UCRL-11739

University of California

Ernest O. Lawrence Radiation Laboratory

GAMMA DEEXCITATION OF FISSION FRAGMENTS.
I. PROMPT RADIATION

TWO-WEEK LOAN COPY

This is a Library Circulating Copy which may be borrowed for two weeks. For a personal retention copy, call Tech. Info. Division, Ext. 5545

Berkeley, California

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

To be published in Nuclear Physics

UCRL-11739

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

GAMMA DEEXCITATION OF FISSION FRAGMENTS. I. PROMPT RADIATION

Sven A. E. Johansson

October 1964

Gamma deexcitation of fission fragments. I. Prompt radiation.

Sven A.E. Johansson*

Lawrence Radiation Laboratory

University of California

Berkeley, California.

Abstract.

The gamma radiation emitted in fission of Cf^{252} has been investigated in coincidence with the fission fragments. The mass ratio of the fragments was recorded, various properties of the radiation being studied as a function of the mass ratio. This paper deals with the prompt radiation, i.e. radiation with a half-life shorter than 10^{-9} sec.

The life-time of the gamma-emitting states was measured by time-of-flight technique. Using a collimator, the gamma radiation was detected after the fragments had travelled different distances. In this way, the decay curve was investigated and the half-life of the radiation was found to be about $1 \cdot 10^{-11}$ sec. Using this information, it was possible to find a collimator setting which permitted a complete separation of the radiation from the two fragments. The properties of the radiation could then be studied as a function of mass. This arrangement was used to record gamma spectra for a number of different masses. It was found that there is a pronounced variation in spectrum shape. The spectra from fragments close to magic numbers are shifted to higher energies. With the same arrangement the yield of gamma rays was determined as a function of mass. The yield curve has a saw-tooth shape.

The significance of the experimental results is discussed. The characteristic feature of the deexcitation process seems to be that it involves high spin states. It is estimated that the average value of the initial spin is about 10. The results of this work indicate that the radiation mainly consists of vibrational transitions. This fact is explained as a consequence of the conditions at scission.

---

* On leave of absence from and now at the Department of Physics, University of Lund, Lund, Sweden.

I. Introduction.

The gamma radiation emitted in the deexcitation of fission fragments is of considerable interest, since it exhibits properties quite different from those found in most other nuclear reactions. The study of fission gamma radiation is beset with some experimental difficulties and the available information is therefore limited. The shape of the spectrum as well as the multiplicity are known.[1-6] The life time of the low energy part has been measured.[5,7] Finally, it has been found that the radiation is not isotropic but is preferentially emitted in the forward and backward directions relative to the motion of the fragment.[8-10]

What is known about fission gamma radiation suggests that the deexcitation process is of a peculiar type. The close analogy with the situation in heavy ion induced reactions makes it very probable that the main characteristics of the process is that it involves high spin states. A study of fission gamma radiation therefore should give information about states which normally are not populated in radicactive decay or nuclear reactions. A limitation is that the radiation is a mixture of gamma rays emitted from a great number of fragments with different nucleon numbers. However, this limitation could be an advantage, if it were possible to separate the radiation according to fragment mass. This can be performed by detecting the radiation in coincidence with fragment pairs with selected mass ratios. The fission fragments cover a large part of the nuclear periodic table. This means that in one single experiment, and under the same conditions, it might be possible to make a systematic study of the deexcitation process for a great number of nuclei of different mass and type, e.g. magic nuclei and deformed nuclei. The aim of the present work was to see to what extent such a program can be carried through. As will be shown below, the results are promising and a systematic study of the fission gamma radiation might open up a new field of nuclear spectroscopy.

During the course of the present investigation, it was found that the radiation could be divided into two classes with greatly different half-lives. The main part of the radiation has a half-life of about 10^{-11} sec. In addition, there is a part with much longer half-life, of about $2 \cdot 10^{-8}$ sec.

A long-lived component has also been reported by Maienschein et al[1] but with a half-life considerably longer than the one found here. This discrepancy will be discussed further in the second part of this paper.

It was found convenient to divide the account of the present investigation into two parts, one dealing with the short-lived component (termed the prompt radiation) and a second one dealing with the long-lived component (termed the delayed radiation). This division has some advantages from a practical point of view, since the experimental technique is different in the two cases. The division has also a deeper significance. The character of the transitions is distinctly different for the prompt and the delayed radiation.

In the first part of the paper, which is presented here, we will discuss the prompt radiation.

II. Experimental procedure.

A. Apparatus.

The experimental arrangement is shown schematically in Fig. 1. A Cf^{252} source, of strength $4 \cdot 10^5$ fissions per minute, was deposited by self-transfer on to a thin nickel foil. The fission fragments were detected by two solid state counters of the surface barrier type placed symmetrically around the source. The source and the counters were enclosed in an evacuated aluminum chamber, which had thin walls in order to minimize the gamma-ray scattering.

The gamma radiation was, in most cases, detected by a 3" x 3" sodium iodide crystal. Sometimes it was necessary to suppress the neutron background. Then a thin crystal, 1" x 1/8", was used. In some of the measurements, it was necessary to select the gamma radiation from only one of the fragments. This was done using a lead collimator, schematically indicated in the figure.

The mass ratio of the fission fragments was determined from the kinetic energies measured by the solid state counters. It is easy to show that the mass ratio is inversely proportional to the ratio of the kinetic energies. Hence the mass ratio can be obtained by dividing the two pulses from the fission counters. This division was performed in a special electronic circuit. Each of the two pulses from the counters charges a condenser, that is discharged through a resistor. The time constant is the same in both

channels. When the voltage over one of the condensers has dropped to one volt, the voltage over the other condenser appears at the output of the divider. It is easily shown that this output voltage is proportional to the ratio between the input pulses. The output pulse goes to a pulse height analyzer, which selects pulses of a certain height, corresponding to a certain mass ratio. The gamma-ray pulses are fed to a stretcher. The output gate of the stretcher is operated by the pulses from the single channel analyzer, so that only gamma-ray pulses, which are in coincidence with a certain mass-ratio, are recorded by the multichannel analyzer.

A simplified block diagram of the electronics is shown in Fig. 2. The coincidence system is of the fast-slow type. The resolving time of the system was 5 nsec. The number of accidental coincidences was very low and could practically always be neglected.

B. Performance.

The pulse spectrum from the fission counters is shown in Fig. 3a. It will be noted that its shape agrees well with the established kinetic energy distribution of the fission fragments. During the course of the experiment, it became evident that the pulse spectrum was not quite constant. After some time, the valley began to be filled in and the spectrum changed in shape. The cause of this change was found to be a considerable reduction of the resolution because of radiation damage. During the experiment the detectors were continously bombarded with fission fragments at a rate of 10.000 per min. It is natural that this leads to severe damage of the crystal material. This difficulty was overcome by frequent changes of detectors and constant monitoring of the pulse spectrum.

The mass ratio spectrum obtained from the divider is displayed in Fig. 3b. It agrees reasonably well with other determinations of the mass ratio in fission of Cf^{252}. This agreement is an indication that the equipment worked properly.

C. Resolution.

It is of some importance to discuss the resolution of the mass ratio measurements. Ideally one would like to be able to select a very narrow mass-ratio interval. Accepting gamma radiation from only one of the frag-

ments, a well-defined mass ratio should enable the study of radiation from fragments of a unique mass. This is, however, impossible for several reasons.

One fundamental difficulty is the dispersion introduced by neutron emission. In determining the mass ratio from the fragment energies, the observed kinetic energies are employed. These differ from the initial energies by the amount carried away by the neutrons. The use of observed energies in the expression for the mass ratio therefore introduces a mass dispersion. This problem has been treated in detail by Terrell.[11]

Another factor influencing the resolution is the mass defect of the solid state counters, which introduces some uncertainty in the energy measurements.

The radiation damage discussed above might also have some influence on the resolution. Frequent replacements of the counters should eliminate this effect, but a small variation of the resolution cannot be excluded.

These various effects have not been studied in detail and no attempt has been made to correct the measurements for the mass dispersion. The reason why it was not necessary to tackle this difficult problem is that the effects studied in the present work do not vary rapidly with mass. As will be shown below, most of the gamma radiation seems to be of collective nature, and it is well known that the parameters of the collective motion vary only slowly with mass.

In this connection it should be realized that, even if one could select a single mass for study, this would not be the same as selecting a single nucleide. The reason is, of course, charge dispersion. The charge distribution is so wide that 2-3 isobars have yields of the same order of magnitude.

D. Corrections.

The main correction in the present measurements was the subtraction of the neutron background. The sodium iodide crystal is an efficient detector for fast neutrons. The predominant reaction is inelastic scattering in iodide. It was found that the relative intensity of the neutron back-

ground varied with energy, but that it was, on average, about one third of the gamma ray intensity.

A helpful circumstance is that the spectrum of the neutron background shows a pronounced structure. It has peaks at 210, 435 and 632 kev, corresponding to transitions in I^{127}. This characteristic shape makes it easy to identify the background, and to check that there is no under- or over-correction of the spectrum.

The determination of the neutron background was made by absorbing the gamma radiation in a lead absorber. The fission gamma rays have a relatively low mean energy and furthermore, the low-energy part of the spectrum was the most thoroughly investigated in the present work. Therefore, in most cases a rather thin lead absorber (less than 1 cm) was sufficient to absorb the gamma radiation. Such an amount of lead will not affect the neutrons to any great extent. In some cases, the measurements required the use of a thicker lead absorber, and then it was necessary to know how it attenuated the neutron flux. This was determined either experimentally, by studying the neutron intensity as a function of absorber thickness and then extrapolating to zero thickness, or theoretically from cross-section data. Satisfactory agreement between the two methods was obtained.

One possibility of eliminating the background is to discriminate between the neutrons and the gamma radiation by time-of-flight technique. Some preliminary measurements along this line were performed with the present apparatus, but the distance between the source and the sodium iodide crystal was too small to give any significant improvement. It was not possible to increase this distance because of intensity considerations. However, a stronger source, a bigger crystal and a shorter resolving time (at the expense of the efficiency at low energies) should enable the use of the time-of-flight technique. This would be of advantage in some of the measurements, but it is no crucial problem, since the subtraction method used here worked satisfactorily.

The rate of accidental coincidences was very low compared to the total counting rate. It could therefore be neglected in most measurements. Only when the decay curve was followed to very low intensities was it necessary to correct for the accidental coincidences. The correction was determined in the usual way by introducing a suitable delay in the gamma-ray channel.

III. Results.

A. Time distribution.

Very little is known about the life-time of the gamma-emitting levels in fission fragments. Skliarevskii et al[5] have estimated the life-time, using a crude time-of-flight method. For radiation below 250 kev they found a half-life between $0.5 \cdot 10^{-9}$ and $2.5 \cdot 10^{-9}$ sec. This assumes, however, a simple exponential decay, but it might quite well be more complex. The life-time of the low-energy radiation, in this case between 25 kev and 100 kev, was also investigated by Dési et al[7] using the method of delayed coincidences. The half-life was found to decrease with increasing energy; the measured values range from 10^{-9} to 10^{-10} sec.

