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MASS RATIOS AND ENERGY RELEASE IN THE FISSION OF U^{235}, U^{233} and Pu^{239} BY THERMAL NEUTRONS

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ABSTRACT

The pairs of simultaneous fission fragments were observed in a double ionization chamber using electron collection. The most probable energy of the heavy fragments is for U^{235}: 60 Mev; U^{233}: 59 Mev; Pu^{239}: 65 Mev. The most probable energy of the light fragments is in the same order, 94, 95 and 93 Mev. The overlap of the two groups is about equal in all three cases and may well be entirely instrumental. The most probable total-kinetic-energy release is, for the three substances in the above order: 153 Mev, 151 Mev, and 156 Mev. The most probable mass after emission of the prompt neutrons for the heavy fragment in all three cases seems to be about 140 mass units. The most probable mass of the light fragment for U^{235} is about 93; for U^{233}: 91; for Pu^{239}: 98. The highest fission yield for a single mass number should be about 5 1/2 % in all three cases. The energy release for splitting in a given ratio seems to be nearly the same for the three substances.

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MASS RATIOS AND ENERGY RELEASE IN THE FISSION OF U^{235}, U^{233} and Pu^{239} BY THERMAL NEUTRONS

The kinetic-energy release and the mass ratios occurring in the fission of U^{235}, U^{238} and Th^{233} has been studied by Jentschke[1] and for U^{235} by Flammersfeld et al.[2] Brolley[3] attempted a preliminary study in the case of Pu^{239}. In the present experiment we studied the fission process in the case of thermal fission of U^{235}, U^{233} and Pu^{239} by a method very similar to that of Jentschke.

EXPERIMENTAL METHOD

Fig. 1 shows a diagram of the ionization chamber used. The material under investigation - pure isotopes in the case of U^{233} and Pu^{239} and about 80 percent U^{235} - were used in the form of nitrates. These were dissolved in a mixture of alcohol and "Zapon" thinner. This solution was added to an appropriate amount of dilute (1 to 50) solution of "Zapon" lacquer. About 0.2 cc of this solution was spread over an area of about 40 cm^2 on freshly cleaved mica. After drying for an hour at 60°C this film was floated off on water and immediately lifted out on a fine nickel grid. The operation had to be carried out quite rapidly since otherwise a large fraction of the active salt dissolved in the water. Attempts to spread the films on other liquids, including mercury, failed for various reasons. The nickel grids were stamped with square holes about 0.15 mm to the side and about 0.15 mm thick. Their optical transmission was about 20 percent. Particles leaving the film on the side on which it was supported were therefore quite well collimated. The grid, supporting the film was then mounted in the chamber in the place indicated in Fig. 1.

The total thickness of the film never exceeded 1.2×10^{-5} gm/cm^2. The amount of active material was about 5×10^{-7} gm in the entire film (40 cm^2) in the case of Pu^{239}, about

1. Zeit. fur Physik, 120, 165 (1943).
2. Zeit. fur Physik, 120, 450 (1943).
3. CN 1840

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3×10^{-6} gm in the case of U^{233} and about 4×10^{-5} gm in the case of U^{233}.

The assemblies of screen and collecting electrode were mounted on the lids of the chamber, identical on both sides of the high-voltage electrode. The screens were made of parallel nickel wires, 0.001" thick spaced 0.030" apart. They served to shield the collecting electrodes against charges induced by the positive ions. This was necessary because of the high low-frequency cutoff of the amplifiers used. By applying pulses to the high-voltage electrode it was shown that the screens were over 99 percent effective. The amplifiers used were of the type known as "Model 100" on the project, stabilized by inverse feedback and had a frequency response such "step" pulses showed a rise time of 0.5 microseconds and a decay time of about 35 microseconds. The chamber was filled to about 50 cm Hg with argon to which about 2 percent CO_2 was added. The gas was purified continuously by circulating over calcium metal at about 250°C. No organic materials were used anywhere in the chamber, other than the film containing the active material, in order to maintain gas purity. The addition of the carbon dioxide was required to speed the collection of the electrons to permit use of the high-frequency circuit, needed in turn to reduce noise due to ionization by the gamma rays from the neutron source and by the alpha rays from the active material. The observed collection time was slightly over a microsecond. The gas pressure was high enough to ensure that all of the ionization took place in the space between the high-voltage electrode and the screens. The high voltage electrode was kept at - 4200 volts while the screens had about -1700 volts applied to them. This makes the field between screen and collecting electrode somewhat higher than that outside the screen. This seems to be necessary in order to "funnel" the electrons through the screen. If the voltage on the screen is too low a large fraction of the electrons fails to reach the collecting electrode. The pulse height is however quite independent of screen voltage once it exceeds a certain minimum value, about 1500 volts in our case.

