GAMMA RAY RESPONSE OF A CsI(Tl) CRYSTAL

TO 14 MEV NEUTRONS

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THESIS

Presented to the Graduate Council of the
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By

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CHAPTER I

INTRODUCTION

The purpose of this paper is to study the possible excited states in the nucleus of Cs^{133}. At North Texas State College a 100-Kev Cockcroft-Walton accelerator has been constructed for use as a neutron source for exciting various nuclei and for studying the gamma spectra obtained to determine their energy levels. The accelerator was designed to employ the $H^3(d,n)He^4$ and $H^2(d,n)He^3$ reactions to obtain a high yield of approximately 14 Mev and 2.5 Mev neutrons respectively. W. W. Givens (14) used this method with the 14 Mev neutrons in his study of As^{75}.

In this author's search for a suitable element to study it was decided to restrict the possible elements to the medium range of nuclides due to the extensive research which has been done on the light elements. A choice element would have the characteristics of a 100 per cent abundance (thus eliminating expensive separation methods), and unknown energy levels. It was noted that cesium had all of these characteristics and that its use as a detector in the form of a thalium-activated CsI scintillator would greatly simplify the experimental arrangement. The use of the scintillation counter as a radiation detector and gamma-ray spectrometer requires an abundant knowledge of a particular scintillator's actions while in use and under bombardment.

Since the time NaI(Tl) crystals were first used successfully as radiation detectors, many other crystals have been developed for this purpose and are being tested and used with continually increasing applications.

The CsI(Tl) crystals have many uses in the detection of various kinds of radiation. They may be used exposed in a vacuum, and may be handled and worked with ease. The current use of crystals has been greatly accelerated by a high sensitivity, an output proportional to the energy of the incident radiation, and a variety of sizes and shapes which are available commercially. The biggest advantage the CsI(Tl) has over the NaI(Tl) crystal is that it is not so hygroscopic and appears to have a slightly better efficiency. However, the cost of the CsI(Tl) crystal at this time is two or three times the cost of a NaI(Tl) crystal of the same size.

The North Texas State College 100-Kev Cockcroft-Walton accelerator was employed in this experiment to obtain neutrons with an approximate energy of 14 Mev. In the $H^3(d,n)He^4$ reaction the energy of the neutrons was calculated to be about 14.82 Mev in the forward direction using the "Q" value given by Van Patter and Whaling (11). The spread of energy over the face of the crystals due to the difference in angle was found to be 0.054 Mev. This is of the same order of magnitude as the spread due to the thickness of the tritium target.

The reactions possible when Cs^{133} is bombarded with 14 Mev neutrons are shown below and the "Q" values given are from calculations using data of atomic masses by Duckworth (3) and beta decay energies of Lidofsky (7):

1. $Cs^{133} + n \longrightarrow Xe^{132} + n + p - 6.692$ Mev
2. $Cs^{133} + n \longrightarrow Xe^{132} + H^{2} - 4.025$ Mev
3. $Cs^{133} + n \longrightarrow Cs^{134} + 6.824$ Mev
4. $Cs^{133} + n \longrightarrow Cs^{132} + 2n - 7.724$ Mev
5. $Cs^{133} + n \longrightarrow Xe^{133} + p + 0.948$ Mev
6. $Cs^{133} + n \longrightarrow I^{130} + He^{4} + 4.27$ Mev
7. $Cs^{133} + n \longrightarrow Cs^{133'} + n + \gamma$.

The first two reactions above give rise to stable isotopes, but the energy of the bombarding particle and the coulomb barrier to the proton, which is about 10.6 Mev, would probably cause the first reaction to go by electromagnetic radiation in preference to proton emission. Concerning the second reaction it appears, in general, that deuteron formation would be unlikely in this region of the table of nuclides. One would conclude, then, that both these reactions would have a very low yield. The next three reactions (3, 4, 5) all give rise to radioactive isotopes and a measure of half-life activity would make it possible to detect the particular activity of the products of these reactions. As for the sixth reaction, the coulomb barrier to the alpha particle is about 21 Mev and I^{130} is a beta emitter which has a half-life that could be detected in the same manner as those above. As for the last reaction (7), it appears to be the most likely to give a large yield. The studies made by many investigators of the reactions of the NaI(Tl) crystal (2, 5, 6, 8, 12) allowed this author to use it as a means of checking the background for this experiment. Consideration was given to an elastic scattering of the neutrons from Na^{23}, and it was found that the maximum energy obtainable for the sodium nucleus would be about 0.224 Mev.

Wolf's (13) paper which reports the use of a NaI(Tl) crystal to study the gamma-ray spectrum of sodium metal will be of interest here due to the similarity in the determination of background, and the values obtained for the energy levels of sodium and iodine. Wolf used 2.5 Mev neutrons on a NaI(Tl) crystal and showed values of the pulse-height distribution taken both with and without sodium metal packed around the crystal. She concluded that the peaks which rose in intensity were probably due to the sodium and those which did not rise were due to iodine. She gave the values for iodine to be about 0.2, 0.41, 0.64 and 1.01 Mev. For sodium she obtained the values 0.45, 1.69 and 2.2 Mev. Even though there is a great difference in the energy of the neutrons used by Wolf and those used by this author, the difference in gamma-ray intensities would not be expected to be too great for the energy range of the gamma rays studied here and one would hope that a comparison of data would be significant.

There were no data obtainable in the literature involving the bombardment of CsI(Tl) crystals with high energy neutrons.

However, Fagg (4), using coulomb excitation with alphas of about 5 Mev, reported cesium to have gamma peaks at 0.082, 0.302 and 0.379 Mev, while Temmer and Heydenburg (9,10), using the same method, reported the 0.082 peak and also found one at 0.163 Mev.

BIBLIOGRAPHY FOR CHAPTER I

Articles

1. Bashkin, S., Carlson, R. R., Douglas, R. A., and Jacobs, J. A., "Response of CsI(Tl) Crystals to Energetic Particles," Physical Review, CIX (1958), 434.

2. der Mateosian, E., Mckeown, M., and Muehlhause, C. O., "Response of NaI Crystals to Alpha Particles and Electrons as a Function of Temperature," Physical Review, CI (1956), 967.

3. Duckworth, H. E., "Mass Spectroscopic Atomic Mass Differences #2," Reviews of Modern Physics, XXIX (1957), 767.

4. Fagg, L. W., Bulletin American Physical Society, II, 4 (1957), 207.

5. Kelly, G. G., Bell, P. R., Davis, R. C., and Lazer, N. H., "Intrinsic Scintillator Resolution," Nucleonics, XIV (4) (1956), 53.

6. Lazer, N. H., Davis, R. C., and Bell, P. R., "Peak Efficiency of NaI," Nucleonics, XIV (4) (1956), 52.

7. Lidofsky, L. J., "Beta Disintegrations Table #2," Reviews of Modern Physics, XXIX (1957), 773.

8. Milton, J. C. D., and Frazer, J. S., "Response of NaI(Tl), KI(Tl), and Stilbene to Fission Fragments," Physical Review, XCVI (1954), 1508.

9. Temmer, G. M., Heydenburg, N. P., "Coulomb Excitation of Heavy and Medium Heavy Nuclei by Alpha Particles," Physical Review, XCIII (1954), 351.

10. Temmer, G. M., "Coulomb Excitation of Medium-Weight Nuclei," Physical Review, CIV (1956), 967.

11. Van Patter, D. M., and Whaling, W., "Nuclear Disintegration Energies," Reviews of Modern Physics, XXVI (1954), 402.

12. Van Sciver, Wesley, "Spectrum and Decay of NaI," Nucleonics, XIV (4) (1956), 50.

13. Wolf, Elizabeth A., "Gamma Rays from Inelastic Neutron Scattering in Sodium and Iodine," Philosophical Magazine, I (1956), 102.

Unpublished Materials

1. Givens, W. W., "An Investigation for Gamma Rays Resulting from the Bombardment of As^{75} with 14 Mev Neutrons," unpublished master's thesis, Department of Physics, North Texas State College, Denton, Texas, 1957.