Hence nothing has been reported about the life-time of the bulk of the gamma radiation. One can expect that it is shorter than for the low-energy part of the spectrum, probably below 10^{-10} sec.

For the short time intervals involved here, the best method is the time-of-flight technique. A flight path of 1 cm for a fission fragment corresponds to about 10^{-9} sec. and, if collimating systems with a definition of 0.1 mm can be used, one should be able to measure decay-times of the order of 10^{-11} sec. This is considerably better than what can be achieved by electronic methods. A further advantage is that the gamma radiation can be recorded by a sodium iodide crystal, which makes it possible to obtain good energy spectra.

The experimental arrangement is shown schematically in Fig. 1. The collimator, with the sodium iodide spectrometer attached to it, can be moved along the direction of flight of the fragments. In the first experiment, the collimator had a thickness of 5 cm and the slit was 3 mm wide. The counting rate as a function of the collimator position is shown i Fig. 4 (circles). The neutron background has been subtracted. The curve shows clearly that the life-time of the gamma radiation is very short. The width and shape correspond closely to the geometrical dimensions of the collimator slit. The tail of the curve can largely be accounted for by edge penetration and scattering. To see if any finite decay time could be detected with this equipment, the following method

was used. A thin aluminium foil, which stopped the fission fragments, was mounted 0.5 mm from the source foil. The distribution shown in Fig. 4 as triangles was obtained. There is indeed a change of the type that is expected if the gamma radiation has a measurable life-time. With the collimator slit placed right above the source, the counting rate increases, since the gamma radiation previosly emitted outside the view-field of the collimator is now emitted in the catcher foil. With the collimator slit to the side of the source, the catcher foil prevents the fragments from reaching this region thereby decreasing the counting rate. It is obvious that this arrangement is too crude to give any details of the decay curve. Some important information can be obtained, however. The difference in counting rate with the slit right above the source shows that 13% of the gamma radiation is emitted after the fragments have travelled 1.5 mm. With the assumption of an exponential decay with only one component this corresponds to a half-life of $4 \cdot 10^{-11}$ sec. Obviously the decay might be more complex. A fast decay followed by a long-lived component with an intensity of 13% would give the same experimental results.

To obtain an improved decay curve a better collimator was used. It had a thickness of 7 cm and a slit width of 0.8 mm. Here the neutron background presents a serious problem. Since the slit is so narrow, only a very small fraction of the gamma radiation reaches the detector, but some of the neutrons can pass throught the lead and therefore give a high background counting rate. The background is roughly proportional to the volume of the sodium iodide crystal and it could be minimized by choosing the crystal as thin as possible. With a crystal thickness of 3 mm, the bakcground was 40% of the total counting rate with the slit placed above the source.

The interpretation of the time distribution requires knowledge of the distribution for a point-source. This distribution can be calculated from the geometry of the collimator, but an experimental determination is safer. To do this, the fission source was replaced with a similar source of Hg^{203}. The gamma radiation of Hg^{203} has an energy of 275 kev, which is close to the effective mean energy of the fission gamma radiation. The distribution obtained for Hg^{203} is shown in Fig. 5 a. The points are the measured values, the line is the calculated distribution. The good

agreement shows that the collimator is working properly. The distribution obtained with a fission source is shown in Fig. 5 b. A comparison of the distributions in Fig. 5 clearly shows the effect of the life-time of the gamma radiation.

It is clear, however, that the life-time is so short, compared to the time definition of the collimator, that it is impossible to see any details in the time distribution. This has the effect that, to correct for the width of the slit, one has to assume a certain functional form for the decay curve. One can, for example, assume a simple exponential decay. After correction for the width of the collimator slit, the decay curve in Fig. 6 is obtained. The decay seems to be a pure exponential with a half-life of $2.3 \cdot 10^{-11}$ sec. The accuracy of this value is about ± 50%. The fact that the decay mainly consists of a single exponential component is noteworthy. With a multiplicity of about ten, one would expect a rather wide spread in life-time of the gamma-rays and consequently a complex decay curve. Great caution is necessary in this connection, however. In correcting for the resolution of the collimator, it was assumed that the decay was a simple exponential. The correction tends to force the curve to assume this shape. However, this effect cannot produce a simple curve if the decay consists of components which differ widely in life-time, and we can therefore conclude that the gamma-emitting states involved here have half-lifes of the same order of magnitude.

In the decay curve of Fig. 6, there is also some sign of a slower component, but the accuracy is not high enough to resolve it. As mentioned above, half-lives between 10^{-9} and 10^{-10} sec. have been measured for the low energy part of the gamma spectrum. The slow component seen here might have a half-life in this range. In the measurements with the wide collimator, it was found that 13% of the radiation was emitted after $1.25 \cdot 10^{-10}$ sec. The total intensity of the slow component depends on the half-life, which cannot be determined very well. A rough estimate based on Fig. 6 gives a value of 20%.

A more realistic analysis of the decay curve has to take into account the multiplicity of the radiation. On the average 4-5 gamma rays are emitted from each fragment. Such a cascade cannot give a simple exponential decay unless the first member of the cascade is considerably slower that the other ones. This is a very unlikely situation. If there is a considerable

difference in life-time, the slowest decays should come late in the cascade. However, the shape of the decay curve shows that it is not composed of several components with greatly different half-lives. We have therefore analyzed the decay curve assuming that the decay consists of a cascade of four gamma-rays with the same half-life. After a correction for the collimator resolution the decay curve in Fig. 7 is obtained. Here, also, we note a fairly good fit to the experimental points and, again, the presence of a long-lived component. If one instead assumes a multiplicity of 3 or 5 the results are similar. The half-life of the gamma-emitting states is then found to be $1.3 \cdot 10^{-11}$ sec., and this value does not depend very much on the multiplicity.

Evidently one can obtain a good fit assuming either a simple decay or a cascade of gamma rays. This illustrates the fact that the definition of the collimator is not good enough to see the details of the decay curve. Unfortunately, for intensity reasons, it is very difficult to improve on this point.

The two cases treated here are, of course, only two of the many possible combinations in a cascade of gamma-rays. However, they serve the purpose of demonstrating that the life-time of the states involved does not depend too much on the assumptions of the analysis.

Another difficulty connected with lack of sufficient time resolution is that one cannot exclude the existence of a very fast component in the decay curve. One can only set an upper limit, which is determined by the statistical errors of the measured points. This limit depends on the shape of the main part of the decay curve. If there is a simple exponential decay, the upper limit for the fast component is 15%, but if there is a cascade of gamma-rays with equal life-times, the limit increases to 25%.

The results of this analysis can be summarized as follows. The gamma radiation, which corresponds to the main part of the decay curve, has a half-life of about $1 \cdot 10^{-11}$ sec., the exact value depending somewhat on the multiplicity. This component comprises 60-70% of the prompt gamma radiation. In addition there is a slow component comprising about 15% with a half-life in the region 10^{-9} - 10^{-10} sec. Finally, there might be a fast component with a half-life considerable shorter than 10^{-11} sec. The upper limit for this component is 15 - 25%.

It is, of course, self-evident that this analysis does not imply that one can divide the decay curve in well-defined components with certain half-lives. There is probably a smooth transition between the components and within each component there is a certain spread in life-time. The purpose of this analysis has only been to emphasize that the decay curve is not as smooth as one would expect from a statistical distribution of life-times, but shows a certain amount of structure.

B. Energy spectra.

An inspection of the decay curve in Fig. 5 shows that it should be possible to obtain a complete separation of the gamma radiation from the two fragments. A choice of 0.5 mm for the collimator setting gives complete separation, while keeping the contribution of the slow component to a minimum. This arrangement was used to record the energy of the gamma radiation as a function of mass. The output of the divider was displayed on one dimension of a two-dimensional pulse-height analyzer and the spectrum of the scintillation spectrometer on the other dimension.

Fig. 8 shows the spectra for a number of fragment masses. Unfortunately one cannot expect to get high quality spectra with the present arrangement. As discussed above, it is necessary to use a thin sodium iodide crystal in order to suppress the neutron background. With such a crystal the correction for the response of the spectrometer is rather uncertain. Furthermore, the thin slit will introduce a considerable scattering, which distorts the spectra. The neutron background is not subtracted in Fig. 8. It is mainly concentrated at low energies, and there the scattering gives such a large uncertainty that it did not seem worthwhile to make a separate determination of the background.

It is also important to realize that the collimator position has a great influence on the spectrum shape. With the setting used here, the collimator accepts gamma radiation emitted in the time interval $(1\text{-}7) \cdot 10^{-11}$ sec. If the half-life of the transitions in the cascade is about 10^{-11} sec., most of the radiation is emitted within this interval. However, if the transitions are considerably faster or slower, only part of the radiations is accepted by the collimator. Hence one would expect that in the spectra those parts are enhanced which

correspond to transitions having a half-life of 10^{-11} sec. The bump at 700 kev can probably be accounted for in this way.

Because of these uncertainties, not too much attention should be paid to the general shape of the spectra. It is mainly variations with mass that are significant, and they will be discussed below.

C. Gamma-ray yield.

The same experimental arrangement, which was described above in connection with the energy spectra, can be used for determining the yield as a function of mass. By setting the collimator in the positions + 0.5 and - 0.5 mm one can select the radiation from the heavy and light fragment, respectively. The mass ratio distribution from the divider was recorded on a pulse height analyzer for the coincident events. Dividing this distribution, after correction for the neutron background, with the normal mass ratio distribution recorded for all fission events, gives the relative gamma-ray yield per fragment.

The difficulty with this experiment is that the effect found is rather small. Furthermore, the variations in yield are such that the changes they produce are the same as a shift of the curve caused by a variation in amplifier gain. Hence it was necessary to have the electronics very stable and to make frequent checks. The experiment was performed so that the collimator was repeatedly alternated between the two positions. A change in collimator position always produced a distinct change in the mass ratio curve. Finally, all the curves were added separately for the two positions.

The neutron background was determined as described above by blocking the collimator slit with a piece of lead. This was done for the two collimator settings. One should expect no difference in the mass ratio curve for the two settings. Since the slit is so narrow, most of neutrons giving the background in the sodium iodide crystal penetrate through the lead and the position of the slit is therefore of little importance as far as the background is concerned. This expectation was borne out by the experiment; the neutron background turned out to be practically independent of collimator position. This fact is a strong indication that the changes observed

with the slit open indeed can be attributed to a variation of gamma-ray yield as a function of mass.

The final result is shown in Fig. 9. The yield curve has a saw-tooth form. It resembles the neutron yield curve. The significance of this similarity will be discussed below.

It is worth mentioning that the yield determination here refers not to the total gamma radiation but to the part selected by the particular collimator setting used here. It corresponds to radiation emitted in the time interval $(1\text{-}7) \cdot 10^{-11}$ sec. Hence it should be representative for the main component of the decay curve. However, since this component comprises the major part of radiation, the yield curve obtained here should be typical also for the total radiation.

