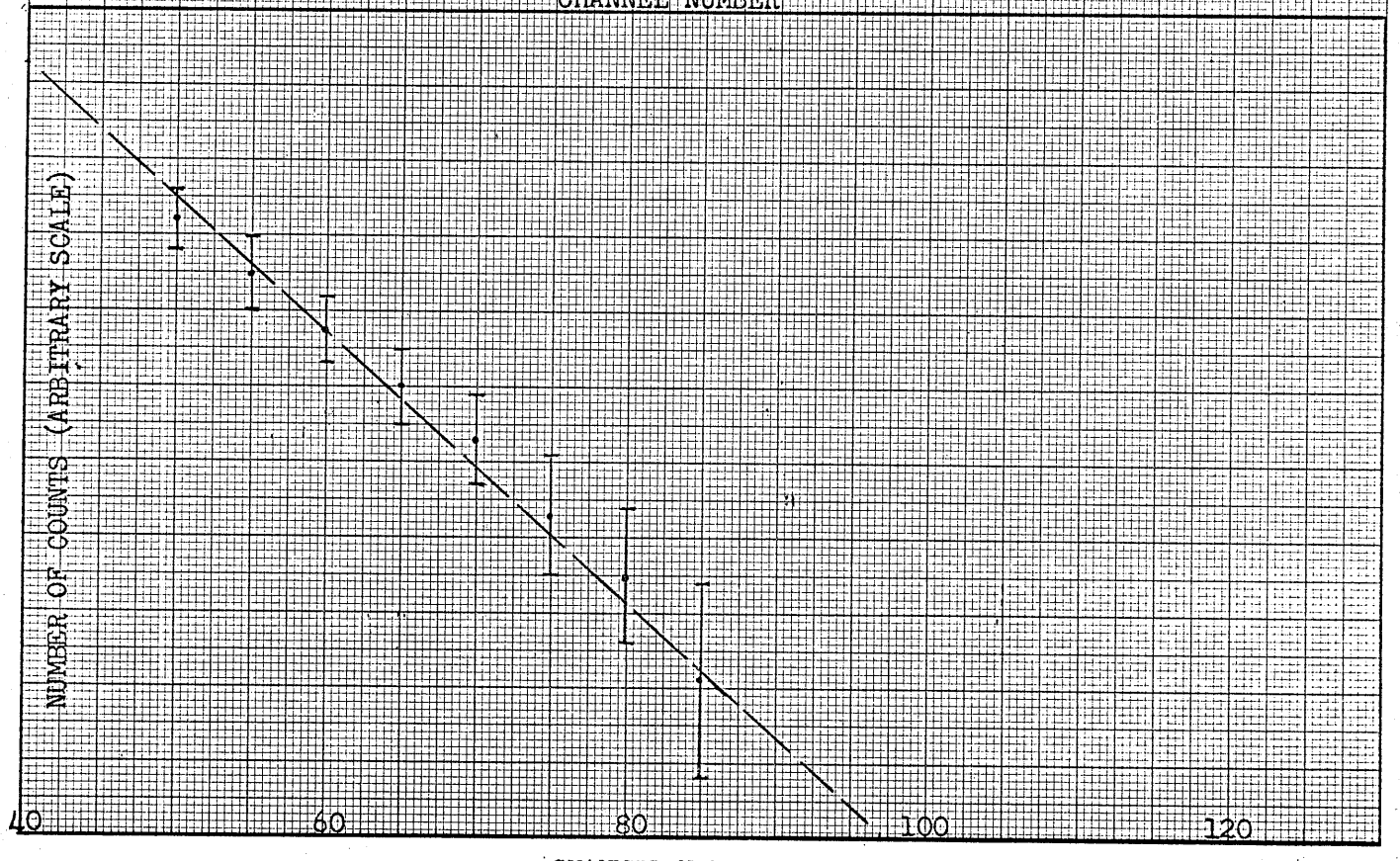
Figure 11

Bremsstrahlung Spectrum from Sn^{113}

a - Linear Plot

b - Kurie Plot





illimeters to the Centimeter

total conversion factor, and using our determined end-point energy, the intensity of the bremsstrahlung radiation relative to the 393 Kev. value being 100, is found to be of the order of 0.1. An estimate of the relative intensity, using the assumed bremsstrahlung spectral shape of $X(1-X)^2$, where X is the ratio of the energy to the end-point energy, yields a value in the present spectrum of approximately 0.4 relative to the 393 Kev. value of 100. The fact that the experimental and theoretical values are within an order of magnitude gives support to the view that the bremsstrahlung spectrum is associated with decay to the 393 Kev. level in In^{113} and not to the ground state. The expected intensity of a ground-state to ground-state transition would be much smaller than the presently found value due to the larger spin change.