CHAPTER II

INSTRUMENTATION

The gamma-ray spectra shown in the Appendix were taken using a single-channel scintillation spectrometer. The spectrometer consisted of a radiation detector, linear amplifier, pulse-height analyzer, and scaler. For the bombarding runs on cesium, the radiation detector consisted of a CsI(Tl) crystal, a 6292 Dumont photomultiplier tube, and a cathode-follower type pre-amplifier designed by the Oak Ridge National Laboratory. The pre-amplifier had been modified to shift the high potential from the cathode to the first dynode. A circuit diagram is shown in Figure 1 (Figure 1 and all other figures will be found in the Appendix). The CsI(Tl) crystal was a Harshaw hermetically sealed product, a cylinder one and one-half inches in diameter by one inch thick. The high voltage for the photomultiplier was supplied by a home-built high-voltage power supply similar to the Atomic Instruments Model 312. The output of the cathode follower in the pre-amplifier was fed into an Al linear amplifier, Atomic Instruments Model 218. This signal in turn was fed through an Atomic Instruments Model 510 single-channel differential pulse-height analyzer. The pulse-height analyzer fed an Atomic Instruments Scale of 64 Model 101-M scaler, the output of which went to a mechanical register. A block diagram of the spectrometer is shown in Figure 2.

In order to have the same number of neutrons passing through the crystal for each interval of counting, an alpha monitor was designed to count the number of alphas given off by the $H^3(d,n)He^4$ reaction at right angles to the beam (Figure 3). The alpha monitor consisted of a Harshaw CsI(Tl) crystal blank which was cut and polished to a diameter of one inch and thickness of 0.25 mm. It was mounted to the lucite light-pipe using Dow-Corning high-vacuum grease for optical coupling. The lucite light-pipe was mounted on a Dumont 6292 photomultiplier in a similar way. The output of the photomultiplier was fed into a cathode-follower type pre-amplifier, National Radiac Model sc-2d. The signal from the pre-amplifier was fed into an A1 type linear amplifier, Atomic Instruments Model 218. The amplified pulse here was fed into an Atomic Instruments Model 510 single-channel pulse-height analyzer to discriminate against any pulses of lower height than the alpha particle pulses. The output of the pulse-height analyzer was fed into a scaler and the central control unit described by Givens (1). The alpha monitor was shielded from fluorescence on the target by a 0.00005-inch nickel foil which was placed over the 0.5 cm. opening in the collimator shown in Figure 3. The tritium target was mounted at a 45° angle to the beam of deuterons so that the alphas formed would not have to traverse any more of the target's thickness than was necessary. The solid angle subtended by the alpha monitor to the target was 0.075 steradians.

BIBLIOGRAPHY FOR CHAPTER II

Unpublished Materials

1. Givens, W. W., "An Investigation for Gamma Rays Resulting from the Bombardment of As^{75} with 14 Mev Neutrons," unpublished master's thesis, Department of Physics, North Texas State College, Denton, Texas, 1957.

CHAPTER III

EXPERIMENTAL PROCEDURE

The background runs were made using the instrumentation previously described with the exception that the CsI(Tl) crystal was replaced by a NaI(Tl) crystal in the spectrometer setup. The NaI(Tl) crystal was also a Harshaw product, which was identical to the CsI(Tl) crystal in dimensions and mounting.

The use of the NaI(Tl) and CsI(Tl) crystals as detectors eliminated the necessity of scatterers. Even though the two crystals were identical in size and shape (one and one-half inches in diameter by one inch thick), the difference in density and the size of the molecules in CsI and NaI made the number of iodine atoms in each crystal different. The ratio of the iodine atoms in the NaI(Tl) to those in the CsI(Tl) was calculated to be about 1.41 to 1.

The geometry of the experiment is shown in Figure 4. The detector was hung from an aluminum frame by thin wire to help reduce scattering of the neutrons. The aluminum frame was made of two 0.75 inches o.d. tubing 42 inches long mounted in 4.75X1.75X1.75 inches aluminum blocks at each end. A similar aluminum block was mounted in the center and could be moved forward or backward along the two tubes. The detector was hung from the center block, while the whole frame was suspended from the ceiling.

Calibration of the equipment was obtained by using Cs^{137} (0.661 Mev), Na^{22} (0.51 and 1.28 Mev) gamma-ray sources. The curves shown in Figure 5 show the spectrum of Cs^{137} and Na^{22}, superimposed, for both the CsI(Tl) and NaI(Tl) crystals. These curves were also used to match the relative intensities of the curves shown in Figure 10.

The resolutions (pulse-width at half maximum divided by the base-line setting) for the 0.661 Mev gamma of Cs^{137} were determined by the curves in Figures 6 and 7 for the CsI(Tl) and NaI(Tl) crystals and were found to be 10.12 and 11.6 per cent respectively.

At the start of accumulation of data for this experiment, it was noted that some form of activity was interfering with the measurements. In order to determine whether this activity was due to any of the possible reactions mentioned in Chapter I, the curves in Figure 8 were plotted from data obtained by shutting off the accelerator and counting the activity in the CsI(Tl) and NaI(Tl) crystals. Counts were taken for intervals of 50 seconds, with intervals of 50 seconds between counting periods. It was impossible to determine the half-life of the interfering radiation accurately with the equipment used due to its reaching background very rapidly. The activity seemed to decay with an approximate half-life of 16 minutes. The origin of this activity, if it is due to a single source, is somewhat obscure. However, since both crystals showed the same half-life, it is probably due to some form of background external to the crystal. It was noted, though, that I^{128} has a 25-minute half-life and, since the reaction $I^{127}(n,\gamma)I^{128}$ is energetically possible, one might think that this could cause some of the activity observed. In order that this activity would not affect the final data, several half-lives were allowed to pass, after the beam was turned on, before any measurements were taken.

CHAPTER IV

ANALYSIS OF DATA

The curve of Figure 9 shows the gamma-ray spectrum of a CsI(Tl) crystal. This curve was taken while exploring the energy range from about 0.25 Mev up to about 2.5 Mev. An expanded search of the area between the arrows for the CsI(Tl) crystal and the background spectrum which was obtained from the NaI(Tl) crystal are plotted in Figure 10. Outside this region no appreciable difference in the two spectra was noted. A combined spectrum for Na^{22} and Cs^{137} sources was plotted to the came coordinates for both the NaI(Tl) and CsI(Tl) crystals in Figure 5 and these were used to normalize the curves of Figure 10 as was mentioned in Chapter III.

It is readily seen in Figure 10 that there is a prominent peak in the NaI(Tl) curve that did not show in the CsI(Tl) curve. It can be seen, in comparison with the data obtained by Wolf (4), that this peak could be the 0.41 Mev gamma given off by iodine. Figure 11 shows the two peaks obtained by Wolf in this range and they appear to agree with those obtained here. One would expect the iodine peaks to show up more in the NaI(Tl) than in the CsI(Tl) due to the higher abundance of iodine atoms in the NaI(Tl) crystal.

In Figure 12 a difference of the two curves of Figure 10 is shown. Since there will be peaks and valleys in a difference of the two curves, one may possibly attribute all the valleys to the peaks which are due to sodium and iodine and all of the peaks will most likely be attributed to cesium. All the reactions outside the crystals resulting in gamma rays should be identical in both curves.

Conclusion

It is seen that there are two valleys in Figure 12 which are in good agreement with Wolf's results for iodine. Also in Figure 12 the valley corresponding to the 0.41 Mev energy has a tendency to spread toward a higher energy. The article by Wolf and one by Morgan (3) show that Na^{23} has an excited state of 0.44 Mev. This excited state of Na^{23} could be the cause of the spreading. It appears that the two positive peaks in Figure 12 would be due to gammas of approximately 0.56 and 0.75 Mev from Cs^{133}. The rise in Figure 12 to the left of these two peaks was on a portion of the curves of Figure 10 which has an almost vertical slope rising to background. Due to this high background this author feels that no statements can be made of energies below 0.35 Mev, and energies up to about 0.4 Mev would be in doubt. The sharp rise on the low-energy side of the 0.41 Mev valley might be due to the 0.379 Mev peak given by Fagg (1), although this author would not be able to make this assignment from these data alone.

It was noted in an article by Keister, Lee and Schmidt (2) that Cs^{134} has a gamma of 0.563 Mev but, as stated in the Introduction, if the reaction $Cs^{133}(n,\gamma)Cs^{134}$ were very prominent there would be a half-life (which is about 3.15 hours for beta emission) that is measurable. There was no half-life measured which was comparable to that of Cs^{134}. The reactions which are shown below have half-lives of 7.1 days, 2.3 days and 12.5 days respectively:

$$Cs^{133} + n \rightarrow Cs^{132} + 2n - 7.724 \text{ Mev}$$
$$Cs^{133} + n \rightarrow Xe^{133} + p + 0.948 \text{ Mev}$$
$$Cs^{133} + n \rightarrow I^{130} + He^{4} + 4.27 \text{ Mev.}$$

No half-lives comparable to these were measured and, as was pointed out, the reactions

$$Cs^{133} + n \rightarrow Xe^{132} + n + p - 6.692 \text{ Mev}$$
$$Cs^{133} + n \rightarrow Xe^{132} + H^{2} - 4.025 \text{ Mev}$$

are improbable. Thus, the results of this investigation point to the conclusion that the gamma rays of energies 0.56 Mev and 0.75 Mev are due to excited states in the Cs^{133} nucleus resulting from the inelastic scattering of neutrons.