Alpha ray pulses were saturated with a total voltage of about 250 volts on the chamber - about 100 volts on the screen. Fission pulses however required about 1500

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volt for saturation. This was tested by placing a somewhat heavier film of active material in the chamber so that the fission counting rate was high enough to measure pulse heights conveniently with an electronic differential pulse height section. This difference in saturation voltage is undoubtedly due to the much higher ionization density in the case of the fission fragments. Since the voltage used in the actual experiment was almost three times as high as required for saturation, this should not cause any difficulty, however. The purity of the gas was tested before every run by checking the saturation voltage for alpha particles.

The output of each amplifier was put onto the horizontal plates of a 5" oscilloscope tube. The operating voltages of the tubes including the high voltage were supplied by a special stabilized supply. The two tubes were photographed simultaneously on "S.B. Pan" film by a General Radio oscillograph recorder camera. No sweep was used on the oscilloscope. Successive pulses were displaced by the motion of the film, which moved about one foot a minute. There are of course more than five times as many pulses from the half of the chamber facing the unsupported side of the active film than from the half facing the supporting grid. In order to avoid confusion which might be caused by a large number of pulses which do not have mates, the intensifiers of both oscilloscopes were triggered by the amplifier carrying the smaller number of pulses. The photographic tracks were projected in a microfilm reader and measured with a transparent scale.

The neutron source used for these measurements was the chain reacting "water-boiler" of Los Alamos. A beam of thermal neutrons was allowed to emerge from the face of the graphite column after passing through 4" of bismuth to reduce the gamma ray ionization. About 30 pairs per minute were observed with Pu^{239} and about 100 with U^{235}. In the latter case the intensity of the source was reduced somewhat below its maximum value to prevent occasional overlapping of pulses in the chamber half facing the unsupported side of the film.

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Several films, each running about 100 minutes, were taken with each of the three substances and the best ones selected for final evaluation. The criteria for selection were absence of extensive straggling similarity of the data for both chamber halves and good clear photographic traces. The differences between various records were small, but some of the active zapon films gave consistently cleaner results than others.

The evaluation of the observed ionization by the fission fragments in terms of kinetic energy was based on the assumption that the same energy is expended by them in producing an ion pair as in the case of alpha particles. The gain of the amplifiers was set so that the output pulse height of the alpha particle pulses due to the activities of the foils was approximately the same as that of the fission pulses during neutron exposure. These pulses were photographed on the double oscilloscope. "Step" pulses from a pulse generator of the type known on the project as "Model 100 Precision Pulser" were then put on the screen grids of the chamber and photographed in the same fashion. In this manner it was found that 1 Mev of particle energy corresponded to 0.842 millivolt applied to the screen. This value agreed within the uncertainty of the calculation with that expected from the calculated capacity of the chamber. It was identical within the experimental uncertainty of about one percent for both sides of the chamber. At the beginning of each neutron exposure the calibration was verified using an electronic pulse height measurement which has been found to agree with the value found by the photographic method. Then, with the amplifier gain set at the same value as used to observe fission pulses, pulses from the precision pulser between 20 and 120 millivolt were photographed on the same film as the fission pulses. This served as calibration for this particular run and eliminated uncertainties of all factors from amplifier gain to those of magnification of the microfilm projector. It also checked the linearity of the amplifier and oscilloscope. The response of the entire amplifier - ocilloscope arrangement varied by less than 3 percent during several weeks. No correction was made for the energy loss of the fragments in the foil. This

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was probably on the average slightly less than one percent, and almost half of this is compensated by the energy loss of the alpha particles used for calibration. A correction of about 0.5 percent should perhaps be applied to all our energy values but this is considerably less than the uncertainty in the absolute values. The comparison of the three fissionable substances is, of course, not affected by this.