IV. Interpretation of the data.

A. Life-time and energy of the radiation.

The results of the present investigation show that the gamma deexcitation proceeds in an ordered fashion. Both the decay curve and the energy spectra are not such as one would expect if the deexcitation in the normal way took place via statistically distributed levels. Such normal cases are, for example, the gamma emission in (p,γ) and (n,γ) reactions. There, the energy spectra peak at 2-3 Mev. The different situation in fission might be explained by the assumption that the fission fragments are formed with a high spin. The neutron emission takes away most of the excitation energy but little angular momentum. Hence, at the beginning of the gamma-ray cascade, the nucleus is left with relatively little energy but with a high spin, which during the cascade must decrease to the ground state spin. This situation naturally has a profound influence on the deexcitation path. However, one cannot from these general ideas draw any conclusions about the detailed properties of the radiation.

The fact that the deexcitation seems to be more ordered than expected for single-particle transitions between statistically distributed levels suggests that collective transitions may play an important rôle. Rotational transitions have such a low energy that they cannot account for any greater part of the spectrum. Furthermore, most of the fragments

have a spherical equilibrium shape and hence have no rotational transitions. However, vibrational transitions might well be of importance. We will now proceed to show that the various properties of the gamma radiation investigated here agree with what can be expected for vibrational transitions.

Practically all information about vibrational transitions comes from studies of the transition from the first 2+ state in even-even spherical nuclei. Since most of the fission fragments are spherical, we will at first limit the discussion to spherical nuclei. Fig. 10 shows a plot of the half-life of the 2+ transitions against the energy for those cases where both quantities are known, the data being limited to regions where the fission fragments occur. The main component of the fission gamma radiation has been found to have a half-life of about $1 \cdot 10^{-11}$ sec. From Fig. 10 it can be seen that this corresponds to an energy of about 600 kev which is close to the mean energy of the fission gamma radiation.

We will now see if the assumption that a great part of the radiation is of vibrational type is consistent with the shape of the total gamma-ray spectrum. First we have to find the energy of the vibrational transitions for all the fission fragments. In the harmonic approximation the members of the vibrational cascade have the same energy, which then simply can be found from the well-known energy of the first 2+ state in even-even nuclei. However, for actual nuclei, this is not exactly true. An inspection of some well studied level schemes shows that there is a tendency of increasing energy for the higher members of the vibrational band. For magic or near-magic nuclei, on the other hand, the sharp increase in energy of the first 2+ state is not accompanied by a corresponding increase for the higher transitions. It is impossible to take these effects into account, however, and, in order to get a rough idea about the situation, the energy of the first 2+ state is a reasonably good measure of the mean energy of the vibrational transitions. Therefore the location of the 2+ state was determined from the well-known systematics of this state. Since the fission fragments have a neutron excess, it is necessary to perform a small extrapolation of the experimental data, but the energy varies so smoothly with nucleon number that this extrapolation cannot produce any error of importance.

Furthermore, we have to know the gamma-ray yield as a function of

fragment mass. It is evident from Fig. 9 that the yield per fragment is not constant. The difficulty here is that the curve gives the yield for only part of the radiation. However, it is obvious that the variations must be small compared to the variations in the mass yield curve. Hence the gamma-ray yield is mainly determined by the yield of the fragments. This means that most of the radiation is emitted by fragments at the peaks of the mass yield curve, i.e. in two rather narrow regions around $A \sim 108$ and $A \sim 142$. An inspection of Fig. 9 shows that the gamma-ray yield per fragment in these two regions is very nearly the same and that the variations within these two regions cannot be of any importance. Hence we take the yield of a certain mass as a measure of the gamma ray yield associated with that mass.

Sorting the gamma radiation in a number of energy intervals and adding the yields in each interval gives the spectrum shown as a histogram in Fig. 11. In the same figure is shown the experimental gamma spectrum taken from the investigations of Smith et al[2] and Bowman and Thompson[3]. It will be noted that in the energy range 500-1500 kev the shape of the calculated and experimental spectra agree very well. This shows that, from this point of view, the assumption of the vibrational character of the gamma radiation is not met with any difficulty.

Further information can be obtained from the measurements of the energy spectra for different masses (Fig. 8). Before interpreting the results, it is worth while discussing what kind of changes one can expect when the mass is changed. First of all it must be realized, as discussed above, that the shape of the spectrum is determined to a great extent by the collimator setting. A certain setting favours transitions with a certain half-life and this means an enhancement at the corresponding energy in the spectrum. The peak at about 700 kev can therefore not be expected to change too much. However, despite this distortion of the spectra, any appreciable changes in shape should be clearly visible.

The type of change one expects is an increase of the mean energy in the vicinity of magic numbers. An inspection of Fig. 8 shows that the predicted changes indeed occur. For $A = 95$ the fragments are close to $N = 50$ and for $A = 128$ close to $Z = 50$ and $N = 82$. The spectra for these masses exhibit a decrease at the low energy end and an increase

at the high energy end exactly as expected. The spectrum for A = 153 is especially interesting. Although the statistics are poor, it gives a clear indication of the same increase in energy as for the magic fragments. The mass 153 corresponds for fission fragments to $Z \sim 60$, $N \sim 93$. Nuclei of this constitution must be deformed. For deformed nuclei vibrational excitation is of two different types, β- and γ-vibrations. It is difficult to say which one is most likely to occur in excited fission fragments, but for the present discussion the distinction between the two types is not so important. The main point is that they have roughly the same energy variation. The energy increases, when the mass increases from the borderline of the deformed region towards the center. For fission fragments with A = 153 the energy can be estimated to be 1200 kev. This is not inconsistent with the experimental spectrum.

If one assumes that the deexcitation goes via a statistical distribution of quasi-particle levels no such variations with mass are expected. A theoretical study of the spectrum shape in this case shows that the mean energy is proportional to the temperature at the initial excitation energy. For a given energy the temperature is higher for magic nuclei. However, this tendency is counteracted by the fact, discussed below, that the total gamma-ray energy has a minimum for magic fragments. A closer study of this problem shows that the experimental variations cannot be accounted for in this way. Hence the energy spectra support the assumption that vibrational transitions play a predominant rôle in the deexcitation of fission fragments.

B. Yield.

The interpretation of the yield curve is made somewhat difficult by the fact, pointed out above, that it refers to only part of the gamma radiation. However, this part is as great as 75%. It seems very unlikely that the remaining part of the radiation could have a yield with such large fluctuations that the yield curve of the total radiation could be basically different from the curve in Fig. 9. In the following, we will therefore assume that this curve represents the total yield.

More important than the yield of gamma rays is the total gamma-ray energy. A calculation of the total energy requires knowledge of the mean

gamma-ray energy as a function of mass. The difficulties in interpreting the spectra in Fig. 8 have been discussed. It was only possible to find some general trend, for example, that the mean energy is higher for magic and near-magic nuclei. This brings up the question if the structure in the yield curve is, perhaps, just a reflection of the differences in the mean gamma-ray energy. The minima at mass 90 and 130 could be connected with the magic numbers N = 50 and Z = 50, N = 82, respectively. If the total gamma-ray energy is constant, minima would appear because of the higher mean energy. A closer inspection shows that it cannot be so. The minima are not at all as narrow as one would expect on such a view. A more detailed examination of the curve shows this more clearly. It is obvious from Fig. 8 that the gamma-ray spectra for A = 102 and A = 118 are practically identical. The mean-energy must be closely the same. Yet the yield curve shows a considerable increase between A = 102 and A = 118. The same is true if one compares the masses 128 and 153. Undoubtedly, the variations in gamma-ray energy will be of some importance, but in its general appearance the total energy curve will have the same saw-tooth shape as the yield curve in Fig. 9.

In order to explain the variations of the total gamma-ray energy it is necessary to take up the problem of the competition between neutron emission and gamma emission. Normally, it is assumed that neutron emission always takes place, whenever it is energetically possible. Calculations of Leachman and Kazek[12] and Terell[13] based on this assumption and statistical theory show that roughly an energy amount equal to the neutron binding energy or about 5 Mev is left for gamma emission, whereas twice as much is found experimentally.[1-3,6] It is very probable that this discrepancy is caused by the neglect of spin effects. Each emitted neutron carries off a considerable energy (binding energy + mean kinetic energy) but only little angular momentum. If the excited fragment starts with a rather high spin, the result will be, that at the end of the neutron emission the energy is relatively low but the spin still high. This makes it increasingly difficult for the last neutrons to find suitable high spin states for the transitions. Quantitatively this situation can be treated by introducing a level density formula including the proper spin dependence. The spin dependent part has the form:

$$(2I+1) \exp - \left[I^2 \hbar^2 / JT \right] \quad \ldots\ldots\ldots \qquad (1)$$

where J is the moment of inertia and T the temperature. Obviosly this formula leads to a decrease in the density of high spin states, especially at low energies. Inserting the rigid body value of J and a resonable value for I (about 10) shows that the situation is not basically improved by this modification. Only if a value about one tenth of the rigid body value is used for J will any significant improvement occur. It is very interesing to note in this connection that, in several cases, it has been found that the value of the moment of inertia appearing in the level density formula turns out to be remarkably small. This has been noted by Vandenbosch and Huizenga[14] in their work on isomer production, by Leachman and Sanmann[15] in an analysis of the angular distributions in fission and also in some nuclear reactions studies. The cause of this discrepancy is that the nuclear spin is a result of the coupling of a small number of nucleon spin vectors. At each energy there is a maximum spin value corresponding to a complete alignment of the spin vectors of the unpaired nucleous. This effect leads to a lower density of the high spin states than given by eq. (1). Unfortunately nothing is known quantitatively about how much the moment of inertia has to be reduced in order to account for this effect.

For our purpose it is better to study directly the maximum possible spin values at different energies. Knowing this, one can estimate to what extent the neutron emission is reduced. The average spin at the beginning of the gamma-ray cascade should be about 10 (see the subsequent discussion). States closely above the neutron emission threshold in a nucleus have to decay to the ground state or to low-lying state in the daughter nucleus. These states never have spin values as high as 10. This means that the neutrons have to carry off several units of angular momentum. The penetration of the low energy neutrons through the resulting centrifugal barrier is so small that neutron emission is practically forbidden. Only for states, which are so high above the neutron emission threshold that their position corresponds to such energies in the daughter nucleus, that levels with spin about 10 begin to appear, can neutron emission take place unhindered. An inspection of known level schemes shows, that practically no levels with spin 10 or higher are known. This is partly explained by the fact that such levels are very difficult to populate. On the other hand, high spin states often give rise to isomers, which are easy to detect experimentally. In any case, it seems obvious that states of spin 10 or higher are very

rare for the first two Mev of excitation. A very interesting case has recently been found by Perlman et al.[16] It is a state with spin 18 appearing at 3 Mev in Po^{212}. In an illuminating theoretical study of this problem, Glendenning[17] has shown how the two protons and the two neutrons outside the core combine to give spin values up to 18. In the mass region of the fission fragments the single particle spins are lower, but the same principles apply. One can easily see that, in order to get a spin of 10, in general at least three nucleon spin vectors have to be combined. This means that in an even-even nucleus at least two pairs have to be broken up, and in other nuclei at least one pair. Hence on the average one should expect states of spin 10 to start to appear at about 2 Mev. Gamma emission will therefore be able to compete succesfully with neutron emission in an energy interval of about 2 Mev above the neutron emission threshold. The previosly mentioned estimates of the total gamma-ray energy have therefore to be encreased by 4 Mev, which is roughly what is required to bring them in agreement with experiments. For other spin values the energy values will, of course, be different. If the initial spin is smaller the total energy will be smaller, and if the spin is higher the energy will be higher.