Further investigations of these levels are contemplated by using the following methods:

1. Bombardment of CsI(Tl) crystal with 2.5 Mev neutrons (the background would be expected to be lower).

2. Bombardment of the CsI(Tl) crystal with both 2.5 Mev and 14 Mev neutrons when the crystal is surrounded with a cesium compound in a method similar to that described by Wolf for sodium.

The results of these experiments would make possible more definite assignment of these levels.

BIBLIOGRAPHY FOR CHAPTER IV

Articles

1. Fagg, L. W., Bulletin American Physical Society, II, 4 (1957), 207.

2. Keister, G. L., Lee, E. B., and Schmidt, F. H., "Radioactive Decay of Cs^{134} and Cs^{134m}," Physical Review, XCVII (1955), 451.

3. Morgan, I. L., "Inelastic Scattering of Neutrons," Physical Review (1956), 1031.

4. Wolf, Elizabeth A., "Gamma Rays from Inelastic Neutron Scattering in Sodium and Iodine," Philosophical Magazine, I (1956), 102.

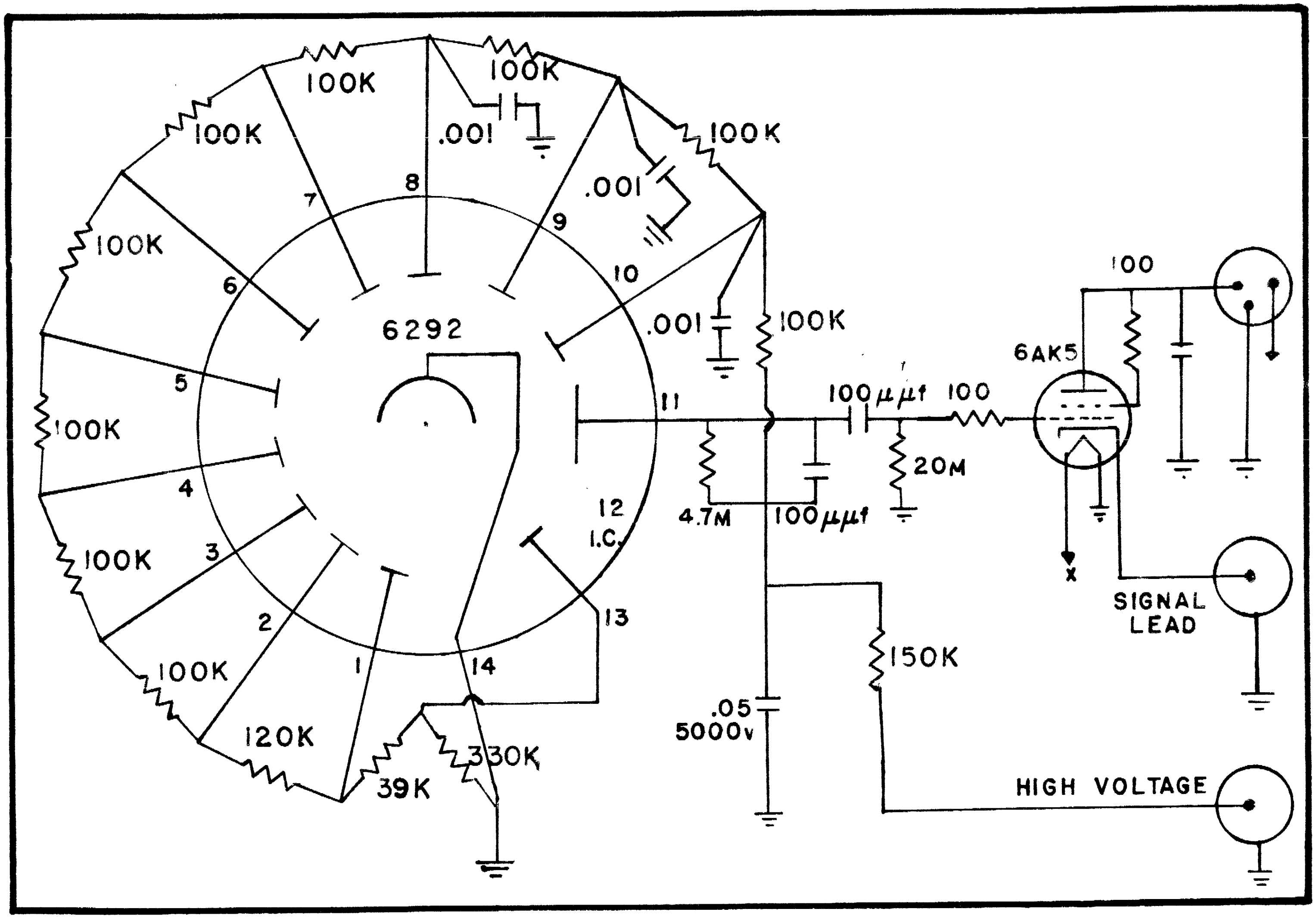


FIG.1 - - MODIFICATION OF GAMMA RAY SPECTROMETER PRE-AMP.