The ionization "noise" due to radiations from the neutron source was equivalent to a particle energy of about 1.5 Mev.

A measure of the resolution of the measurements is obtained from the fact that the alpha particle groups from the foils showed a width at half maximum of about 2.5 percent. About 97 percent of the pulses showed energies in the peak; the remaining 3 percent straggled over a considerable range. This is perhaps not as good as one might hope for, but the difficulty in keeping the films and grids free from dust may account for most of the straggling.

One phenomenon which remained without a completely satisfactory explanation was the occurance of a considerable number of fission pulses on the side supported by the grid which did not have mates on the unsupported side. Their number varied from film to film between 5 and 50 percent of the number of paired pulses. There was no relation between the fraction of such single pulses and the amount of active material or the amount of straggling. They were disregarded in evaluating the data. The most reasonable explanation seems to be that there is a creepage of active material onto the surface of the grid, including the lower surface, perhaps while the film is wet. It must be remembered that because of the rather low transmission of the grid, only a few percent of the active material need spread onto the supporting grid to cause the observed effect. A number of other possible explanations were ruled ~~out by~~ experiments.

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RESULTS

About 1600 pairs were measured for U^{235}, about the same number for U^{233}, and about 900 for Pu^{239}. Figs. 2 to 13 show histograms of our results. Figs. 14,15 and 16 show smoothed curves drawn from the same results.

The single-fragment distributions (Figs. 2 to 7) summarized in Fig. 14 were obtained in each case by combining the results of both halves of the chamber. The distributions on the side which faced the unsupported surface of the film generally showed less tailing off, possibly because of scattering in the grid. The energy values at the peaks are

	U^{235}	U^{233}	Pu^{239}
Heavy fragment (Mev)	60	59	65
Light fragment (Mev)	94	95	93
Ratio	1.57	1.61	1.43

In the case of the light fragments the three substances give very similar distributions although the shift towards lower energies for Pu^{239}, shown in Fig. 14, seems outside the experimental error. The heavier fragments show nearly identical distributions for the two uranium isotopes but in plutonium the peak is clearly shifted to higher energies. The ratios of the peak energies seem to indicate that the fission of plutonium is appreciably more symmetrical than that of U^{235} and that of U^{233} is slightly more asymmetric. We shall presently see other evidence for this.

Another point of interest in connection with the single-fragment curves is the amount of overlap of the two groups. It amounts to about 4 percent of all fragments for the uranium isotopes and to about 6 percent for plutonium. This is the order of magnitude of the straggling observed with alpha particles in our arrangement. Jentschke found appreciably less overlap and Snyder (LAMS 299) found no pulses at all at the bottom of the "valley" for U^{235}, using a single ionization chamber. It seems therefore likely that a large fraction of the overlap observed by us is instrumental in origin.

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In that case it would be expected that the overlap is greater in the case of plutonium since the "valley" is narrower so that the same experimental broadening will have a more pronounced effect. Probably there is very little true overlap in either of the three substances.

The total kinetic-energy release (Figs. 8, 9, and 10), i.e. the sum of the energies of the two fragments of each pair, is summarized in Fig. 15. The most probable energy release is for U^{235}: 153 Mev; for U^{233}: 151 Mev; and for Pu^{239}: 156 Mev. In the latter case the peak seems to be shifted somewhat towards higher energies with respect to the wings of the distribution. This again is connected with the fact that the more symmetrical modes of fission are more probable in plutonium than in the uranium isotopes, since these modes show a higher energy release. The mean energies, in the smae order as the most probable energies given above, are 153, 151, and 153 Mev, respectively, clearly indicating the asymmetry in the distribution for Pu^{239}.

The ratios of the kinetic energies of the members of the pairs of fragments (Figs. 11, 12, and 13) are summarized in Fig. 16. These distributions show more clearly the degree of asymmetry of the fissions in the three cases. The most probable ratios are for U^{235}: 1.49; for U^{233}: 1.52; and for Pu^{239}: 1.32. These show exactly the same trend as the ratios of the peaks in the single particle curves. Because of the conservation of momentum the ratios shown in Fig. 16 should also represent the mass ratios of the pairs. This statement must be modified somewhat because of the momentum imparted by the prompt neutrons. These neutrons are probably emitted isotropically from the moving fragments with velocities comparable with those of the fragments themselves. Thus their effect will be to cause one particular initial mass ratio to appear as a spread of ratios of kinetic energies, the maximum spread being about 7 percent in the ratio for the cases where all of the neutrons come from the slow fragments in the forward direction, compared with those where they are emitted from the fast fragment in the backward direction. These extreme cases are certainly quite rare. If there is a definite tendency for the neutrons to come from the ~~heavy or from the light~~ fragments, and the

