APPLICATION OF THE SCINTILLATION SPECTROMETER IN NUCLEAR PHYSICS

A Thesis

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at the

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by

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PREFACE

The work to be described in this paper was carried out at the University of Manitoba during 1951 - 1952. It forms a part of a research programme begun in 1949 by Dr. R. W. Pringle.

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INTRODUCTION

The construction of suitable counters for the detection of elementary particles and electromagnetic radiation has been, and is yet, one of the principal tasks of the experimental nuclear physicist. The Geiger counter, proportional counter and the boron-trifluoride neutron counter have been used extensively for many years for the detection of charged and neutral particles. However, due to the extremely low efficiency of the Geiger and proportional counters for the detection of γ-rays, the standard method for studying such radiation has been to use a magnetic β-spectrometer for the analysis of conversion electrons produced by the γ-radiation. It was not until the advent of the scintillation counter in 1947,[1] that an efficient detector of γ-radiation became available to the nuclear physicist. It would not be an exaggeration to state that the scintillation counter has revolutionized counter technology.

The uses to which scintillation counters have been put may be divided into two distinct classes:

1. they are used as detectors - both for particles and electromagnetic radiation, and
2. they have been successfully employed as proportional devices for energy measurements.

The first class of uses is expanding at such a rapid rate that only through constant perusal of current

literature can one hope to keep in touch with the latest applications of the scintillation counter. Reference will be made to a few such applications in later sections of this work. In the second class, where one is concerned with accurate measurement of γ-ray energies, technical developments, which lead to improvements in performance, also appear in current journals in large quantity. Since the results to be presented in following sections were obtained by means of a 'scintillation spectrometer', this aspect of scintillation counter technology will be dealt with in some detail. Wherever possible, however, reference will be made to any uses of the counter as a detector only that are of current interest.

The simplest type of scintillation counter employs a solid or liquid scintillator, which is placed on the window of a photomultiplier tube. When subjected to γ-radiation, electrons are released in the scintillator which excite centres of fluorescence in the crystal lattice (for a solid), and the scintillator emits radiation that is subsequently detected by the photomultiplier. A cathode follower circuit transmits the pulse from the photomultiplier tube to a linear amplifier. A scaler, or other suitable device, records the number of pulses coming from the tube. If the counter is to be used as a spectrometer, a differential discriminator, of one or more channels, is inserted in the circuit between the amplifier and the recording device.

The suitability of a scintillator depends largely on whether or not the radiation it emits lies in the spectral range for which the photomultiplier was designed. The most common crystal scintillators in use are anthracene, stilbene, sodium iodide (NaI(Tl)), and lithium iodide (LiI(Sn)), which has recently been found to be an excellent detector of slow neutrons through the reaction:

$$Li^6 + n^1 \longrightarrow H^3 + He^4 + Q$$

Many liquid scintillators are known,[2] but the most successful one is a solution of terphenyl (p-diphenylbenzene) in zylene. This liquid scintillator is widely used in cosmic ray work as a detector but is apparently of little use in spectrometry.

Sodium iodide with a 1% thallium impurity is the most satisfactory phosphor for γ-ray spectroscopy and since it was used almost exclusively in the work to be described in following sections, it will be discussed in detail. Unfortunately, its hygroscopic nature makes handling awkward. The crystals have to be stored in light oil (preferably mineral oil) and when in use on a photomultiplier tube, they must be covered by thin aluminum foil which acts as a reflector and also protects the crystal from the atmosphere. A NaI crystal so prepared, usually requires inspection every three or four days. One recent development in the use of reflectors for NaI crystals is of particular interest. The crystal is polished in a dry atmosphere and then placed

on the photomultiplier window with a small quantity of clear vacuum grease forming the crystal-window bond. A plastic or glass cylinder with open ends is then cemented onto the photocell. This forms a container around the crystal into which magnesium oxide powder is poured until the crystal is completely covered. The open end of the cylinder is now covered in such a way that the interior is air tight. Preliminary data indicates that increases of from 30% to 60% in the height of the pulses from the crystal can be realized. Further work is being carried out at present on this new type of reflecting surface.

Cylinders or cubes of NaI(Tl) ranging from 1/2 in. to 1 in. on the major dimension have been used with equal success in scintillation spectroscopy. It is felt, however, that low energy events (e.g. X-radiation) are best studied by means of smaller crystals, with a depth of about 3 or 4 mm. If the source to be investigated was extremely weak, a larger crystal was found to be desirable because of the subsequent increase in the counting rate. In the near future, a cylinder of NaI(Tl) 1-1/2 in. diam. x 1-1/2 in. will be used to study low-activity sources.

When a γ-ray quantum penetrates the crystal, several interactions are possible, depending on the energy of the incident quantum. Since about 85% of the weight of a NaI(Tl) crystal is due to iodine, it is convenient to consider

as a first approximation, interactions between γ-ray quanta and the iodine constituent only. In Fig. 1, the absorption coefficient is shown plotted against γ-ray energy for the three interactions that can occur between radiation and matter. Clearly, the Compton and photoelectric effects are the predominant interactions for γ-ray energies in the range common to natural and artificial radioactive elements. Pair production becomes important for energies greater than 2 Mev. If a γ-quantum interacts with an atom of iodine in the crystal lattice through the photoelectric effect, the whole energy of the γ-ray is ultimately absorbed by the crystal. The low energy X-rays produced after an electron has been ejected from an atom, are easily absorbed and so contribute to the total excitation of the crystal. Hence, the result is the production of electrons in the crystal with the full energy of the incident γ-ray quantum. The distribution of the pulses obtained from the photomultiplier tube is Gaussian, and an examination of the pulses with a differential discriminator gives a photoelectric peak on the pulse height distribution curve. This peak will have a characteristic resolution defined as

$$\frac{\text{width of peak at half height - volts}}{\text{pulse height at centre of peak - volts}} \times 100$$

which depends almost exclusively on the crystal-photomultiplier combination. A γ-ray can also interact with the crystal through Compton scattering. In this case, electrons

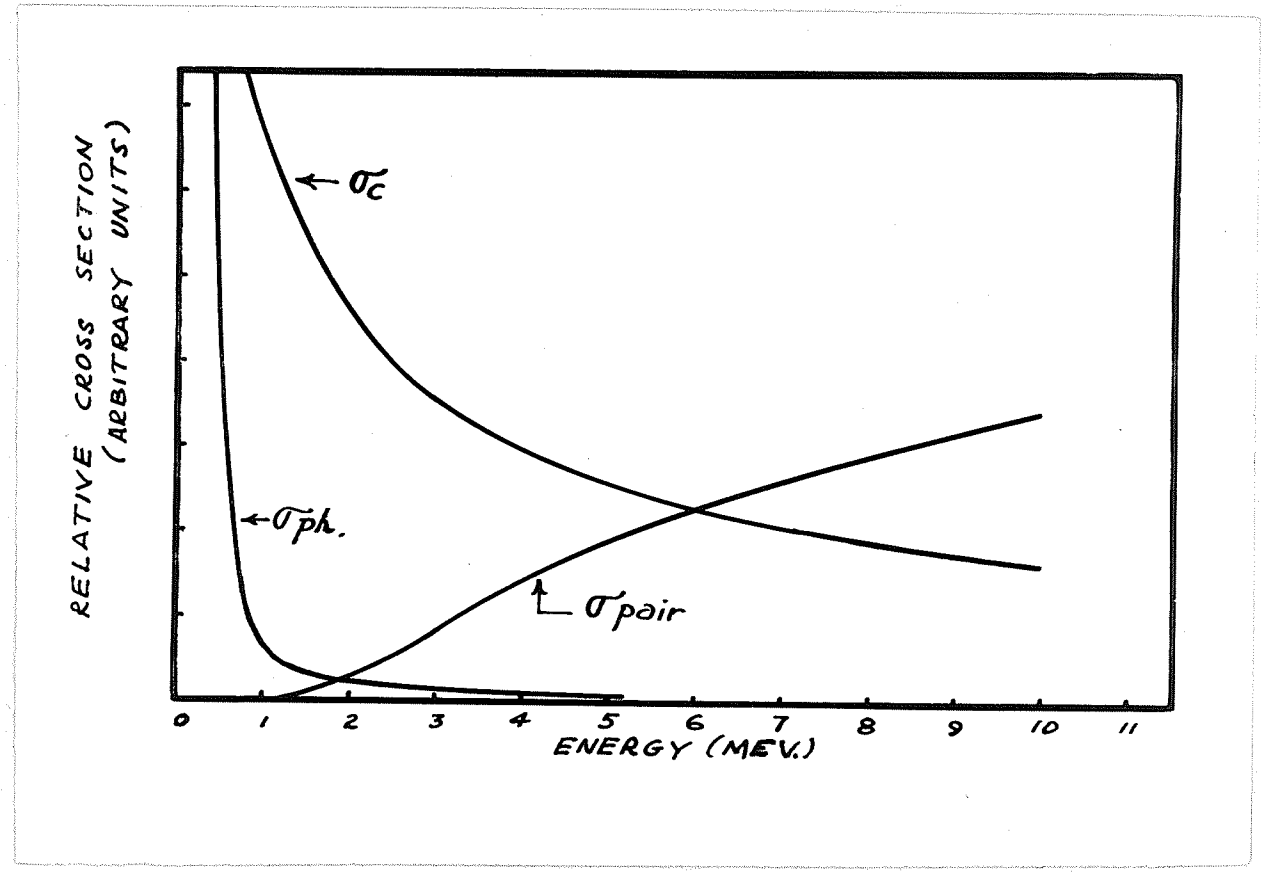


FIG. 1

RELATIVE CROSS SECTIONS FOR PHOTOELECTRIC (ph.), COMPTON (c), AND PAIR PRODUCTION (pair) PROCESSES IN IODINE

are liberated with a range of energies approaching a maximum given by

$$E_c = \frac{E_\gamma}{1 + \frac{0.511}{2E_\gamma}}$$

where E_γ = γ-ray energy in Mev.

E_c = maximum energy of Compton distribution in Mev. The scattered quantum from such an interaction can also be absorbed through a repetition of the process, or it can escape from the crystal. A high energy γ-ray ($E_\gamma > 2.0$ Mev.) will produce a pair line on the pulse height distribution at an energy of (E_γ - 1.02) Mev. This may be followed by two subsidiary peaks corresponding to the additional capture of one or two annihilation quanta from positron interactions in the crystal. The second subsidiary peak would be equivalent to that due to the photoelectric effect in which the entire energy of the high energy γ-ray is captured.

Anthracene crystals have been shown to be efficient detectors for neutrons and β-particles. Neutrons which impinge on anthracene give rise to recoil protons which excite the atoms of the crystal and give rise to light pulses. β-particles react with the atoms of anthracene in the same manner as the aforementioned recoil protons, with the energy of the pulse depending on the energy of the incident electron. Unfortunately, the relationship between pulse size and electron energy is not strictly

linear for electron energies less than 125 kev. The percentage variation is small, however, so that the difficulties which arise are not too serious. By using an anthracene crystal as the phosphor in a scintillation spectrometer, one can analyze the energy distribution of the electrons from a β-active source directly. Preliminary investigations of β-spectra with such a unit have been carried out and will be discussed in a following section.

From what has already been said, it is clear that the quality of the photomultiplier tube determines the value of the scintillation counter as a γ-ray spectrometer. Selected tubes of the R.C.A. type 5819 have been used with considerable success by workers in the United States and Canada, although the earlier models were far from satisfactory. The results to be described in later sections were obtained with two British photomultiplier tubes - the E.M.I. 5311 (1" cathode) and the E.M.I. VX 5055 (1-3/4" cathode). (The older R.C.A. photomultipliers - 931A, 1P21 and 1P28 - have now been abandoned by most workers.) These tubes have gains of the order of 10^6. New tubes have been developed in both Britain and the United States with extra large end windows. Some of them (E.M.I. 6260, and R.C.A. 4646) have 14 stages of amplification or have a gain of about 10^9. With such tubes, one

can analyze the pulses directly without employing an amplifier. Unfortunately, these are not, as yet, generally available to research workers. Scintillation spectroscopy has brought to light many hidden defects in standard photomultipliers. The requirements of higher collection efficiency, regularity of the photosensitive surface, greater amplification, etc., have raised new design problems. At the present time, it appears that many of these problems are nearing solution, and it is to be hoped that the photomultipliers soon to be released for distribution will greatly reduce the difficulties experienced in spectrometry. The specific technical details of the use of the tubes described above (E.M.I. 5311, and E.M.I. VX 5055) will be given in later sections.

We have seen that by a critical analysis of the pulse height distribution obtained with a differential discriminator, one can identify the features which correspond to the three interactions of radiation with a NaI crystal outlined above. Since the relationship between the pulse height and the energy of the incident quantum is a linear one, taking into account the statistical spread of all such features, the energy of the incident γ-ray can be determined directly from the shape of the pulse height distribution. The net result is that one has a simple γ-ray spectrometer which is extremely efficient and compact. While it is true that the resolution of such a device depends on the energy of

the incident quantum, and that it is, in general, very much poorer than that obtained with a β-ray spectrometer, the determination of X-ray and γ-ray energies ranging from 2 kev. to 9 Mev. has been successfully carried out by workers in the field. The worth of the instrument has been thoroughly proven in all respects.

For a further discussion of the more general aspects of scintillation counters and their applications, the reader is referred to current papers which attempt to review the whole field of scintillation spectroscopy.[3,4,5,6]

APPLICATION OF THE SCINTILLATION SPECTROMETER TO THE STUDY OF GAMMA RAY SPECTRA

INTRODUCTION

The results to be described in this section form a part of an extensive research project now being carried out at the University of Manitoba. Since the data to be presented were obtained, several modifications in the equipment and techniques have been carried out. These will be mentioned at appropriate places in the discussion to follow.

The study of γ-ray spectra with scintillation spectrometers was started early in 1949,[7] and since that time, the number of such investigations has increased rapidly. Many nuclear decay schemes have been studied and several unique problems have been solved. One such problem is the study of low-activity sources, where many grams of the material may be used but the source strength is only of the order of a few milli-microcuries (1 microcurie = 3.7×10^4 disintegrations per second). In this section, two pieces of research will be described in detail, one of which is an example of low-activity work.

THE NATURAL ACTIVITY OF LANTHANUM

The theory of beta-decay, as it is presently accepted, indicates that of two neighbouring isobars (nuclei with the same mass number), the one of greater mass will be unstable against beta-decay to the other. Since the conclusions which follow from beta-decay theory have, in general, been confirmed by experiment, the existence of pairs or triplets of isobars of neighbouring atomic number Z, can only be explained by assuming that there are natural activities present in such anomalies, but that their extremely long half-lives make detection difficult. Examples of such long-lived activities which have been discovered in the last few years are K^{40} (1.4×10^9 years), Re^{187} (4×10^{12} years), Lu^{176} (2.4×10^{10} years), Rb^{87} (6.3×10^{10} years)[8] and In^{115} ($6 \pm 2 \times 10^{14}$ years)[9]. An activity in In^{113} of the isobaric pair (Cd^{113}, In^{113}) has been suggested.[10] Until recently there were two sets of isobaric triplets - (Ba^{138}, La^{138}, Ce^{138}) and (T^{50}, V^{50}, Cr^{50}). The natural activity of La^{138} was first reported by this laboratory in 1950.[11] At that time, the activity was attributed to the K-capture process, resulting in the formation of Ba^{138}, with the emission of a 1.05 Mev. γ-ray. These preliminary results have now been extended and the decay scheme clarified.[12]

DESCRIPTION OF THE APPARATUS

The scintillation detector used for low activity work consisted of an E.M.I. 5311 photomultiplier tube mounted on an aluminum chassis and surrounded by a light-tight metal cannister. It has a 1 in. diam. photo-sensitive semi-transparent cathode followed by ten dynodes. The wiring of the tube was conventional and employed 10 megohm resistances between adjacent dynodes. The gain of such a tube varies as the seventh power of the voltage across it, hence a highly stabilized supply of high voltage was required. The power supply used was manufactured for the A.E.R.E. Harwell project, Eng. (Type 1007, No. 3). The actual voltage across the tube was controlled by a precision resistance network placed across the 2200 volt supply. The output voltage from the supply was accurate to 1 part in 2000 for an input voltage fluctuation of 10%. One of the modifications referred to earlier consisted of placing 20 megohm resistors between the first two dynodes, thereby securing a larger voltage drop across the first two stages. It was felt that the resolution of the tube could be improved slightly by such a change. The improvement was found to be small, but nevertheless, desirable. With the earlier resistance network, there was approximately a 90 volt drop across each stage, resulting in an amplification of about 10^6. A standard cathode follower circuit was used to feed the pulses from the tube to an amplifier.

A 3/4 in. cylindrical NaI(Tl) crystal was mounted on the end window of the E.M.I. tube (See Fig. 2). The crystal was highly polished and liberally coated with mineral oil to prevent deterioration. A thin aluminum foil covered all the surfaces but that placed against the window of the tube. Such a crystal can produce about 4×10^4 photons of light in the neighbourhood of 4000 Angstroms by absorbing a 1 Mev. electron released from the crystal lattice. Since the photomultiplier is very sensitive to radiation in this range, and the crystal has a low self-absorption at 4000 Angstroms, a substantial pulse can be delivered by the tube for such a primary event in the crystal.

The pulses from the counter are fed into a linear amplifier (Atomic Instrument Co. Linear Amplifier 204-C). After suitable amplification, they are analyzed by means of a Pulse Amplitude Analyzer (Murphy Radio Ltd., Eng. Type 1074A, No. 3) (see Fig. 3). This analyzer has four 1/2-volt channels, and it can analyze pulses spread over a voltage range from 5 to 35 volts. The maximum allowable counting rate, without undue losses, is 2000 counts per minute per channel.

The pulses from the amplifier were also analyzed by means of a cathode ray oscillograph (Tektronix Oscillograph 511A) and a camera (f/4.5 lens). A light-tight metal cone was fastened over the oscillograph screen, with the camera lens placed at the other end. The pulses were displayed on the

