GAMMA RAYS FROM NEUTRON EXCITATION OF Cs^{133}

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CHAPTER I

INTRODUCTION

The energy levels of the Cs^{133} nucleus may be studied by several methods. The first to be employed was the gamma ray spectrum of radioactive Ba^{133} (1, 2, 6, 7, 8, 9). The Ba^{133} nucleus decays to Cs^{133} by capturing an orbital electron, resulting in an excited state of Cs^{133}. The Cs^{133} nucleus will then proceed to the ground state, or level of zero energy, by occupying successively lower energy levels. At each transition, the excess energy is released in the form of a gamma ray. The limitation of this method of study is twofold. First, there is no possibility of exciting levels of greater energy than 0.44 Mev, since this is the highest level to which Ba^{133} decays by electron capture. Second, considerations of spin and parity forbid certain transitions so that there might be levels which would not be observed because they are not accessible from Ba^{133} except by forbidden transitions.

Levels not accessible by allowed transitions in radioactive decay might be observed by direct excitation of the Cs^{133} ground-state nucleus. This excitation may be accomplished by bombarding Cs^{133} with nuclear particles. One

method which has been employed is coulomb excitation (4), which involves bombarding the target nucleus with a charged particle of insufficient energy to penetrate its coulomb field. However, the field of the bombarding particle may transfer energy to the target nucleus. The target nucleus thus elevated to an excited state will then decay in the manner previously described.

There exists an even more powerful method of excitation. The neutron, being uncharged, can readily penetrate the coulomb field of the nucleus. It may form a compound nucleus too short-lived to be observed, and then emerge with a slightly lower energy leaving the target nucleus in an excited state. Unfortunately, the neutron is also a powerful tool for inducing a multitude of other reactions so that results may be difficult to interpret. Nevertheless, this method, known as (n,n') or neutron inelastic scattering, can produce valuable information, particularly where some data are already available to aid interpretation. All previously published work on Cs^{133} was done by one of the first two methods.

The North Texas State College 100 Kv Cockcroft-Walton accelerator [for operation details see reference (3, 5)] can produce 14 and 2.5 Mev neutrons from the $D(t,n)He^4$ and $D(d,n)He^3$ reactions, respectively. These are referred to as the D-T and the D-D reactions, respectively. It was therefore decided to investigate Cs^{133} with both of these neutron

energies in the hope of finding new levels and clarifying some points concerning previously reported transitions.

A recently published (9) energy-level diagram of Cs^{133} is shown in Figure 1. (This and succeeding figures will be found in the appendix.) The horizontal lines are known energy states. The excitation energy appears at the right. Arrows indicate observed transitions as measured by the gamma rays emitted. The dotted arrows are transitions not observed by all investigators. Table I gives experimental details and additional data. Throughout this paper this level diagram will be used to identify transitions. Where investigators report rays of different energies those energies are given, but in identifying the transitions, reference is made to Figure 1. Since the various values reported for the levels are close enough that no ambiguity results, the necessity of reporting the various level schemes is avoided.

The first phase of the project here, an investigation with the 14 Mev D-T neutrons, was completed by Young (10) in August, 1958. Young's results are included in Table I. The present paper deals with the investigation using the 3.3 Mev D-D neutrons. The spectrum is thus extended to lower energy, since the less energetic D-D neutrons produce less background in that region. As in Young's work, the cesium is present in the form of a CsI(Tl) scintillation crystal. For comparison, a NaI crystal was also used, since it allowed iodine rays to be identified by their appearance in the spectra of both

crystals. Also, the response of NaI crystals to neutrons in this energy range has been published. (See Chapter IV.)

The background consists primarily of gamma rays from neutron-induced reactions in the structures surrounding the crystal, reactions for which the less energetic D-D neutrons are less effective, especially with the rather low neutron flux used. Since the rays are produced within the crystal, almost all of them will pass through part of the crystal, making weak rays more easily observed than with an external scatterer. With this arrangement it is possible to apply shielding to the detectors since there will be no attenuation of the rays which are of interest. The neutrons, of course, can penetrate matter easily, and there will be no difficulty on their account if it can be established that no neutron-induced reactions occur in the shielding material.

The conditions discussed in the preceding paragraph allowed the spectrum to extend as low as .1 Mev. The upper limit of .51 Mev was established by the difficulty of maintaining instrumental stability over the long counting intervals required for good statistics. Preliminary checks above .51 Mev showed very low counting rates, and no prominent features on the spectrum.

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CHAPTER II

INSTRUMENTATION

The de-excitation gamma rays were detected, and the energies determined by single-channel scintillation spectrometers. The counting interval was determined by a neutron monitor. Figure 2 shows the instrumentation in block form, with the components listed in Table II. Figure 7 shows the detector heads consisting of the crystals with their photomultipliers and preamplifiers, the lowermost of which is the monitor. The detector head assembly was suspended as shown in Figure 8, in which the target has been replaced by a glass plate. A lead sheet which hung between the detectors and target has been omitted from this photograph. The use of this shield was later discontinued as will be explained later.

The single-channel pulse analyzers were operated with a counting channel width of one volt and the gain set to give an energy calibration of 42 V = 0.51 Mev. It was considered advisable to have the same calibration in order to obtain approximately the same resolution on both channels, and to allow rapid comparison when taking the sodium and cesium spectra simultaneously. Because of a difference in

the pulse heights from the two crystals, and a failure of the amplifier gain ranges to overlap, this was the only setting where the energy calibration on both channels would be the same. The improvements in resolution which might be expected at higher gains were not obtained because of amplifier instability. The amplifiers were set for delay-line clipping producing a square pulse of one microsecond duration. Resolution of the 0.66 Mev Cs^{137} peak was 10 per cent.

The monitor amplifier was operated with a bandwidth setting of 0.1 Mc. Since no pulse-height analyzer was employed, the amplifier's discriminator output was used. The amplifier was operated at maximum gain so that there would be a comparatively wide range between noise and the neutron pulses, with the discriminator set in this range so that gain drift would have little effect. Since the noise was well below the discrimination level, and the neutron pulses were well above it, an upward drift would merely saturate the amplifier. This was of no consequence without pulse-height analysis. The monitor was checked with Co^{60} and Na^{22} gamma sources and had negligible response. Also, as operated, the monitor showed no response to bombardment of a bare target plate which duplicated all operating conditions except neutron flux.