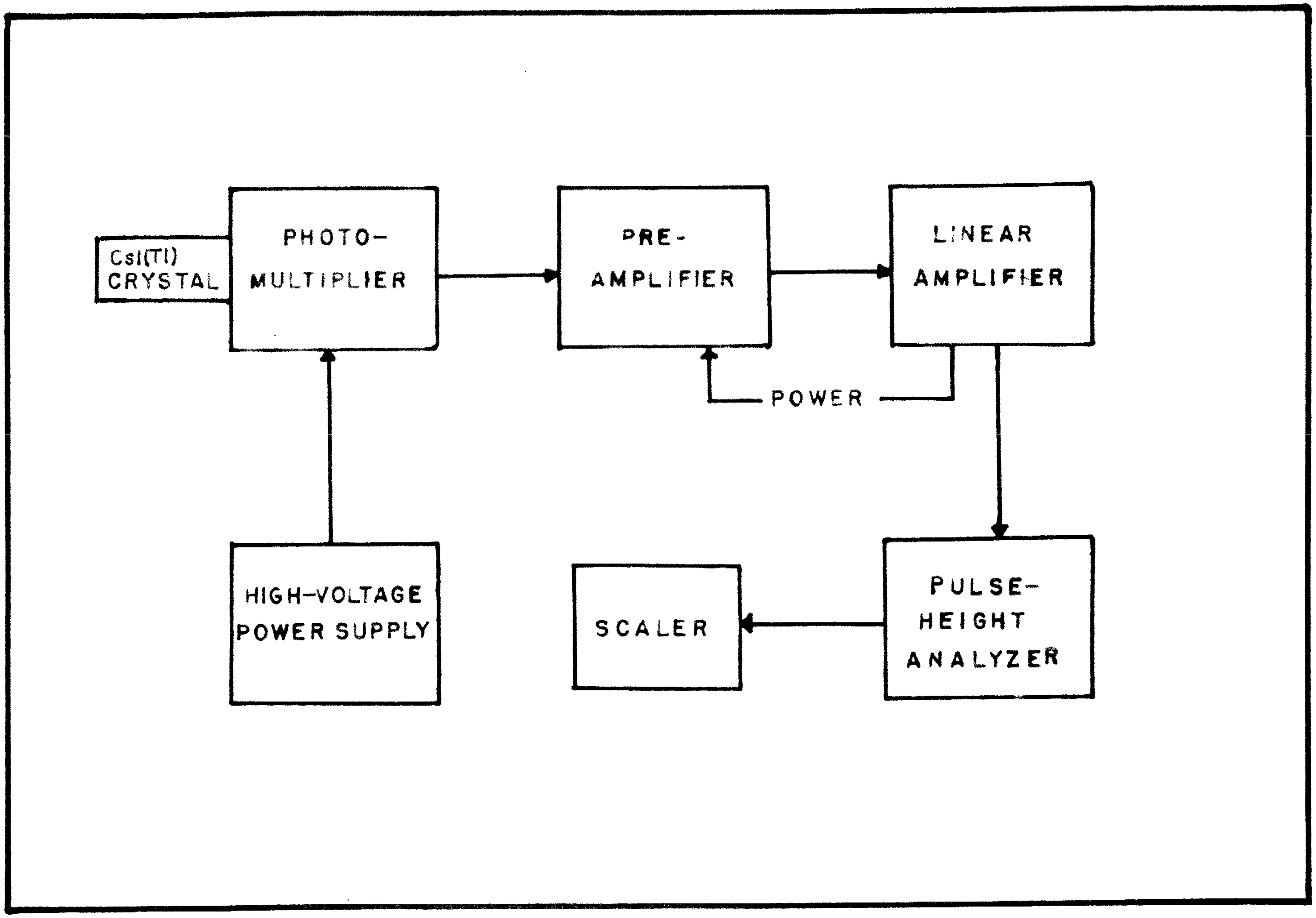


FIG. 2--GAMMA RAY SPECTROMETER BLOCK DIAGRAM

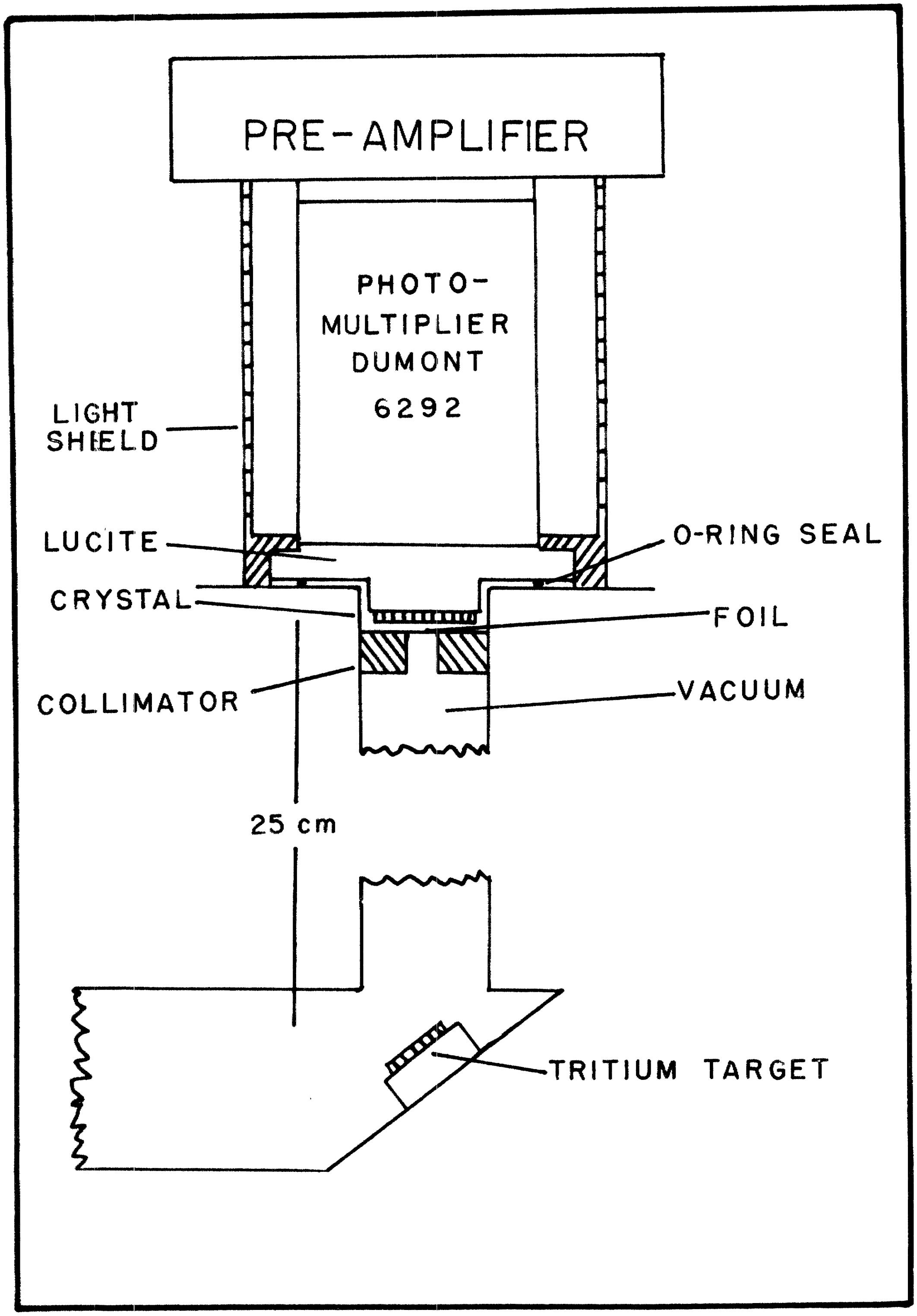


FIG. 3--ALPHA MONITOR

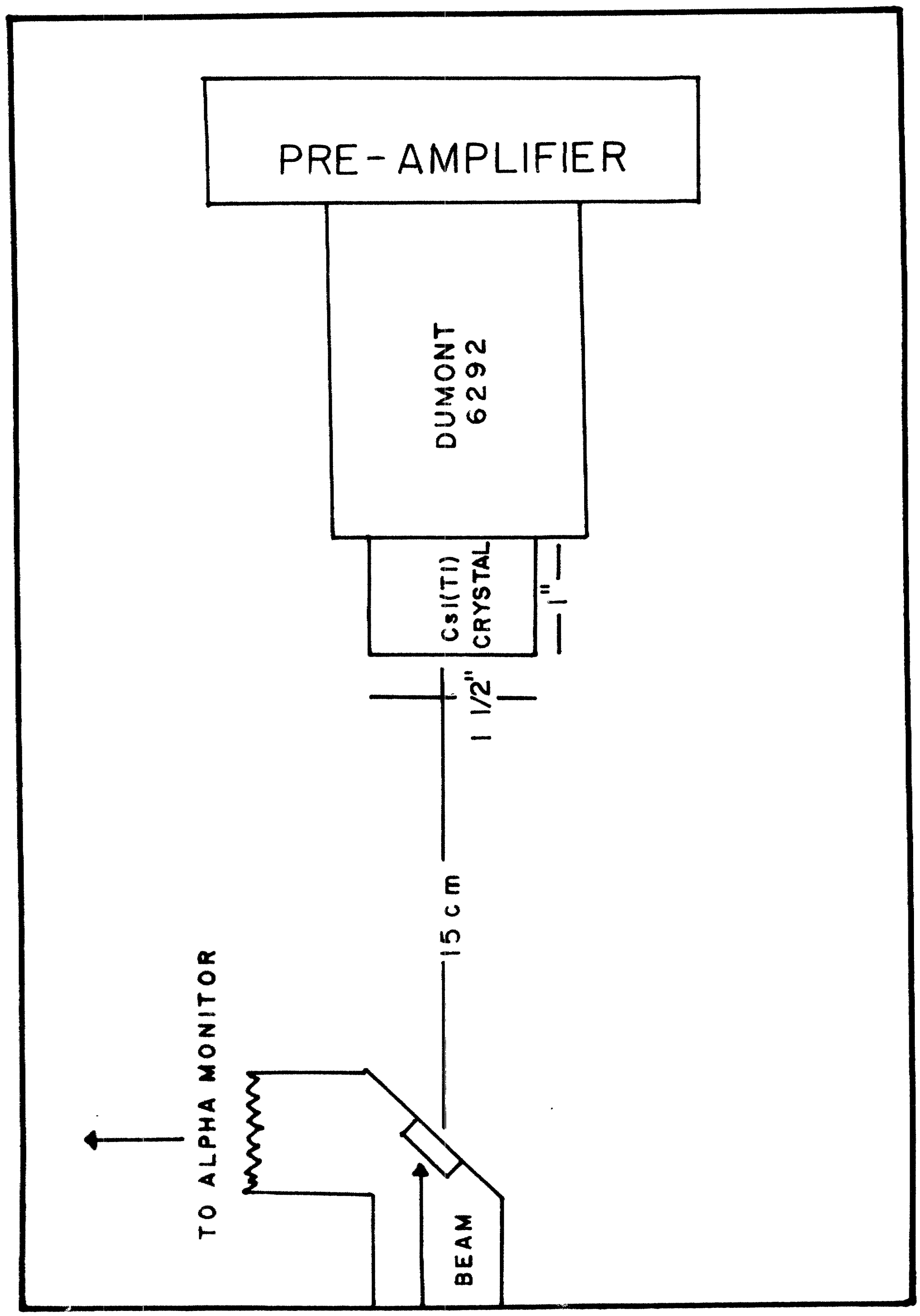


FIG. 4--EXPERIMENTAL ARRANGEMENT