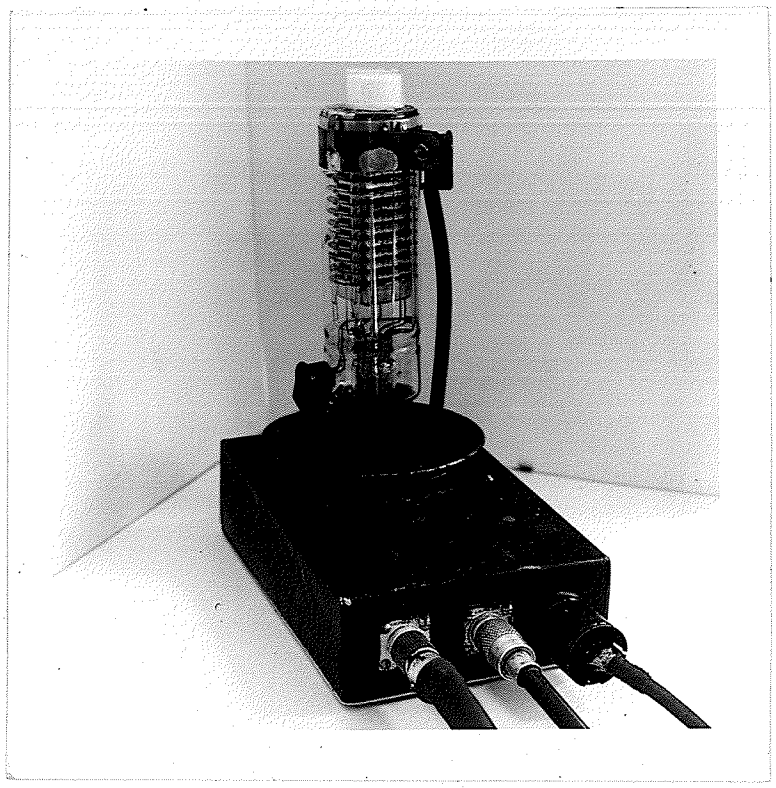


FIG. 2

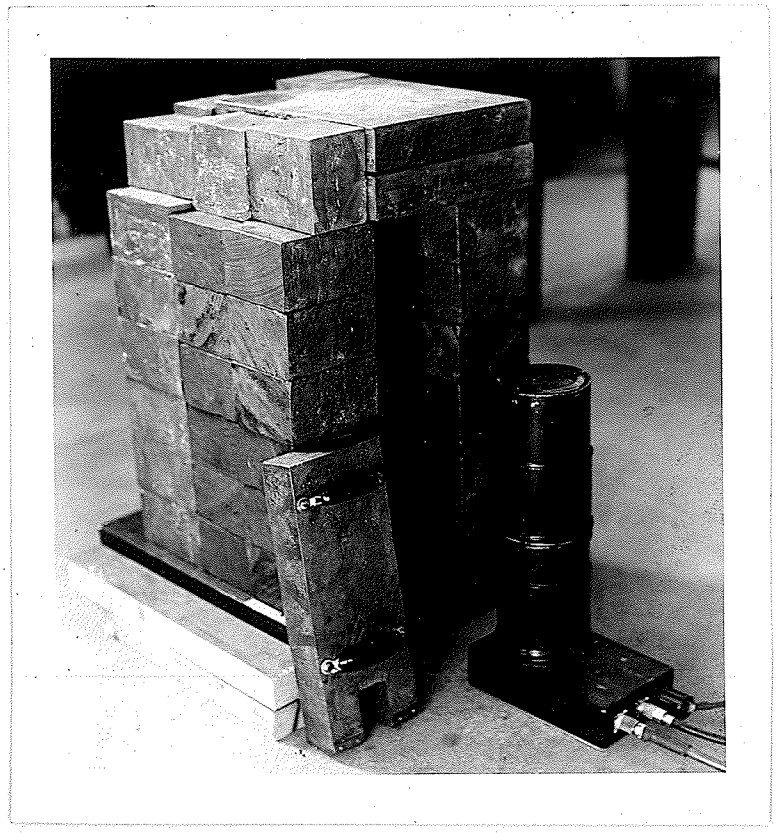


FIG. 4

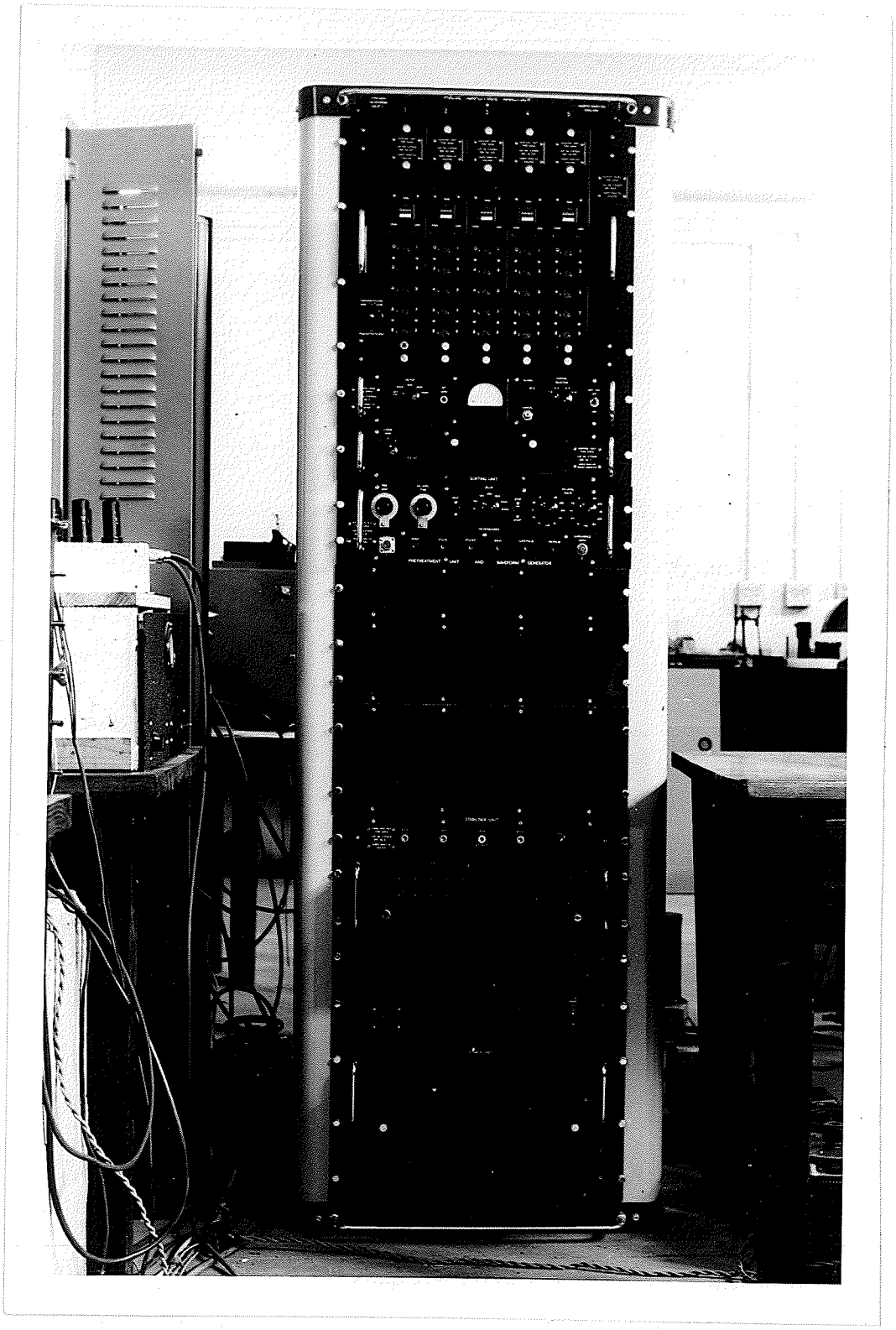


FIG. 3

oscillograph screen and photographed over a suitable length of time. Since the height of a pulse on the screen is proportional to the energy of the primary event in the crystal, an analysis of the pulses on a negative can lead to the determination of the energies of γ-rays incident on the crystal. The film used for this photographic analysis was Kodak Contrast Process Ortho.

EXPERIMENTAL RESULTS

The source used was 39 grams of highly purified lanthanum oxide (La_2O_3) (not enriched in La^{138}). It was placed in a container designed to fit snugly over the crystal. The cannister was sealed and the counter placed in a 3 in. lead 'castle' similar to the type illustrated in Fig. 4. The pulse height distribution obtained with the pulse analyser is shown in Fig. 5. Features A, B and D were identified as the photoelectric peaks due to three γ-rays of energies 535 $\pm$ 15 kev. (A), 807 $\pm$ 15 kev. (B) and 1390 $\pm$ 30 kev. (D). The 363 kev. γ-ray of I^{131} and the 1170 kev. and 1330 kev. γ-rays of Co^{60} were used to calibrate the energy scale. Feature C was identified as the maximum of a Compton distribution arising from the high energy γ-ray. This maximum is believed to correspond to that obtained in the earlier investigation mentioned previously.

The pulse height distribution was verified by studying the pulses as displayed on the oscillograph screen. Fig. 6

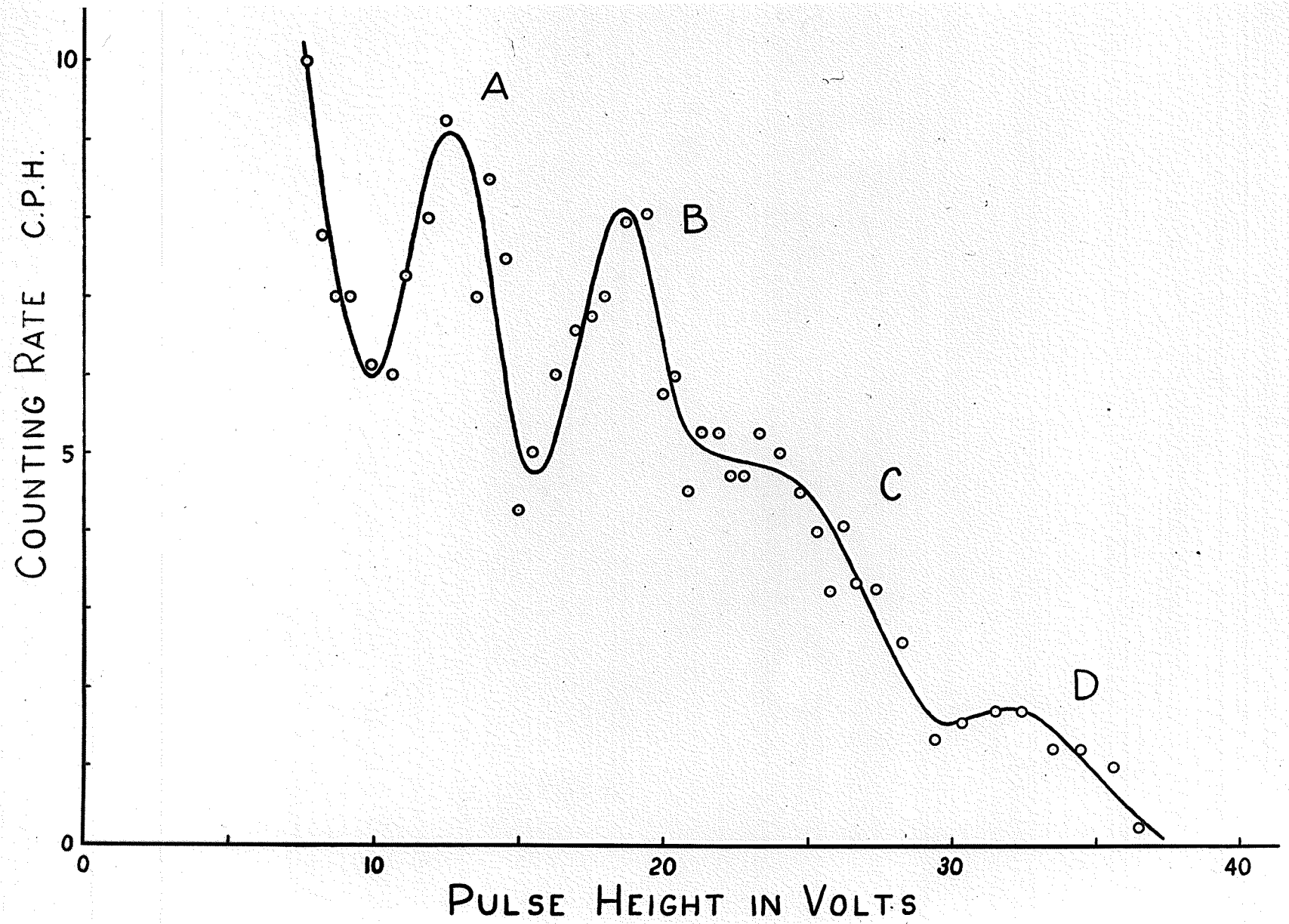

COUNTING RATE C.P.H.
10
5
0
A
B
C
D
0
10
20
30
40
PULSE HEIGHT IN VOLTS

FIG. 5

shows the result of a 40 hour exposure at f/4.5. The features mentioned above were easily identified in the photograph and energy determinations made. The values quoted in the preceding paragraph are, in reality, means of values obtained from these two distinct analyses. (The halo appearing in the photograph is an unavoidable consequence of long exposures. The filament of the cathode ray tube emits light in the red part of the spectrum and gives rise to such effects. By placing the pulses at a suitable position on the oscillograph screen, obliteration of the spectrum can be avoided.)

A further study of the source was made to verify the proposed K-capture process of the first investigation. A thin window Geiger counter (1 mg/cm^2) was placed over a thin sample of La_2O_3, the entire unit being enclosed by 3 in. of lead, and the source was suitably studied. It was concluded that the number of electrons or positrons emitted per gram of natural lanthanum having energies greater than 100 kev. was less than 0.2/sec. The variation of the sensitivity of the detector with γ-ray energy was known with sufficient accuracy to be able to estimate the relative intensities of features D, B and A as 1.0 : 0.65 : 0.30. From these data, and observations on a weighed sample of K^{40} (specific activity of 3.3 γ-rays per gm. per sec),[13] it was estimated that natural lanthanum gave 0.6 γ-quanta per sec. per gram. The specific activity of the sample was found to be 0.45 disintegrations per sec. per gram of natural lanthanum.

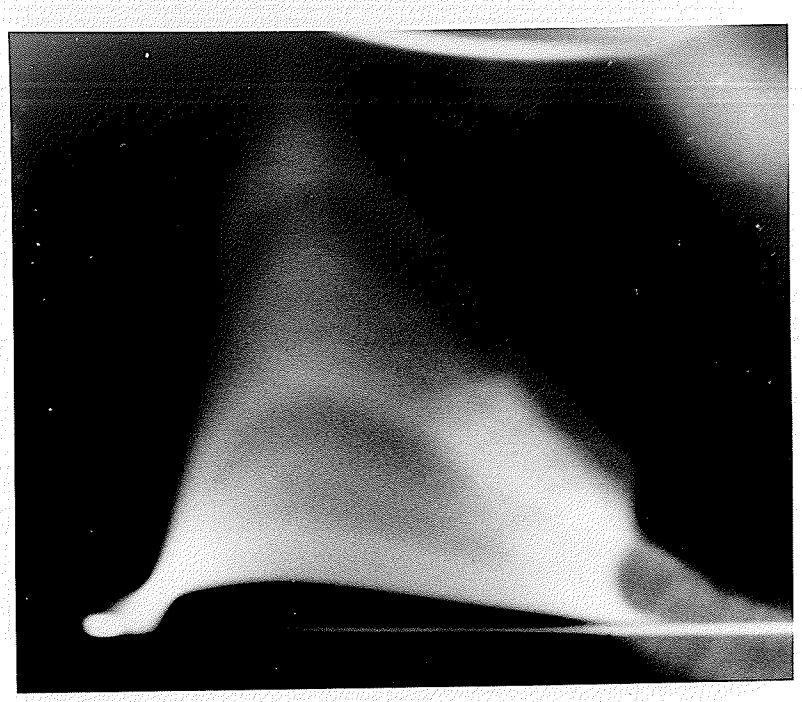


FIG. 6

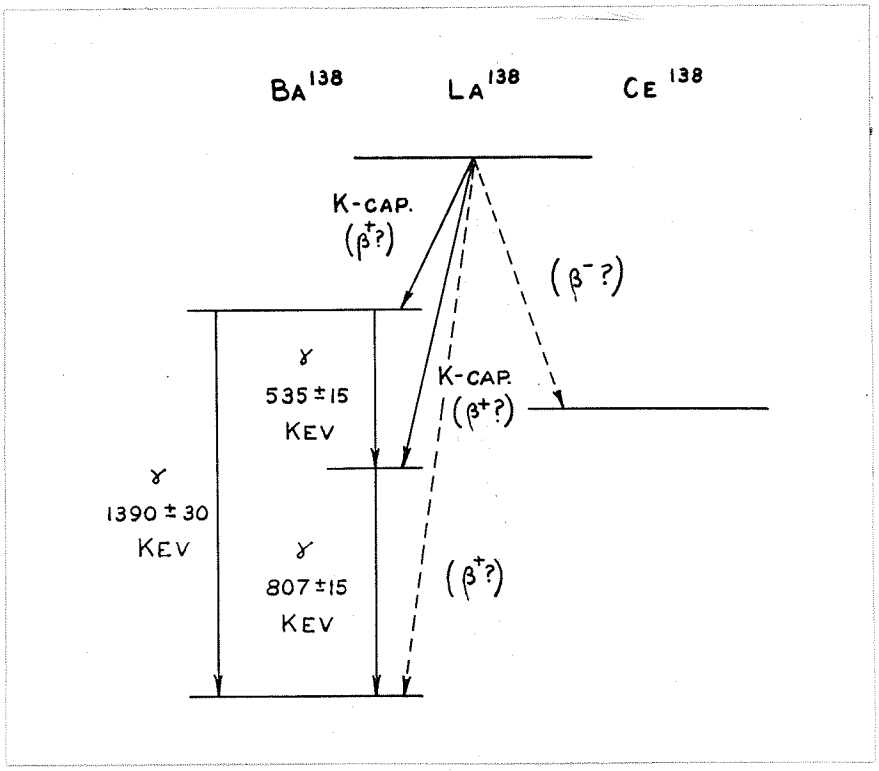


FIG. 7

The existence of the 807 kev. level in Ba^{138}, and the direct transition to it by the K-capture process was suggested by a consideration of the intensities quoted on P.15 and the relative intensities of the features in Fig. 6.

The relative intensities of the three features as exhibited on the photographic plate, coupled with the lack of evidence for emitted particles, led to the proposed decay scheme of Fig. 7. The two alternative modes of decay follow from the intense line in the photograph which corresponds to feature B. It was recognized, however, that further evidence must be sought in order to strengthen the argument for the K-capture process.

When a nucleus captures an orbital electron to form a product nucleus, the corresponding atom that is formed is lacking one of its innermost electrons. In the re-orientation of the electronic system, the characteristic X-rays of the product atom will be emitted. With this process in mind, it was decided to look for the X-radiation from Ba^{138}. To this end, the low energy radiations of RaD and I^{131} were examined with the same spectrometer. The γ-ray spectrum of RaD is shown in Fig. 8. The resolution of the line, which is associated with the 46 kev. γ-ray of RaD, was estimated as 37%. The low energy radiation associated with the decay of I^{131} is shown in Fig. 9. The first peak is due to the K X-radiation of Xe^{131}, which is the product formed by the beta-decay of I^{131}. The photoelectric peak at 80 kev. is due to a well-known γ-ray from I^{131}.[15] In Fig. 10, a photographic analysis of these features is shown. The 29 kev. X-ray can be seen at the bottom of the photograph, followed by the 80 kev. γ-ray, and the 164 kev.

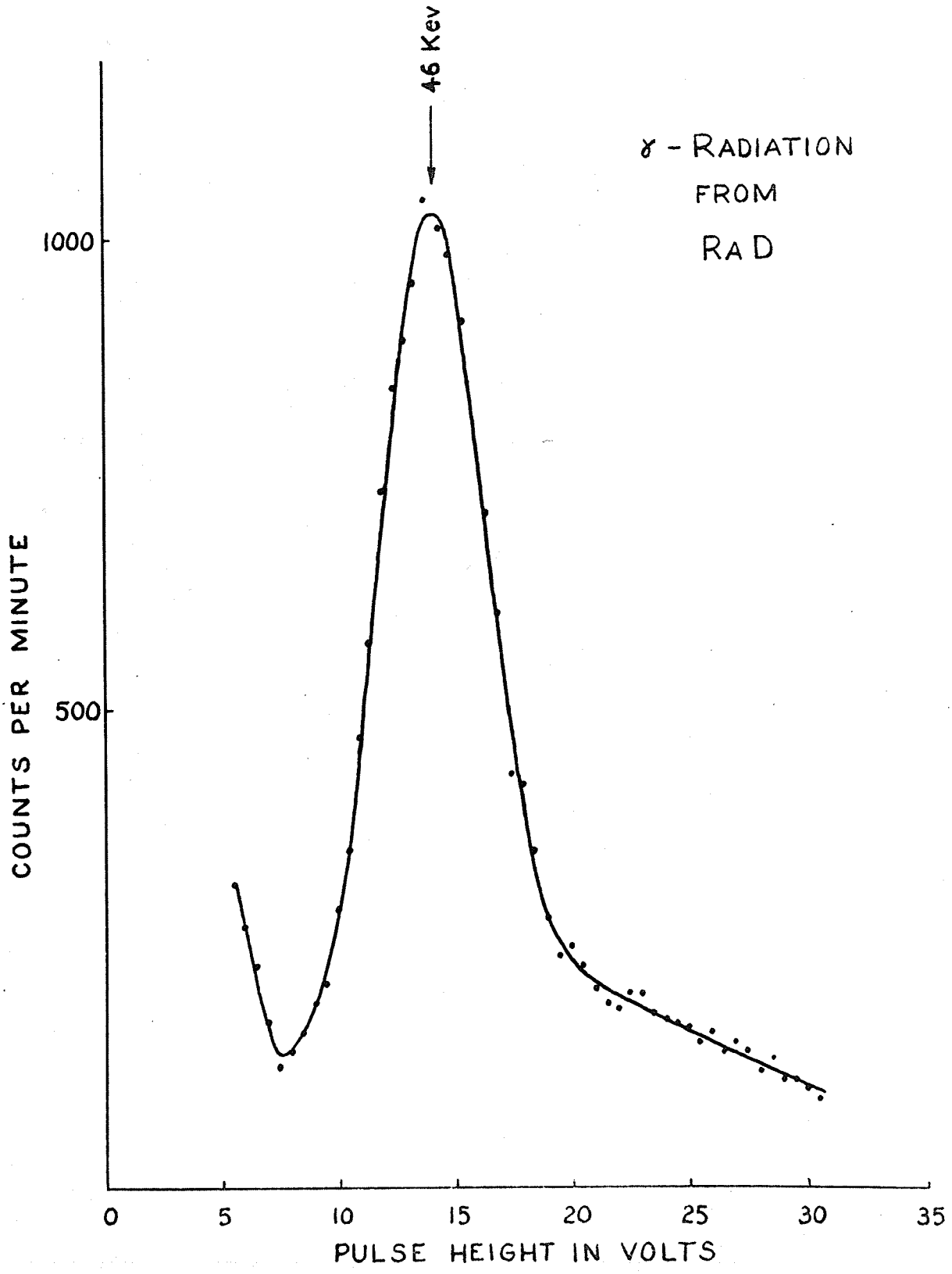

46 Kev
γ - RADIATION
FROM
RA D
1000
500
COUNTS PER MINUTE
0
5
10
15
20
25
30
35
PULSE HEIGHT IN VOLTS

FIG. 8

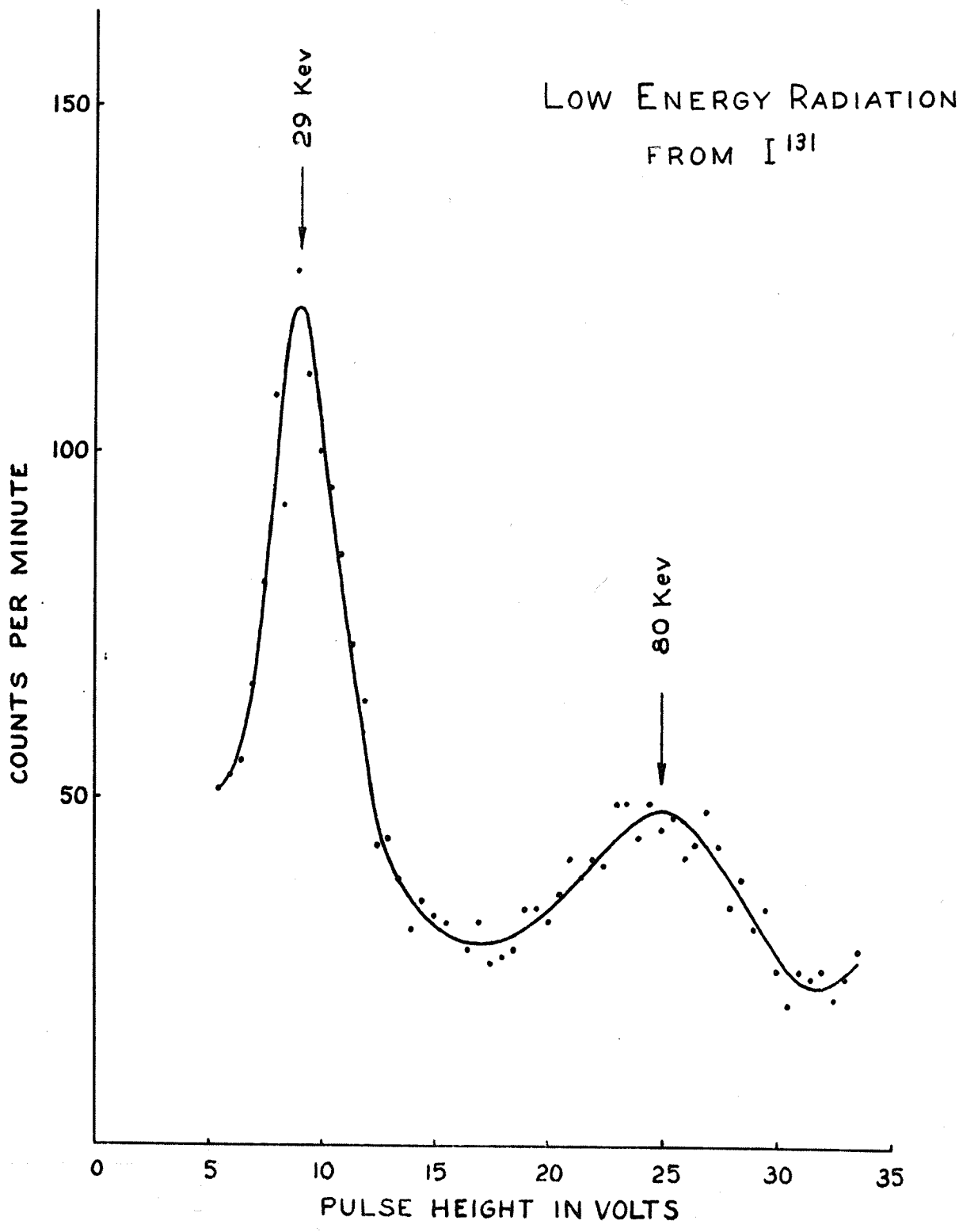

LOW ENERGY RADIATION
FROM I^{131}
29 Kev
80 Kev
150
100
50
COUNTS PER MINUTE
0
5
10
15
20
25
30
35
PULSE HEIGHT IN VOLTS

FIG. 9

γ -ray which runs into the trace of the blocking pulses at the top of the spectrum.