The gamma channels were calibrated with Na^{22} (0.51 Mev annihilation). A pulse generator was used to calibrate the

monitor amplifier, the signal being applied simultaneously to the previously calibrated gamma channel. The neutron crystal does not produce a sharp peak, so that calibration of the detector was not possible for the monitor. However, it was found that large drifts had no noticeable effect on the counting rate of the monitor for reasons already stated. An automatic switch activated by the monitor register stopped all three channels and the timer after 6400 monitor counts.

CHAPTER III

EXPERIMENTAL PROCEDURE

The first preliminary run was taken without any shielding of the detectors from the high-voltage power supply of the accelerator. Background completely obscured the spectrum below 0.1 Mev, and it was discovered that X-rays from the accelerator power-supply rectifiers were largely responsible. A lead shield was hung on the safety screen surrounding the high-voltage supply, reducing the background so that the spectrum could be taken down to zero energy, whereas the registers had previously overloaded at about 0.10 Mev. The X-rays seemed to have a maximum energy corresponding to the peak inverse voltage on the rectifier tube. While intense enough to saturate the scintillation counters, these X-rays were not found to be dangerous to personnel when checked with an ionization survey meter.

With this shielding, the obscuring effect was not serious above 0.10 Mev but the X-ray peak still appeared both with and without the accelerator target. This peak obscured the region below 0.10 Mev. The background was generally high in this region so that even more shielding would not have improved matters appreciably. For this reason spectra were not extended into this region on subsequent runs.

For the first series of spectra the cesium iodide and sodium iodide crystals were used simultaneously. The deuteron beam was 100 Kev and calibrations were made every hour on the gamma channels. Nine curves were obtained in this run. The greatest problem was the prevention of instrumental fluctuations since a spectrum from ten volts pulse height to forty-five volts required about eight hours. Most of the curves were free from sudden shifts, but there were slow variations which made normalization impossible although they could not give rise to spurious peaks. This series of runs was made with the lead shield present between target and detectors. The failure of dropping resistors on the accelerator column terminated this series.

The second series of runs was made with only one gamma ray channel. After a linearity check with Pb^{210} (.047 Mev) revealed that the CsI channel was non-linear, it was removed since the defect was not readily remedied. Also, checks showed that the shield between target and detector had no effect, and it was removed. This allowed the single detector to be placed more favorably with respect to the target, and slightly higher counting rates were obtained. The stability and resolution were improved by taking the most reliable units from the two channels. This series yielded two complete curves and one partial one. An improved beam gave higher neutron fluxes so that the time required for a spectrum was

reduced to about four hours. The effects of drift were not noticeable on the curves.

A final run was made with the gain doubled and the counting channel width unchanged in the hope of securing better resolution. This was not successful, and the curve was poor due to erratic performance of the amplifier on high gain (near maximum).

If calibration checks at the beginning and end of a period showed a drift of more than one volt, or if arcs were observed during the period, then the data taken during that period were rejected. It was observed that drifts of several volts occurred after all but the smallest arcs. Even the use of a Sola saturable core transformer and a Sorenson voltage regulator in series on the power line did not prevent this. A malfunction of the ion-source oscillator concluded the experiment.

CHAPTER IV

RESULTS AND CONCLUSIONS

A typical cesium iodide spectrum is shown in Figure 3. This curve was taken during the second series. A typical sodium iodide spectrum is shown in Figure 4. This spectrum was taken during the first run. (Note that the NaI crystal was on the linear channel during this series.) The counting interval was 6400 neutron counts, so that the numbers plotted in these figures are 1/64 of the total counts observed. Since a valid normalization procedure could not be found, the curves were not combined. The principal cause of this difficulty in normalization was the variation of the neutron flux, which allowed the counting interval to vary, admitting a variable amount of background. These variations were slow, and did not produce peaks, but made it impossible to find a normalization factor for an entire curve. However, even though the statistics on a single curve are not ideal, it is significant to note that the features in Figure 3 were defined on all CsI curves and no feature not shown appears on two curves. The same consideration applies to Figure 3. Since the CsI curves from the earlier runs are non-linear, they are not included in actual calculations, their energy

calibrations being uncertain, but they do serve to confirm the existence of the features reported.

The possibility of interference by the neutron-induced reactions mentioned in the introduction can be eliminated by considering the possible reactions. Those in cesium are:

$$Cs^{133} + n \rightarrow Xe^{133} + P + 0.948 \text{ Mev}$$

$$Cs^{133} + n \rightarrow I^{130} + He^{4} + 4.27 \text{ Mev}$$

Any other reactions which are energetically possible would appear on both NaI and CsI spectra and would cause no confusion. Both of the reactions above have activities with half-lives long enough to be easily observed, and no such activity was detected.

All features on the curves are taken to be photopeaks; at the energies and intensities encountered it is unlikely that other effects would be prominent. Only the peak at 23 volt pulse height in Figure 3 is suspect; it is in the proper position for a Compton edge from the 33 volt peak, but if it were a Compton edge the sodium iodide curve in Figure 4 should show one and it does not.

The features on the cesium iodide spectrum which are sufficiently well defined to require identification are those at pulse heights 14.5V, 17V, 23 to 25V, and 30 to 40V. The assignment to cesium is on the basis of comparison of the sodium iodide spectrum in Figure 4. Transitions are identified by comparison with the known level structure (Figure 1) and published experiments (Table I).

The 14.5 volt peak corresponds to a gamma of 0.175 Mev. This is taken to be the 0.16 Mev ray reported by others (4, 6, 9, 10, 14) representing the 0.16 Mev level-ground state transition. Both coulomb excitation (6) and the present experiment show a prominent .16 Mev ray, as compared to the Ba^{133} studies, such as that by Crasemann et al.(4), where it was faint on conversion spectra, and was completely obscured on scintillation spectra. This indicates that the .16 Mev level is directly excited by neutrons rather than being fed by the .44 Mev level as in Ba^{133} decay.

The peak at 17.5 volts is attributed to iodine. It corresponds to a 0.2 Mev gamma ray reported by Wolf (15) and others (1, 5, 7, 8, 9, 11, 13). Wolf used a sodium iodide crystal and 2.5 Mev neutrons; hence a similar response would be expected here. Wolf confirmed this assignment by the use of a sodium scatterer which did not increase this peak. It is interesting to note that in the present experiment this peak was much more prominent on the sodium iodide spectrum. The Cs^+ ion is larger than the Na^+ ion, so that the cesium iodide crystal contains fewer iodine atoms (0.7 times as many). This would indicate a relative intensity of 0.7 for this ray in CsI as compared to NaI which obviously does not account for the observed difference. The responses of the two crystals to other rays in the experiment and to calibration sources do not lead one to expect any such difference in intensity for the 0.20 Mev iodine ray.