Recent conversion-electron work by Durosinmi-Etti, Brundrit, and Sen (Durosinmi-Etti, Brundrit, and Sen 1965) indicates the energy difference between the ground state of Sn^{113} and the 393 Kev. state of In^{113} to be $280\ ^{+350}_{-100}$ Kev., or a total decay energy of $680\ ^{+350}_{-100}$ Kev. Their work is in agreement with that of Bhatki et al who report a decay energy between the ground state of Sn^{113} and the 648 Kev. state of In^{113} of 36 Kev., and hence a total decay energy of 684 Kev., by a method related to that of Sen.

There exist, therefore, two rather widely differing groups of decay energy values. Those obtained by any one method appear to be quite consistent, but the two groups agree only marginally. The reason for this poor agreement is at the present unknown, but may be due to insufficient accuracy in the theoretical development.

BARIUM 131

A large amount of data on the decay of Ba^{131} into Cs^{131}, and the resulting gamma ray de-excitations in Cs^{131} have been gathered. Canada and Mitchell (Canada and Mitchell 1951) reported the existence of six gamma rays in their spectra. The number of reported peaks was increased to thirteen following the work of Cork et al (Cork et al 1953). Many rather drastic changes in the proposed decay scheme of the decay have been required as additional data became available. Recently Kelly and Horen (Kelly and Horen 1963), based on their high resolution conversion electron measurements, have proposed a decay scheme into which they fit a total of thirty-four gamma ray transitions.

The sum-coincidence method, as was earlier noted, has its greatest value for those cascades which result from decays between the uppermost excited level and the ground state. Due to the large number of cascades present in the decay of Cs^{131}, and the wide variation in the relative intensities, a sum-coincidence examination of those cascades which begin at intermediate levels yields an extremely complicated spectrum. A two parameter sum-coincidence spectrum of the upper energy region of the Cs^{131} gamma ray spectrum was undertaken. A total analysis time of eight days

was required in order to obtain a useful number of counts in the spectrum.

In Figure 12 the contour and isometric displays of the spectrum obtained are shown. For the contour display, a slight change in the coordinate system defined earlier needs to be made. Let the X axis again be along the bottom of the display, with the Y axis running up it. Increasing X and Y correspond to increasing sum and singles energies respectively. The coordinates of the isometric display are as before.

Examining the contour display, seven ridges are immediately evident. Those ridges which are at corresponding distances from the central one are sum compliments. Spectra taken over different time intervals using the same source indicate that the central ridge is due largely, and probably entirely, to chance coincidences of two 496 Kev. gamma rays which result from an intense transition between the 620 Kev. and 124 Kev. levels of Cs^{131}.

In Figure 13 is shown the proposed decay scheme for the 1040 Kev. level of Cs^{131}. Also shown is one of the typical sum lines obtained.

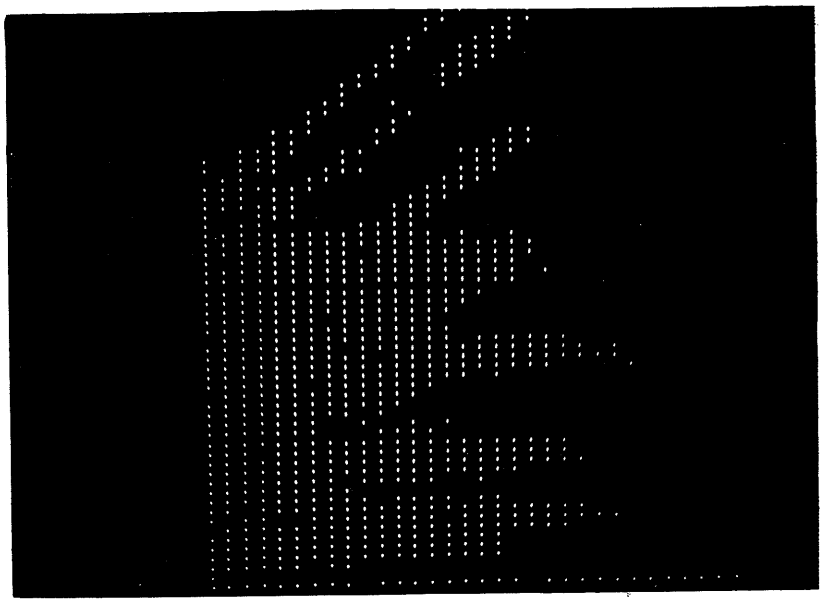
The six genuine ridges in the contour display correspond to cascades involving the following pairs of gamma rays: 916 Kev. with 124 Kev., 824 Kev. with 216 Kev. and 670 Kev. with 373 Kev. The first two cascades have been