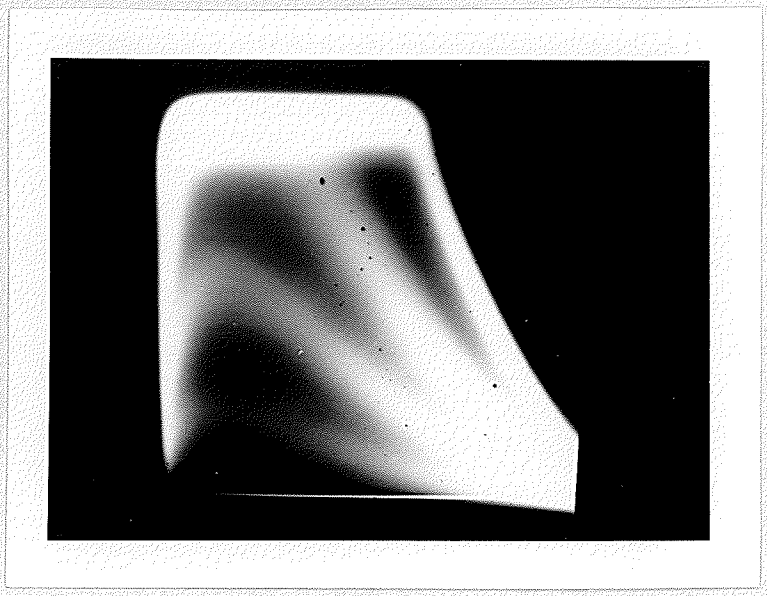


Fig. 10

Using these three prominent features to calibrate the energy scale, the X-ray region around 30 kev. was examined. In Fig. 11 the K X-ray peak associated with Ba^{138} is shown. The energy of the X-ray was found to be 32 $\pm$ 1 kev. in better agreement with the barium K X-ray (31.4 kev.), than that due to cerium (33.7 kev.).

From the data collected, it was possible to estimate the half-life for the activity of La^{138}. As mentioned above, the specific activity was found to be 0.45 disintegrations per second per gram of natural lanthanum. Using the known abundance of La^{138}, (0.089%), and the well-known relationship between specific activity and half-life,[16] La^{138} was found to have a half-life of 2.0×10^{11} years, a result that

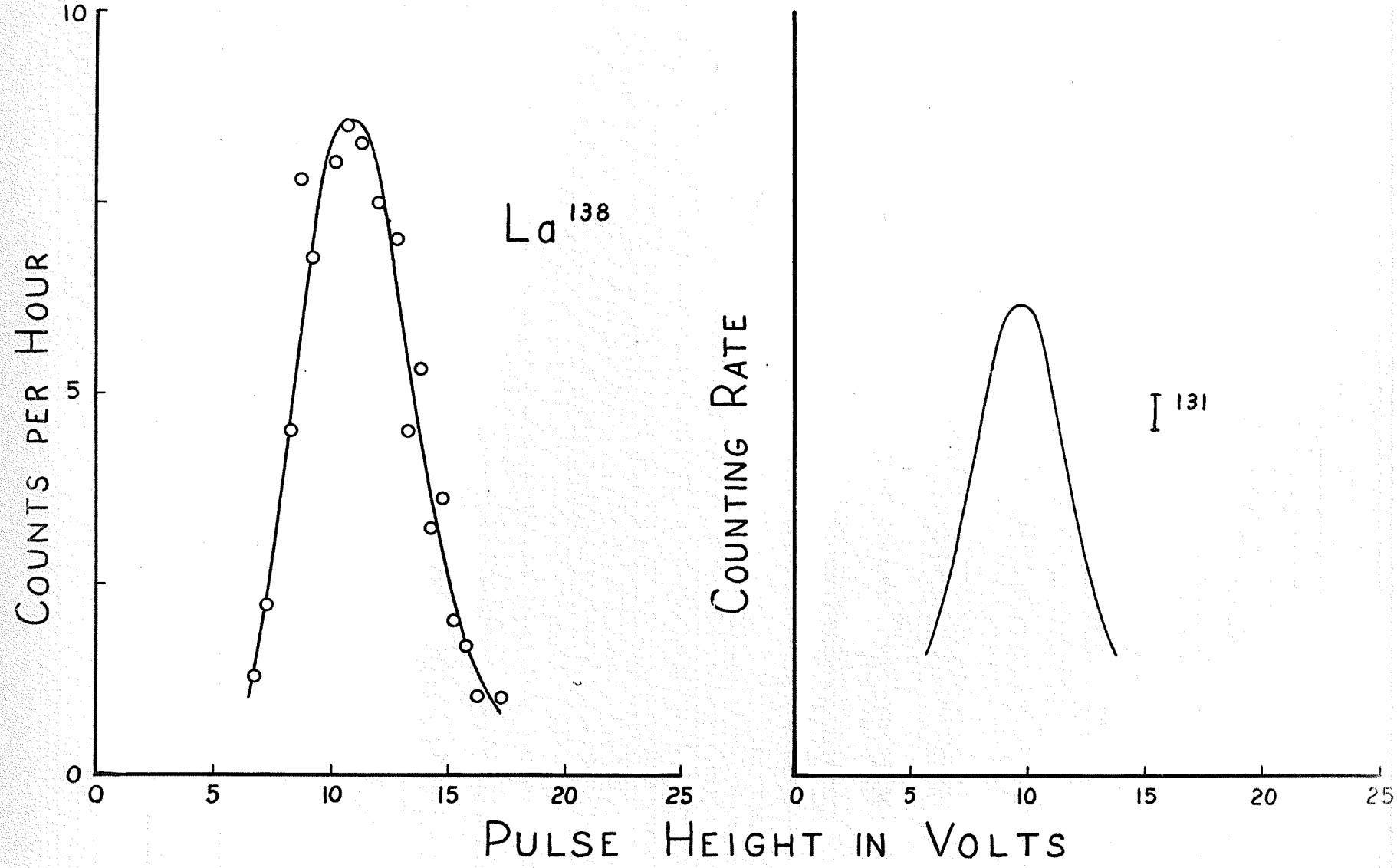

10
5
0
0
5
10
15
20
25
COUNTS PER HOUR
La 138
COUNTING RATE
I 131
0
5
10
15
20
25
PULSE HEIGHT IN VOLTS

FIG. 11

was found to be in excellent agreement with the earlier value (see reference 11.).

The evidence presented above seems to be complete in all respects, yielding as it does a decay scheme that is consistent with all the experimental data. The observations themselves represent a rather unique result in γ-ray spectroscopy, for the strength of the lanthanum source was estimated to be only 0.5 milli-microcuries in spite of its bulk. In all low activity work, the stability of the equipment used is the factor which ultimately determines the success of the undertaking. A search for γ-radiation from In^{115} will be undertaken in the near future, using improved shielding and a large (1-1/2 in. diam. x 1-1/2 in.) NaI(Tl) crystal in the spectrometer.

Before leaving the subject of La^{138}, it will be appropriate to mention at this point some interesting low energy studies that arose as a consequence of the search for the barium X-ray mentioned above.

RaD has long been known to have a γ-ray of very low energy (46 kev). In order to get information on the resolution of the spectrometer at very low energies, it was decided to study RaD in some detail. In Fig. 12, the shape of the RaD spectrum is shown under different conditions. Fig. 12a shows the pulse height distribution obtained when there is no absorbing material (except the thin aluminum foil on the crystal) between the source and the crystal.

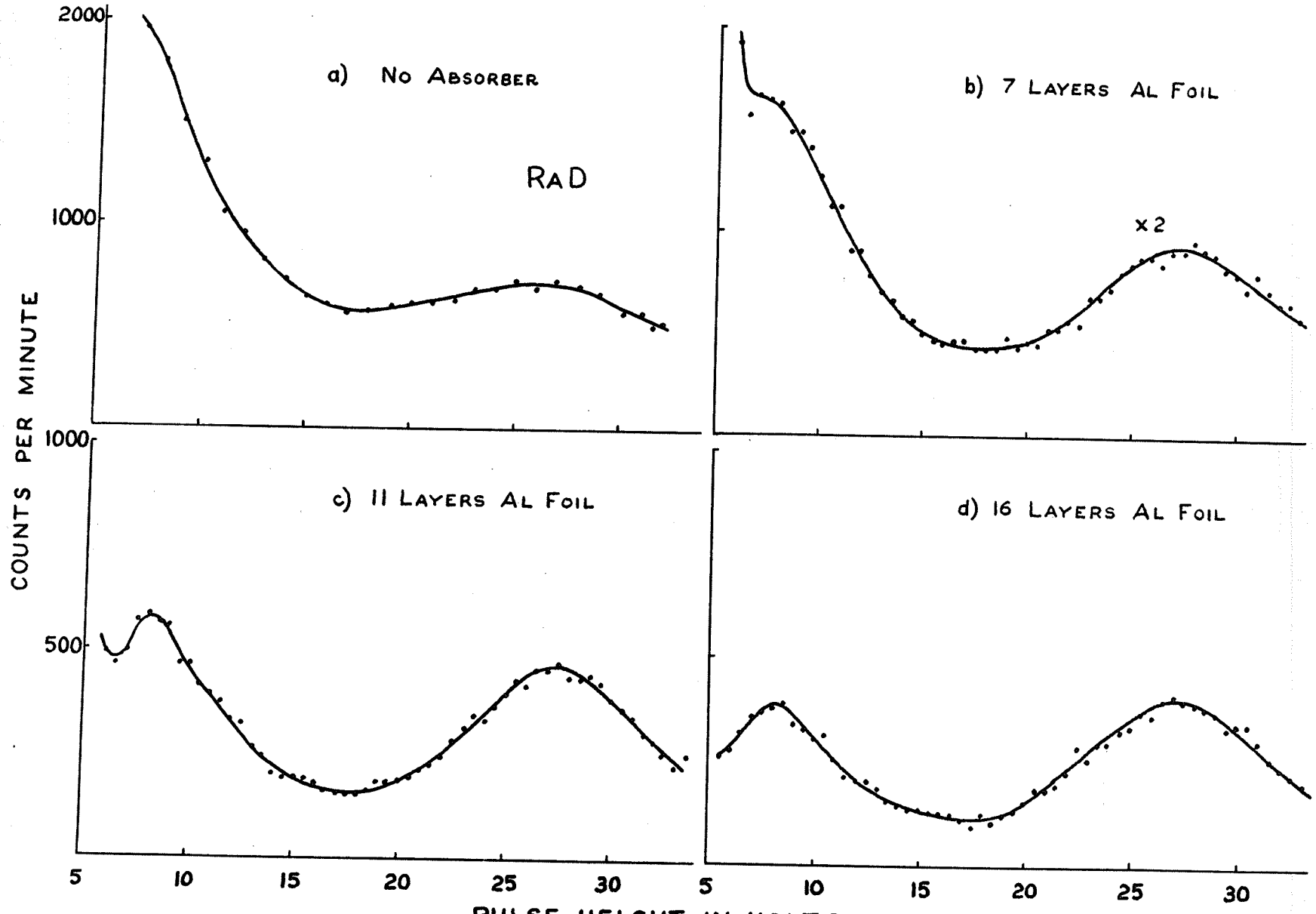

a) No Absorber
RaD
b) 7 Layers Al Foil
x2
c) 11 Layers Al Foil
d) 16 Layers Al Foil
2000
1000
1000
500
COUNTS PER MINUTE
5
10
15
20
25
30
5
10
15
20
25
30
PULSE HEIGHT IN VOLTS

FIG. 12

The spectrum is really a ($\beta + \gamma$) spectrum and the 46 kev. peak is not too prominent. As thin aluminum foil is placed between the source and the crystal, and the distribution re-examined, pronounced changes occur. The low energy β-particles ($E_{max} \doteqdot 18$ kev.) are gradually absorbed and the γ-ray spectrum is clearly exhibited. The 46 kev. peak becomes prominent and symmetric in shape. The low energy peak is due to the L X-ray of Bi^{210} (~ 13.5 kev.). The curves illustrate how vital it is when working with a counter that is extremely sensitive to low energy particles and radiations, to eliminate any unwanted radiations by proper absorption in foils.

Fig. 13 is another pulse height distribution of the γ-radiation from RaD. As indicated in the diagram, the photomultiplier voltage was raised by 60 volts in order to facilitate a closer study of the X-ray at 13 kev. It was found that the noise level of the tube started to become troublesome at from 2 - 5 kev., so that the range of use of this particular spectrometer was limited to energies of 5 kev. and greater. Due to the extremely poor resolution of the spectrometer at 46 kev. ($\sim 40\%$), early photographs of low energy spectra left much to be desired. As an illustration, the γ-ray spectrum of RaD is shown in Fig. 14a. The crystal used for this photograph was coated with oil and covered with aluminum foil. In Fig. 14b, the

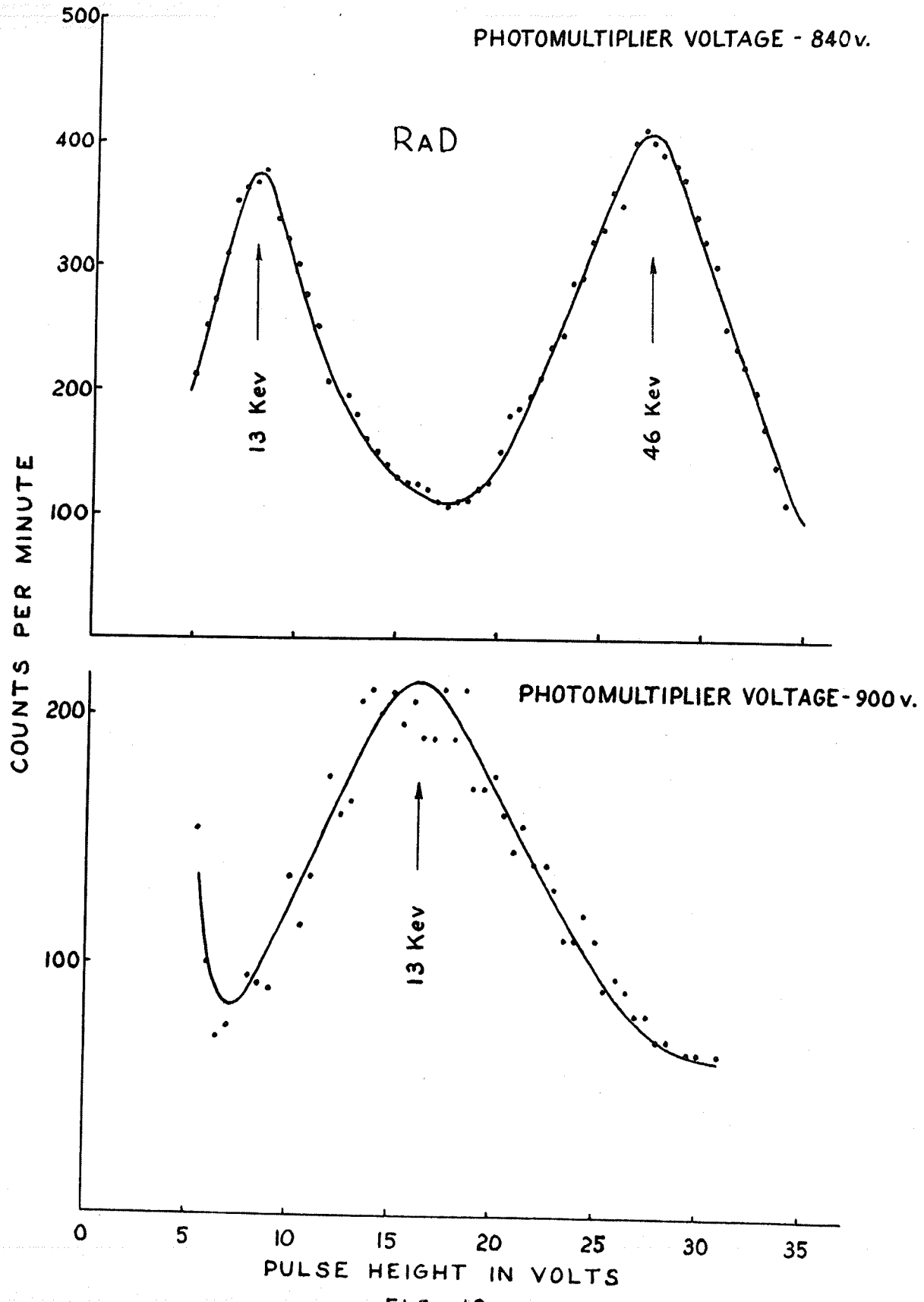
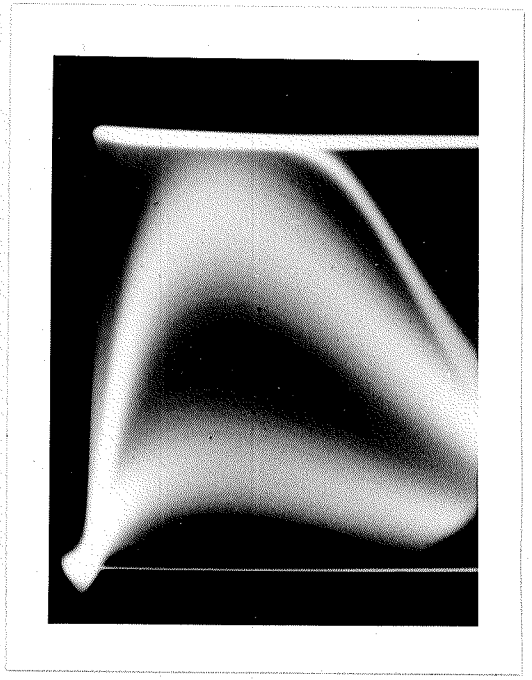
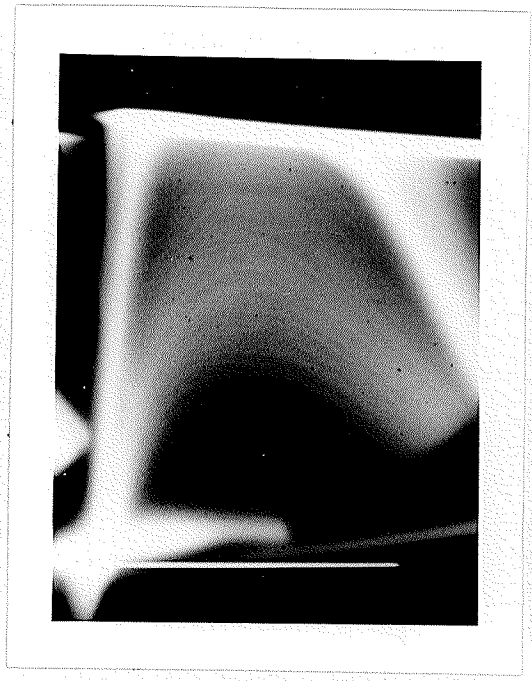

PHOTOMULTIPLIER VOLTAGE - 840 v.
RaD
500
400
300
200
100
13 Kev
46 Kev
COUNTS PER MINUTE
PHOTOMULTIPLIER VOLTAGE - 900 v.
200
100
13 Kev
0
5
10
15
20
25
30
35
PULSE HEIGHT IN VOLTS

FIG. 13



(a)



(b)

FIG. 14

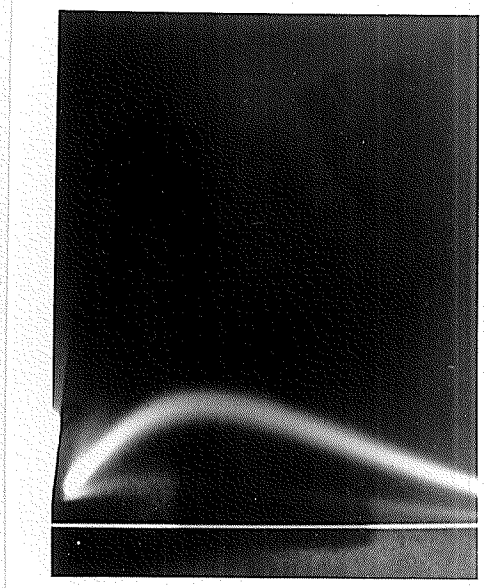


FIG. 15

13 kev. Bi^{210} X-ray only is shown. Up until recently, these photographs were the best that could be expected in this low energy region. Shortly before the time of writing, the same crystal was mounted on the E.M.I. 5311 tube, using the technique outlined briefly in the introduction. This consisted of using magnesium oxide powder as a diffuse reflector for the crystal. In Fig. 15, a photograph of the 46 kev. γ-ray of RaD is shown, utilizing this improvement in technique. A detailed study of this method has recently been undertaken at this laboratory (see Roulston PhD. Thesis 1952, University of Manitoba). It is hoped that wherever possible, future work to be done at this laboratory will be carried out with this new modification in technique.