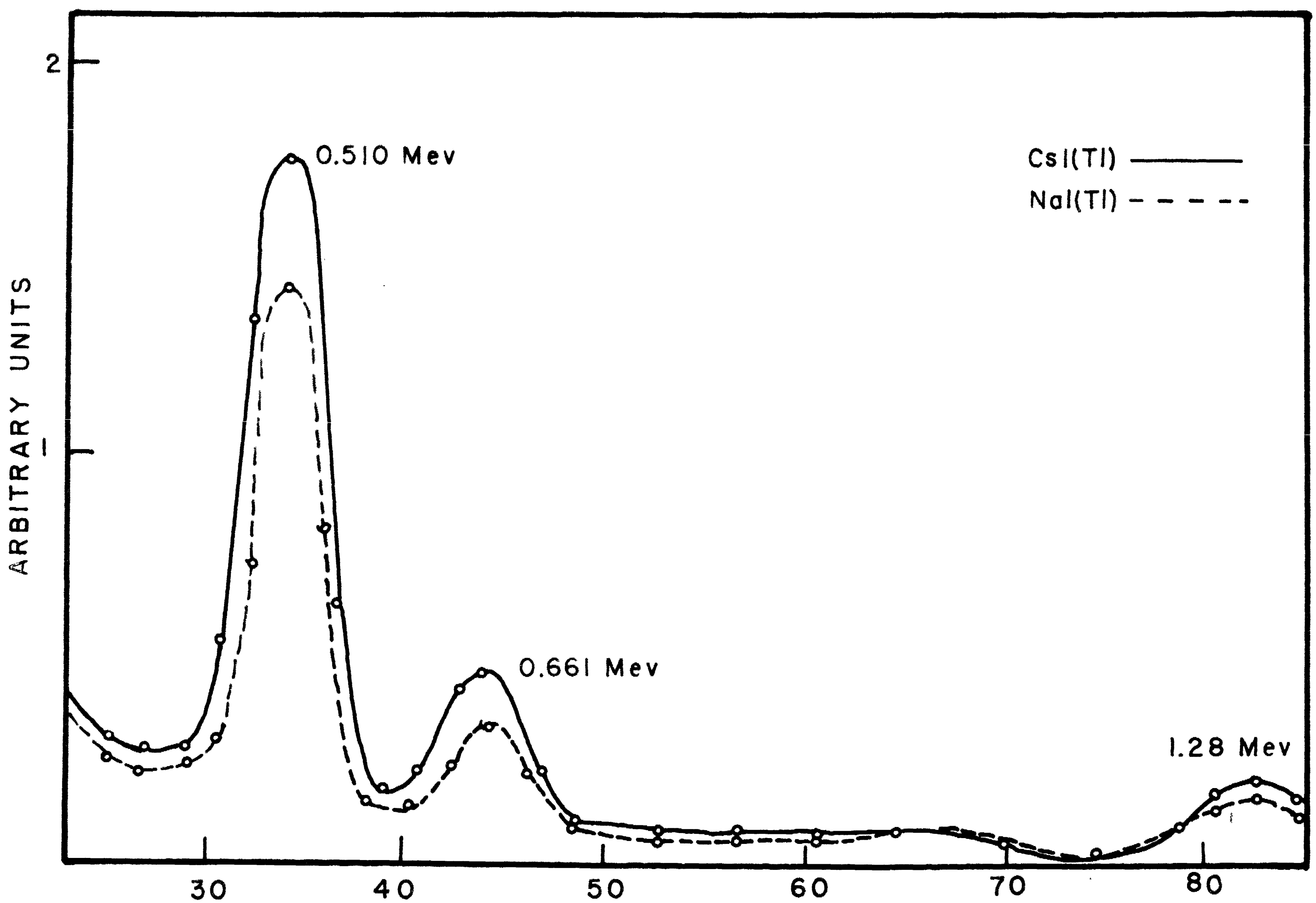


FIG. 5--PULSE-HEIGHT DISTRIBUTION FOR Na^{22} & Cs^{137}

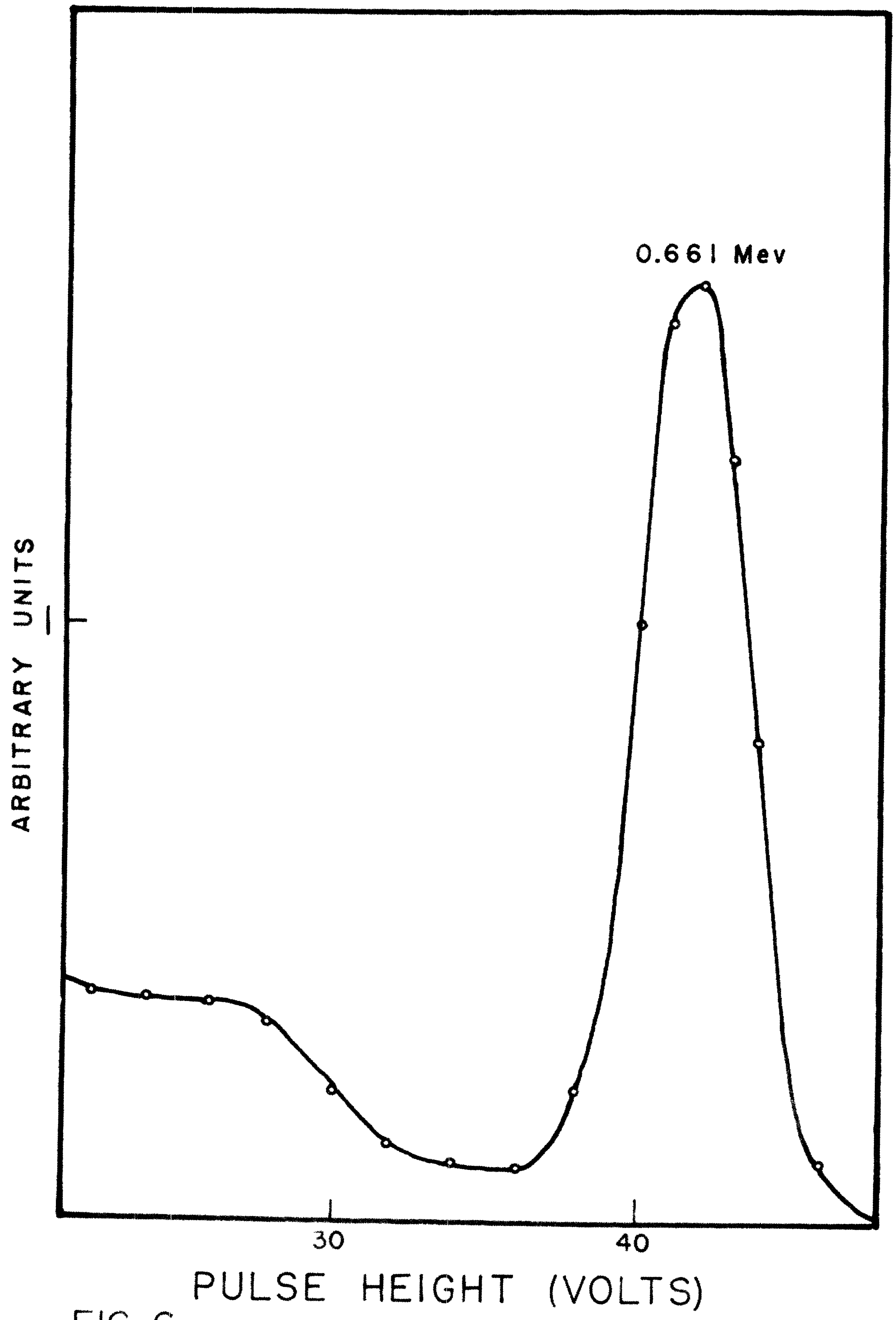


FIG. 6--PULSE HEIGHT DISTRIBUTION FOR Cs^{137} USING THE CsI(Tl) CRYSTAL