We are now able to discuss the saw-tooth curve of the total gamma-ray energy. It resembles the curve for the total excitation energy. However, high excitation does not in itself give a high gamma-ray energy. On the contrary, a high excitation means more neutrons, which can carry off slightly more angular momentum and that favours neutron emission over gamma emission. Therefore, the only way to account for the variations of the gamma-ray energy appears to be, to assume that it is caused by a similar variation of the spin of the fragments so that a high total gamma-ray energy corresponds to a high spin. Since the curves for the total gamma-ray energy and the total excitation energy are similar, the interesting conclusion is that spin and excitation energy vary in the same way with mass.

The relation between spin and excitation energy established here should be of value for the understanding of the conditions during scission. At present our knowledge of this stage of the fission process is so limited that any interpretation of the data necessarily will be very speculative, and therefore we refrain from discussing various possible explanations of this experimental results.

V. Discussion.

With the results of the present paper added to what has been known before, one can attempt to get a consistent picture of the deexcitation process.

It is instructive to compare the fission gamma radiation with the radiation emitted in two other processes, neutron capture and heavy ion induced reactions. In neutron capture the multiplicity is low and the spectra peak at 2 - 3 Mev. In heavy ion reactions the multiplicity is high (10-20) and the spectra peak at low energy. The gamma radiation in fission seems to be an intermediate case, but is closer to the second type. The basic difference between these two reactions is the amount of angular momentum of the excited nucleus. In the first case it is low (up to 4 $\hbar$) but in the second case it can be very high(20-40 $\hbar$). A closer comparison of the fragment deexcitation with the other two reactions gives as a rough estimate of the spin the value 10-15.

A better estimate could, in principle, be obtained from an analysis of the angular distribution of the gamma radiation. Unfortunately, the analysis of the experimental results is beset with many difficulties. One such difficulty is that the results depend on various assumptions concerning the orientation of the initial spin and about the path of deexcitation. Strutinski[18] derived a formula based on the assumption of a deexcitation via a statistical distribution of levels according to eq.(1). The spin dependence of the level density has the effect that, in each step of the deexcitation, there is a tendency of the transitions to go to the levels with the lowest possible spin compatible with the multipolarity of the radiation. The difficulty is that, with the high spin and relatively low excitation occuring in fission, the assumption of a statistical distribution of levels is questionable. The limitations on the maximum possible spin, discussed above, come in here, too. The result is that the value of the moment of inertia, which appears in the expression for the anisotropy, is very uncertain. The spin values then have a corresponding uncertainty.

Hoffman[8] fitted his measured angular distributions with a theoretical expression containing a dipole and a quadrupole term. With some assumptions about the orientation of the spin vector he could deduce a spin value

by varying the parameters to get the best possible fit. It turned out that the quadrupole term was the dominating one.

Unfortunately, it seems that the analysis of the angular distributions are of a somewhat limited value for providing information about the fragment spin. However, in this connection it should be noted that, despite the difficulties of the analysis, the angular distribution strongly indicates that the greater part of the radiation consists of quadrupole transitions.

The best way of estimating the fragment spin appears to be to use the information about the multipolarity and the multiplicity of the radiation. Both the angular distribution of the radiation and the life-time determination in the present work show that we mainly have quadrupole transitions. The multiplicity is known to be about ten. The yield curve of Fig. 9 shows that for the most probable mass ratio the yield is the same for the two fragments. Hence the average multiplicity per fragment should be about five. For quadrupole radiation this gives a maximum initial spin of 10. This corresponds to the case in which the spin is always decreasing regularly. However, this is probably also the most likely case. The scarcity of high spin states, expecially at low excitation gives a strong tendency favouring transitions in which the spin is decreasing. Therefore the initial spin is probably not too much below the upper limit. On the whole a spin of about 10 is in good agreement with all experimental facts.

When the quadrupole character of the transitions has been established, the problem is, if it is possible to say anyting further about the properties of the transitions. As has been discussed in some detail above, the experimental results seem to indicate that the transitions are of the vibrational type. The strongest evidence is probably the life-time. It was found that the main part of the radiation has a half-life of about $1 \cdot 10^{-11}$ sec. The average energy of the spectra in Fig. 8 is about 500 kev. An inspection of the relation between half-life and energy for known vibrational transitions (Fig. 10) shows, that these values agree very well with this relation. On the other hand the single particle estimate for the half-life of a quadrupole transitions of 500 kev is $5 \cdot 10^{-9}$ sec. Therefore the fission gamma rays seem to be enhanced quadrupole transitions and vibrational transitions is then the only possibility. As discussed above this interpretation is further supported by the shape of the total gamma-ray spectrum and by the variation of the energy spectra as a function of mass.

The interesting question is now, why vibrational transitions are preferred in the deexcitation of the fission fragments. The most likely explanation can be found in the conditions during scission. At that point, the electrostatic repulsion deforms the fragments. When they separate, a vibrational motion will be set up. In addition the fragments can obtain a rotational motion, corresponding to a high spin. Finally, it is likely that there is not time enough for the fragments to adjust adiabatically to the rapid changes and that therefore some nucleons are left in higher levels giving an intrinsic excitation of the fragments. Therefore, at the moment of scission, the fragments are formed in very complex states with intrinsic, vibrational and rotational excitation. Most of the excitation energy is then taken away by the neutrons. It is likely that the energy needed to evaporate the neutrons is taken at first hand from the intrinsic excitation. The reason is, that the intrinsic excitation corresponds to a small number of excited nucleons, whereas many nucleons take part in the collective motion. Since neutron emission requires that a large part of the excitation energy is concentrated on a single nucleon, it should take place mainly at the expense of the intrinsic excitation. This implies that, at the end of the neutron emission, the fragments are left in states having mainly or perhaps even exclusively collective excitation. In this connection, vibrations are the most important mode of collective excitation (in spherical fragments it is the only possibility). It is therefore quite natural that the gamma deexcitation proceeds via a cascade of vibrational transitions.

If the vibrational transitions account for the main part of the spectrum from 300 to 1500 kev, it is interesting to ask about the character of the remaining parts. In the low energy range rotational transitions can be expected to be of importance. This question will be treated in the second part of this paper. Very little can be said with certainty about the high energy gamma rays. They could be single particle transitions. Another possibility is that they are octupole vibrations, The energy of the octupole vibrations (2-3 Mev) is in agreement with this view. As a matter of fact one would expect to get octupole vibrations for the same reasons as one expects quadrupole vibrations. At scission the deformation of the fragments is probably not of the second order only. It is more likely that the fragments are slightly pear shaped. This would tend to induce octupole vibration as a part of the very complex excitation of

the fragments. It should be possible to get some experimental information on this point by investigating the slow component of the gamma radiation more in detail.

VI. Conclusions.

The present investigation has given some information about the deexcitation of the fission fragments. These results are of value for the understanding of the conditions at scission. Perhaps more important than the results themselves is the fact that they show that the technique used here is capable of giving detailed information about the deexcitation process. The present work can only be considered as a preliminary survey and various improvements are possible. The crucial point is the time resolution of the collimator. With some effort it should be possible to achieve an improvement with a factor of 2 or 3. This would make it possible to study the decay curve in much greater detail. A systematic study of the radiation for different collimator settings would then bring forth a wealth of information about decay times, energy spectra, and yields of the various components of the radiation. Another improvement would be to study the gamma radiation also as a function of the excitation energy of the fragments.

Investigations of the present type lead to a new type of nuclear spectroscopy dealing with the properties of highly excited collective states in a great number of different nuclei. The information which is obtained should be of value for the systematics of the collective states as well as for the understanding of the fission process. The results of this work indicate that it should be worth while to push this technique as far as possible.

Acknowledgements.

The author is greatly indebted to Dr S.G. Thompson for his continous interest in and support of the work reported here. Many thanks are due to Mr L. Gibson for helping with the electronic circuitry and to Mr R. Gatti and Mr L. Phillips for preparing the sources. The author also likes to express his gratitude to Professor I. Perlman for the pleasant hospitality of the Chemistry Division of the Lawrence Radiation Laboratory.

Figure captions.

Fig. 1. Schematic picture of the experimental arrangement.

Fig. 2. Block diagram of the electronic circuits.

Fig. 3. a. Pulse spectrum from the fission detectors.
b. Pulse spectrum from the divider.

Fig. 4. Decay curve determined with the 3 mm collimator.
Circles: Counting rate as a function of collimator setting.
Triangles: Counting rate with a catcher foil in front of the source.

Fig. 5 a. The response of the 0.8 mm collimator for a Hg^{203} source. The line is the calculated response.
b. The decay curve of the fission gamma radiation determined with the 0.8 mm collimator.

Fig. 6 Analysis of the decay curve assuming a simple exponential decay.

Fig. 7. Analysis of the decay curve assuming a cascade of four gamma rays with equal life-time. The curve shows the theoretical decay curve for such a cascade.

Fig. 8. Energy spectra of the radiation for a number of different masses.

Fig. 9. The gamma-ray yield as a function of mass.

Fig. 10. Half-life of the first 2+ state in even-even nuclei as a function of the energy of the state. Data for the following elements are included: Se, Mo, Ru, Pd, Cd, and Te.

Fig. 11. A comparison of the spectrum of the total gamma radiation (the curve) with the spectrum calculated on the assumption that the gamma radiation consists of vibrational transitions (the histogram).

References.

1. F.C. Maienschein, R.W. Peele, W. Zobel and T.A. Love, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy 15, Paper P/670 (Geneva, 1958) 366.

2. A.B. Smith, P.R. Fields and A.M. Friedman, Phys. Rev. 104 (1956) 699.

3. H.R. Bowman and S.G. Thompson, Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy 15, Paper P/652 (Geneva, 1958) 212.