Even at these energies, the relationship between pulse height and energy of the primary event in the crystal is still a linear one. The study of low energy radiation has been advanced to the point where 2 kev. X-radiation has been analyzed without tube noise affecting the results.[17] From these results, one can see the wide range of energies open to study with the scintillation counter. The proportionality of the device has been found to hold from the low energy X-ray region to γ-ray energies of several Mev. Evidence for the proportionality at high energies will be presented in a later section.

Another result of interest was secured by placing the RaD source over the lanthanum source used in the low activity

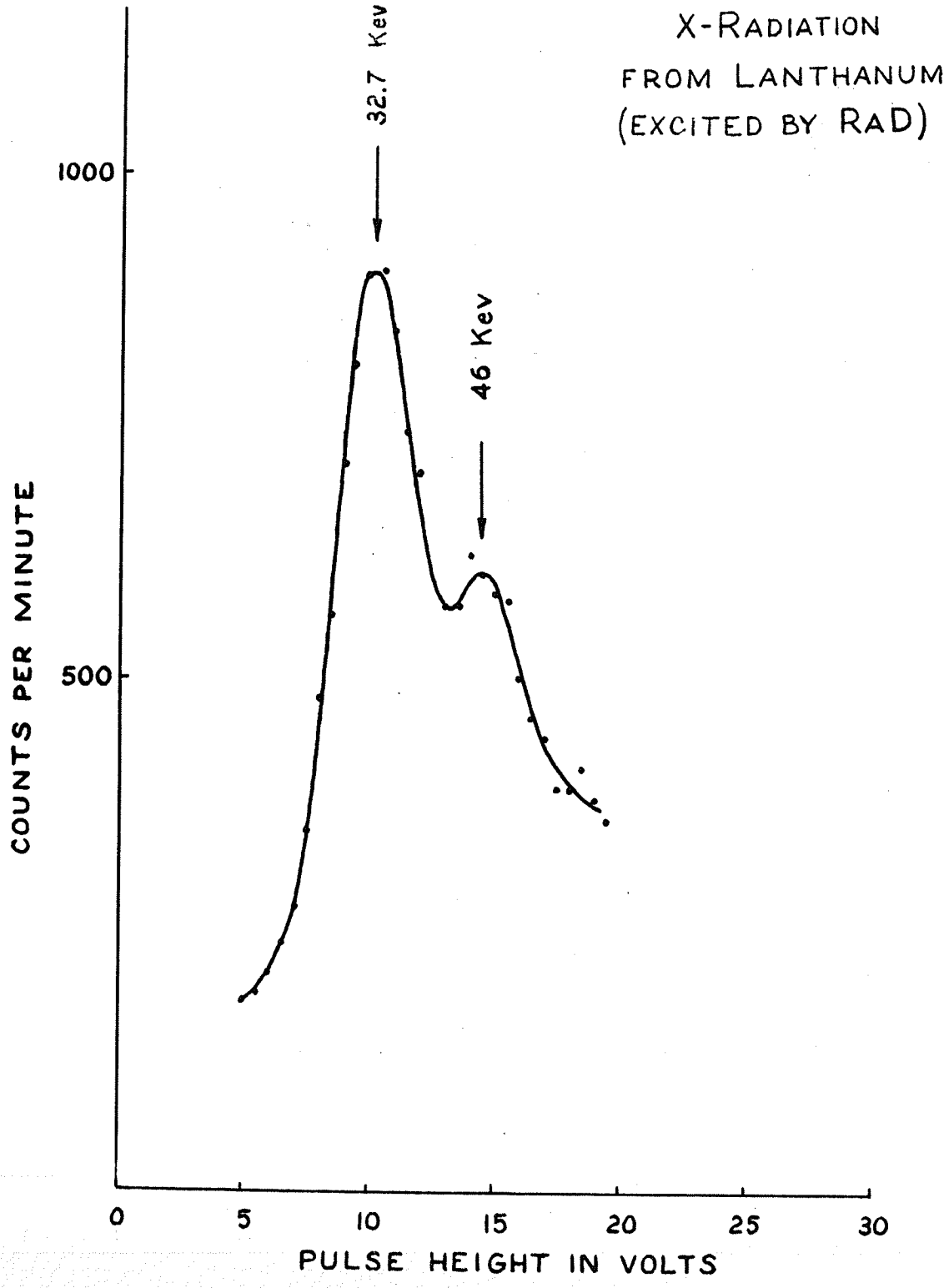

X-RADIATION
FROM LANTHANUM
(EXCITED BY RaD)
32.7 Kev
46 Kev
1000
500
COUNTS PER MINUTE
0
5
10
15
20
25
30
PULSE HEIGHT IN VOLTS

FIG. 16

work previously described. In this way, it was found possible to excite the K X-radiation of lanthanum (32.4 kev.). Fig. 16 shows the pulse height distribution of the excited radiation. The residue of the 46 kev. γ-ray from the RaD is still clearly visible. The possibility of using this technique for the identification of unknown substances is evident. By excitation of, and measurement of the energy of the characteristic X-radiation of an unknownsample, one could at least state to within two or three elements, what the main constituent of the sample was. At best, the method could only be considered as a first approximation, to be followed by suitable chemical procedures where these are possible.

We shall now proceed to discuss the γ-ray spectrum of polonium (RaF) as determined by scintillation spectrometer.

THE GAMMA RAY SPECTRUM OF POLONIUM

Because of its membership in the radium family, the activity of polonium 210 has been the subject of numerous investigations. It is known to decay by α-particle emission to stable Pb^{206}. In 1930, Bothe[18] showed that γ-radiation accompanied the decay of polonium, although further details concerning this γ-radiation were not forthcoming until 1947. At that time DeBenedetti and Kerner,[19] using an absorption technique, reported the existence of a single γ-ray of energy 800 kev. Seigbahn and Slatis[20] also measured the energy of this γ-ray and found it to have an energy of 773 kev. They also showed that there were no other γ-rays with energies exceeding 150 kev. This revived interest in polonium was brought about by some spurious results concerning the α-particle spectrum which were reported in 1946 by Chang.[21] The γ-ray spectrum was re-investigated in 1948 by Zajac, Broda and Feather,[22] who confirmed the high energy γ-ray and presented evidence for a soft component at 84 ± 4 kev. However, a recent investigation[23] by a group of workers at Oxford indicated that this low energy component was probably not nuclear in origin, but in reality, was only the K X-radiation from lead excited by the internal conversion of the high energy γ-ray. On the other hand, earlier experiments performed in this laboratory[24] with a scintillation spectrometer had given evidence of γ-rays

and 82 ± 1 kev. Recent work by Alburger and Friedlander[25] confirms the new value of the high energy component.

In spite of the numerous investigations of the polonium γ-ray spectrum, the existing discrepancies suggested that further study was necessary. In view of the results obtained in the early investigation carried out at this laboratory, it was decided that a complete re-examination of the problem should be undertaken.

EXPERIMENTAL PROCEDURE AND RESULTS

The scintillation spectrometer used consisted of an E.M.I. VX 5055 photomultiplier tube (1-3/4 in. diam. cathode) and a 1 in. cube of NaI(Tl) crystal. The crystal was highly polished, liberally coated with mineral oil, and covered with thin aluminum foil. The electronic equipment was the same as that described in the preceding section. A 3 in. lead castle was used to reduce the background counting rate.

The source of 138-day polonium was obtained from the Eldorado Mining and Refining (1944) Ltd., and consisted of 4 millicuries of polonium nitrate crystals. A preliminary investigation of the source was carried out to determine whether or not radioactive impurities were present. The results shown in Fig. 17 indicated that the polonium source contained a large quantity of 22-year RaD. The low energy region shown in Fig. 17a was studied and then the RaD spectrum was normalized to the same counting rates and fitted over the polonium spectrum. The difference between the two distributions is quite evident. It was decided to attempt a chemical separation of the polonium from the RaD present in the source.

The Chemistry Department of the University of Manitoba carried out the separation. A brief outline of the procedure follows. The polonium nitrate crystals were dissolved in dilute H_2SO_4. The liquid was then taken off by pipette

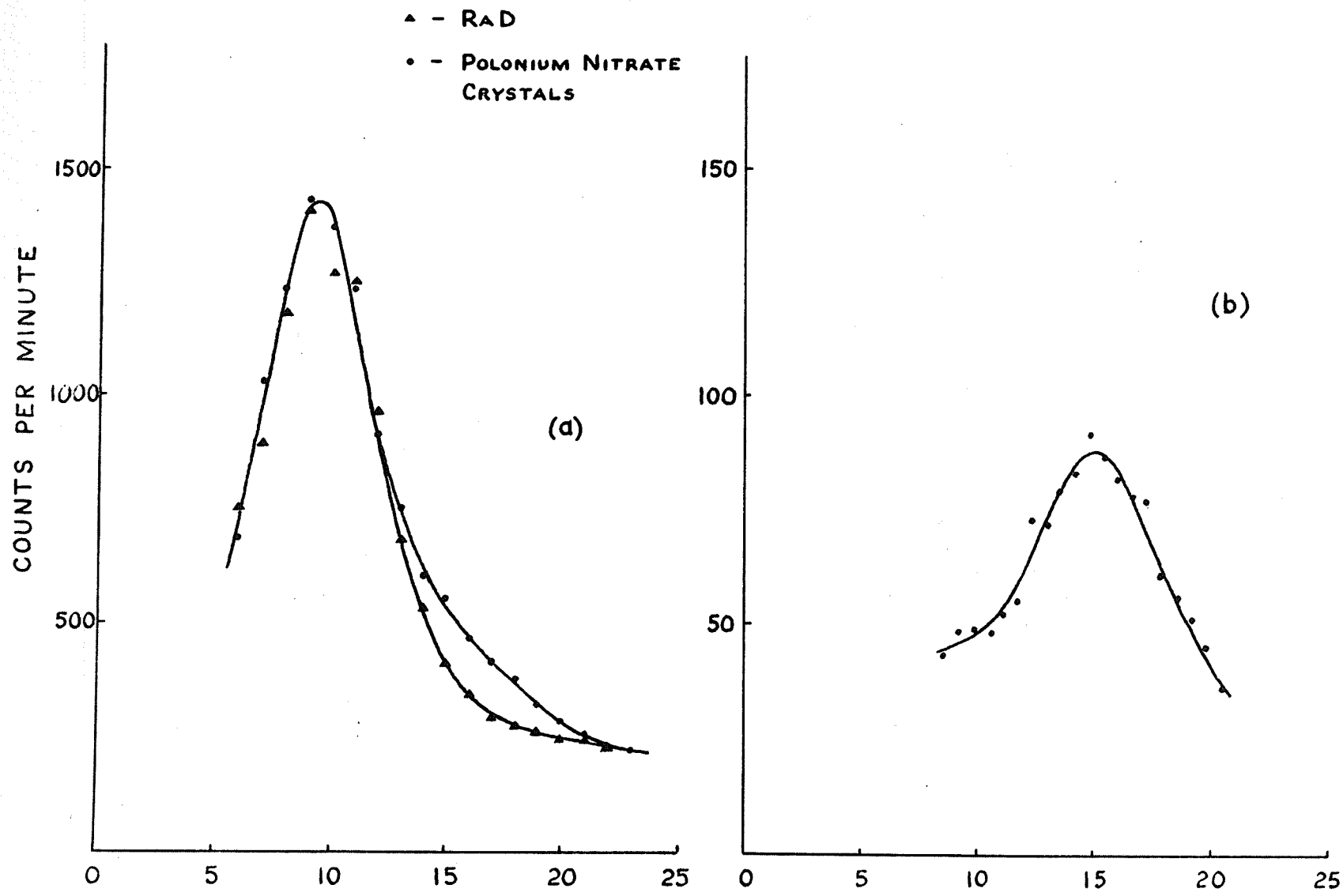

▲ - RaD
• - Polonium Nitrate Crystals
COUNTS PER MINUTE
1500
1000
500
0
5
10
15
20
25
(a)
150
100
50
0
5
10
15
20
25
(b)
PULSE HEIGHT IN VOLTS

FIG. 17

and treated in a centrifuge. After removal from the centrifuge, a trace amount of $BaCl_2$ (0.1 N) was added to the liquid to bring down any radium present as a precipitate. Once again the liquid was taken off and treated in the centrifuge. The remaining solution contained polonium only, in the form of a sulphate. It was placed in a suitable glass container and became the source for subsequent investigations. The total volume of the source was about 2 cm^3. The low energy region of the polonium spectrum was re-examined after the separation had been carried out. The resulting distribution is shown in Fig. 17b. The elimination of most of the RaD contaminant is evident from a comparison of the two graphs.

The high energy section of the polonium spectrum was then studied using the pulse amplitude analyzer. The pulse height distribution is shown in Fig. 18. A polonium beryllium source which contained a radium impurity was used to calibrate the energy axis. This source had had an initial strength of 2 millicuries some 24 months before. Since that time, the polonium content had decreased until only a residue was now present. The peak corresponding to the high energy γ-ray of polonium can be seen even in the radium distribution of Fig. 18. It coincides in position with the feature obtained with the newer polonium source discussed above. Three prominent features of the radium spectrum due to the impurity were identified. The values given to these

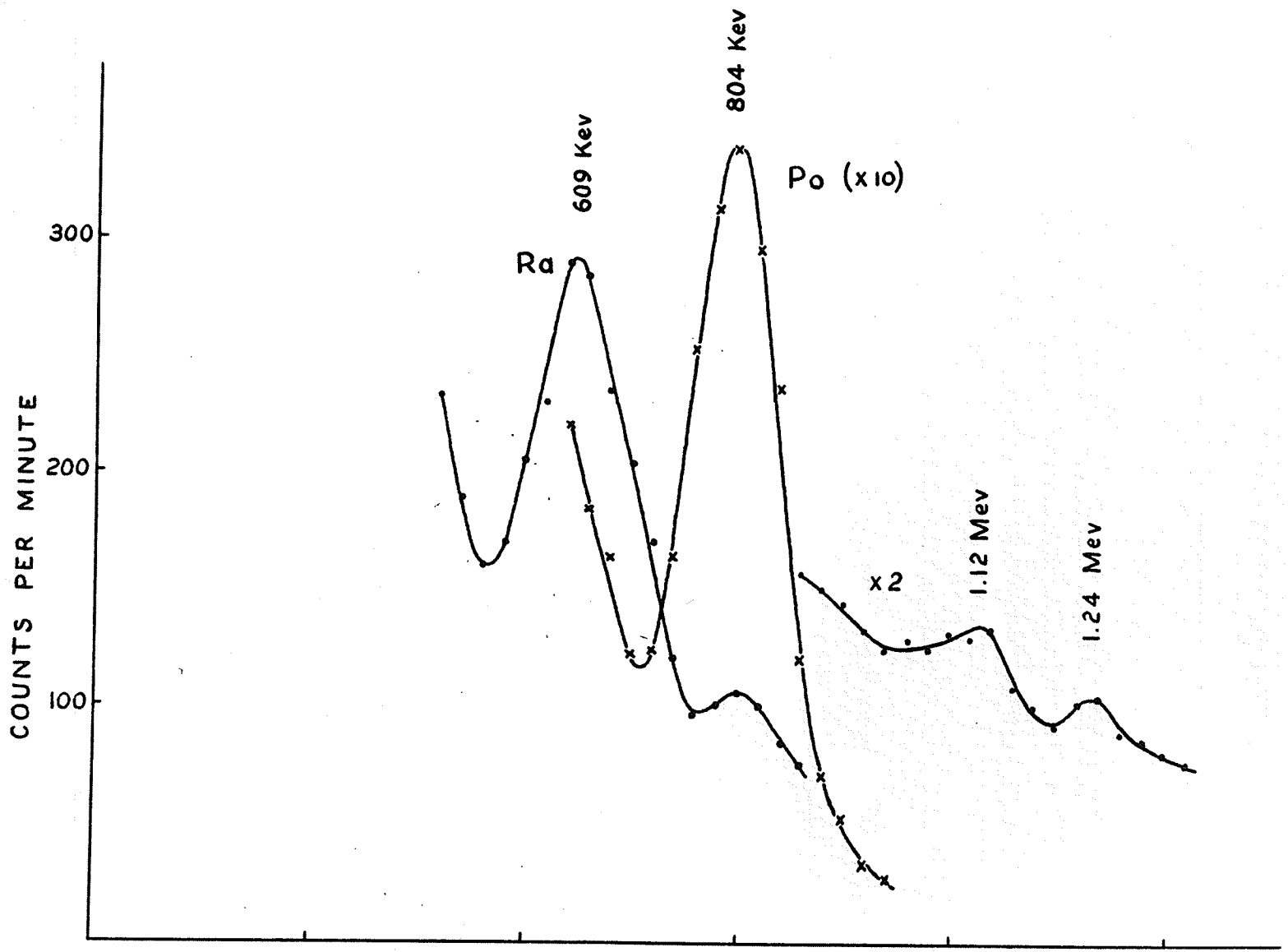
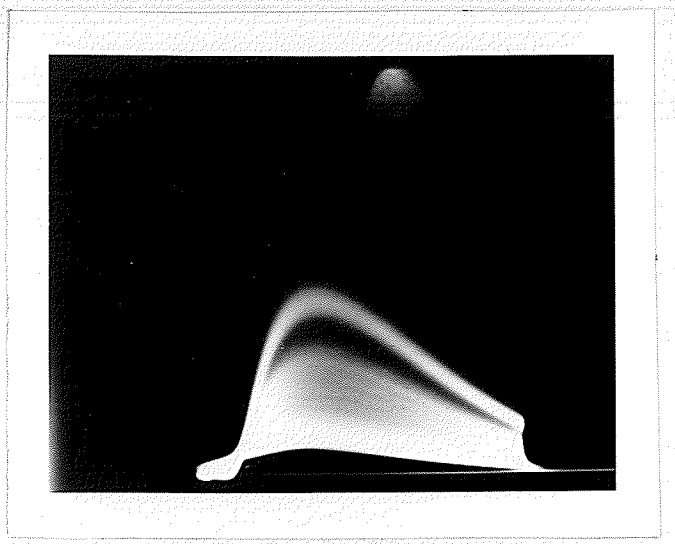

804 Kev
609 Kev
Po (x10)
Ra
300
200
100
COUNTS PER MINUTE
x2
1.12 Mev
1.24 Mev
0
5
10
15
20
25
PULSE HEIGHT IN VOLTS

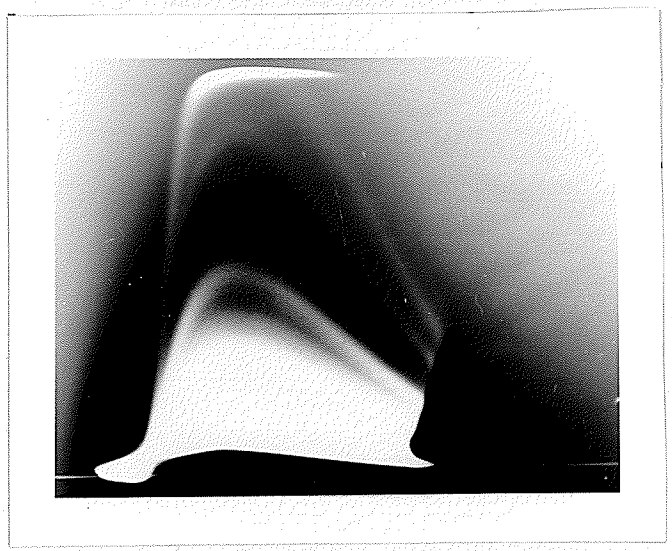
FIG. 18

γ-rays were taken from a recent paper by Cork.[26] The energy of the polonium γ-ray was found to be 804 kev. on the basis of these radium features. The energy was also determined by the photographic technique described earlier. In Fig. 19, photographs of the polonium, thorium and radium spectra are given. In Fig. 19a, the polonium high energy γ-ray is shown. The photograph also indicates the lack of any other γ-radiation above 200 kev., a result in good agreement with the conclusions of Seigbahn. Fig.19b shows the spectrum of thorium. The most prominent feature is a line due to a γ-ray of 960 kev.[27] Another line at 1.60 Mev. is also clearly visible. The radium spectrum is presented in Fig. 19c. The very intense line is due to the well-known 609 kev. γ-ray. By using the energies of these three features, it was possible to estimate the energy of the line in the polonium spectrum. It was found to be 798 kev. In view of the slight difference between this value and that found from the pulse height distribution, it was decided to take a mean value of 801 $\pm$ 4 kev. as the energy of the γ-radiation from polonium. The result disagrees with the work of Seigbahn and Slatis but is in excellent agreement with the results of Alburger and Friedlander who found the energy to be 800 $\pm$ 6 kev.