The peak at 23V is attributed to cesium since it does not occur on the NaI spectrum. It is taken to be the 0.44 to 0.16 Mev transition. The energy of 0.28 Mev obtained here is in excellent agreement with results obtained by Grasemann et al (4) who reported conversion electrons from this ray but did not observe the gamma ray, which is not listed in the summary by Strominger et al (14). Coulomb excitation studies by Fagg (6) with 4.5 Mev alpha particles do not reveal this transition. However, Fagg does not report the .44 Mev level at all. It seems reasonable that neutrons would excite this level more strongly as results of the present experiment indicate.

In the region from 23 to 25 volts pulse height there is the possibility of another gamma ray. To render this peak prominent, it is necessary to correct for the effect of nearby peaks. This is done by extrapolating the 17.5 volt peaks. Reflection of the upslope at about 17.5 volts gives a symmetrical peak. The trough is then extrapolated, and the difference between this and the experimental curve is plotted. This is shown in the insert in Figure 3. Since the prominence of the peak is strongly dependent upon the rather uncertain extrapolation of the trough, it can only be said that the presence of a ray here is possible. A ray of 0.30 Mev would be expected here from the known level structure, and from coulomb-excitation data by Fagg (6). The presence of this ray would imply the presence of a 0.38

Mev ray and vice versa, since the 0.30 ray is a transition from the 0.38 Mev level to the 0.08 Mev level. (Because of the background, nothing can be said about this level from this experiment.) Wherever the 0.38 Mev level has been found, the 0.30 Mev ray has also been present (3, 4, 5, 7, 8, 10).

At pulse heights from 30 to 40 volts there occurs a peak which is too broad to consist of a single gamma ray peak of any reasonable energy. While no spectra were taken above this point, preliminary checks adequately show that there is no photopeak at higher energy large enough to have a Compton edge of any size in the region of this peak. It is therefore assumed that this peak consists of several gamma-rays. Figure 6 shows this region of both the NaI and CsI curves. Rays reported in this region for iodine are .4 Mev (5, 7, 15) and .441 Mev (5), and for sodium a .440 Mev ray. The level structure of I^{127} is somewhat confused (5); the peak on the NaI curve is, however, safely attributed to the .4 Mev iodine and the .440 Mev sodium rays. Wolf (15), in an almost identical experiment, confirmed this assignment by using a NaI crystal with and without a sodium scatterer. Also, experiments with 0 - 2.7 Mev neutrons and an iodine scatterer (7, 11) fail to show the .446 iodine ray. Inspection of Figure 6 indicates a ray in CsI at an energy slightly below .4 Mev, since the CsI peak is displaced toward lower energy. This could well be the .38 Mev cesium ray. A small cesium ray at about .44 Mev is required to explain the

peak observed. The .44 Mev cesium level has not been observed to decay to the ground state directly, and on the basis of tentatively assigned spins (4, 8) would not be expected to do so. However, as will be explained later, a peak at this point due to cesium is possible.

It is of interest to compare the relative prominence of the rays here and in other experiments. Comparison of this experiment, and that of Fagg (6) with the Ba^{133} study by Grasemann et al.(4) shows that both alpha particles and neutrons excite the .16 Mev level directly. Also, the present experiment shows the .36 Mev transition (4) absent, and the .302 Mev transition, if present, much smaller than in other experiments.

Some of the differences may be explained by the detection of both rays of a cascade. Both the .44 Mev and .38 Mev levels cascade through the .08 Mev level. Since these rays originate in the crystal, both must pass through part of the crystal. Since the detection efficiency is high for the .08 Mev ray ($\sim$ 80 per cent), in many cases both rays may be detected simultaneously with the result that their pulses add. Such an effect will result in some of the .30 Mev peak being shifted to the .38 Mev peak. Likewise, part of the .36 Mev rays will appear in the .44 Mev peak. The .44 Mev cascade could account for the weak .44 Mev peak which is required to explain the feature in the 30-40 volt region in Figure 3. In any case, the .38 and .44 Mev levels do not appear to be

strongly excited by neutron bombardment, in contrast to the decay of Ba^{133} to the .44 and .38 Mev levels where the .36 Mev ray is prominent (4), and coulomb excitation (6), which shows a more prominent .30 Mev ray.

Since some of the above conclusions are somewhat tentative, it would be of interest to make future studies using coincidence techniques and a cesium scatterer. It would be necessary to reduce the low-energy background since the .08 Mev ray which would be needed for gating is well obscured at present. It would also be well, if possible, to conduct an investigation at higher energies to observe the rays reported by Young (16).

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TABLE I

GAMMA-RAYS FROM Cs^{133}

Energy (Mev)	Detection Method	Excitation Method	Coincident With	Transition* With Spins If Given	Reference**
0.082	Scint.Spec.	Coulomb	.082	.082-gnd.,.16-.082	Fagg (4)
.163	Scint.Spec.	4.5 Mev	None	.163-gnd.	
.385	Scint.Spec.	. . .	. . .	.385-gnd.	
.302	Scint.Spec.	. . .	.082	.385-gnd.	
.053	Scint.Spec.	Ba^{133} Decay	. . .	.434(1/2)-.381(1/2)	Grasemann et al (2)
.079	Scint.Spec. and Conv.Elec.	. . .	.079,.302	.079(5/2)-gnd(7/2), .158(7/2,9/2,or 11/2)-.079	
.302	Conv.Elec.	. . .	. . .	.381-.079	
.355	. . .	. . .	Not .302	.434-.079	

*Levels as reported, not as in Figure 1.