4. V.K. Voitovetski, B.A. Levin and E.V. Marchenko, Soviet Physics JETP 5 (1957)184.

5. V.V. Skliarevskii, D.E. Fomenko and E.P. Stepanov, Soviet Physics JETP 5 (1957) 220.

6. F.E.W. Rau, Annalen d. Physik 10 (1963) 252.

7. S. Dési, A. Lajtai and L. Nagy, Acta Phys. Hung. 15 (1962) 185.

8. M.M. Hoffman, Phys. Rev. 133 B (1964) 714.

9. S. Dési, G. Graff, A. Lajtai and L. Nagy, Physics Lett. 3 (1963) 343.

10. S.S. Kapoor and R. Ramanna, Phys. Rev. 133 B (1964) 598.

11. J. Terrell, Phys. Rev. 127 (1962) 880.

12. R.B. Leachman and C.S. Kazek, Phys. Rev. 105 (1957) 1511.

13. J. Terrell, Phys. Rev. 113 (1959) 527.

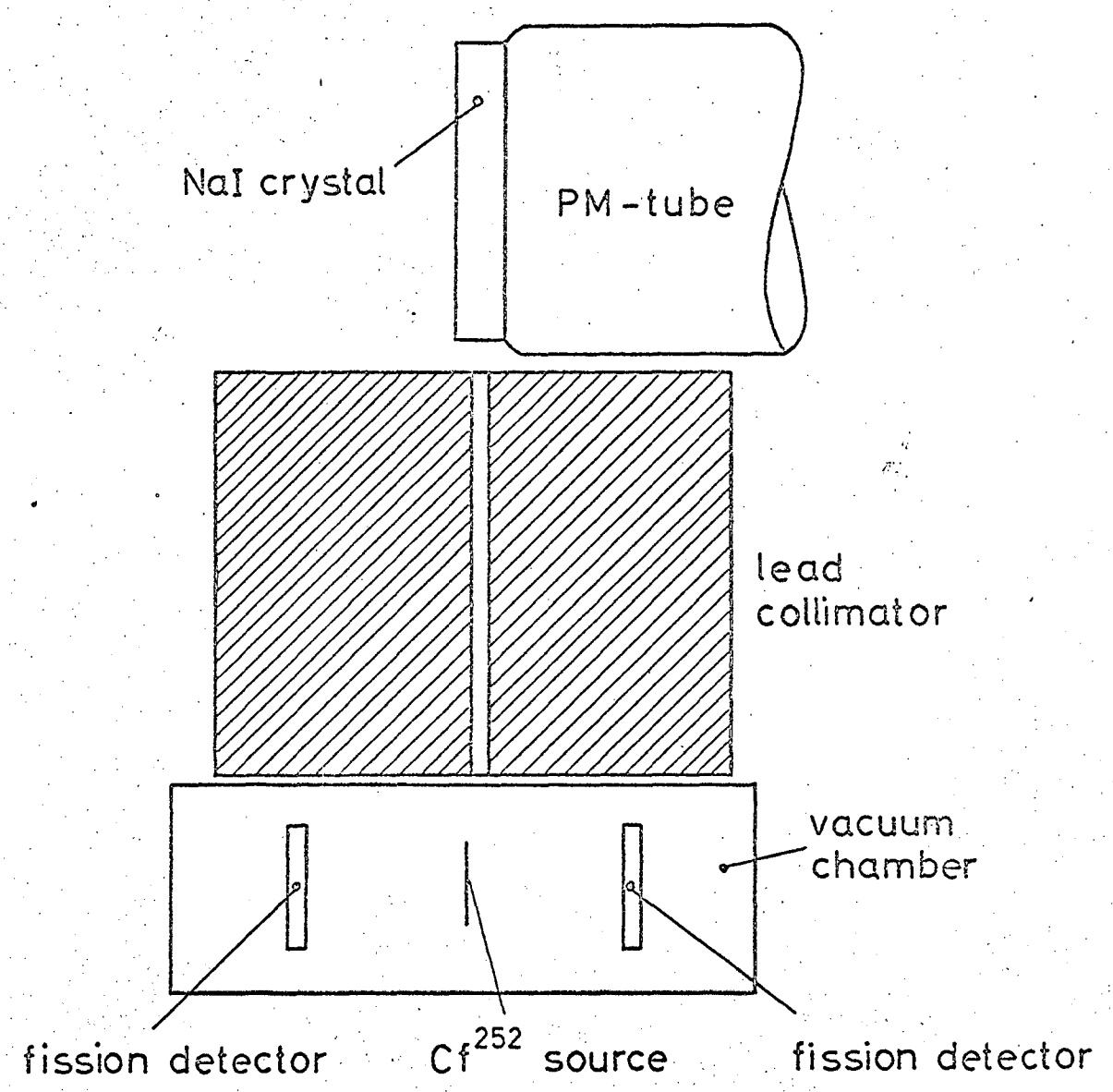
14. R. Vandenbosch and J.R. Huizenga, Phys. Rev. 120 (1960) 1313.

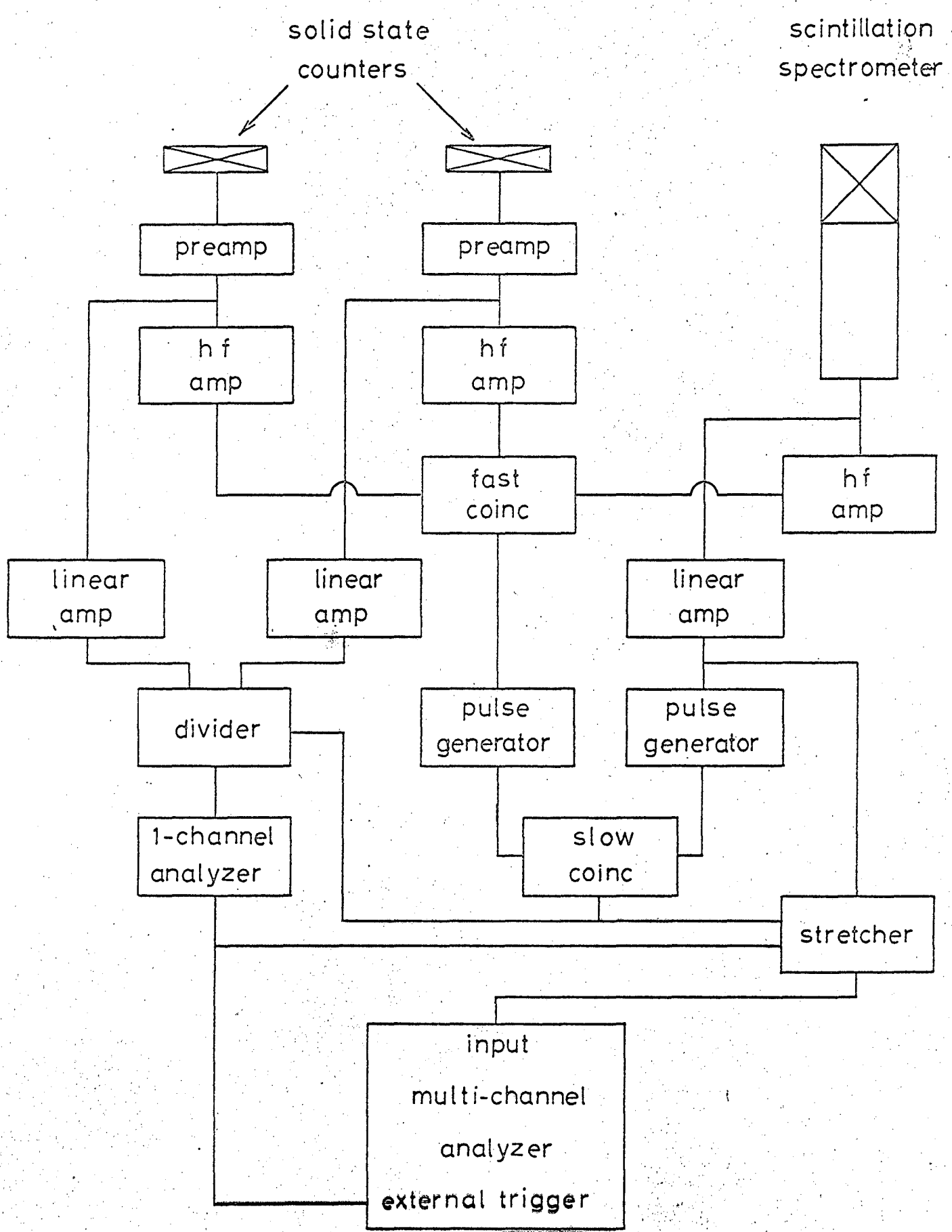
15. R.B. Leachman and E.E. Sanmann, Ann. of Phys. 18 (1962) 274.

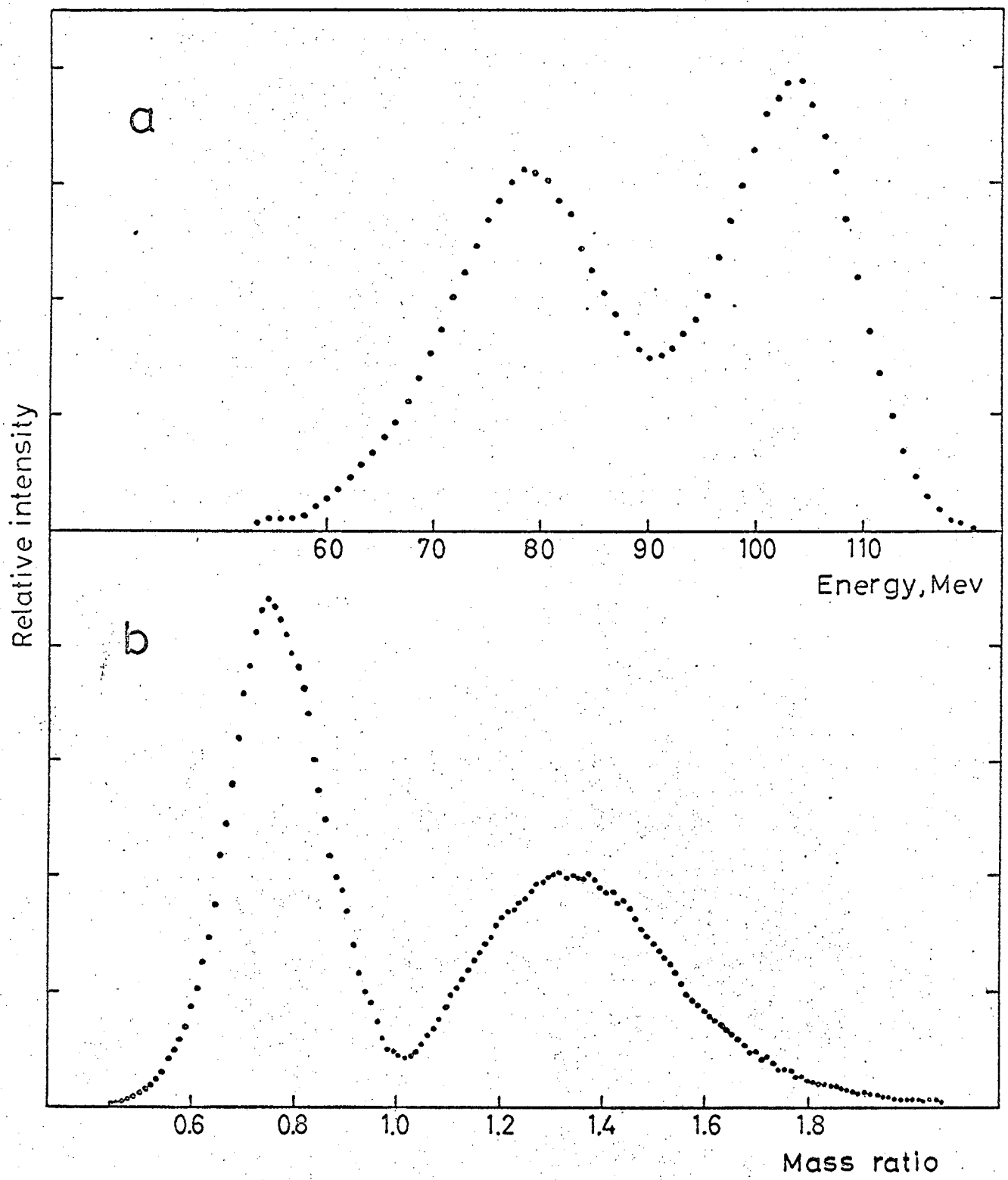
16. I. Perlman, F.Asaro, A. Ghiorso, A. Larsh and R. Latimer, Phys. Rev. 127 (1962) 917.