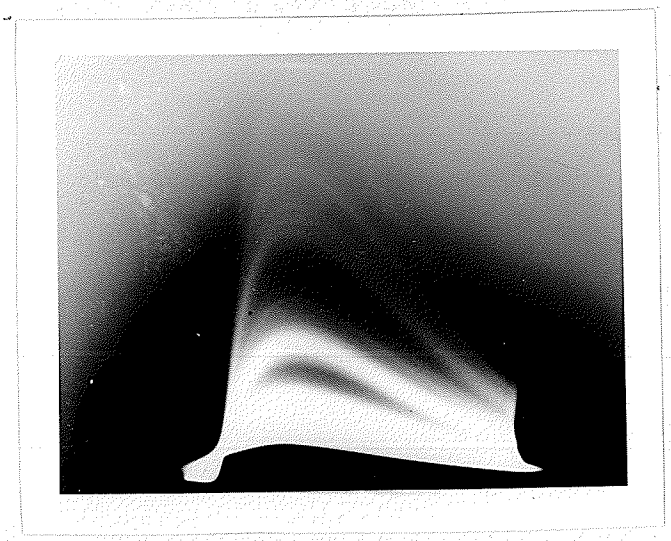
The low energy radiation from polonium was studied using the same spectrometer. Care was taken to have the polonium



(a)



(b)



(c)

FIG. 19

source, and the calibration source (in this case, I^{131}) in completely identical geometries. The I^{131} source occupied a volume comparable to that of the polonium. Its strength was estimated to be a few microcuries. The container used to hold it had about the same wall thickness as that for the polonium. The 80 kev. γ-ray from I^{131} was examined with the pulse amplitude analyser. The polonium low energy feature was then studied, followed by a repetition of the I^{131} curve. The polonium curve was normalized to the same counting rate as the I^{131} distribution and plotted on the same graph. The result is shown in Fig. 20. It is evident from the result that the polonium low energy radiation has an energy less than 80 kev. In a recent discussion of the problem given by Grace et al (see reference 23), the authors concluded that there was no low energy γ-radiation associated with polonium. It was contended that only the K X-radiation of lead was present and that this had been incorrectly identified by the earlier workers. Although the value of the polonium peak shown in Fig. 20 is about 2 kev. higher than the K_{α_2} X-ray of lead, it is in good qualitative agreement with the mean energy of the complete K X-radiation spectrum. A further examination of the low energy spectrum was carried out using the photographic technique. The 80 kev. γ-ray of I^{131} is shown above the 29 kev. X-ray in Fig. 21a, while the low energy radiation from polonium is exhibited in Fig. 21b. Unfortunately, the resolution of the spectrometer makes a precise measurement of the energy of this polonium line

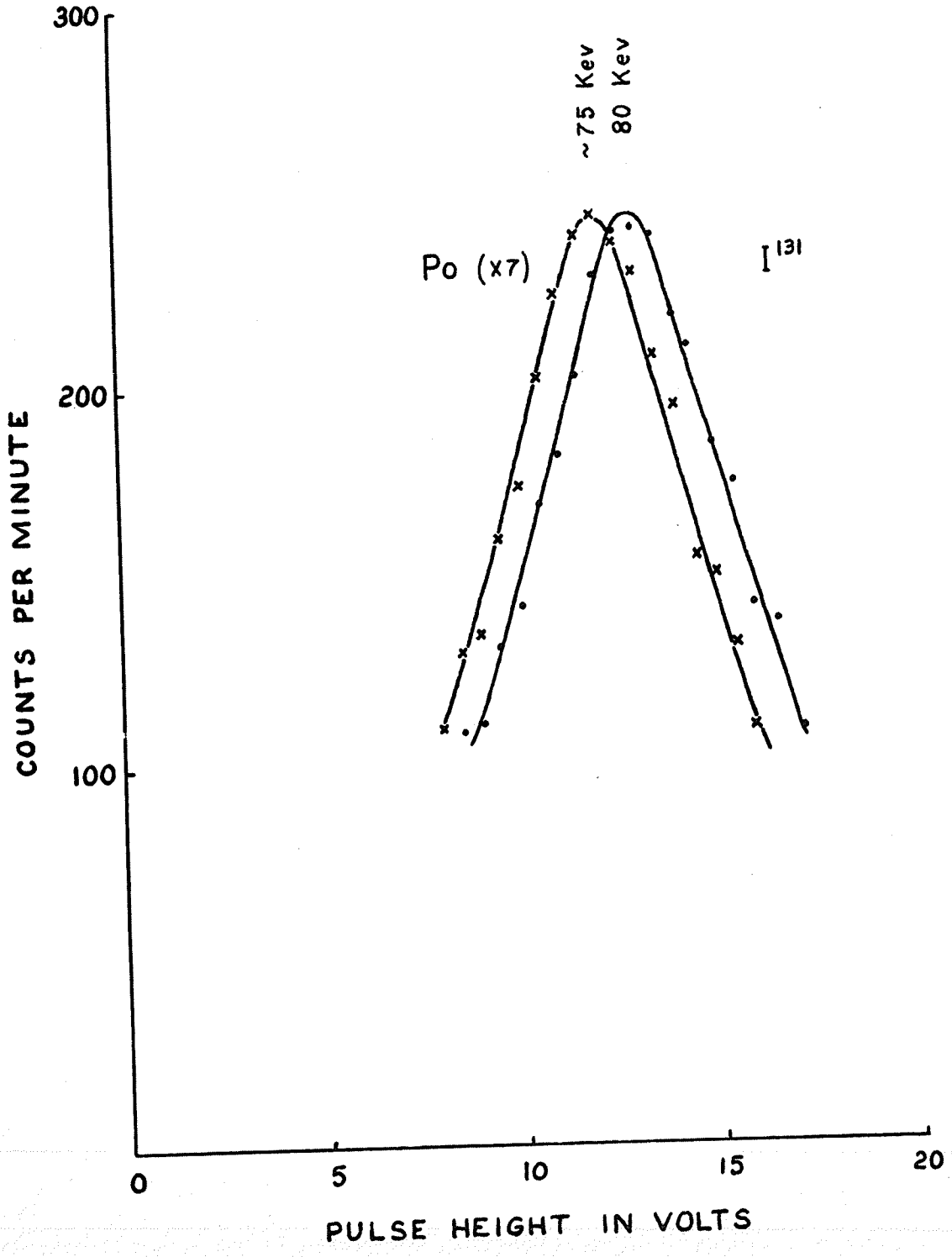
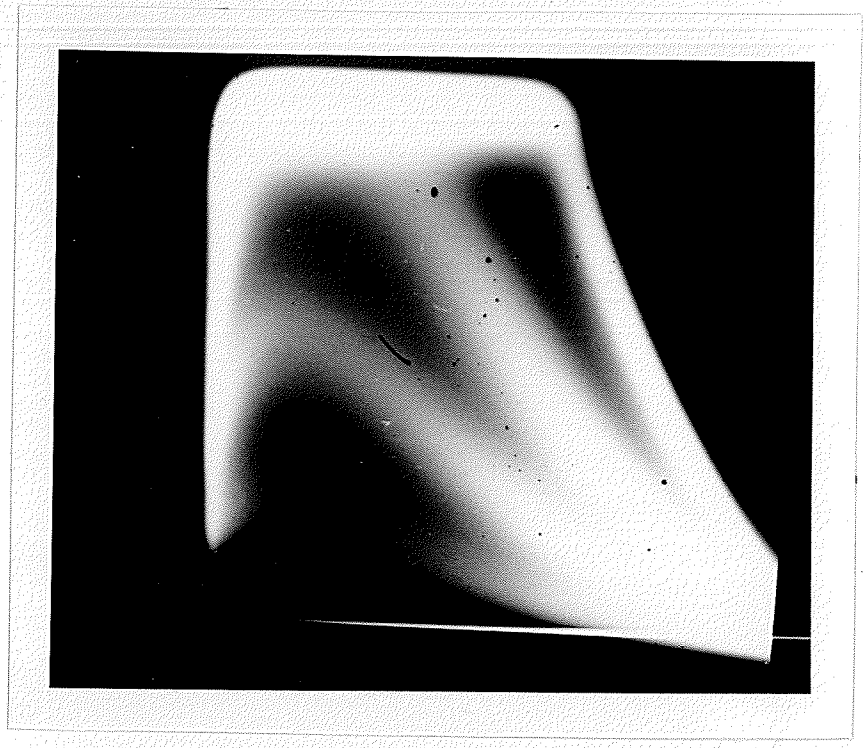
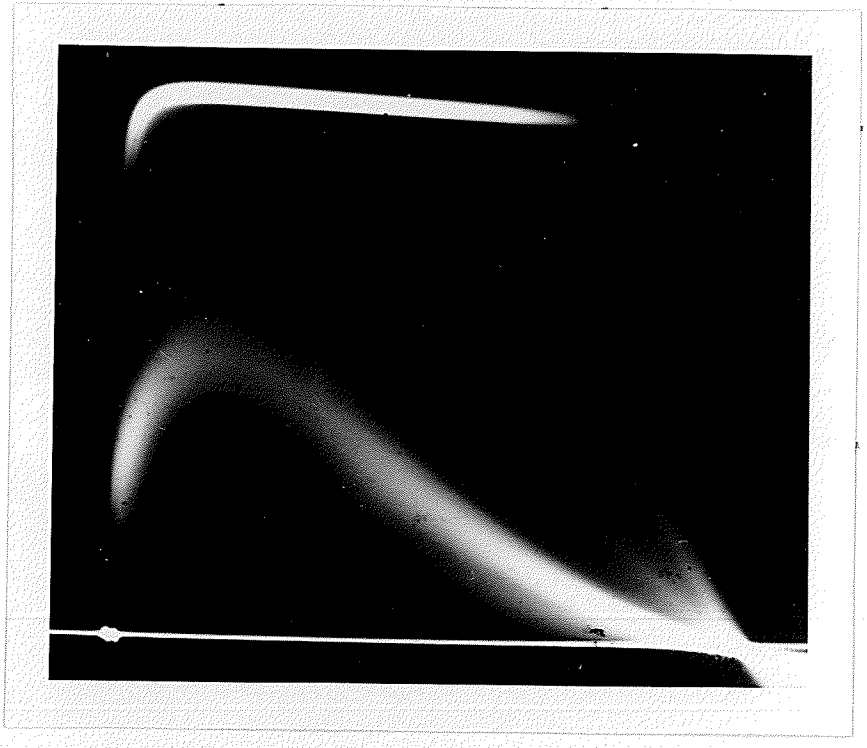

300
200
100
COUNTS PER MINUTE
~75 Kev
80 Kev
Po (x7)
I 131
0
5
10
15
20
PULSE HEIGHT IN VOLTS

FIG. 20



(a)



(b)

FIG. 21

difficult. This difficulty is not experienced to the same extent in a pulse height distribution where, even though the resolution on a line may be poor, its center may be determined with reasonable accuracy. Such measurements as could be made from the photograph indicate that the energy of the polonium line is less than or equal to 80 kev. Keeping the aforementioned difficulties in mind, it is evident that this result is not in disagreement with the distribution in Fig. 20. The evidence contained in that distribution suggests that the earlier results obtained in this laboratory were a little high.

In conclusion, a summary of the present state of the problem may prove useful. The energy of the high energy γ-ray from polonium seems to be 801 ± 4 kev. rather than 773 kev. as is usually quoted. The low energy radiation now appears to be merely the K X-radiation from lead, excited by the internal conversion process. This conclusion certainly accounts for the data collected to date, although the possibility of low energy γ-radiation cannot be entirely excluded on the basis of the evidence presented here.

CONCLUSION

The use of the scintillation spectrometer for the study of γ-ray spectra has been illustrated by means of two examples - La^{138} and Po^{210}. The value of the instrument is apparent both from the results presented here and the numerous papers on scintillation spectroscopy which have appeared in recent journals. Even at this stage of development, the spectrometer has proven to be a useful tool in nuclear physics. With further improvements in resolution, it will be possible to study complex γ-ray spectra with greater accuracy. It is evident, however, that radical changes in the counter will have to be made if the resolution is to approach that obtainable with a conventional magnetic β-ray spectrometer.

AN INVESTIGATION

OF THE

SCINTILLATION BETA-RAY SPECTROMETER

INTRODUCTION

Most of the crystals known to fluoresce when excited by γ-radiation can also be excited through bombardment with charged or uncharged particles.[28] The possibility of using the scintillation counter for the analysis of a beta-ray spectrum was first demonstrated in 1949.[29,30,31] At that time, anthracene was used as the detector. The use of this crystal for the detection of β-particles and neutrons has already been mentioned in a previous section of this paper. The early work showed that the end-point of a beta spectrum could be determined with reasonable accuracy either from the pulse height distribution directly, or from a suitable Kurie plot. The accuracy of the method was first demonstrated by Bell and Cassidy (see reference 30).

It is not to be expected that the scintillation spectrometer will replace the conventional β-ray spectrometer in the study of beta spectra. However, the device does supply a new approach to some of the problems of beta spectroscopy. Beta sources of low intensity can be studied with considerable ease with a scintillation spectrometer, because thin sources of fairly large surface area can be used. The thickness of a low intensity source is usually a serious drawback in conventional β-spectrometers.

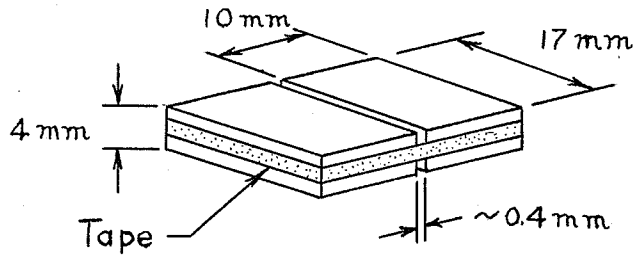
With the rapid advances being made toward improved resolution in photomultiplier tubes, it is probable that the scintillation spectrometer will prove to be of considerable use in the determination of the end-point energies of β -spectra, particularly when low intensities are involved.

In the sections to follow, the beta spectra of phosphorus 32, iodine 131 and iridium 192 will be discussed. The results presented are to be regarded as preliminary in the sense that more extensive investigations are contemplated for the near future.

DESCRIPTION OF THE APPARATUS

In using a scintillation spectrometer for the study of beta spectra, one must use a crystal phosphor that is an efficient detector of electrons. In the work to be described here, clear crystals of anthracene were used.

The E.M.I. 5311 photomultiplier used was mounted inside a large light-tight metal box. The circuits associated with the tube were identical with those previously described (see page 12). The crystal detector consisted of two pieces of anthracene in the form of parallelopipeds with dimensions 4, 10 and 17 millimeters. The crystals were held together by a thin strip of tape, as shown in the sketch below.



After the source (of suitable thickness) had been placed in the narrow gap between the crystals, they were placed on the photomultiplier window with a thin coating of oil supplying the crystal-window bond. The outer faces were covered with thin aluminum foil and a plastic cover which kept the crystal system rigid.

The pulses from the photomultiplier were passed through a cathode follower stage to a linear amplifier similar to Model 204-C mentioned earlier. After suitable amplification, the pulses were fed into a single channel differential discriminator which was designed and built in this laboratory.[32] The channel width was set at one volt and the pulse height distribution was scanned by moving this 'gate' over the range 0 to 100 volts. The pulses from the discriminator were counted with a scaler (Dynatron Radio Ltd.,Eng. - Type 1009A). (A spectrometer similar to the one just described, using two anthracene crystals, has recently been used successfully by Ketelle[33] in the study of the β -ray spectrum of Ca^{45}.)

THE BETA SPECTRUM OF PHOSPHORUS 32

The beta spectrum of 14.3-day P^{32} is simple in that there is no γ-radiation associated with it. For this reason, P^{32} has been studied extensively in order to gain information concerning the fundamental process in beta-decay - the emission of an electron from the nucleus. The energy of the end-point of the spectrum has been measured many times. Values as high as 1.75 Mev. have been quoted in the literature,[34] but recent measurements yield the somewhat lower value of about 1.71 Mev.[35,36,37] In view of the range of values quoted above, it was decided to take 1.72 Mev. as the end-point energy of the spectrum.

The P^{32} source was prepared as follows. Thin rice paper (2 mg/cm^2) was dipped into an aqueous solution containing radioactive phosphorus as a sulphate. The paper was dried, cut to a suitable size, and mounted between two pieces of the same material which were then sealed together to form a thin source with a thickness of about 6 mg/cm^2. Its strength was of the order of one or two microcuries. The source was mounted between the anthracene crystals and studied with the spectrometer described above.

The spectrum of the β-particles emitted by the source is shown in Fig. 22. The end-point was found and an energy of 1.72 Mev. ascribed to it. In this way, the pulse height axis was calibrated for succeeding experiments.

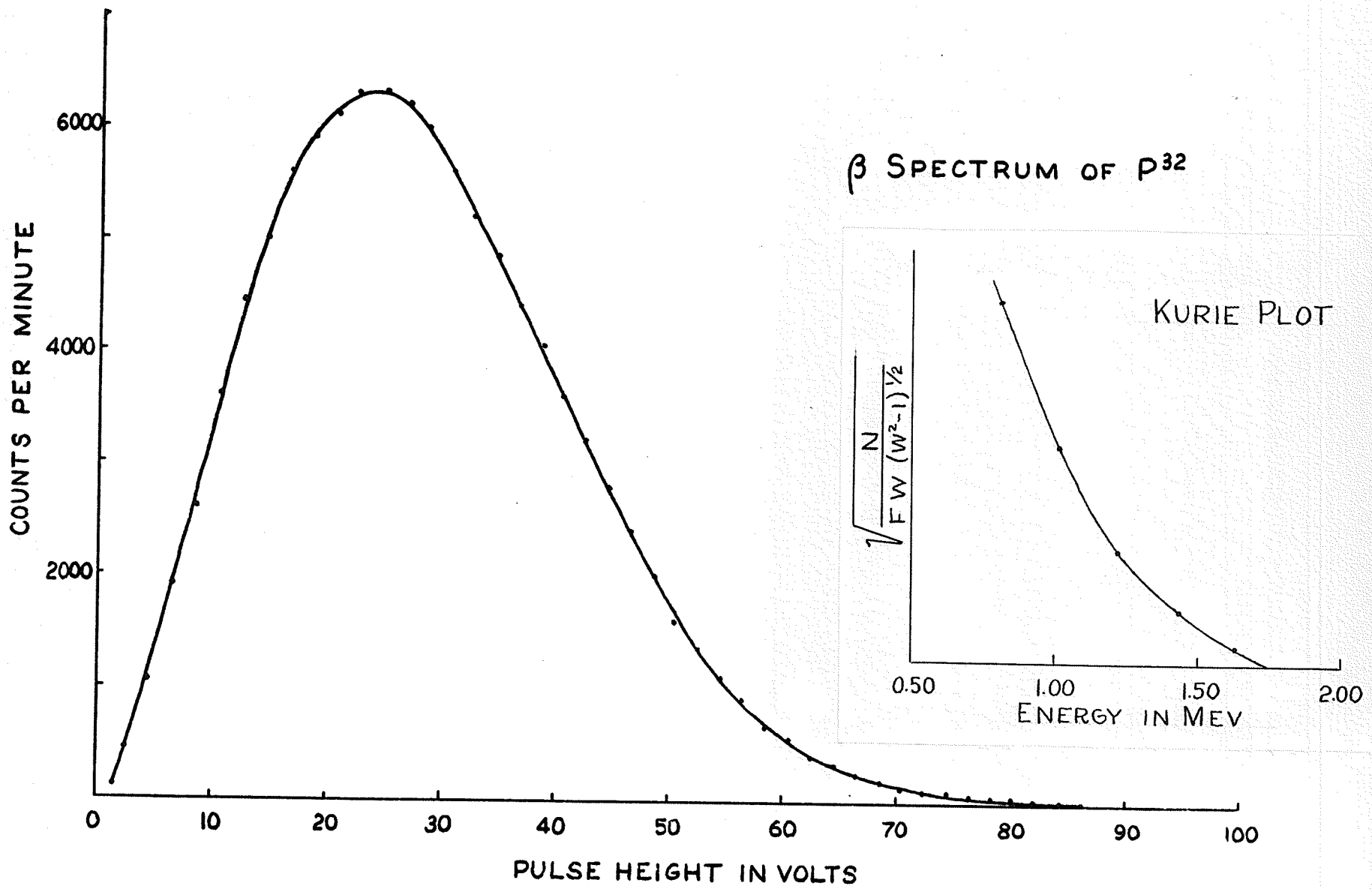

β SPECTRUM OF P32
KURIE PLOT
COUNTS PER MINUTE
6000
4000
2000
0
10
20
30
40
50
60
70
80
90
100
PULSE HEIGHT IN VOLTS
$\sqrt{\frac{N}{F\,W\,(W^2-1)^{1/2}}}$
0.50
1.00
1.50
2.00
ENERGY IN MEV

FIG. 22

The Kurie plot for the spectrum shown does not give a straight line. From this, it was concluded that the spectrometer was actually giving the spectrum a distorted shape, particularly in the high energy region. Due to the loss of high energy electrons from the crystals, one would expect lower counting rates along the tail, resulting in an incorrect approach to the pulse height axis. This effect would also give rise to an excess of lower energy pulses (probably of the order of a few hundred kev.) and a corresponding distortion of the spectrum in that energy region. In order to reduce the losses of high energy β -particles, the crystals used should be at least 0.7 cm. in depth. Unfortunately, such crystals were not available at the time these data were obtained.

It is apparent that the problems mentioned in connection with the P^{32} spectrum would not arise in the study of sources whose end-point energies are only a few hundred kev. For such, the losses from the crystals would be small and the distortions accompanying them would not appear. Other factors can produce distortions, however, as will be shown in the following sections.

THE BETA SPECTRUM OF IODINE 131

Since the advent of tracer techniques, I^{131} has become extremely important in the field of medical research. The details of both the beta-ray and γ-ray spectrum have been studied by many workers. The γ-ray spectrum has been investigated extensively with scintillation spectrometers.[38] The β-ray spectrum has been investigated by means of conventional β-ray spectrometers and also split-crystal scintillation spectrometers. In the case of split-crystal spectrometers, coincidence techniques were employed.[39] A scintillation counter using a split anthracene crystal with the source mounted between the two parts of the crystal, was used to study the β-particles. A counter using a NaI(Tl) crystal detected the γ-radiation from the source. The study of coincidences between β-particles and γ-rays contributed to the knowledge of the decay scheme. Unlike these elaborate techniques, the work to be described here consisted of a simple analysis of the β-particles emitted by I^{131}.

The source of I^{131} was prepared in the same manner as the P^{32} source described in the preceding section and mounted between the crystals. The pulse height distribution of the β-particles coming from the source is shown in Fig. 23. The end-point energy was found to be 805 kev., in good agreement with the end-point energy of the β-component leading to a 12-day metastable state of Xe^{131} (810 kev.).