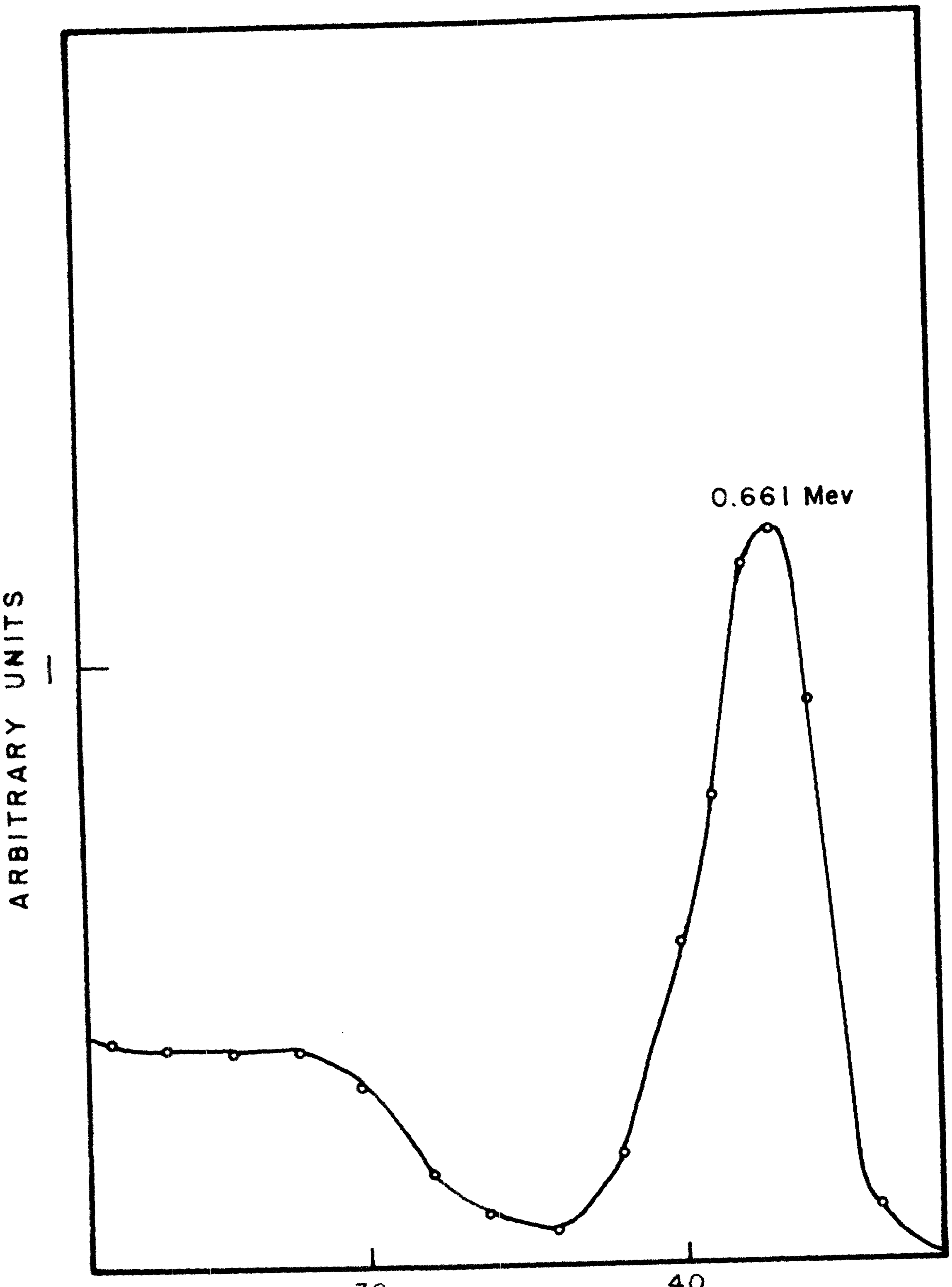


FIG. 7--PULSE HEIGHT DISTRIBUTION FOR Cs^{137} USING THE NaI(Tl) CRYSTAL

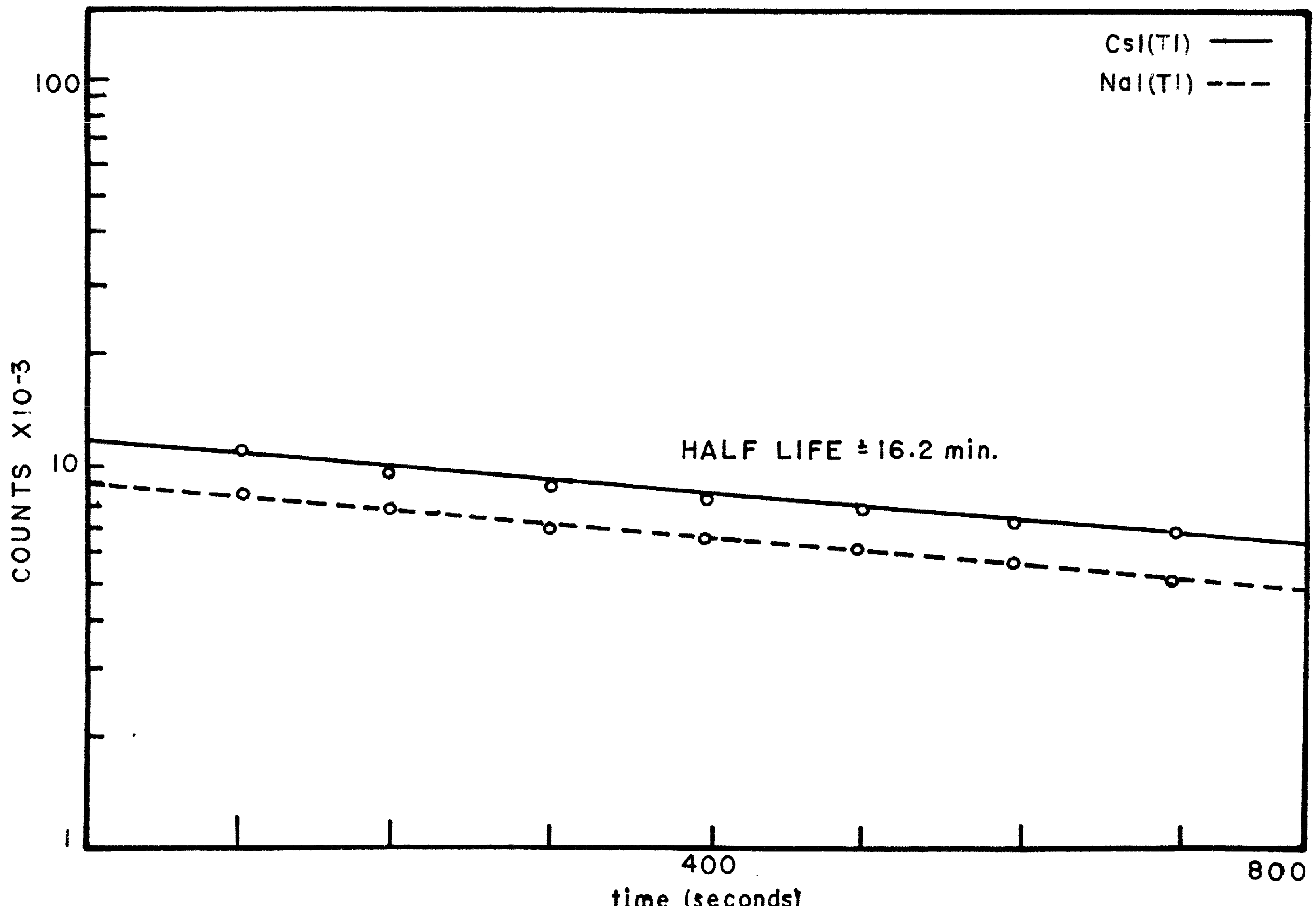
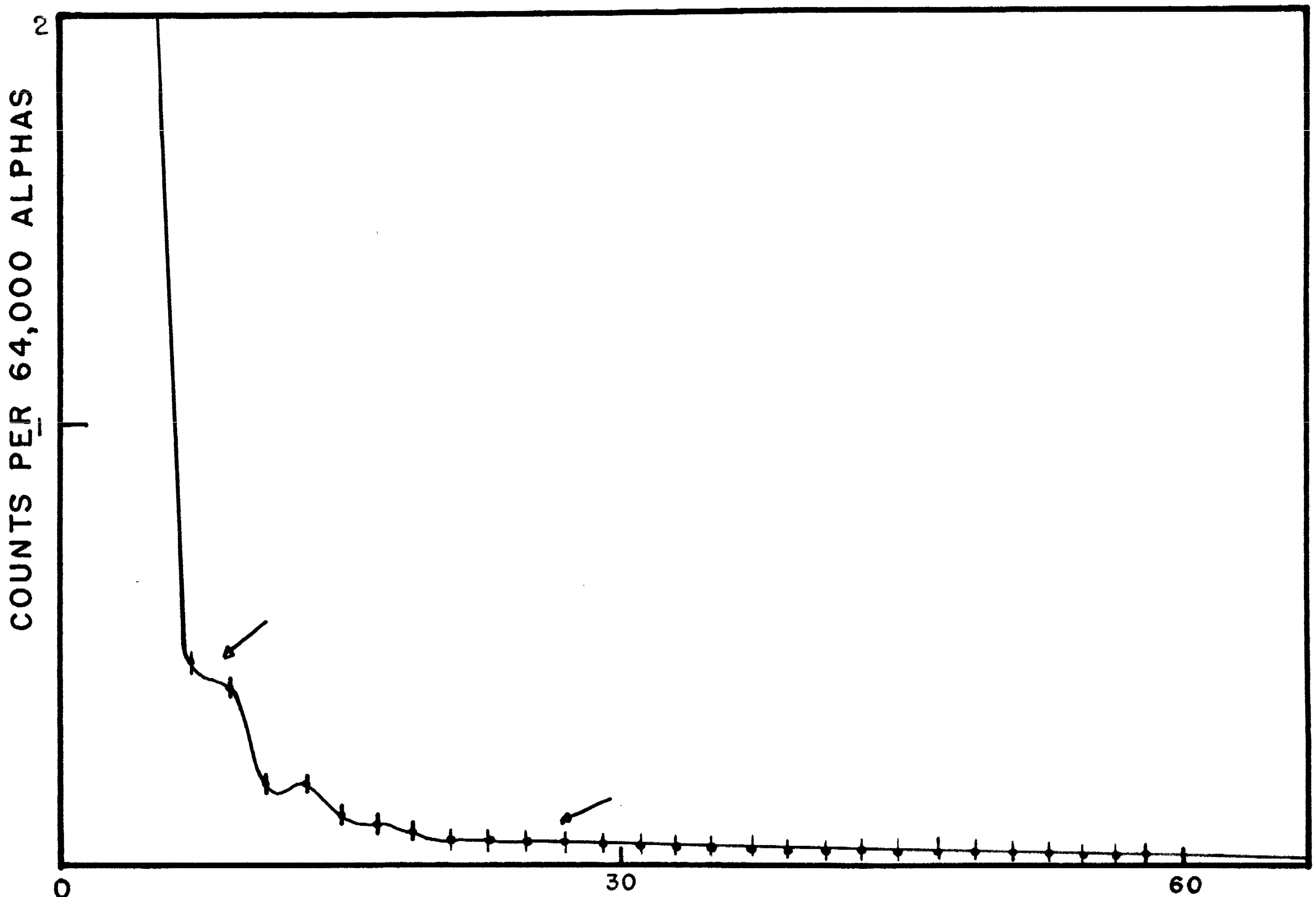