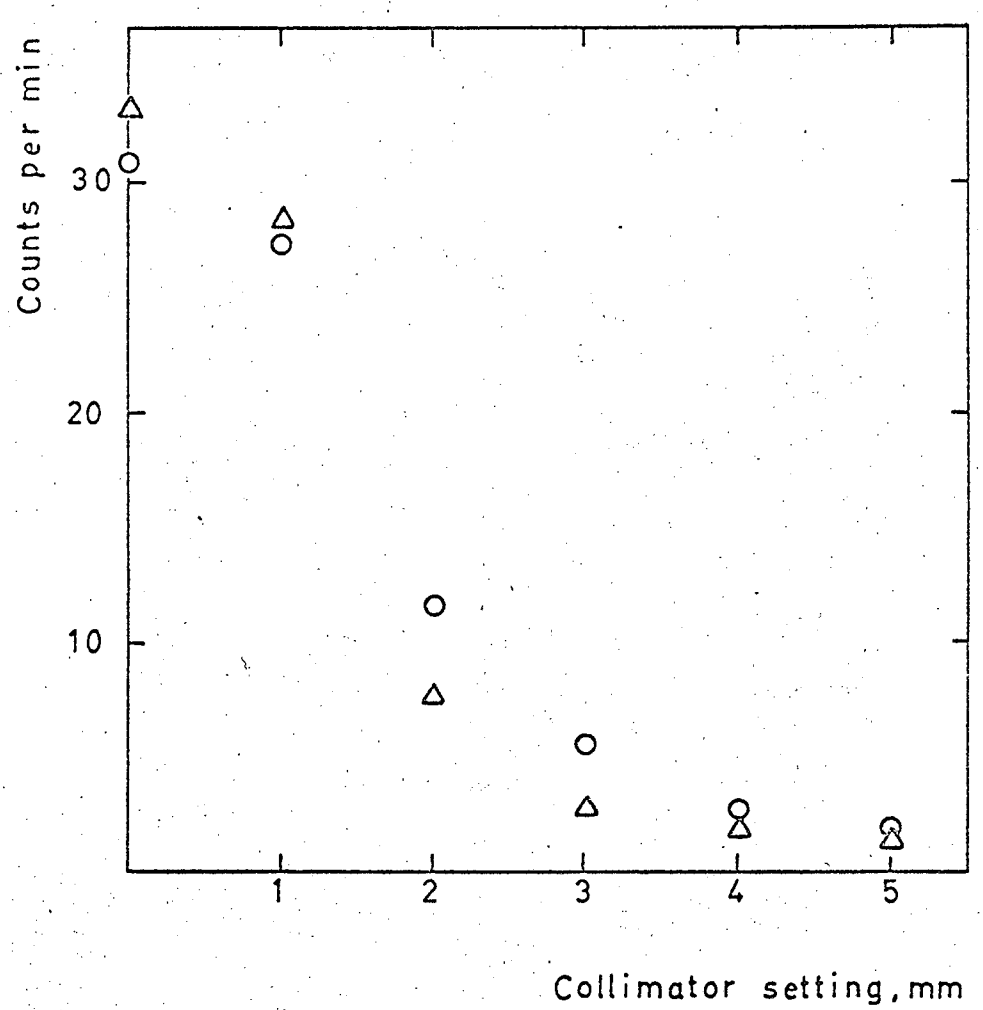
17. N.K. Glendenning, Phys. Rev. 127 (1962) 923.

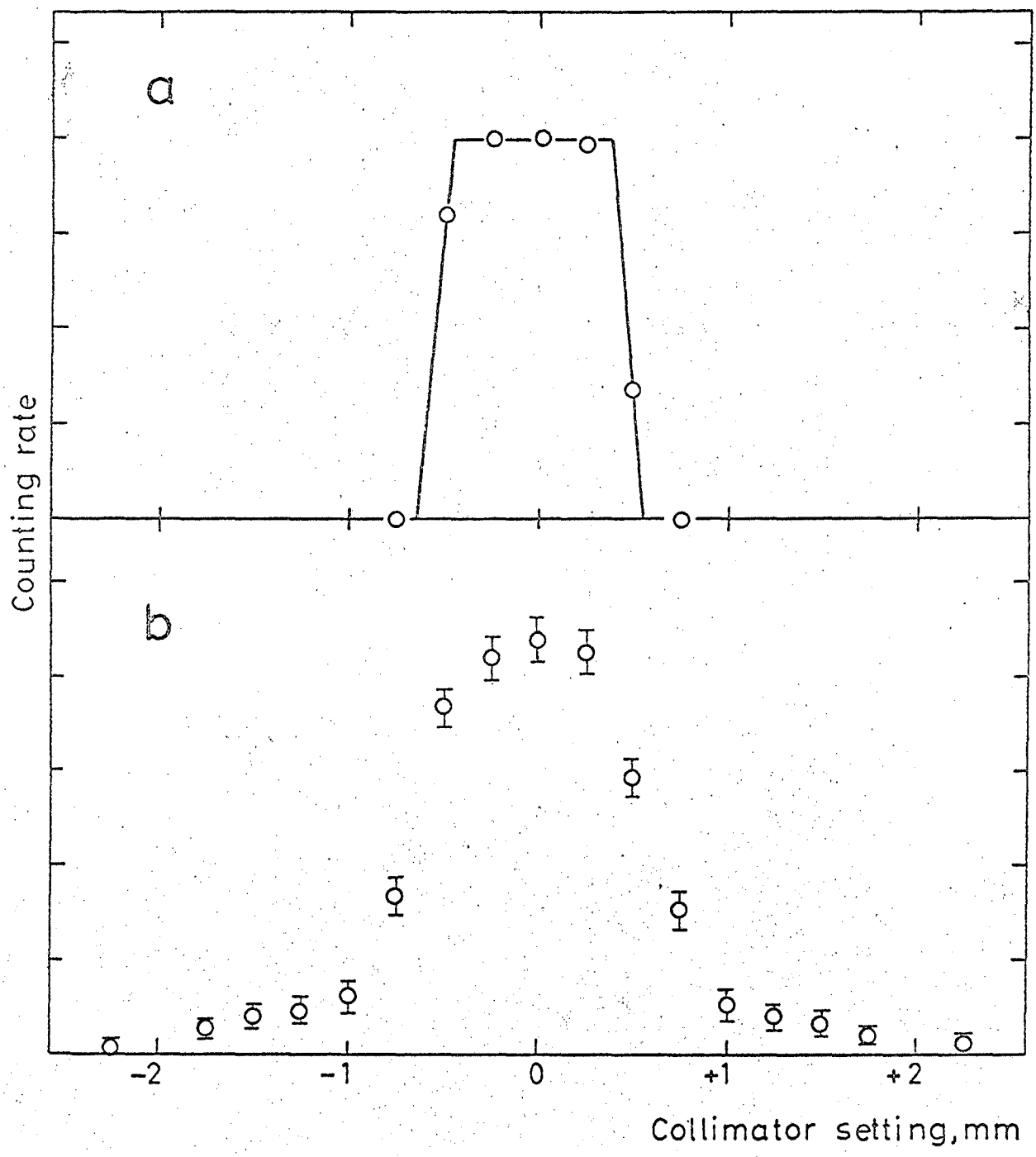
18. V.M. Strutinskii, Soviet Phys. JETP 10 (1960) 614.

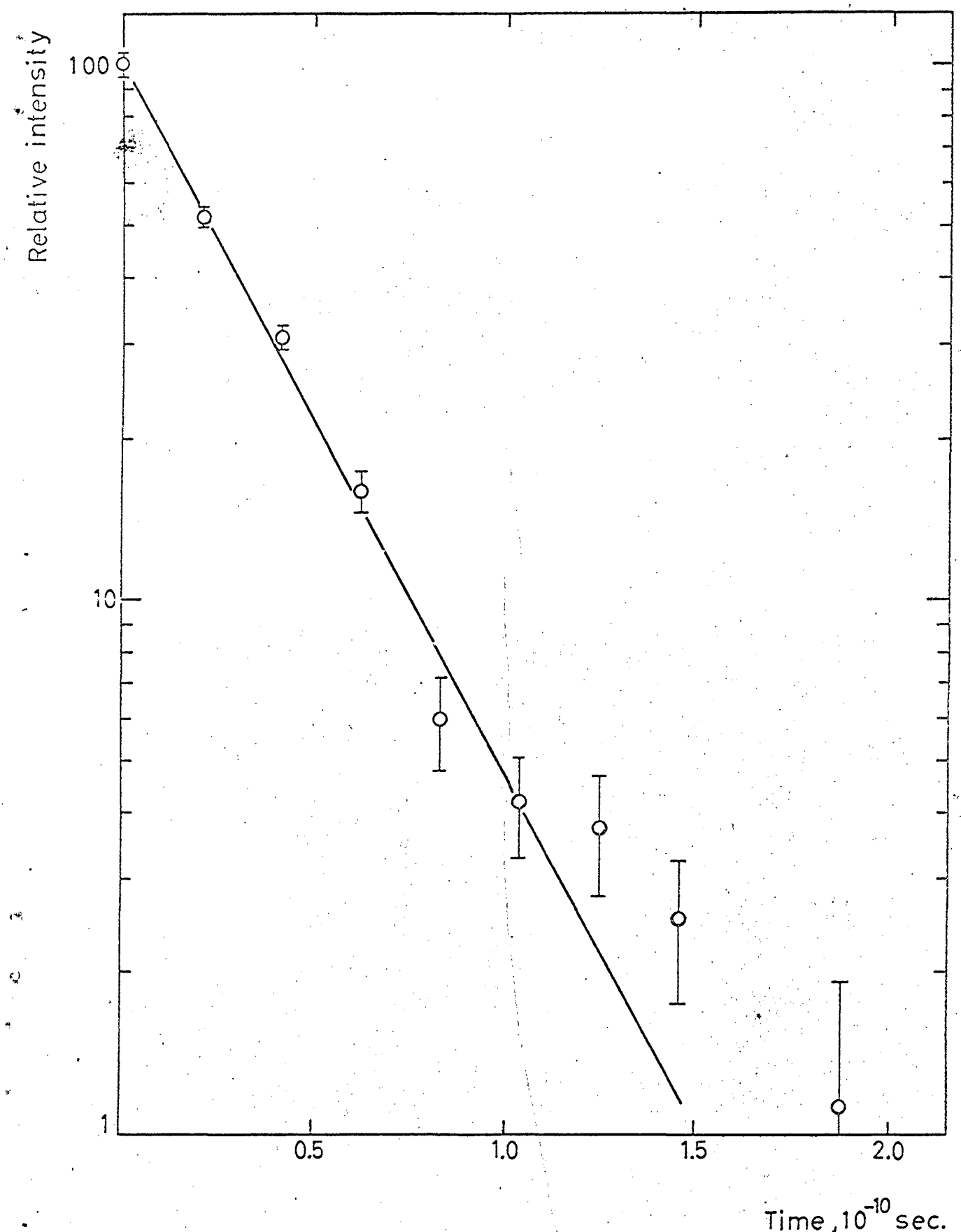

NaI crystal
PM-tube
lead
collimator
vacuum
chamber
fission detector
Cf252 source
fission detector