**See Chapter IV Bibliography for references.

TABLE I--Continued

Energy (Mev)	Detection Method	Excitation Method	Coincident With	Transition With Spins If Given	Reference
.158	Conv.Elec.	. . .	. . .	.158-gnd.	
.276	Conv.Elec.	. . .	. . .	.434-.158***	
0.057	Scint.Spec.	Ba^{133} Decay	Not .357	.439(1/2)-.382(3/2)	Hayward et al (6)
.082	Scint.Spec	. . .	.082,.057, .300	.082(5/2)-gnd.(7/2)	
.300	Scint.Spec.	. . .	Not .357	.382-.082	
.357	Scint.Spec.	. . .	Not .300	.439-.158	
.276	Scint.Spec.	Ba^{133} Decay	. . .	Ba^{133} metastable to Ba^{133} gnd.	Hill and Metzger (7)
.56 .75	Scint. Spec.	14 Mev Neutron Inelastic	. . .	. . . . .	Young (16)

***No Ba^{133} metastable present.

TABLE II

IDENTIFICATION OF COMPONENTS IN FIGURE 2

C	Crystal	Harshaw 6D4 (Gamma) National Radiac MFS-2 (monitor)
PM	Photomultiplier	DuMont 6292
PA	Preamplifier	NTSC Built
PPS	Photomultiplier Power Supply*	NTSC Built, Similar to Atomic Instruments Mod. 312
LA	Linear Amplifier	Atomic Instruments Mod. 218
PHA	Pulse Height Analyzer	Atomic Instruments Mod. 510
S	Scaler	
A	Automatic Switch	
R	Register	

*Monitor PPS is Atomic Instruments Mod. 318.