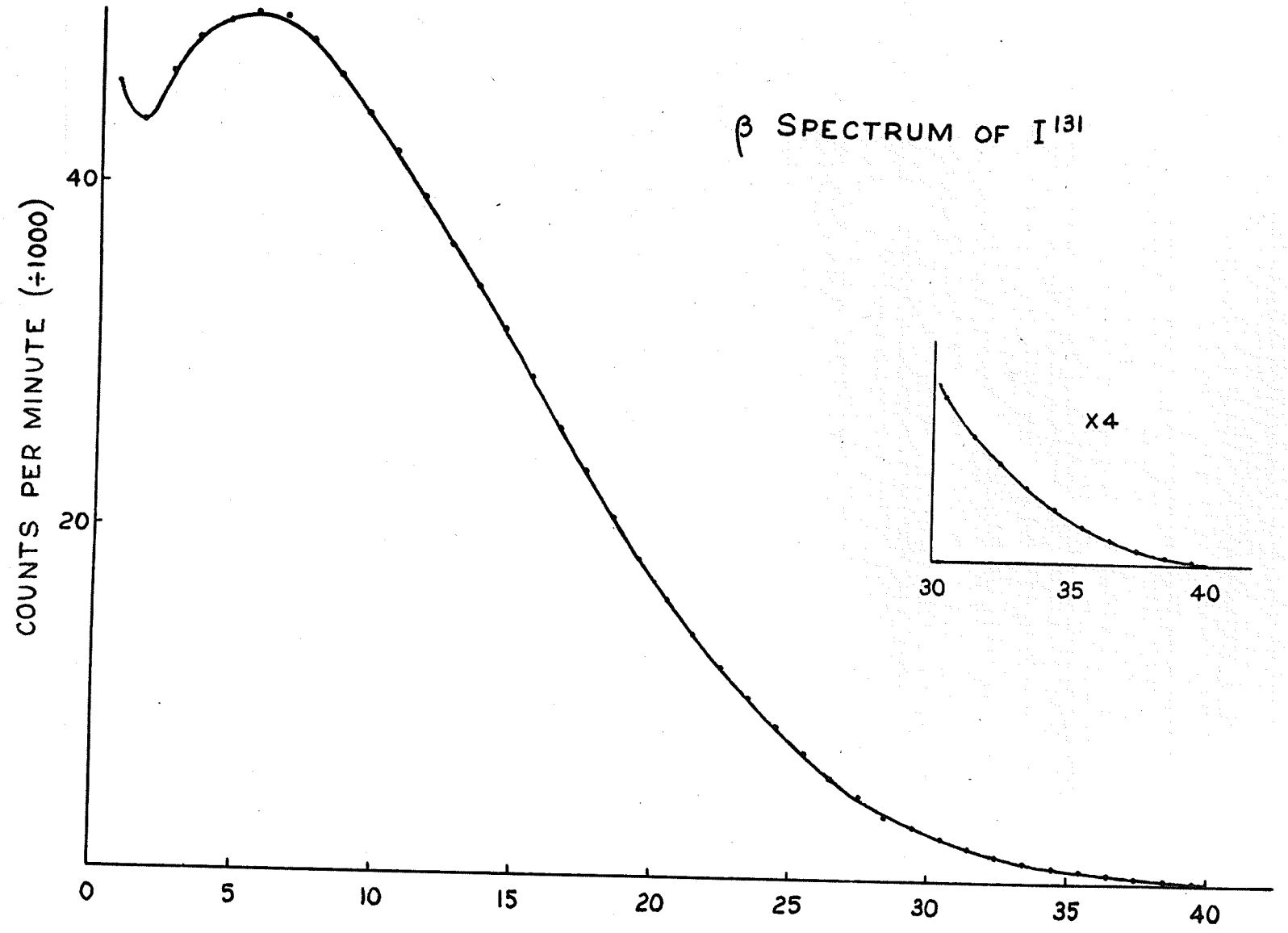

β SPECTRUM OF I 131
COUNTS PER MINUTE (÷1000)
40
20
X4
30
35
40
0
5
10
15
20
25
30
35
40
PULSE HEIGHT IN VOLTS

FIG. 23

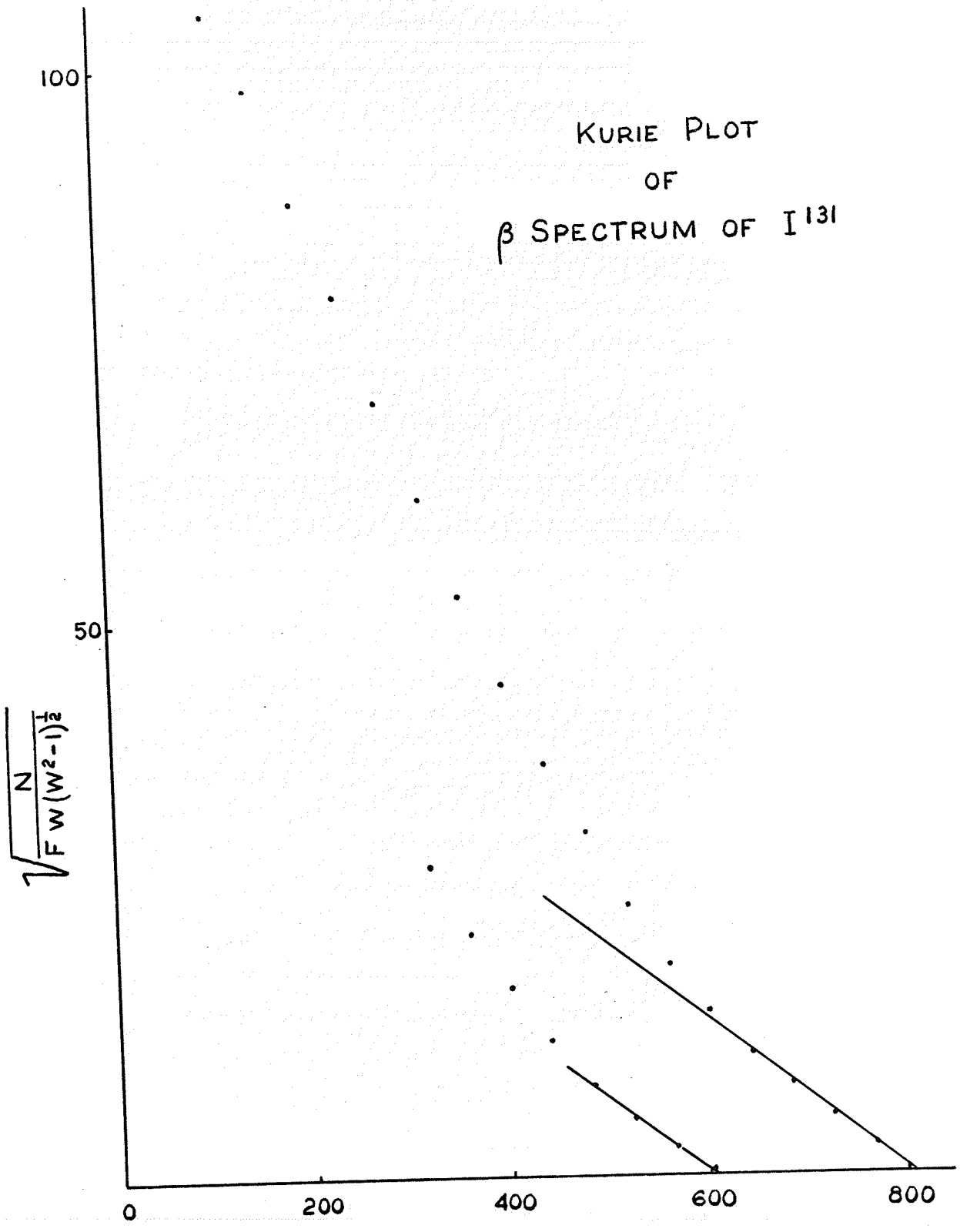

KURIE PLOT
OF
β SPECTRUM OF I 131
100
50
$\sqrt{\frac{N}{F\,W(W^2-1)^2}}$
0
200
400
600
800
ENERGY IN KEV

FIG. 24

The complete beta spectrum of I^{131} has recently been shown to have four components with end-point energies of 244, 315, 600 and 810 kev.[40,41] A Kurie plot of the iodine spectrum was made (Fig. 24). It was possible to identify two components of the beta-ray spectrum with end-point energies of 805 and 605 kev. The curvature of the Kurie plot starting at about 450 kev. seems to indicate that the spectrum has a distorted shape in that energy region. The plot should be linear to about 315 kev., where curvature should begin, indicating the existence of the 315 kev. component. A simple explanation for this distortion is not apparent as yet. The processes occurring in a split-crystal are certain to be complex. Bell et al (see reference 39) encountered similar difficulties due to coincidences between β-particles and γ-rays in the crystal. These gave pulses that were larger than would be expected from the capture of β-particles only. With the split-crystal spectrometer, all possible processes which can occur in the crystal are detected and recorded as part of the β-spectrum. Fortunately, measurement of the end-point energy was not affected by a distortion of the spectrum proper, in the results described above. In using a coincidence technique, Bell found an increase in the end-point energy just equal to the energy of the γ-ray in coincidence with the β-particles studied.

THE BETA SPECTRUM OF IRIDIUM 192

The study of the beta spectrum of Ir^{192} has proven to be one of the most difficult problems encountered in β-ray spectroscopy. Unfortunately, the end-point of the spectrum obtained with a conventional spectrometer is usually masked by internal conversion lines. These are due to the γ-rays which accompany the decay of Ir^{192}. The γ-radiation associated with this activity is highly converted in the source.[42] The percentage of all the γ-rays so converted has been given by Wiedenbeck and Chu[43] as 28.6%. Later estimates tend to be closer to 18%.

The large number of γ-rays present in the Ir^{192} activity makes the determination of a decay scheme a difficult task. Cork[44] has presented a scheme which, as yet, has not met with too much criticism. It is to be hoped that a proper application of coincidence techniques to the problem will yield an unambiguous decay scheme.

The beta spectrum of Ir^{192} as determined with a split-crystal spectrometer is shown in Fig. 25. The tail extends out to 950 kev., which suggests a distortion of the spectrum since the high energy beta component has an end-point at 670 kev. Bannerman[45] has shown that lines due to conversion electrons can be resolved with a spectrometer that uses a single crystal of anthracene as a detector. The results to

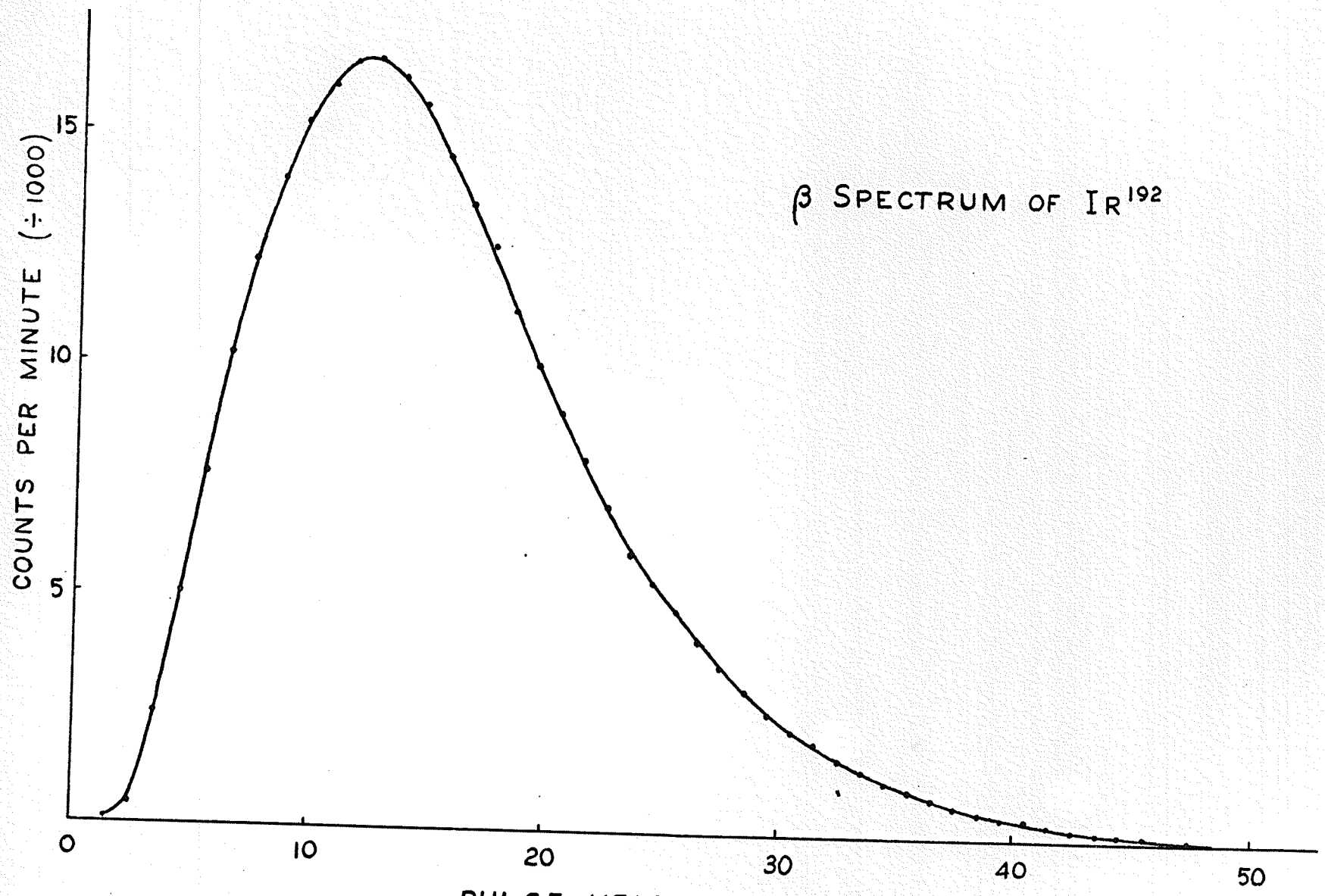
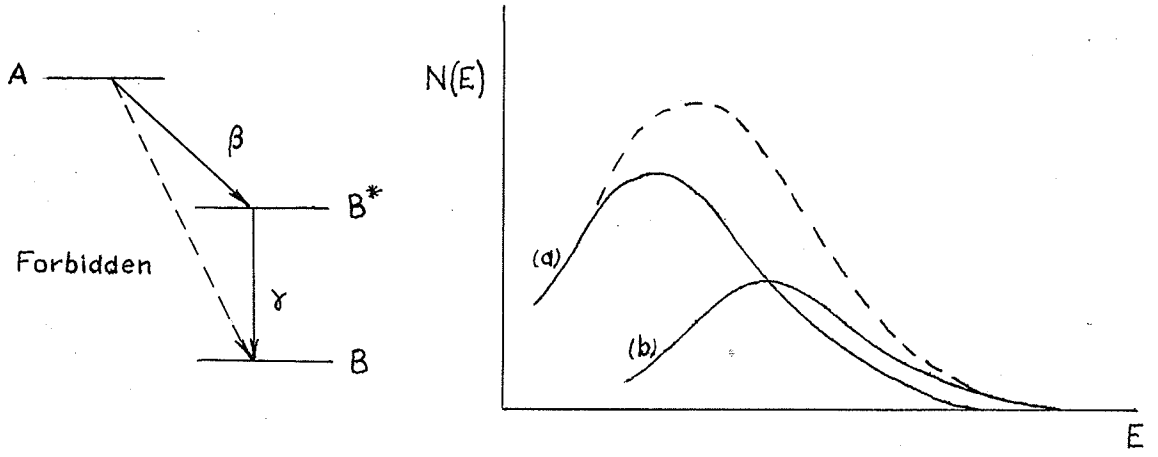

β SPECTRUM OF Ir^{192}
COUNTS PER MINUTE (÷1000)
15
10
5
0
10
20
30
40
50
PULSE HEIGHT IN VOLTS

FIG. 25

be expected with a split-crystal spectrometer are somewhat different, however. With reference to the sketches below, consider the case of a β-activity for which the ground state transition is forbidden.



Suppose that the unstable nucleus A emits a β-particle as shown in the sketch, which is captured by one of the crystals. Assume that the γ-ray following the decay is not absorbed or internally converted. From the study of all such processes, we get an ordinary β-spectrum indicated by (a) in the right-hand sketch. However, if the γ-ray is internally converted, we get two β-particles entering the crystals. These give rise to one pulse from the detector which is larger than it should be by the energy given up to the crystal by the conversion electron. The study of all such processes, taking into account the variable energy of electrons from the primary spectrum, will yield another spectrum which is displaced from the origin, as indicated by (b) in the sketch. When both processes are detected

simultaneously, as in a split-crystal spectrometer, the resulting spectrum is similar to that shown by the dotted curve.

The Ir^{192} spectrum shown in Fig. 25 is of this type. The displacement of the distribution from the origin and the long tail seem to verify the conclusions reached in the preceding paragraph. It is interesting to note that the end-point energy is nearly equal to 670 kev. plus the mean energy of a conversion electron that has been ejected from the K shell by any one of the three intense iridium γ-rays (308 kev., 295 kev. and 316 kev.).

Bannerman (see reference 45) has pointed out that long tails on β-spectra can occur through a piling up of low energy pulses, when strong sources are used. The sources used in the work described above were of the order of a few microcuries so that a piling up of pulses was extremely unlikely, and hence could not account for the observed shape of the spectrum.

CONCLUSIONS

The application of the split-crystal scintillation spectrometer to the study of beta spectra has been described. The difficulties encountered in using anthracene crystals for the study of β-spectra are apparent from the preceding sections. When the β-spectrum has a high energy component, loss of electrons from the crystal can distort the shape of the spectrum. When highly converted γ-rays are present, the beta spectrum is shifted along the energy axis away from the origin. A spurious approach to the energy axis usually accompanies the shift. Such distortions in the distribution are due to the superposition of several different processes which can occur in the crystal. It is to be hoped that further investigation will suggest methods of eliminating at least some of the undesirable properties of the split-crystal spectrometer.

THE SCINTILLATION SPECTROSCOPY

OF

NEUTRON-CAPTURE GAMMA RADIATION

INTRODUCTION

When Rutherford discovered that the structure of nuclei could be changed through bombardment with and absorption of α-particles, he was in effect entering a vast field of research now called the study of Nuclear Reactions. A complete discussion of the topic will not be given here, but since the experimental work to be described involves the process of slow neutron-capture, we shall discuss briefly the principal features of this particular reaction only.

Nuclei are now known to have discrete energy levels. The spacing of these levels is not constant. In fact, the levels of higher energy (of the order of a few Mev.) tend to be so closely packed that we can effectively refer to a continuum of energy states. The probability for the capture of a neutron incident on a nucleus can be stated in terms of the energy of the neutron, and the widths of any nuclear levels in this energy region. The description of the capture process in such terms was originally due to Breit and Wigner.[46] They derived expressions for the probability for capture in terms of the de Broglie wave length and the energy of the incident neutron, and the widths of the energy levels in the target nucleus.[47] Their results explained why some nuclei have extremely large capture cross-sections at certain energies.

It was proposed that certain levels existed in the nucleus at these energies and that a kind of resonance capture occurred, for which the cross-section was unusually high. These conclusions have been shown to be in excellent agreement with cross-section curves for different elements, which show the variation of the cross-section with the energy of the incident neutron.

After the capture of a neutron has occurred, there is formed a nucleus which is referred to as the Compound Nucleus, a concept first proposed by Bohr in 1936. This nucleus is in general unstable and will emit either a particle or γ-radiation. In fact, a competition between alternative modes of decay actually takes place. The emission of a charged particle may be restricted by the potential barrier of the compound nucleus, while selection rules might eliminate other modes of decay. Bohr assumed that there was a tendency for an equipartition of energy in the compound nucleus, i.e. the excess energy due to the captured particle is distributed among all the particles forming the product nucleus. This effect tends to give a non-zero life-time to the compound nucleus. Radiative capture of slow neutrons is that process in which γ-radiation is emitted from the compound nucleus to form the final product. This reaction is denoted by the symbol (n,γ). Clearly, in the $(n\,\gamma)$ reaction, the compound nucleus is just an excited state of the product nucleus,

so that by studying the γ-radiation emitted during the reaction process, one can get valuable information on the energy levels of the product nucleus. The importance of such information is obvious when one remembers that the purpose of nuclear theory is to explain the simpler nuclear structures and to predict the properties of all nuclei using the simple nuclei as a basis. Knowing what levels exist in a nucleus is a vital necessity for the successful formulation of any nuclear theory. For a detailed discussion of the theory of resonance processes, the reader is referred to standard works on nuclear theory.[48]

The study of the γ-radiation emitted in the (n,γ) process has received scant attention until recently. Since 1939, when the possibilities of the fission process were realized, a vast amount of data has been gathered on nuclear reactions in general, and on neutron-induced reactions in particular. The study of the (n,γ) reaction was held back for several years by the lack of suitable neutron fluxes, but since the advent of the neutron reactor, high neutron fluxes with small γ-ray backgrounds have become available for the investigation of this process. The study of capture γ-ray spectra has recently been undertaken at both Chalk River[49,50,51,52,53] in Canada and the Argonne National Laboratory[54,55] in the U. S. A.

By using a suitable source of neutrons (e.g. nuclear reactions which involve the emission of neutrons) one can

study the (n, γ) reaction, provided that the neutrons are first reduced to thermal energies ($\sim$1/30 electron volt) before they are used to bombard the target. In the experimental work to be described here, such a neutron source was used. It must be remembered, however, that the use of neutron sources usually involves undesirable γ-radiation which makes detection of the capture γ-radiation difficult. These points will be discussed in greater detail in the sections which follow.

DISCUSSION OF EXPERIMENTAL PROCEDURE

The use of a neutron source for the study of (n, γ) reactions necessitates certain modifications in the conventional scintillation spectrometer. Some of these changes will now be described. A schematic diagram of the equipment used is shown in Fig. 26.

The counter consisted of the usual E.M.I. 5311 photomultiplier with a 3/4 in. diam. x 1 in. NaI(Tl) crystal mounted on the end window. The output of the tube was amplified with a Model 204-C amplifier. The 4-channel pulse analyzer was employed in analyzing the pulses. In order to reduce the γ-ray background, a 1 in. thick lead cap was placed over the counter cannister. A cylindrical lead plug (1 in. diam., 3 in. long) was placed on the top of the cap and the moderating material (in this case a paraffin block) was placed around it. The neutrons from the source were reduced to thermal energies through elastic collisions in this paraffin.