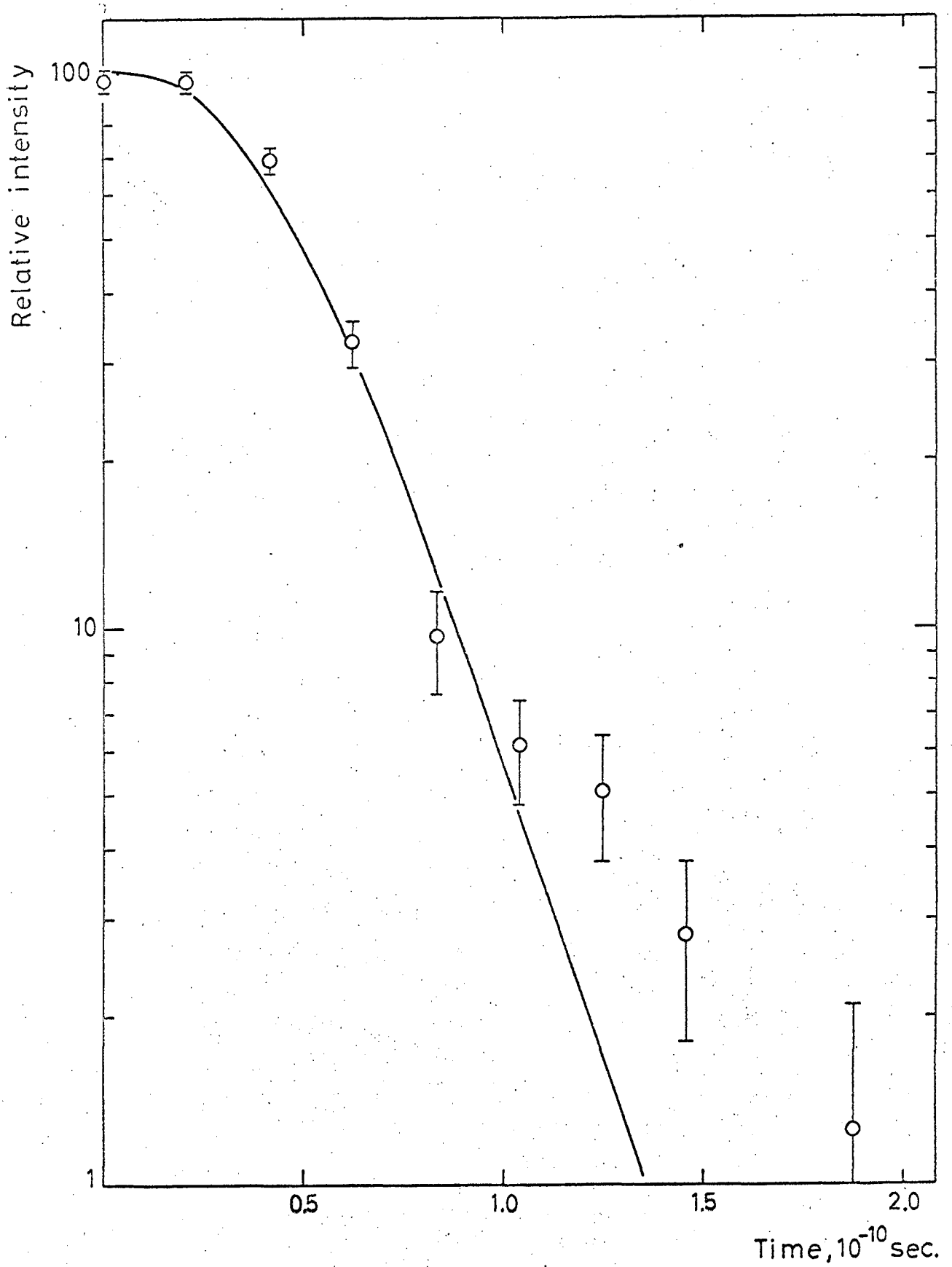

solid state
counters
scintillation
spectrometer
preamp
preamp
h f
amp
h f
amp
fast
coinc
h f
amp
linear
amp
linear
amp
linear
amp
divider
pulse
generator
pulse
generator
1-channel
analyzer
slow
coinc
stretcher
input
multi-channel
analyzer
external trigger


a
b
Relative intensity
60
70
80
90
100
110
Energy, Mev
0.6
0.8
1.0
1.2
1.4
1.6
1.8
Mass ratio


Counts per min
30
20
10
1
2
3
4
5
Collimator setting, mm


a
b
Counting rate
-2
-1
0
+1
+2
Collimator setting, mm


Relative intensity
100
10
1
0.5
1.0
1.5
2.0
Time, 10^{-10} sec.


Relative intensity
100
10
1
0.5
1.0
1.5
2.0
Time, 10^{-10} sec.