Figure 12

Two Parameter Spectral Display of Ba^{131}

a - Contour Display

b - Isometric Display



• MAY • 65

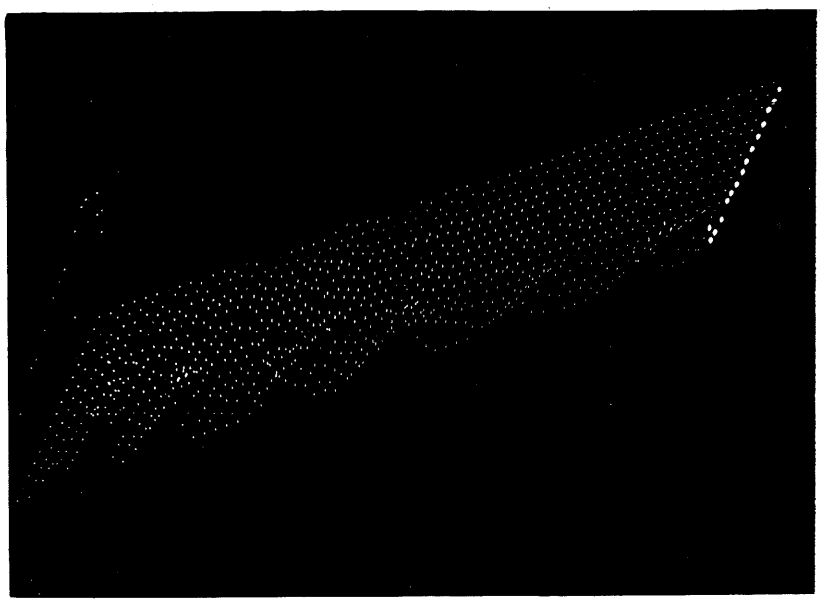


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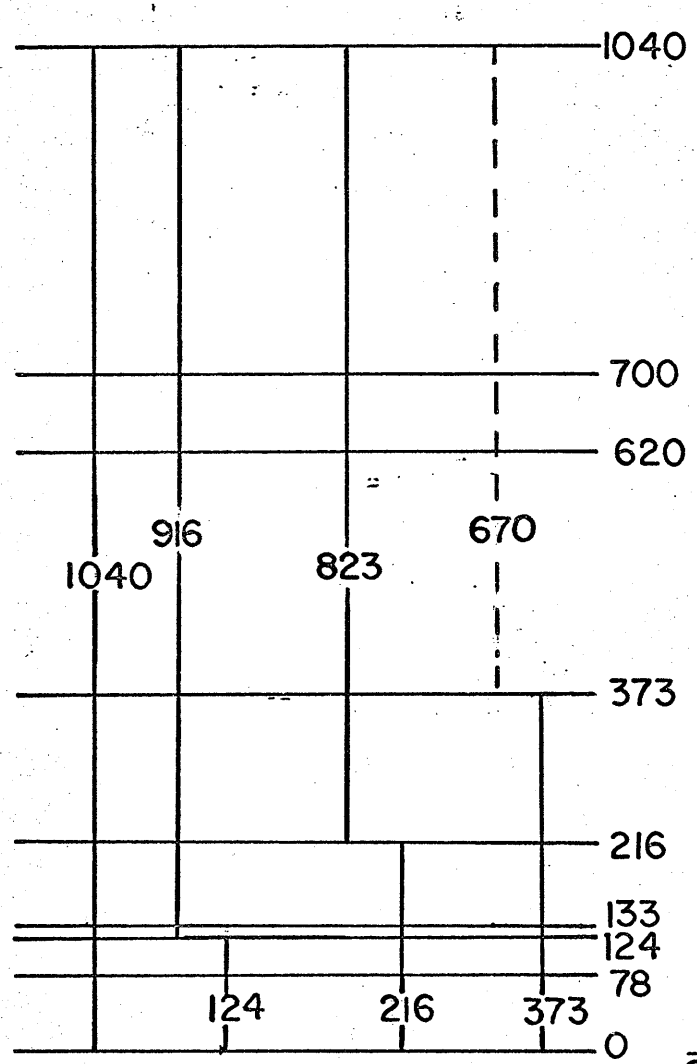
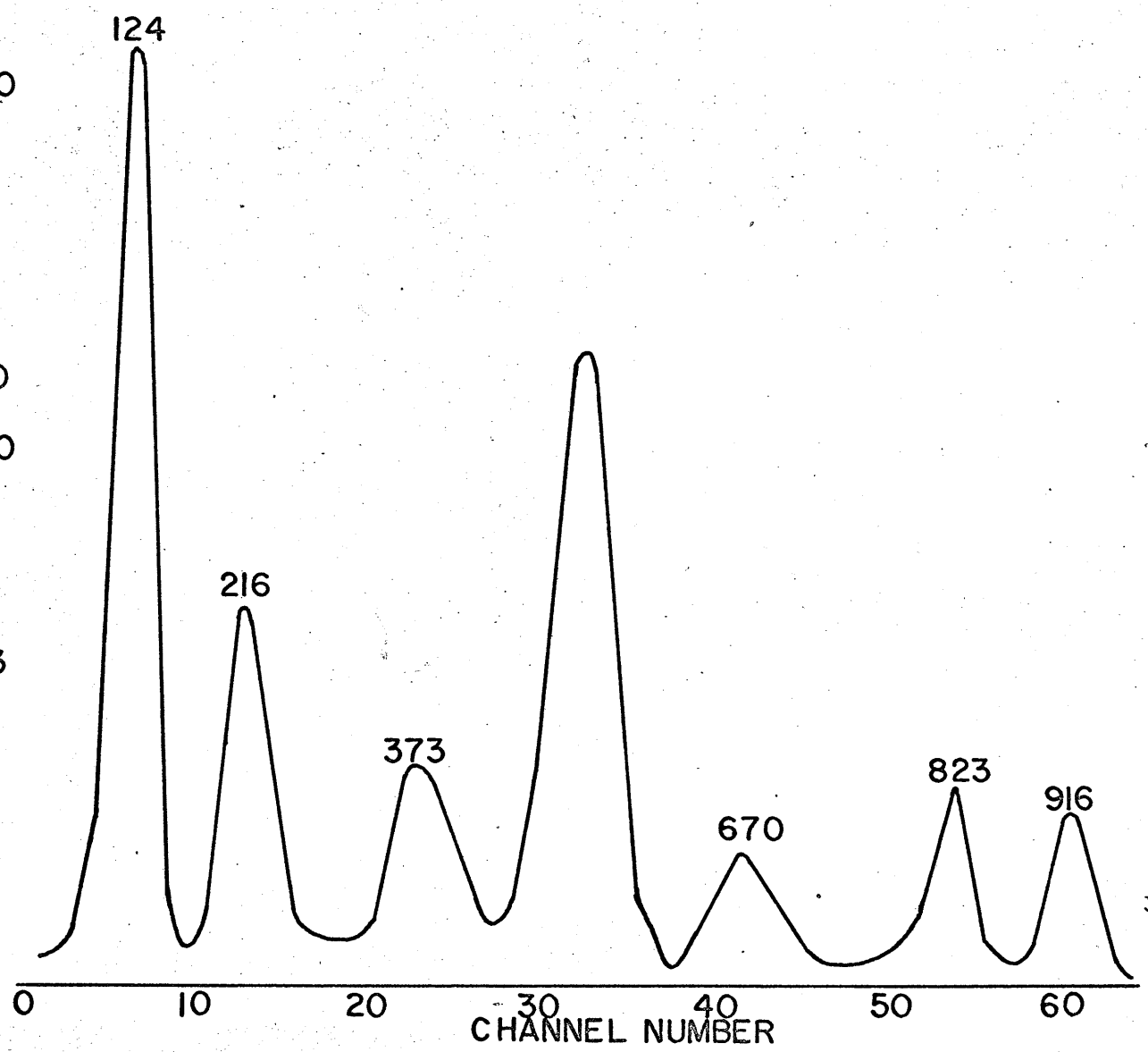
Figure 13