The neutron source was obtained from the Eldorado Mining and Refining (1944) Ltd. It consisted of 400 mc. of RaD mixed with finely powdered beryllium in the ratio

$$\frac{\text{RaD (mc)}}{\text{Be (mg)}} = \frac{1}{15}$$

The source gave 10^6 neutrons per sec.; these neutrons have an average energy of 4.1 Mev. The mixture itself

APPARATUS FOR NEUTRON - CAPTURE STUDIES

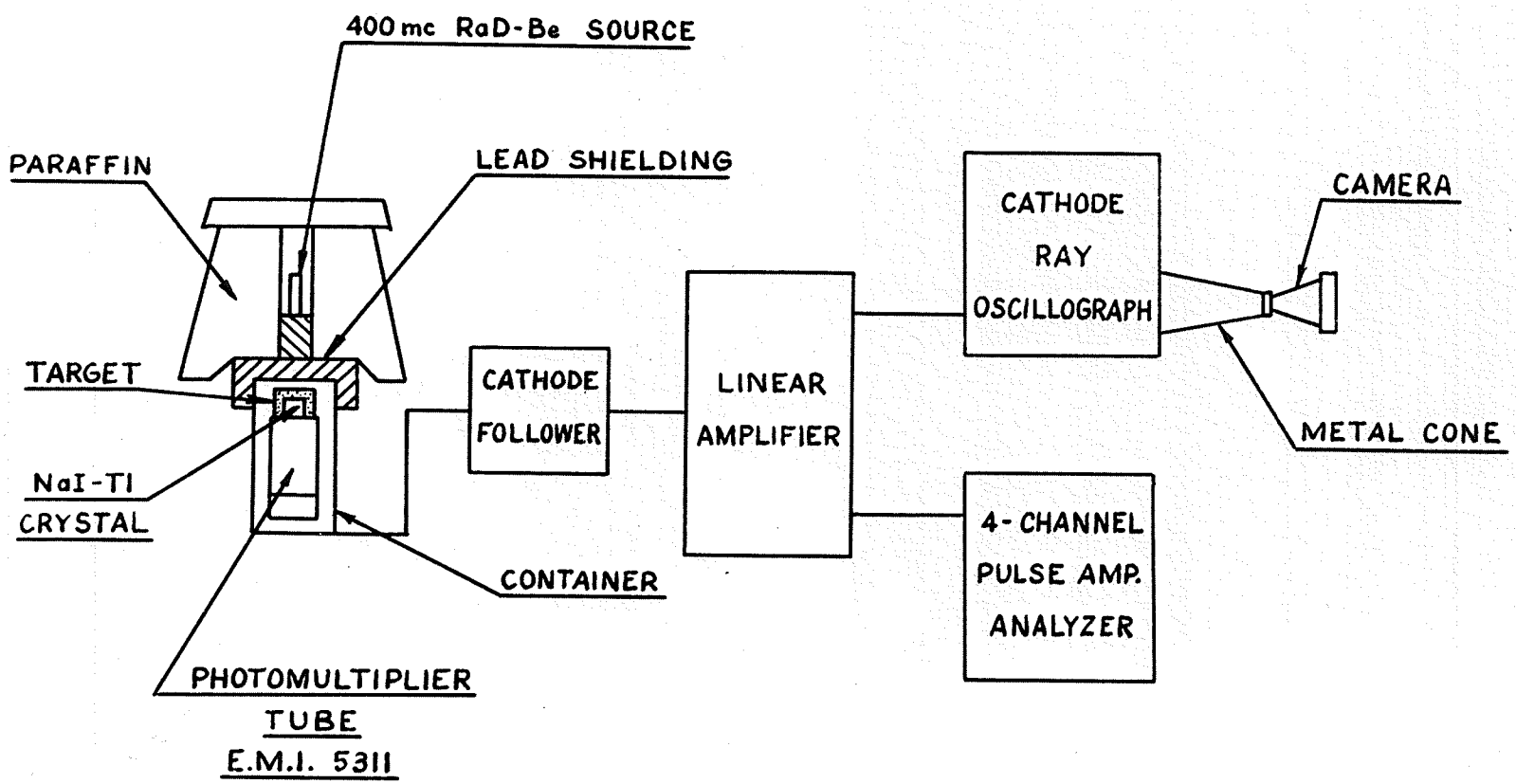


FIG. 26

was contained in a steel cylinder about 2 in. long and 1/2 in. in diameter. The γ-ray background due to the source was equivalent to that from 0.224 mc. of radium. The neutrons are produced primarily by the following reaction:

$$Be^{9} + He^{4} \longrightarrow C^{12} + n^{1} + Q$$

This reaction gives neutrons having a wide range of energies. Suitable moderating material must be used to reduce these neutrons to thermal energies. The paraffin block used for this purpose was constructed so that the source to crystal distance involved no more than 10 cm. of paraffin. This is an important detail of construction, for the number of slow neutrons per cm^3 rapidly diminishes at distances greater than 9 - 10 cm. from the source of fast neutrons. These losses are due to (n,γ) reactions occurring in the paraffin itself.

When a target is placed inside the counter cannister, immediately over the crystal, slow neutrons from the paraffin become incident upon it. (See Fig. 26). The (n,γ) reaction which occurs in the target gives rise to γ-radiation some of which is detected. An analysis of the pulses from the counter yields, after a suitable correction for background has been applied, the neutron-capture γ-ray spectrum of the target material.

There are, however, several complicating factors. There is no reason to believe that every feature on such a

spectrum corresponds to a γ-ray emitted through a transition from an excited state to the ground state of the product nucleus. Transitions between excited states, as well as cross-over transitions are likely to occur and must be taken into consideration when attempting to build up a level scheme for the nucleus. Another complication arises from the fact that most of the features on a high energy spectrum are the pair lines mentioned earlier in this paper. The energy of a pair line is $(E_\gamma - 1.02)$ Mev. where E_γ = the energy of the incident γ-ray. The positron produced by this γ-ray will usually be annihilated in the crystal giving rise to two γ-rays each with an energy of 0.51 Mev. Either one or both of these will generally be absorbed by the crystal. This will tend to give two additional lines on the spectrum at energies of $(E_\gamma - 0.51)$ Mev. and E_γ (the full energy of the incident γ-ray). Such lines must not be confused with the pair lines when γ-ray energies are proposed. In practice, when a spectrum contains many pair lines, these additional features do not complicate the distribution too much. They are usually masked by intense adjacent pair lines. In the case of a simple spectrum, these extra features are easily recognized.

The photographic technique was found to be of little use in the study of the spectra to be described. The complexity of the spectra, combined with the existence of a

background extending to high energies for which correction was difficult, made a photographic analysis impractical. Complete dependence on the pulse height distributions proved to be the most satisfactory method for studying the (γ,γ) process. Needless to say, however, many photographs of spectra were taken during the course of the work in an effort to make use of the technique. For that reason, the photographic unit has been included in Fig. 26. For relatively simple spectra, or for spectra which contain a few very intense features, a photographic analysis can sometimes be carried out.[56]

THE NEUTRON-CAPTURE GAMMA RAY SPECTRUM OF CADMIUM

Before the capture γ-ray spectrum of cadmium is discussed in detail, a few comments will be made on the nature of the background for this work and the method used to calibrate the pulse height axis in Mev.

A background curve gives an analysis of the pulses coming from the counter when no target is used, but the neutrons are incident on the crystal itself. Most of the background is due to γ-radiation coming from the RaD-Be source. A typical distribution is shown in Fig.27. It is not to the same scale as the figures in this section. It illustrates the nature of the background from 1 to 4 Mev. only. The three features A, B and C were found to have energies of 1.12, 1.77 and 2.26 Mev. respectively. These values are in good agreement with the known γ-ray energies of radium - 1.12, 1.76 and 2.208 Mev. The shape of the curve, combined with the fact that the energy of feature C is slightly greater than 2.208 Mev. suggests that radiation other than that due to the RaD-Be source was detected by the counter. It was felt that part of feature C was due to the reaction $H^1(n,\gamma)D^2$ in the paraffin moderator. The capture γ-radiation from hydrogen consists of one γ-ray with an energy of 2.230 ± 0.007 Mev.[57] Since the cross section, $\sigma(n,\gamma)$, for this reaction is about 0.3 barn (1 barn = 10^{-24} cm.^{2}), it is reasonable to expect some

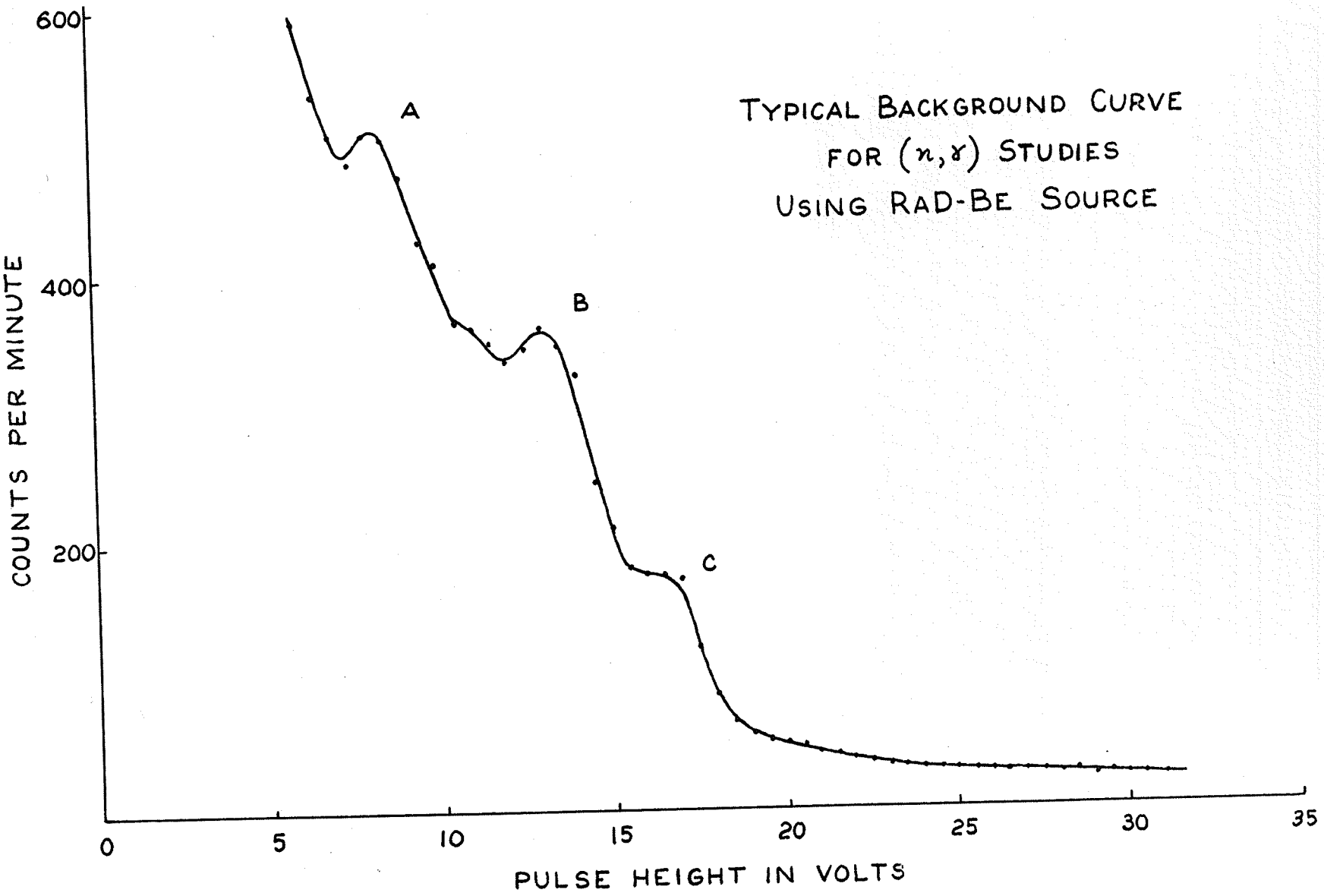

TYPICAL BACKGROUND CURVE
FOR (n,γ) STUDIES
USING RAD-BE SOURCE
COUNTS PER MINUTE
600
400
200
A
B
C
0
5
10
15
20
25
30
35
PULSE HEIGHT IN VOLTS

FIG. 27

contribution from the many pounds of paraffin used. Although the background is not shown for energies greater than 4 Mev. in Fig. 27, it can be stated that the curve is quite flat out to at least 9 Mev.

The pulse height axis was calibrated in Mev. by means of a polonium-beryllium source. This mixture can also be used as a neutron source through the reaction mentioned on page 47. The product nucleus, C^{12}, is formed in an excited state. On de-excitation, a γ-ray of 4.47 Mev. is emitted. It produces an electron-positron pair in the crystal through the interaction discussed on Page 6. The corresponding pulse height distribution for this γ-radiation is shown in Fig. 28. Feature A is the pair line corresponding to the absorption in the crystal of the electron-positron pairs. It has been studied extensively by several groups of workers,[58,59,60] and has been found to have an energy of 3.45 Mev. Feature B corresponds to the simultaneous capture of the pair and one annihilation quantum of 0.51 Mev. Feature C corresponds to the capture of the pair plus two annihilation quanta. By using the energies of the three features in Fig. 28 and assuming the linearity of the spectrometer for high energy radiation, the pulse height axis was calibrated. In succeeding figures, the pair line at 3.45 Mev. is shown without any specific identification. All determinations of the energies of lines in capture γ-ray spectra have been made in terms

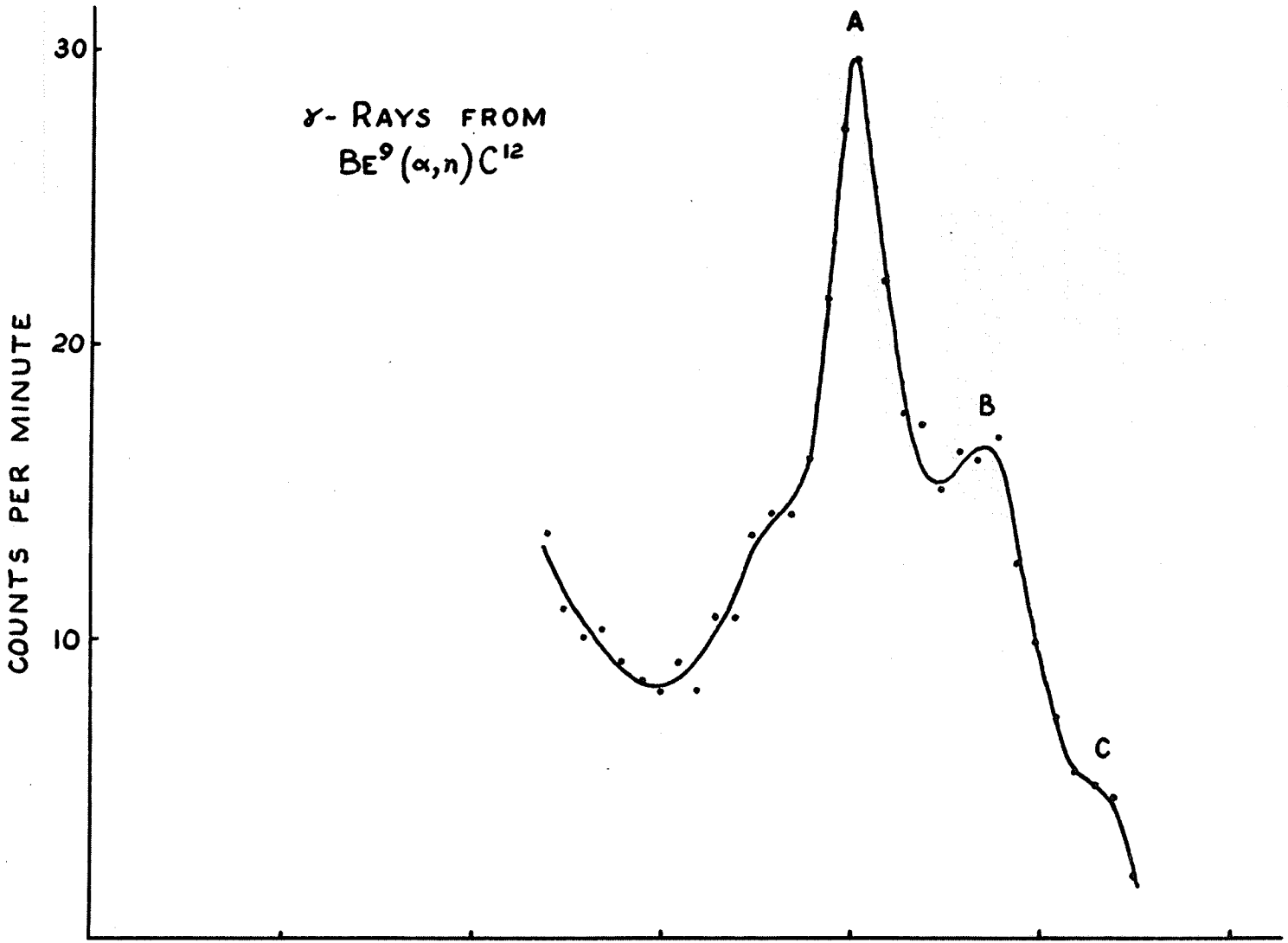

γ- RAYS FROM
$Be^9(\alpha,n)C^{12}$
A
B
C
30
20
10
COUNTS PER MINUTE
0
5
10
15
20
25
30
PULSE HEIGHT IN VOLTS

FIG. 28

of this prominent pair line.

Of the stable isotopes of cadmium, Cd^{113} alone must be considered in the reaction Cd(n, γ). This isotope (12% abundance) has a cross-section for slow neutron capture of 20,000 barns,[61] and consequently it is primarily responsible for the (n, γ) reaction in cadmium. Hence the capture γ-ray spectrum of cadmium will give information on the levels in the Cd^{114} nucleus.

The target consisted of a thin sheet of cadmium metal shaped to fit snugly over the crystal. The capture γ-ray spectrum, corrected for background, is shown in Fig. 29. The general shape of the spectrum was found to agree closely with that found by Hamermesh using a photographic emulsion technique (see reference 51), and Wilson[62], who employed ionization chambers. The spectrum found by Hamermesh gives the γ-ray energies directly, while Fig. 29 gives only the energies of the electrons produced by the γ-rays through the pair production process. One must add 1.02 Mev. to the energy of a feature on the spectrum in order to get the γ-ray energy. For convenience, the corresponding γ-ray energies of the features on the spectrum have been tabulated in Table I together with a set of values supplied by the Chalk River group[63] who have also studied this spectrum. (The workers of this group are using a magnetic pair spectrometer and consequently their investigations

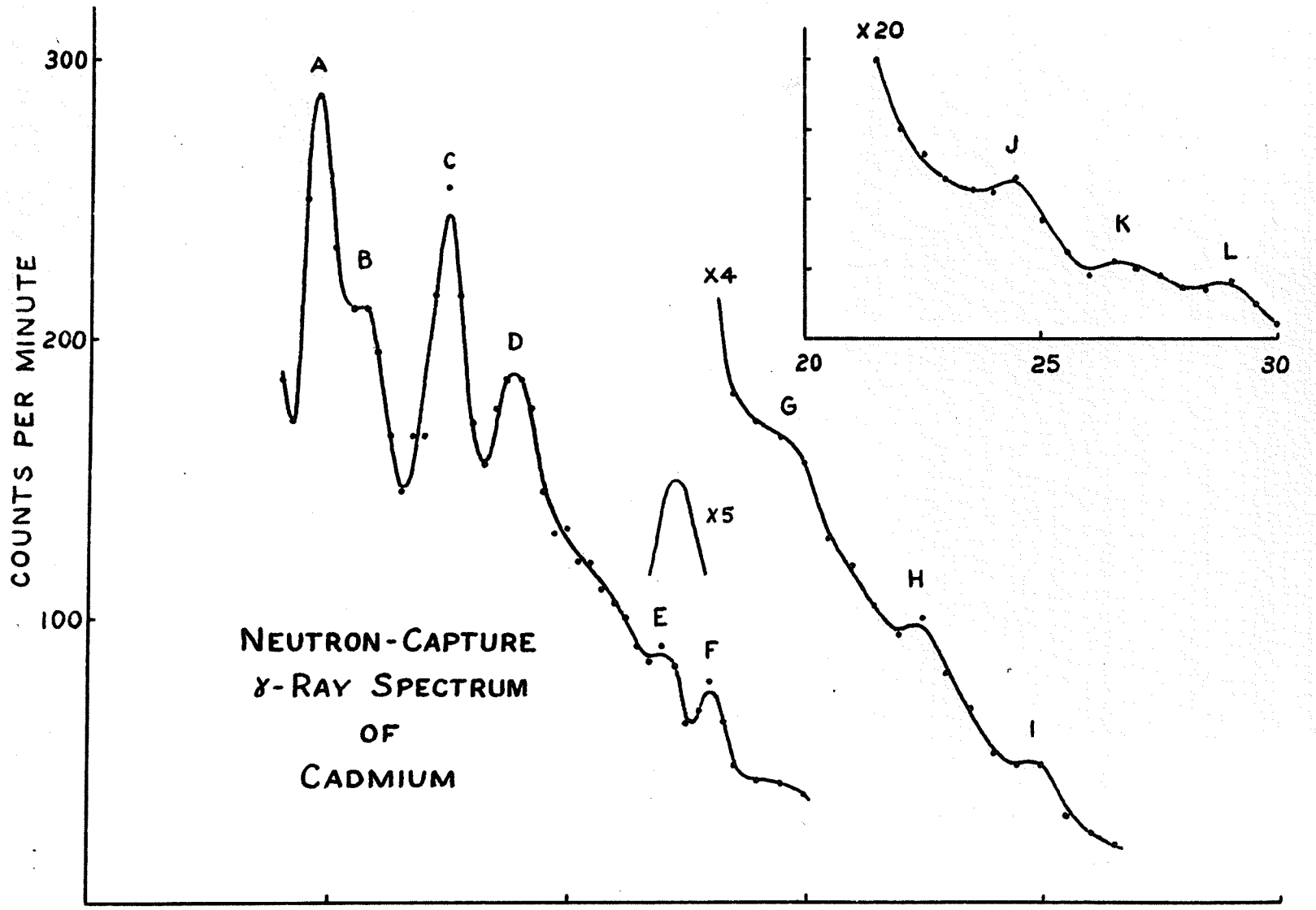

300
200
100
COUNTS PER MINUTE
A
B
C
D
E
F
G
H
I
J
K
L
X20
X4
X5
NEUTRON-CAPTURE
γ-RAY SPECTRUM
OF
CADMIUM
0
5
10
15
20
25
20
25
30
PULSE HEIGHT IN VOLTS

FIG. 29

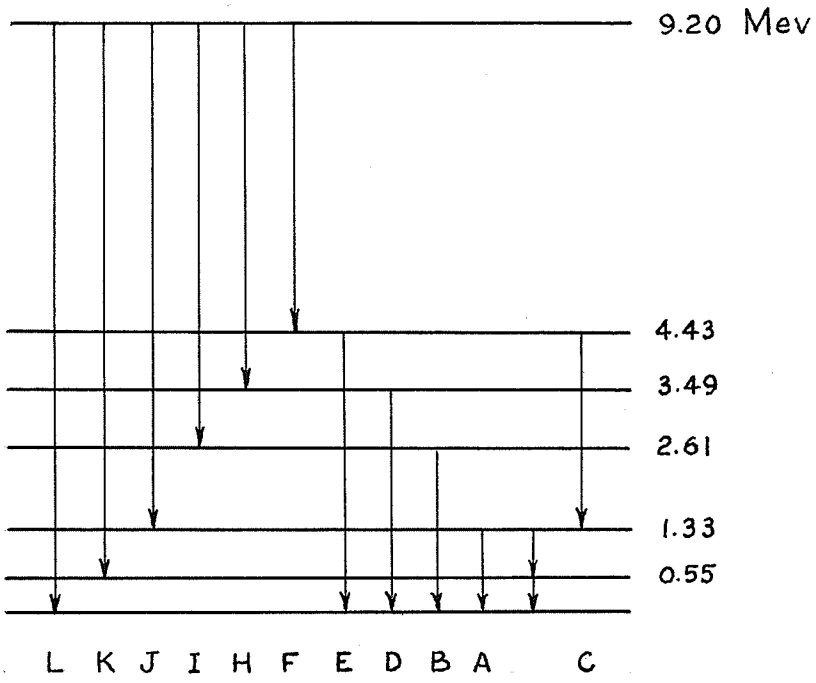
TABLE I

Feature	γ-Ray Energy (Mev)	
	Kinsey	Present Determination
A	-	1.33
B	-	2.63
C	-	3.14
D	-	3.49
E	4.58	4.43
F	4.82	4.71
G	5.26	5.17
H	5.96	5.85
I	6.60	6.61
J	7.81	7.92
K	8.50	8.55
L	9.33	9.20

are limited to energies exceeding 3 Mev. In using a RaD-Be source with its relatively low γ-ray background, it was hoped that the 1 - 10 Mev. region could be studied successfully.)