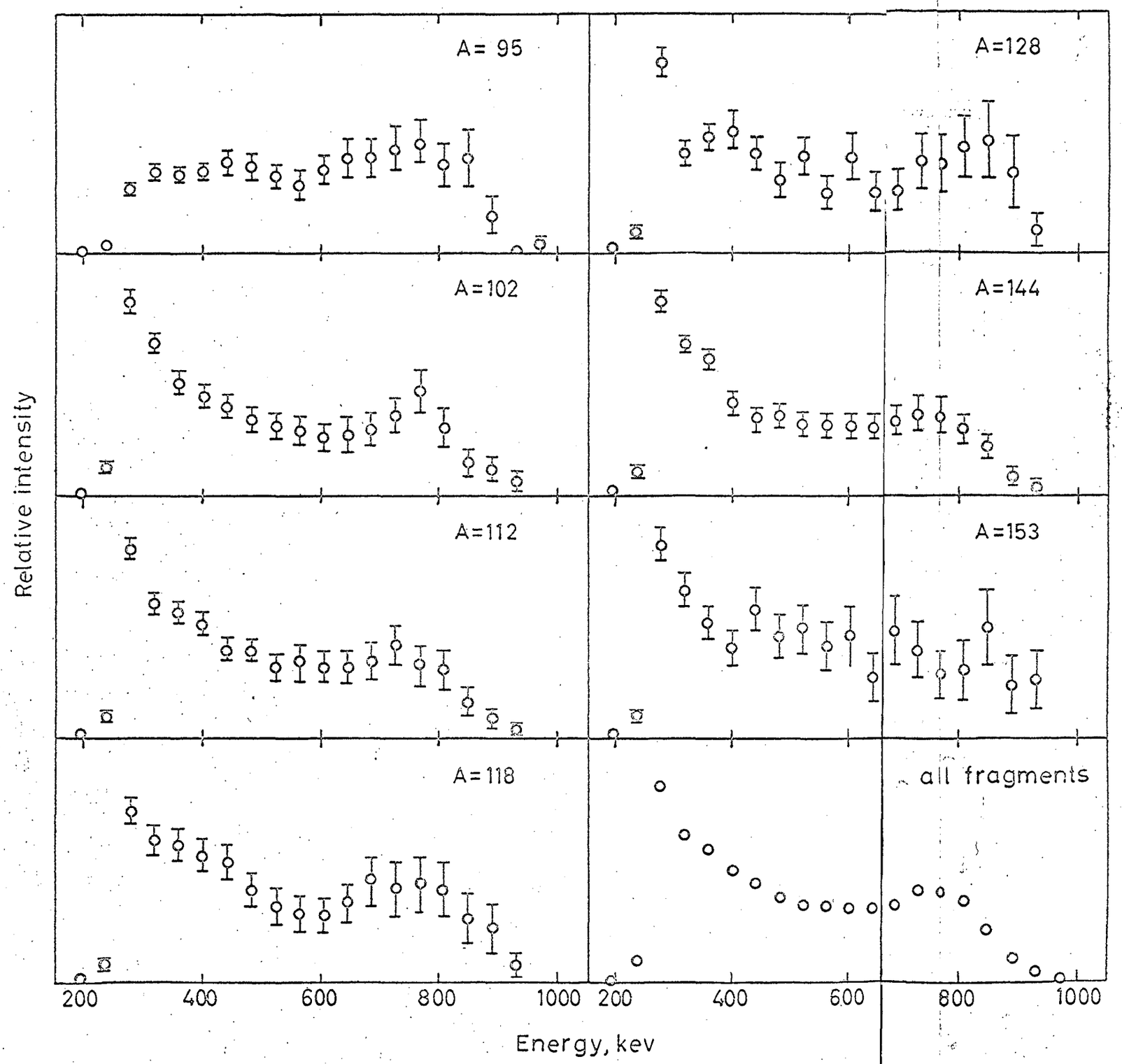
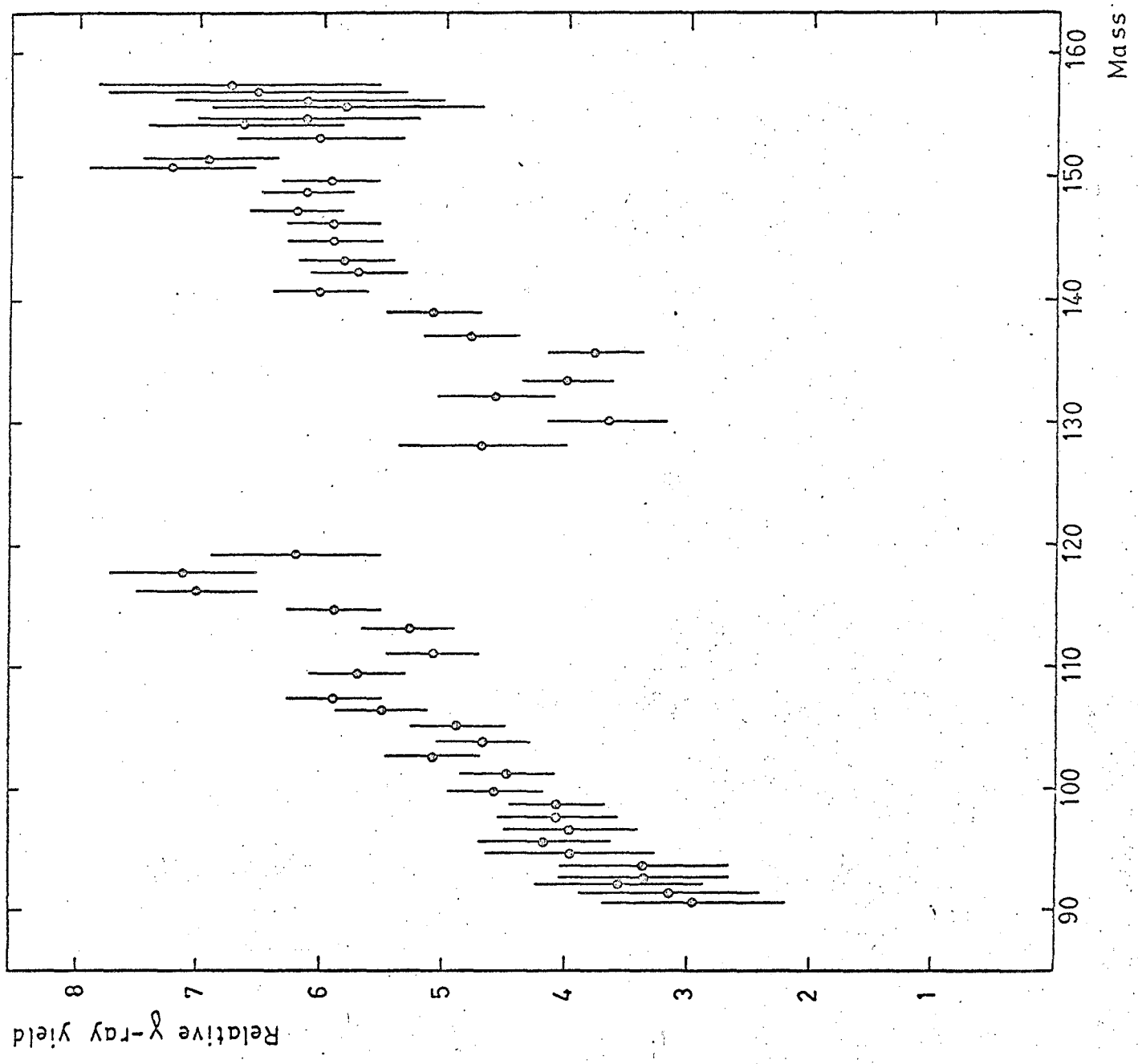
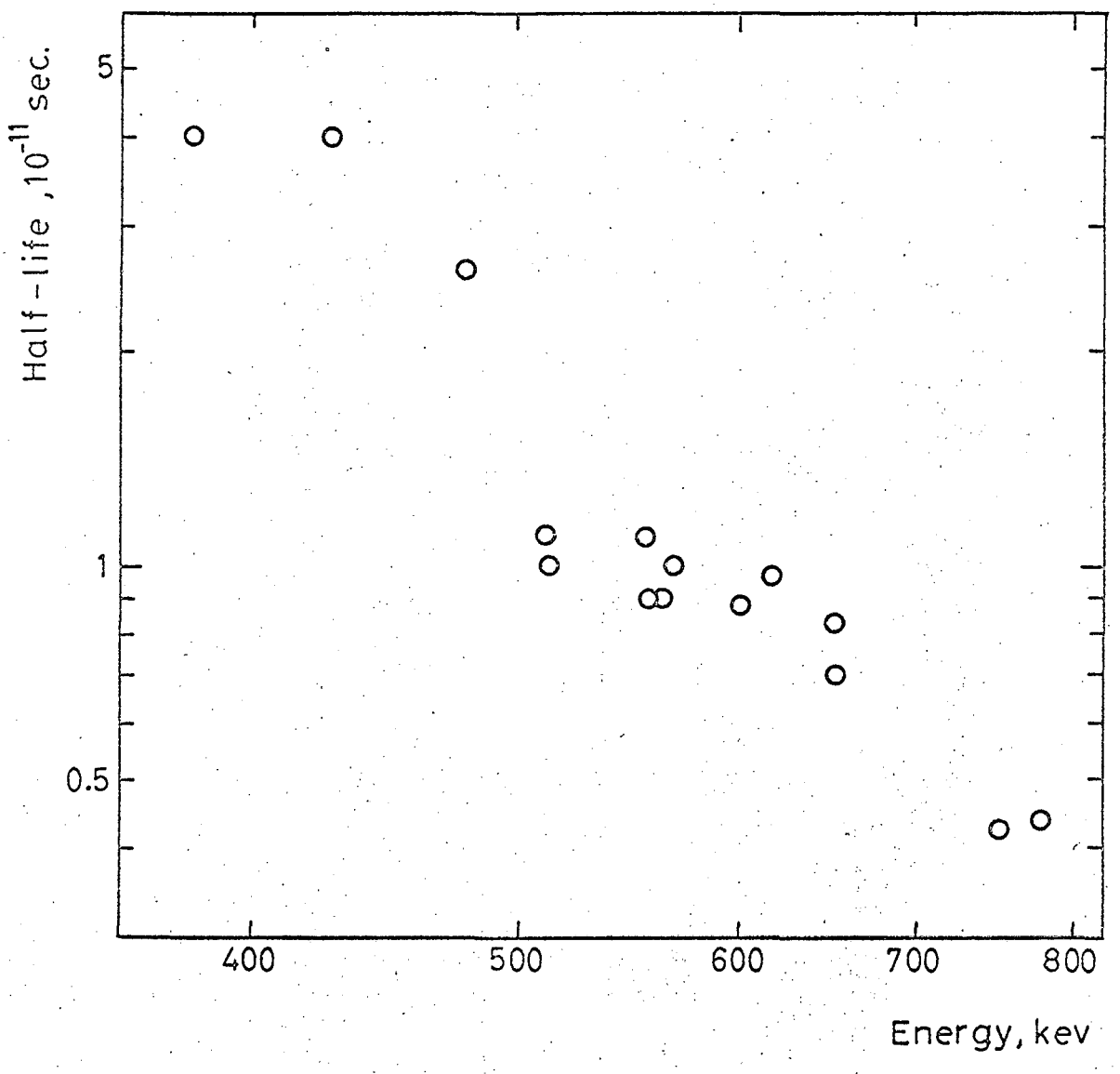
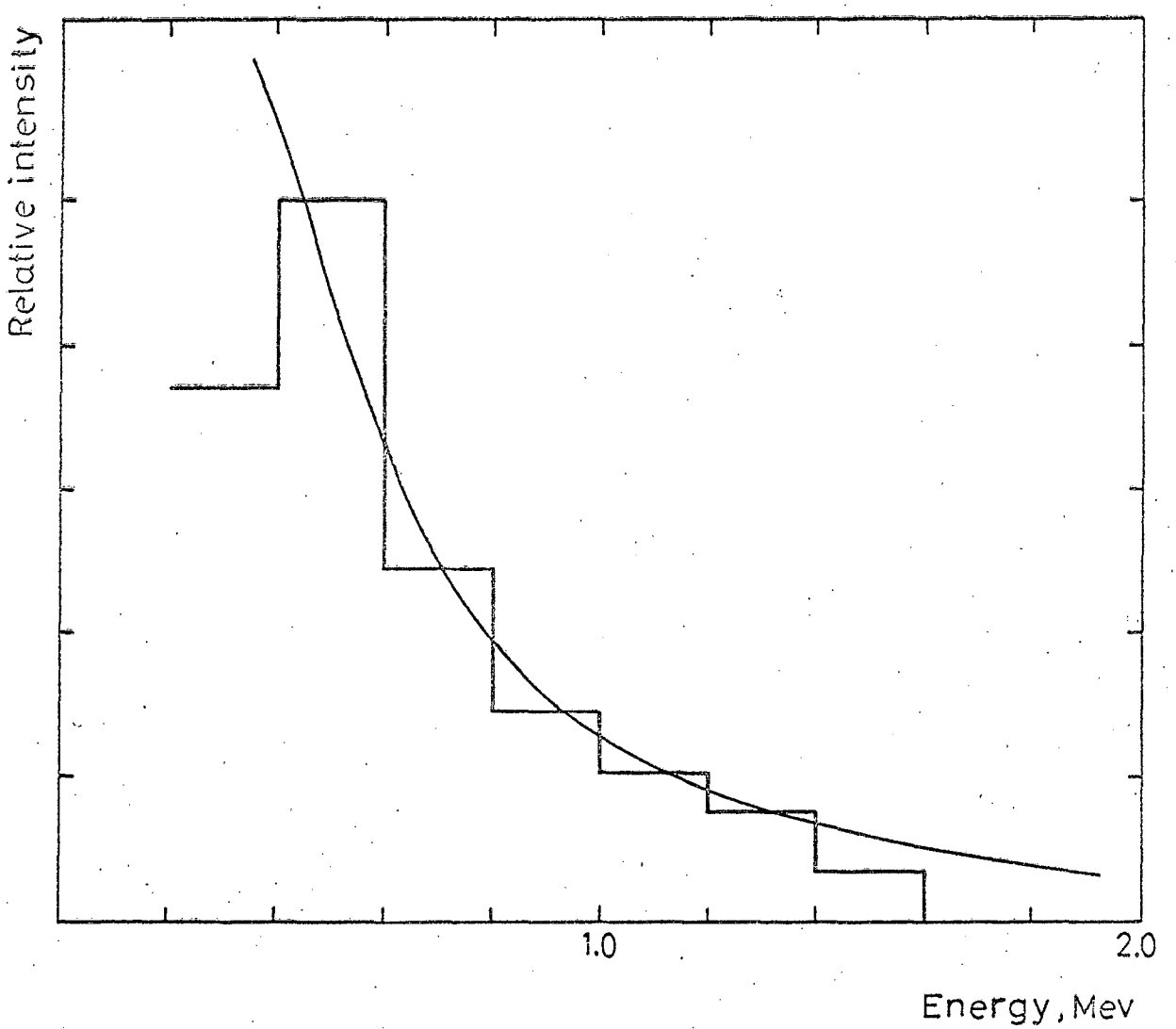

A= 95
A=128
A=102
A=144
A=112
A=153
A=118
all fragments
Relative intensity
200
400
600
800
1000
Energy, kev

Fig 8


Relative γ-ray yield
8
7
6
5
4
3
2
1
90
100
110
120
130
140
150
160
Mass


Half-life, 10^{-11} sec.
5
1
0.5
400
500
600
700
800
Energy, kev


Relative intensity
1.0
2.0
Energy, Mev

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.