FIG. 8.--HALF LIFE OF ACTIVITY PRESENT



PULSE HEIGHT DISTRIBUTION FOR CsI(Tl)

FIG. 9

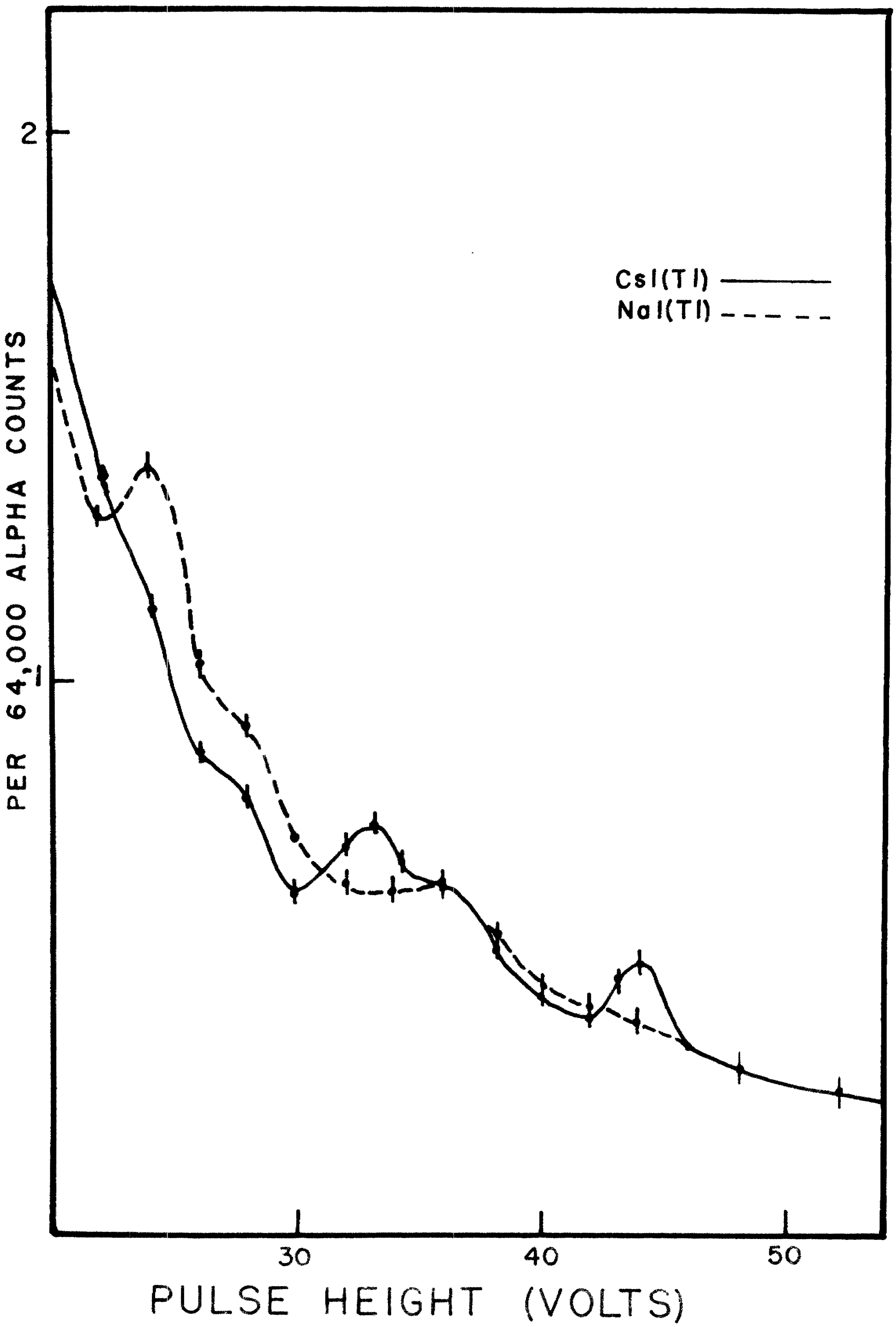
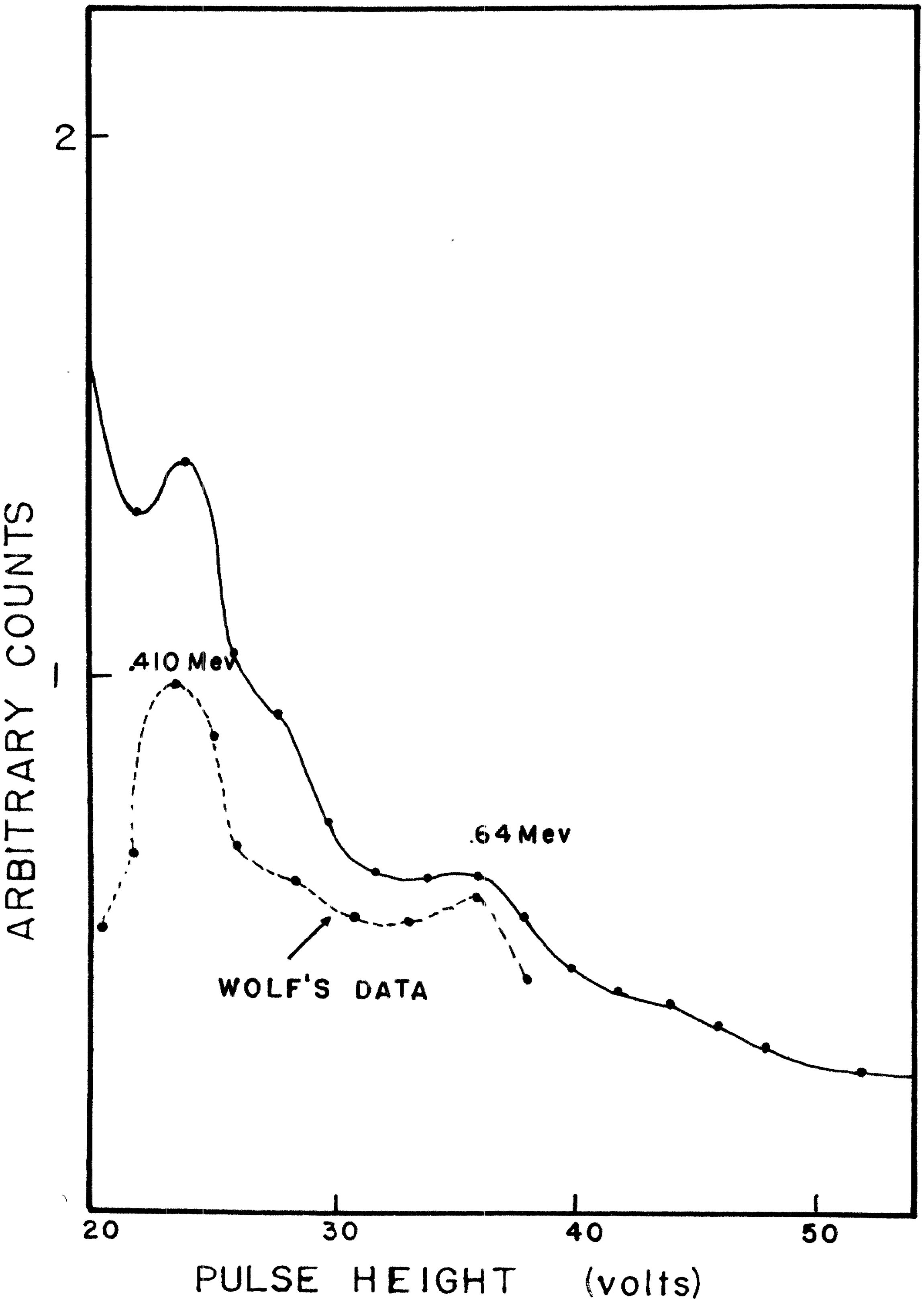