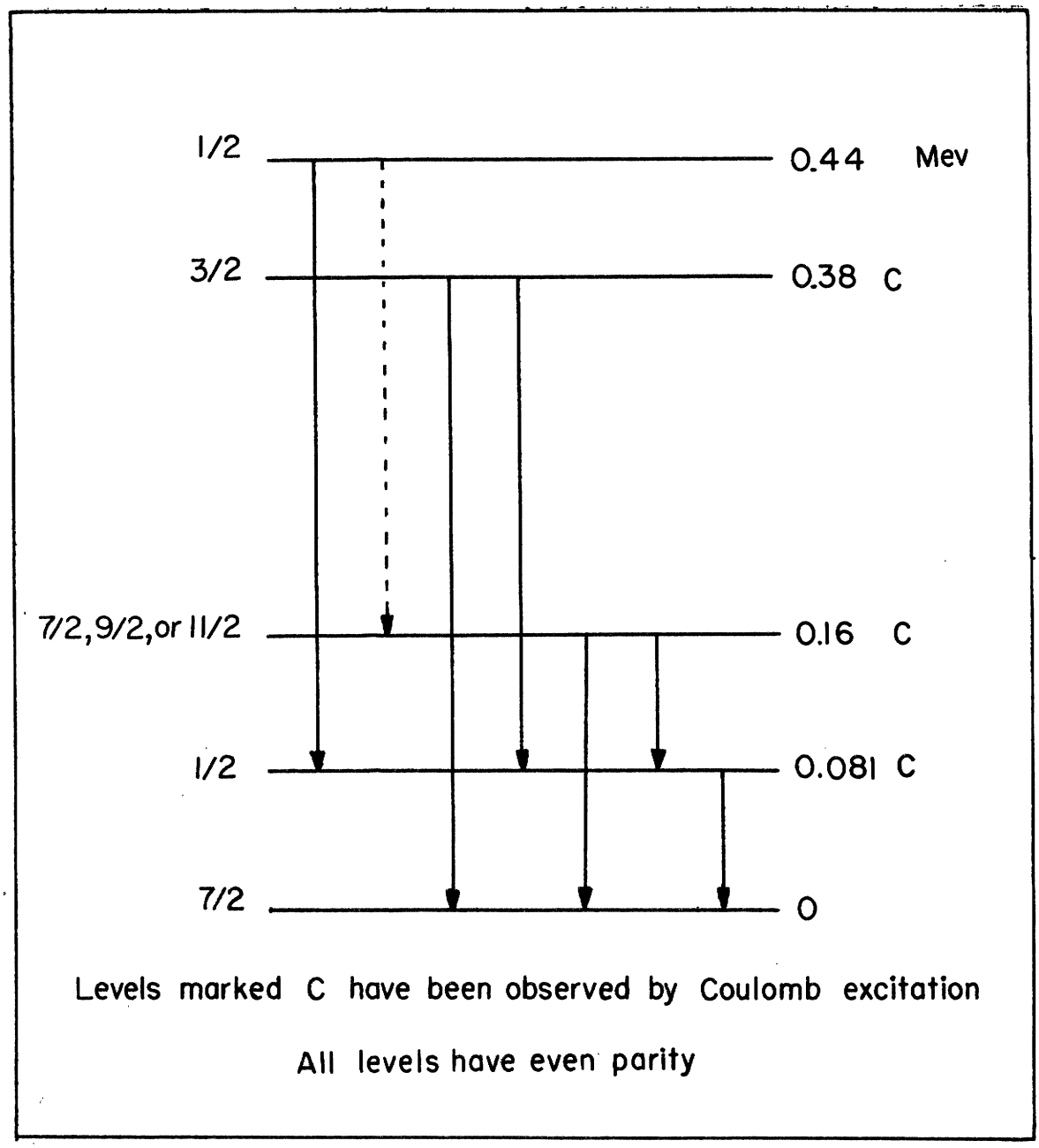

Fig. 1 Energy levels in Cs^{133}

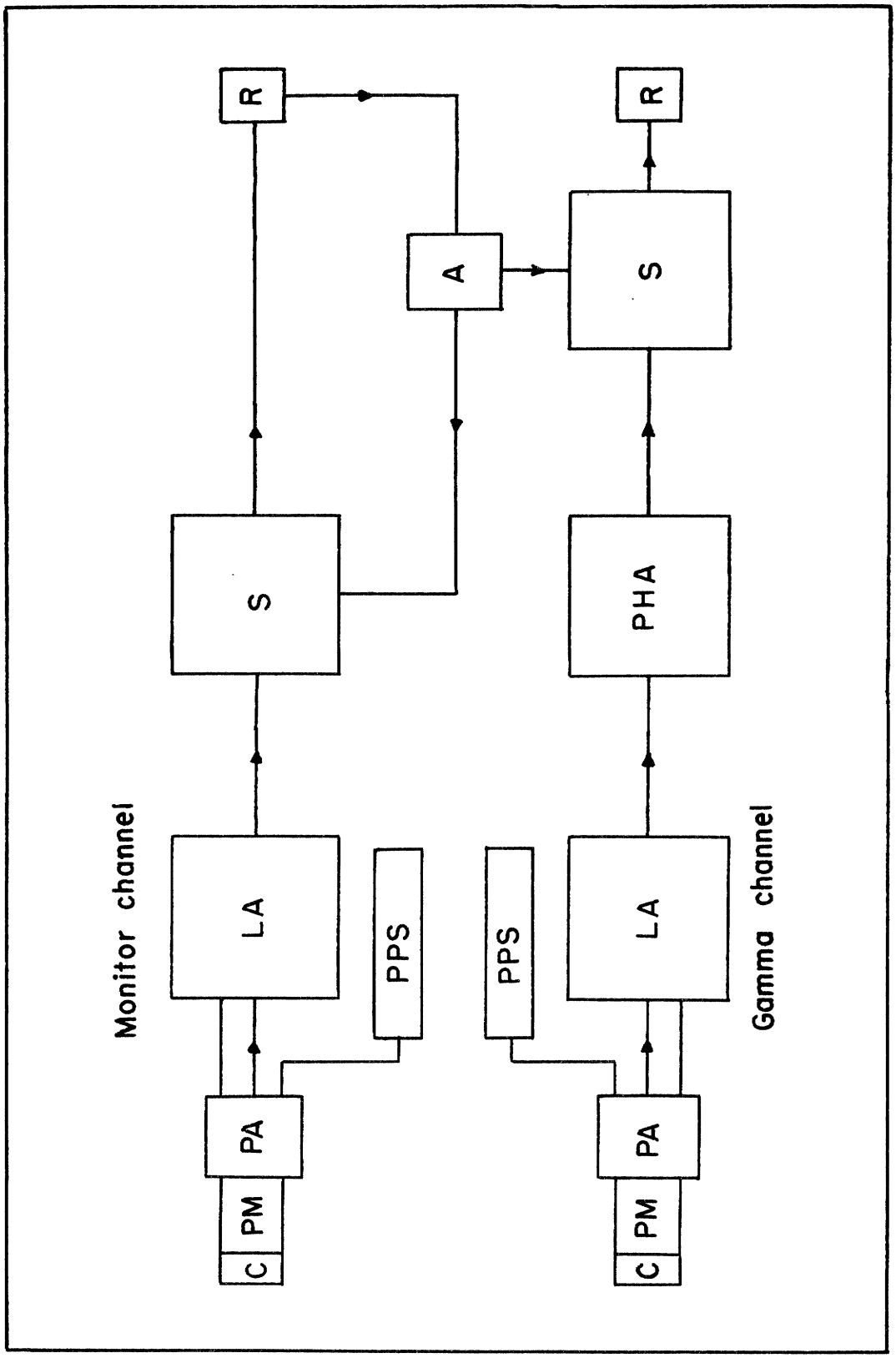

Fig. 2. Detector instrumentation