Feature A was thought to be the photoelectric peak due to a 1.33 Mev. γ-ray, since a nuclear level of Cd^{114} is known to be at 1.27 Mev. (see reference 27). B and C are pair lines which correspond to γ-rays of 2.63 and 3.14 Mev. respectively. The identification of the primary event responsible for a line in the 1.0 - 2.5 Mev. region is difficult because of the competition of the photoelectric and pair production processes in this region. The results presented above seem to be reasonable in view of the relative cross-sections for these processes in the iodine constituent of the crystal. The remainder of the features listed in Table I are evidently in good agreement with the results of Kinsey et al. Feature L, which is due to a γ-ray of 9.20 Mev. is useful in that it determines the binding energy of the last neutron in the Cd^{114} nucleus.

A level in the Cd^{114} nucleus is known to exist at 0.55 Mev. Making use of this, an attempt was made to construct a possible level scheme for this nucleus. The result is shown in the accompanying diagram. All the γ-rays found have been included in this scheme with the exception of the 5.17 Mev. γ-ray. If this represents a



PROPOSED LEVELS IN Cd^{114}

transition from a state at 5.17 Mev. to the ground state, one would expect to find another γ-ray present at 4.03 Mev. The pair line corresponding to this γ-ray would lie between features D and E on the pulse height distribution. The shape of the curve in this energy region suggests that such a component may exist. The variation with energy of the cross sections for the photoelectric and pair production processes (Fig. 1) and the relative intensities of the features of Fig. 29 were used to establish the decay scheme. The assumption that feature C corresponds to a transition from the level at 4.43 Mev. to that at 1.33 Mev.

is consistent with the fact that the intensity of F is somewhat greater than that of E. However, since reference to Figs. 1 and 29 indicates that A and C are of comparable intensity and the 1.33 Mev. level is also fed by the very much weaker transition J, it is reasonable to expect an alternative transition from the second excited state. A study of the K-capture process in In^{114} has revealed the existence of a transition from the 1.33 Mev. to the 0.55 Mev. level.[64] This accounts for the feeding of the 1.33 Mev. level by both C and J in a satisfactory manner. An analysis of the relative cross sections for features D and H leads to the conclusion that H is slightly less intense than D and hence the existence of a level at 3.49 Mev. is indicated. A similar procedure determines the order of transitions B and I in the scheme. The transition from the 0.55 Mev. level to the ground state is known to be more intense than A (see reference 64), and hence the association of feature K with the first excited state of Cd^{114} seems reasonable. The level scheme is by no means complete, but since only the most intense γ-rays are involved, it was felt that the scheme had some validity.

In general, some of the neutrons incident on a target will pass through it and give rise to (n, γ) reactions in the crystal. This process is extremely unlikely for a

cadmium target because of the high capture cross-section of Cd^{113}. The background curve, however, was obtained while neutrons were actually impinging on the crystal, hence the distribution of Fig. 27 does not give a true picture of the background in the cadmium investigation. Unfortunately, no method of correction is available at present so that it was found necessary to use the 'approximate' background curve of Fig. 27. The agreement with Kinsey's results indicated that this procedure was valid, at least for the high energy section of the cadmium spectrum.

The results presented above suggest that this method of studying nuclear energy levels is a useful one, provided that the capture cross-section of the target material is reasonably large. Complications can arise, however, when this condition is not fulfilled, as will be seen in the discussion of the chromium capture γ-ray spectrum which follows.

THE NEUTRON-CAPTURE GAMMA RAY SPECTRUM OF CHROMIUM

The four stable isotopes of chromium are listed below together with their percentage abundances and capture cross-sections:

TABLE II

Isotope	Abundance	Capture Cross-section σ (th.n, γ)
Cr^{50}	4.4%	16 barns
Cr^{52}	83.5%	$\sim$ 0.7 barn
Cr^{53}	9.5%	17.5 barns
Cr^{54}	2.6%	$\leqslant$ 0.3 barn

An examination of this table indicates that each of these isotopes will contribute to the γ-ray spectrum of the Cr(n, γ) reaction. No reference will be made to any particular isotope in the discussion which follows.

The (n, γ) reaction in chromium presents great difficulties due to the existence of several stable isotopes. In fact, very little is known about the energy levels of the product nuclei formed through the radiative capture of slow neutrons. One level in Cr^{54} at 0.835 Mev. has been found, but this represents all the present knowledge of such levels. The results to be described here are preliminary in nature and are not to be regarded as complete. Kinsey, Bartholomew and Walker have attempted to

study this spectrum (see reference 50). They state that Cr gives only a continuous γ-ray spectrum. This is probably due to the superposition of the spectra from the different isotopes.

The target used for the investigation of the Cr(n, γ) reaction consisted of approximately 200 grams of pure metallic chromium flakes. These were closely packed around the crystal. The pulse height distribution was obtained with the pulse height analyser and is shown in Fig. 30. The usual Po-Be pair line (3.45 Mev.) is present also. The five prominent features shown correspond to γ-ray energies of 5.32 (A), 6.07 (B), 7.00 (C), 8.17(D) and 9.35 (E) Mev. The high energy feature (9.35 Mev.) will correspond to the binding energy of the last neutron in one of the four possible product nuclei. The sharp drop in the distribution on the low energy side of feature A is entirely instrumental in nature. The large mass of chromium used supplies effective shielding of the crystal for γ-radiation up to 3 Mev. In fact, it was found that the target supplied fewer counts due to capture γ-radiation than it absorbed from the background in this energy region. Hence, the background curve actually crossed the (n, γ) curve at about 3.0 Mev. making an investigation of the 1 - 3 Mev. region impossible. A comparison of the counting rates of the cadmium and chromium spectra will illustrate the relatively poor yield in the latter case.

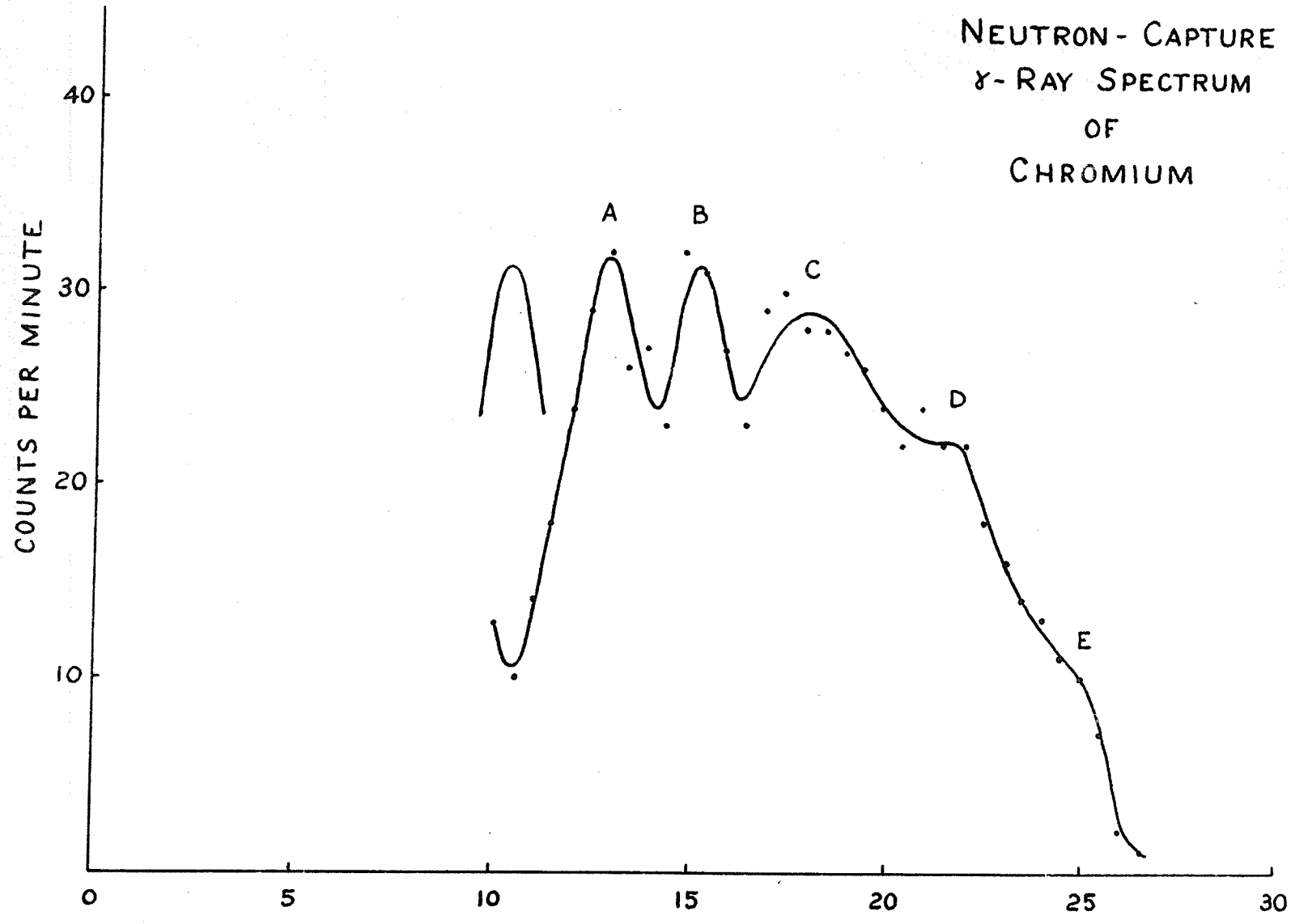

NEUTRON - CAPTURE
γ - RAY SPECTRUM
OF
CHROMIUM
A
B
C
D
E
COUNTS PER MINUTE
40
30
20
10
0
5
10
15
20
25
30
PULSE HEIGHT IN VOLTS

FIG. 30

The complexity of the chromium investigation is evident from the preceding remarks. The existence of several stable isotopes, the shielding of the crystal by the target material and the low counting rates encountered make the study of the Cr(n, γ) reaction an unusually difficult problem. At present, the only conclusion that can be reached is that nuclear energy levels in chromium isotopes do exist out to at least 9 Mev.

THE NEUTRON-CAPTURE GAMMA RAY SPECTRUM OF CHLORINE

Unlike chromium, chlorine has only two stable isotopes which can take part in the (n, γ) reaction. They are Cl^{35} (abundance 75%, σ(th.n, γ) $\sim$ 32 barns) and Cl^{37} (abundance 25%, σ(th.n, γ) $\sim$ 0.5 barn). Clearly the predominant reaction will be Cl^{35}(n, γ) Cl^{36}. Only two levels in the Cl^{36} nucleus are known at present. They are at 0.96 and 4.81 Mev. The less important Cl^{38} (half-life $\sim$ 38 min.) formed by the Cl^{37} (n, γ) Cl^{38} reaction has levels at 1.00, 1.92 and 6.11 Mev.

The target used consisted of approximately 100 grams of sodium chloride. It was felt that the sodium content in the target would not contribute to the spectrum because of its small cross-section ($\sim$ 0.5 barn). The pulse height distribution for the Cl (n, γ) reaction is shown in Fig. 31. The Po-Be pair line is also shown in the figure. Five distinct features were found on the distribution. The line marked A was found to have an energy of 1.93 Mev. If this is a photoelectric line, it could arise through transitions from the 1.92 Mev. level in the Cl^{38} nucleus. A more likely explanation is that it is the pair line corresponding to a γ-ray of 2.95 Mev. from Cl^{36}. Feature B has an energy of 2.69 Mev. corresponding to a γ-ray of 3.71 Mev. A level in the Cl^{36} nucleus at 4.81 Mev. was mentioned above. Assuming a

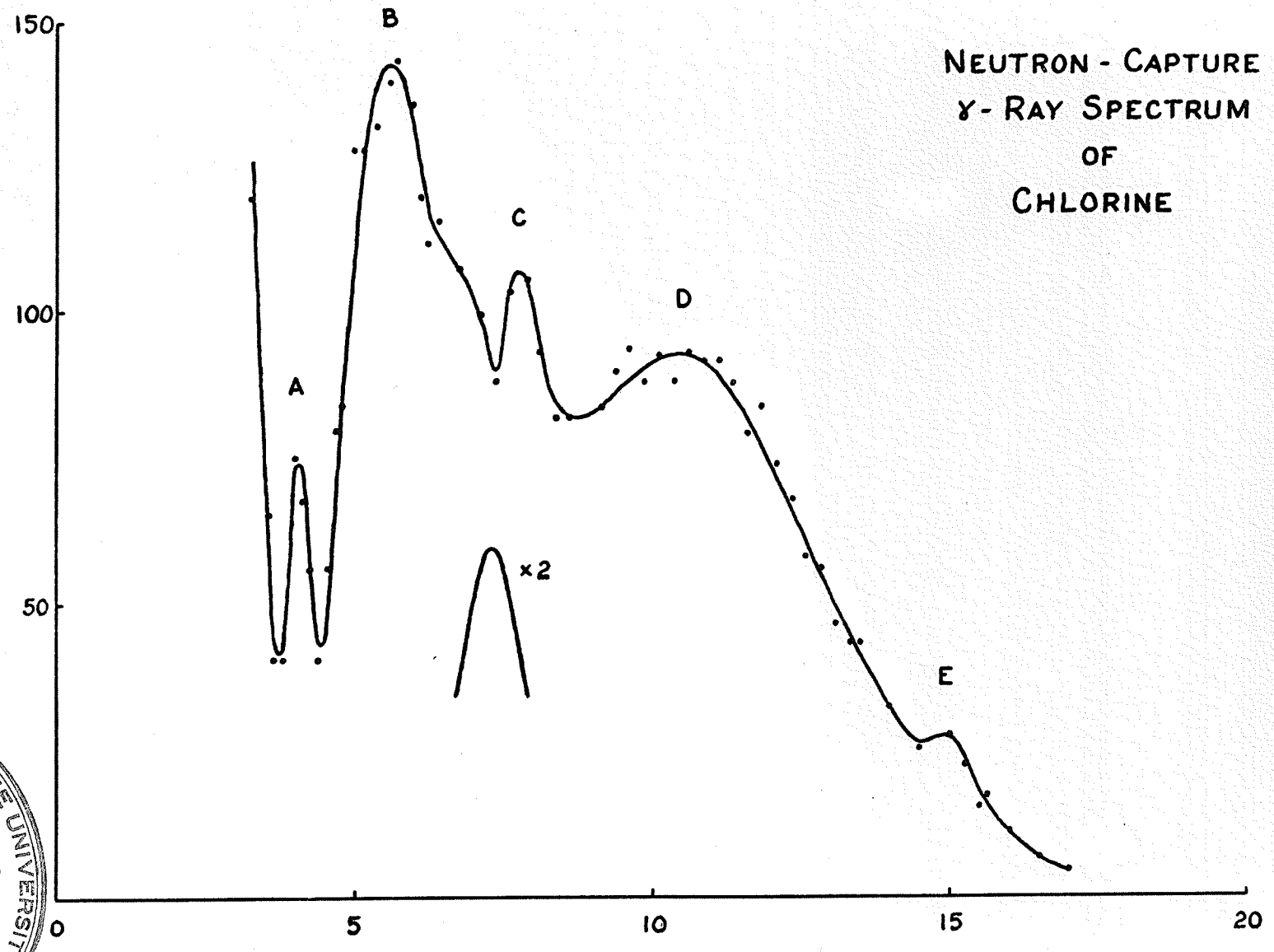


FIG. 31

ground state transition from this level, the γ-ray energy of feature C (4.76 Mev.) was found to be in good agreement with the known existence of such a level. Kinsey et al (see reference 50) have found the end-point energies of the capture γ-ray spectra for Cl^{36} and Cl^{38} to be 8.56 and 6.11 Mev. respectively. The broad feature labelled D in the above distribution is presumably the pair line of a 6.05 Mev. γ-ray, in good agreement with the high energy radiation from Cl^{38}. Its breadth suggests, however, that other pair lines probably exist in that energy region due to γ-rays from excited states in the Cl^{36} nucleus. The resolution of the spectrometer was such that the separation of these components could not be achieved. The last pair line (E) was found to correspond to a γ-ray energy of 8.26 Mev. This agrees fairly well with the result given by Kinsey although it tends to be somewhat lower.

The distribution of Fig. 31 does not agree with the work of Wilson (see reference 62). Using CCl_4 as a target, he finds an end-point energy of 10.5 Mev. Although no attempt was made to identify the source of this γ-ray, it is presumably from Cl^{36}. Hamermesh (see reference 50) gives a spectrum whose shape agrees reasonably well with the curve presented here. The agreement with Kinsey's results seems to be satisfactory.

The peculiar shape of the spectrum in the neighbourhood of feature A was believed to be largely instrumental in origin. Partial shielding of the crystal against γ-radiation with an energy of from 1 to 2 Mev. could result in a shape similar to that shown, although this probably does not account for the entire effect. Feature A was genuine, however, in that it appeared on all the curves taken with NaCl as target. Its inclusion was felt to be justified on this basis.

CONCLUSIONS

A method for studying the excited states of nuclei formed by the radiative capture of thermal neutrons has been outlined. It has been illustrated with three examples - cadmium, chromium and chlorine. The results indicate that useful information can be obtained using relatively weak neutron sources. The method is subject to limitations, however. The resolution of the photomultiplier tube, low counting rates and partial shielding of the crystal by the target reduce the effectiveness of the technique at present. Improvements in resolution are expected in the latest photomultipliers. As yet, there appears to be little that can be done to increase counting rates without increasing the strength of the source. The use of more target material increases the shielding effects mentioned above.

The γ-ray background due to the neutron source itself can be reduced by using a polonium-beryllium source. Unfortunately, the short half-life of polonium makes impossible the maintenance of a constant neutron flux over a period of months, unless the longer lived members of the radium family are introduced into the source.

GENERAL DISCUSSION

The versatility of the scintillation spectrometer has been demonstrated through its application to the study of γ-ray spectra, beta spectra and the nuclear energy levels excited by neutron capture.

The study of γ-ray and beta-ray spectra by this means is now an established technique in nuclear physics. The study of the radiative capture of slow neutrons by means of a scintillation spectrometer, however, is a recent innovation. It is undoubtedly the most significant aspect of scintillation spectroscopy mentioned in this paper, for it supplies a method of studying nuclear energy levels without involving the use of high energy accelerators. The results presented suggest, however, that the use of the scintillation spectrometer in conjunction with a nuclear reactor will be the most efficient method of studying (n, γ) processes. Improvements in resolution are necessary of course. It is hoped that these will be forthcoming in the near future with the appearance of new photomultiplier tubes and improved methods of utilizing the crystal detectors.

The scintillation counter has now become a basic tool for the nuclear physicist, whether it be used for spectrometry or for detection purposes only. Its advent has

completely altered the attitude toward the study of natural activities, while the use of large volumes of liquid scintillating materials has greatly increased the efficiency of coincidence and anti-coincidence techniques in cosmic ray research. Undoubtedly many new applications will yet be found. Some of these will supercede current techniques, while others will lead to fields of activity as yet untouched.

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