FIG. 10--PULSE HEIGHT DISTRIBUTION FOR CsI(Tl) & NaI(Tl) CRYSTALS



PULSE HEIGHT DISTRIBUTION FOR NaI(Tl) SHOWING DATA GIVEN BY WOLF

FIG. 11

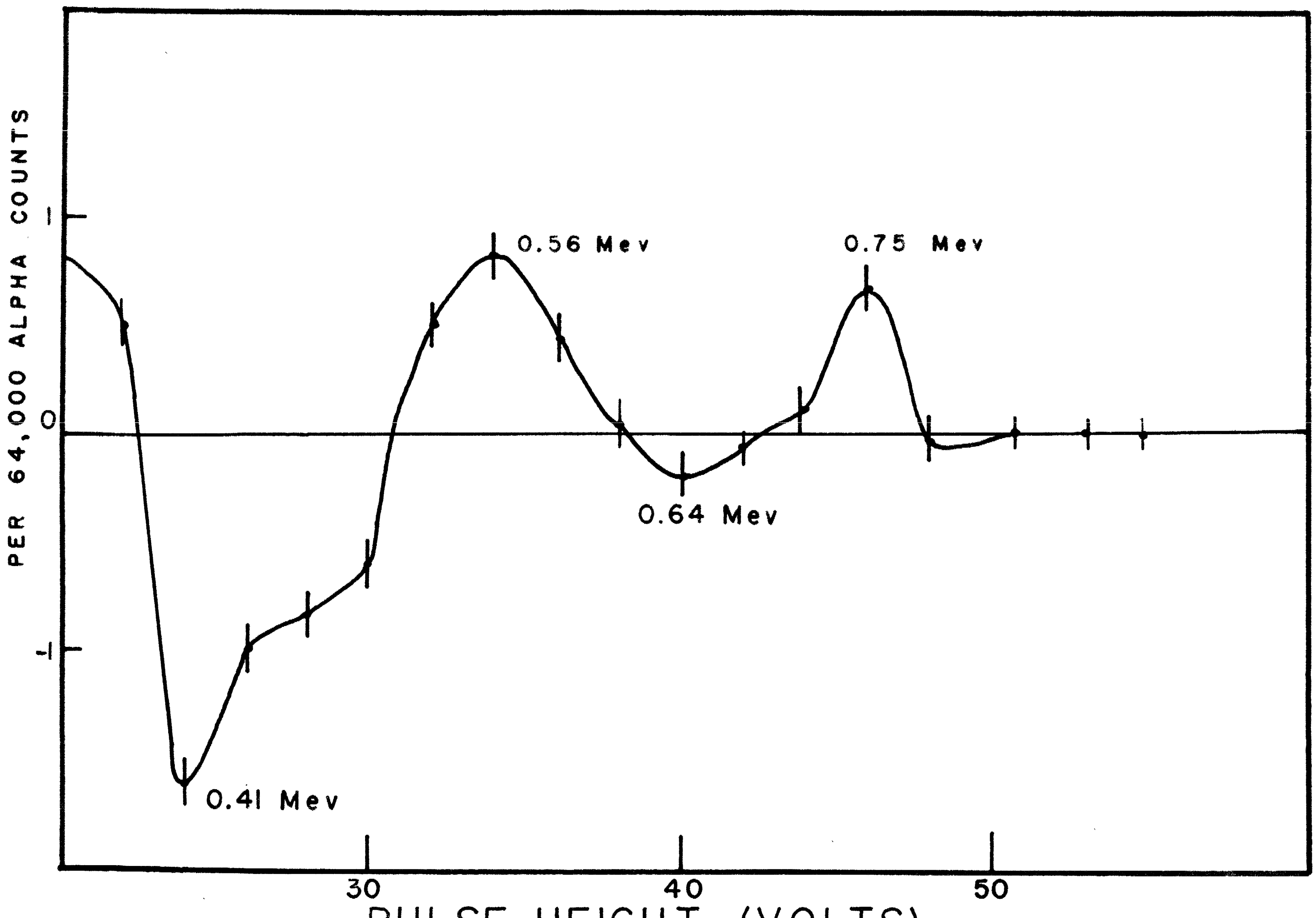


FIG. 12--CORRECTED PULSE-HEIGHT DISTRIBUTION FOR Cs^{133}

BIBLIOGRAPHY

Articles

Bashkin, S., Carlson, R. R., Douglas, R. A., and Jacobs, J. A., "Response of CsI(Tl) Crystals to Energetic Particles," Physical Review, CIX (1958), 434.

der Mateosian, E., Mckeown, M., and Muehlhause, C. O., "Response of NaI Crystals to Alpha Particles and Electrons as a Function of Temperature," Physical Review, CI (1956), 967.

Duckworth, H. E., "Mass Spectroscopic Atomic Mass Differences #2," Reviews of Modern Physics, XXIX (1957), 767.

Fagg, L. W., Bulletin American Physical Society, II, 4 (1957), 207.

Kelly, G. G., Bell, P. R., Davis, R. C., and Lazer, N. H., "Intrinsic Scintillator Resolution," Nucleonics, XIV (4) (1956), 53.

Keister, G. L., Lee, E. B., and Schmidt, F. H., "Radioactive Decay of Cs^{134} and Cs^{134m}," Physical Review, XCVII (1955), 451.

Lazer, N. H., Davis, R. C., and Bell, P. R., "Peak Efficiency of NaI," Nucleonics, XIV (4) (1956), 52.

Lidofsky, L. J., "Beta Disintegrations Table #2," Reviews of Modern Physics, XXIX (1957), 773.

Milton, J. C. D., and Frazer, J. S., "Response of NaI(Tl), KI(Tl), and Stilbene to Fission Fragments," Physical Review, XCVI (1954), 1508.

Morgan, I. L., "Inelastic Scattering of Neutrons," Physical Review, CIII (1956), 1031.

Temmer, G. M., and Heydenburg, N. P., "Coulomb Excitation of Heavy and Medium Heavy Nuclei by Alpha Particles," Physical Review, XCIII (1954), 351.

Temmer, G. M., "Coulomb Excitation of Medium-Weight Nuclei," Physical Review, CIV (1956), 967.

Van Patter, D. M., and Whaling, W., "Nuclear Disintegration Energies," Reviews of Modern Physics, XXVI (1954), 402.

Van Sciver, Wesley, "Spectrum and Decay of NaI," Nucleonics, XIV (4) (1956), 50.

Wolf, Elizabeth A., "Gamma Rays from Inelastic Neutron Scattering in Sodium and Iodine," Philosophical Magazine, I (1956), 102.

Unpublished Materials

Givens, W. W., "An Investigation for Gamma Rays Resulting from the Bombardment of As^{75} with 14 Mev Neutrons," unpublished master's thesis, Department of Physics, North Texas State College, Denton, Texas, 1957.