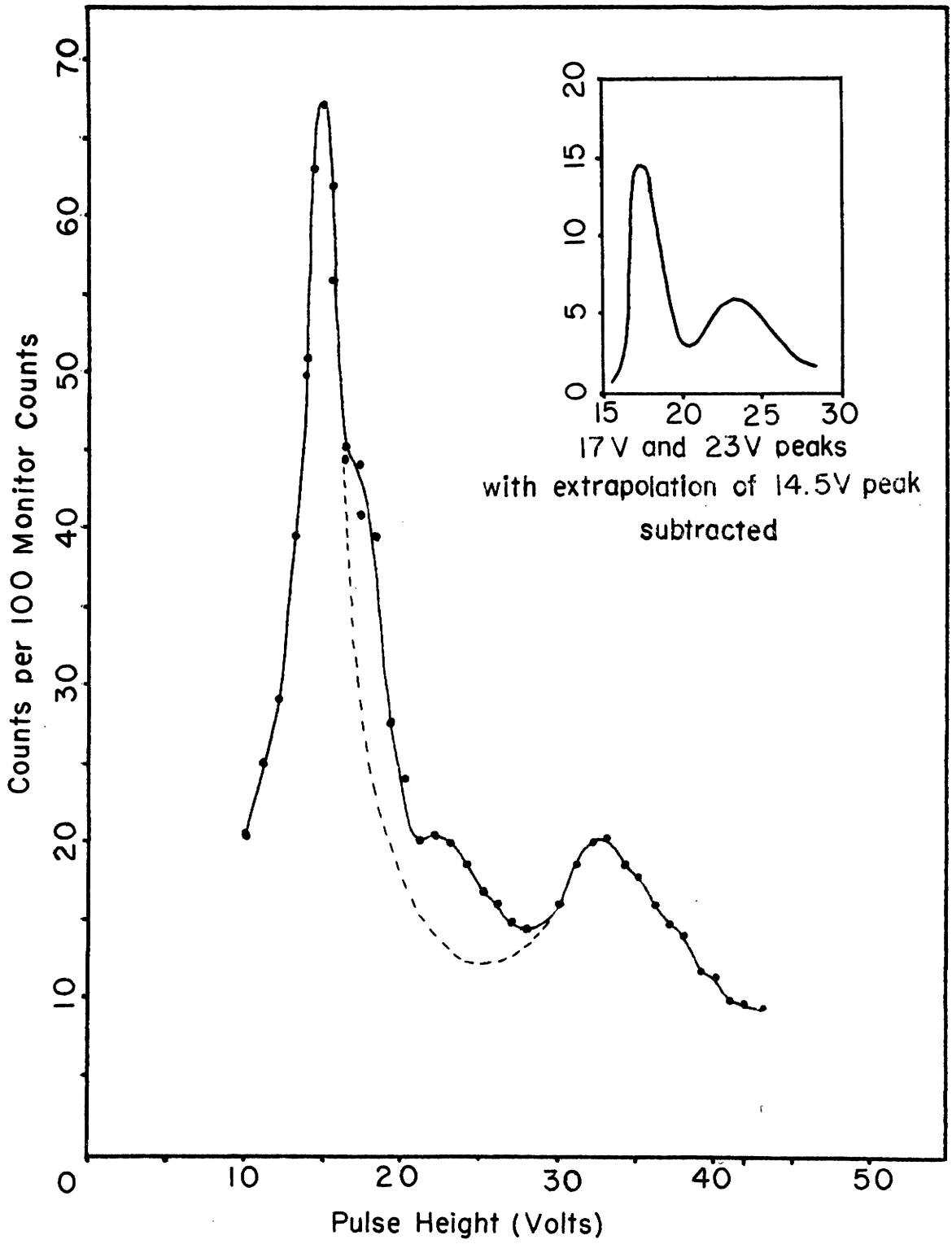

Fig. 3 Typical CsI spectrum

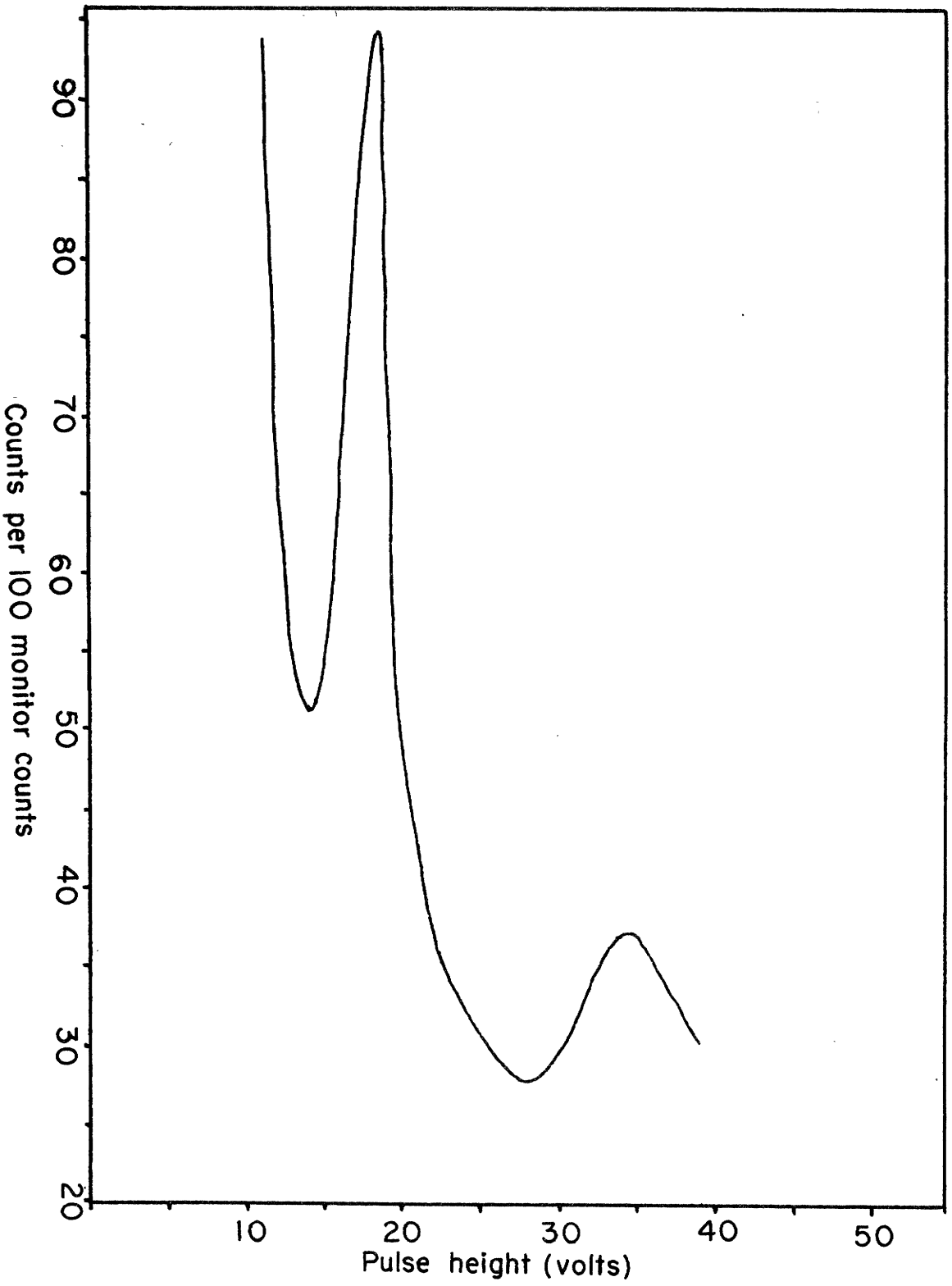

Fig. 4 Typical NaI spectrum