Decay of 1040 Kev. Level in Cs^{131}

Energies in Kev.

DECAY of 1040 Kev. LEVEL (Cesium 131)





previously observed and are reported in Nuclear Data Sheets. No report of the 670 Kev. transition, or of the 670 Kev.-373 Kev. cascade could be found in the literature previous to the article by Kelly and Horen.

Comparing the areas of the peaks obtained in the spectra for the 670 Kev.-373 Kev. cascade and the 216 Kev.-824 Kev. cascade, and using the reported intensities of the other gamma rays, the intensity of the 670 Kev. transition is estimated to be 0.2 to 0.3%. This is in agreement with the 0.21% intensity reported by Kelly and Horen.

Some question as to the spin and parity of several of the levels of Cs^{131} still exists. The two-parameter method of sum-coincidence analysis lends itself very well to angular correlation studies. Since all the cascades are analyzed at one time, if spectra be taken at various inter-detector angles, the correlation functions for all the cascades may be obtained simultaneously. Furthermore, if one particular correlation function is known, it may be used as a calibration function for the other cascades, and corrections for unstable experimental geometry, if they are not too large, may be made.

The results of the angular correlation work of Bodenstedt et al on the 912 Kev.-124 Kev. cascade in Cs^{131} are given in the report of Kelly and Horen. An attempt was made in the present project to determine the correlation

functions for the remaining two cascades which have been observed in the decay of the 1040 Kev. level. Due to the weak intensity of the cascade, counting periods of several days were required for each angle. Instability of some of the electronic circuitry and also of the source during these periods were found, and no consistent results can be reported. Confirmation of the 670 Kev.-373 Kev. cascade, however, was again made using the spectra obtained.

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