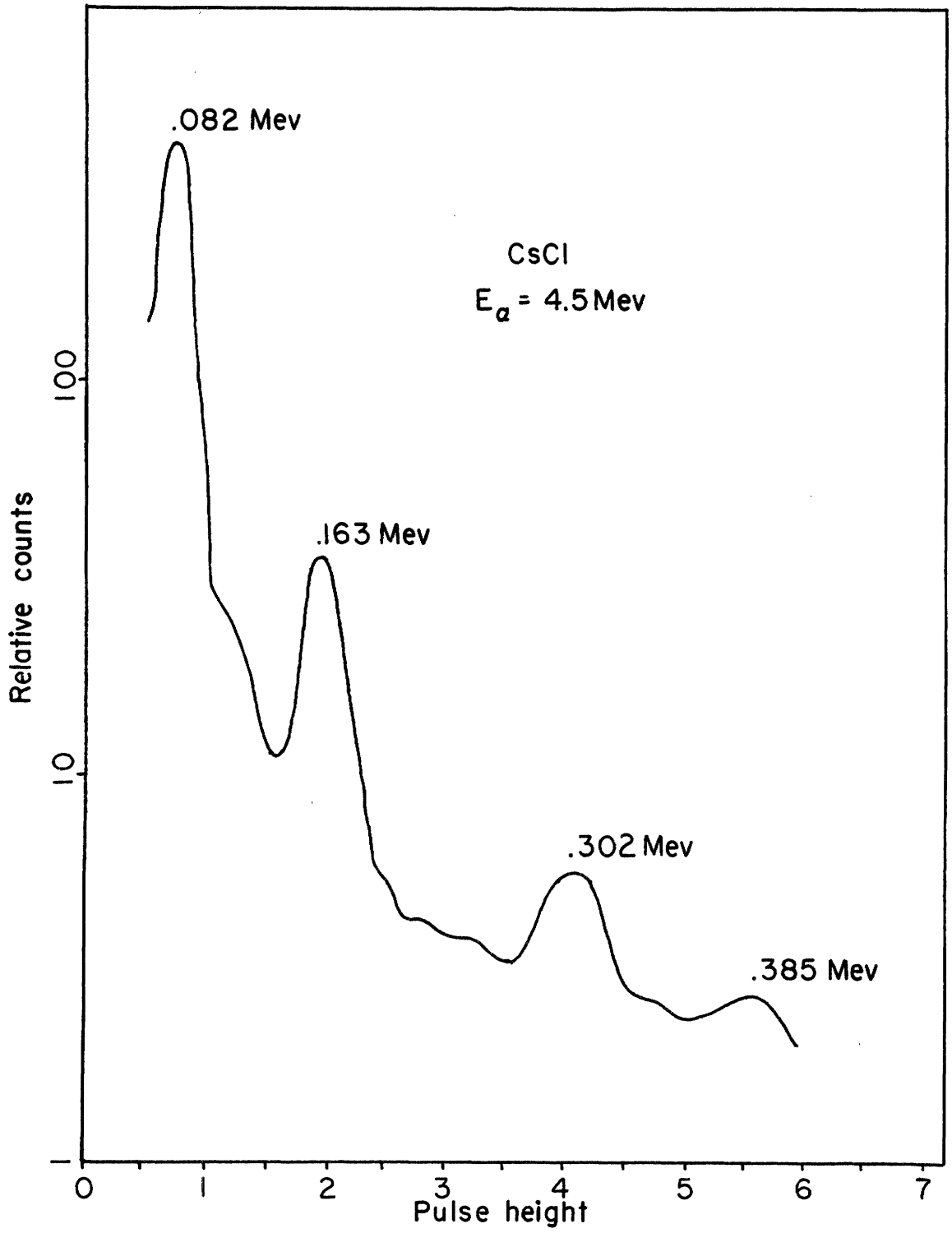

Fig. 5. Coulomb excitation spectrum by Fagg (6)

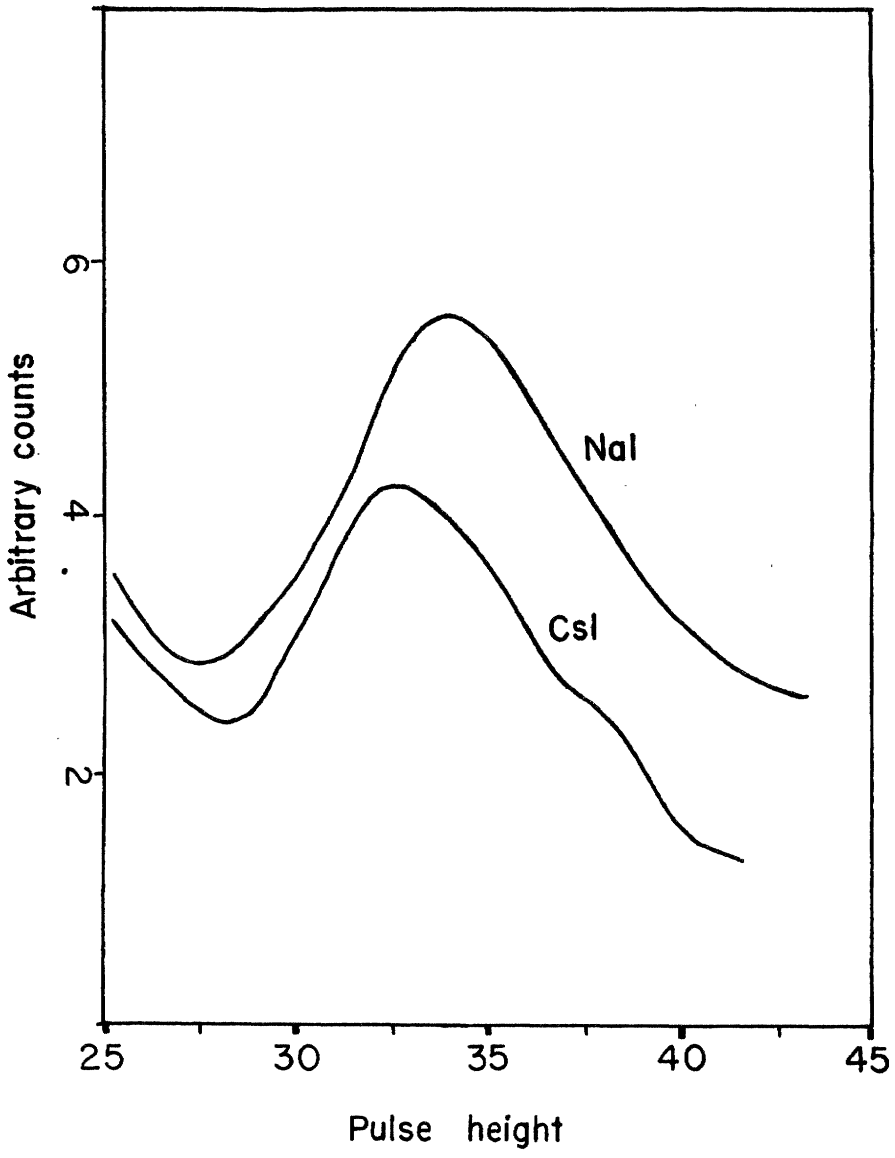

Fig. 6. Enlargement of
fig.3. and fig. 4.

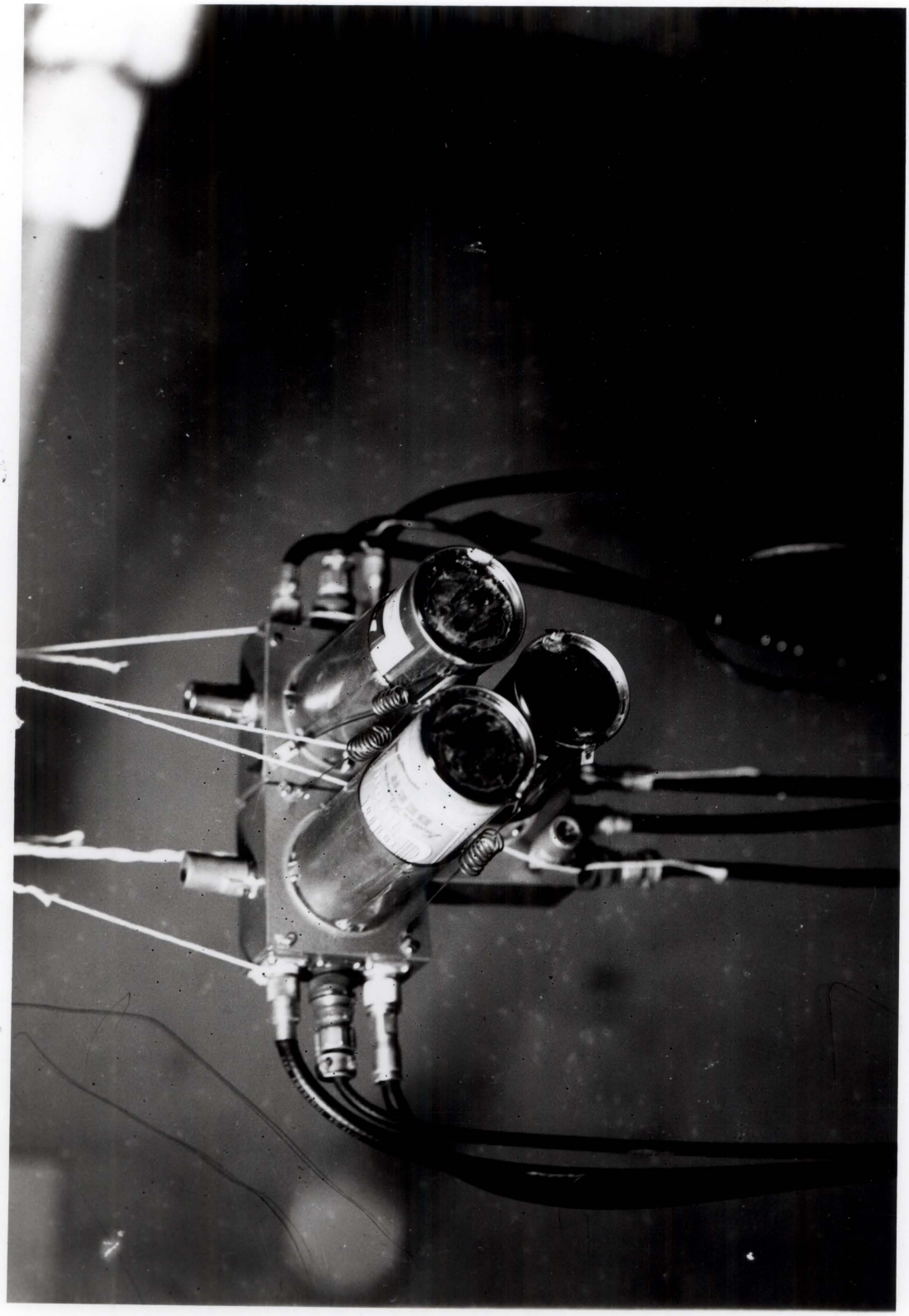

Fig. 7--Photograph of Detector Head Assembly

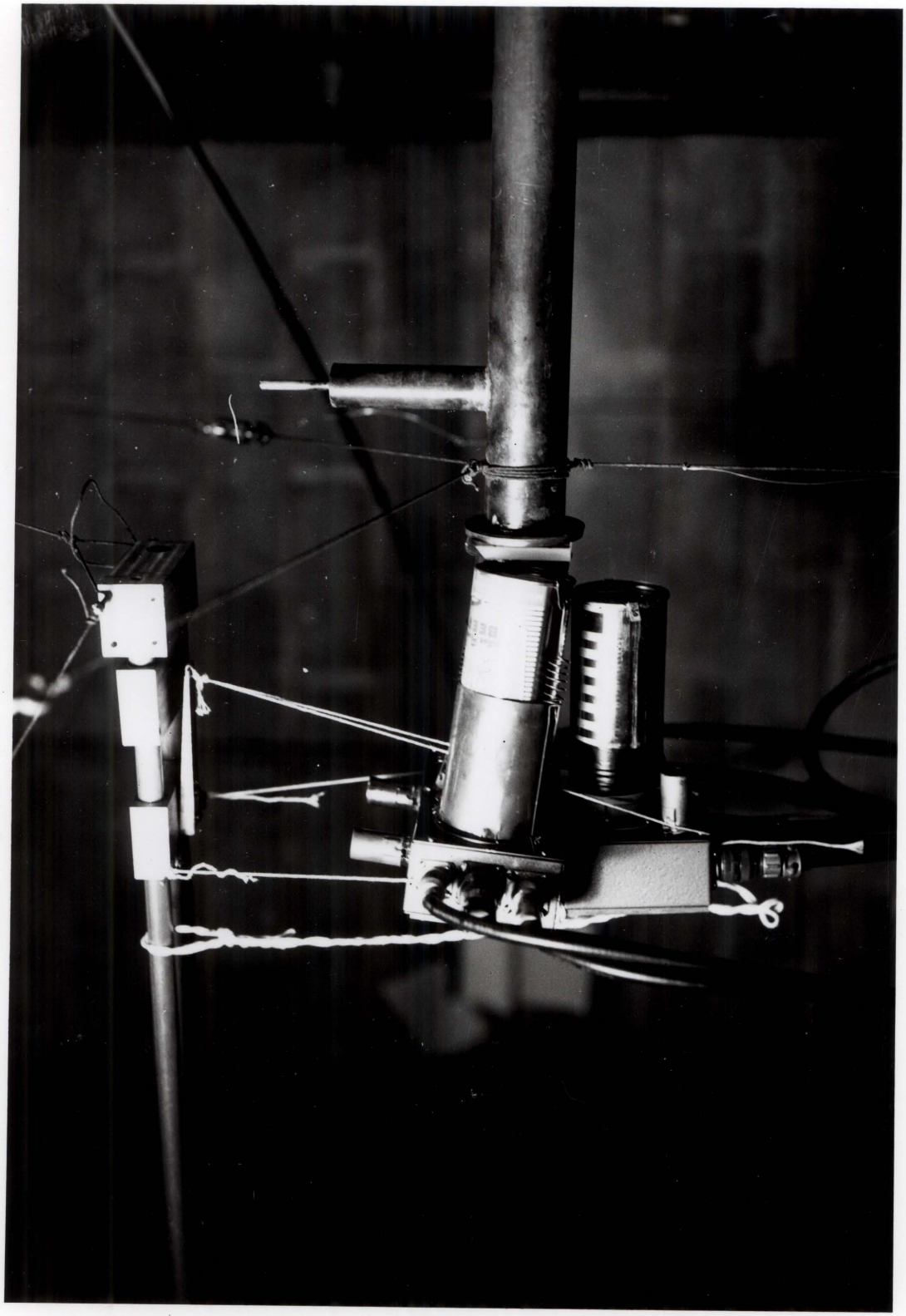

Fig. 8--Detector Head in Place

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