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final mass ratios will be different from the initial ratios deduced from the kinetic energies. The curves of Fig. 16 do not quite indicate the relative probabilities of certain mass numbers in the fission of the three substances since the ordinates are drawn for equal intervals instead of equal mass number intervals. In Fig. 17 we have drawn the expected fission yields for various mass numbers from the three substances. These curves were obtained by redrawing Fig. 16 for equal mass intervals, and using the final total mass, i.e. the mass after emission of the average number of prompt neutrons. Although the figure is somewhat confusing, closer inspection shows the following features: the distribution of the heavy group is almost identical for all three substances; the most probable heavy mass is about 140, perhaps a little lower in the case of plutonium; the most probable light mass for U^{235} is about 93 (the distribution for U^{233} is shifted by two units towards lower masses and that for Pu^{239} is shifted almost 5 units toward heavier masses). The highest expected yield for one mass number is about 5 1/2 percent in all cases. Also on Fig. 17 we have indicated by the dashed and dotted lines the fission yields observed by chemical methods. We are indebted to Dr. Sugarman for these curves. It can be seen that the agreement is very good both for U^{235} and plutonium. There is no chemical evidence in the case of U^{233}. The location height of the peaks is almost identical by the two methods. The greater width of the curve obtained by the fragment energy method is due partly to the effect of the recoil from prompt neutrons as discussed above and partly to experimental uncertainties. There is some indication that the chemical method indicates slightly greater symmetry of fission than do our experiments. This may be explained by assuming a tendency for the neutrons to leave the heavier rather than the lighter fragments. Or it may be due to experimental effects such as a slight difference in the energy required to make an ion pair in argon between the faster and the slower fragments. It may be noted in passing that if there were a difference in the probability of prompt neutron emission between the more and the less symmetrical modes of fission, the "heavy" and "light" peaks as found by chemical methods should show different heights and widths. No such difference seems indicated.

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Figs. 18, 19 and 20 show a somewhat different ~~presentation of our~~ results. The abscissa shows the total energy release, the ordinate, the energy ratio; while the density of the dashes indicates the frequency of occurrence of a particular combination of ratio and energy. This presentation is the equivalent of a three-dimensional plot. In Fig. 19 we have also drawn in the lines corresponding to definite values of the energy of the light or the heavy fragments separately. These plots really summarize the results completely. Adding "columns" on the graphs we should obtain the curves of Fig. 15; adding "rows", the curves of Fig. 16. Adding strips along the lines of the grid drawn in Fig. 19, we get the curves of Fig. 14.

We can see at a glance that one particular energy of a light fragment may correspond to all possible values of the mass ratio. On the other hand there is a definite correlation between the energy of the heavy fragment and the mass ratio, low values of the energy corresponding to high mass ratios, and vice versa. The graphs are cut off arbitrarily at energies below 130 Mev to save space. Only a very small number of pairs had energies less than this value and these showed all possible mass ratios.

In Figs. 21 and 22 we plot the average and maximum energy release, respectively, for various mass ratios for the three substances. The smooth curve in Fig. 22 represents the calculated maximum energy for U^{235} as given Jentschke[1]. Although slightly better packing fraction values are now available it did not seem worth while to recalculate this curve. Unfortunately the average energy (Fig. 21) is greatly influenced by the small number of straggling tracks, while the maximum energy (Fig. 22) is a somewhat accidental quantity since it depends on the last track in a distribution. The following conclusions seem fairly reliable, though: the dependence of maximum energy release on mass ratio follows roughly the expected behaviour; the observed variation is about 20 percent for the extreme modes of fission; the energy release for the same ratio of splitting does not differ greatly for the three substances investigated. There is some indication that the energy release depends somewhat more steeply on mass ratio in the case of plutonium than in the case of uranium. Some crude considerations of packing fractions indicate that plutonium should release about 4 Mev more energy

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on fission than U^{235} for the same mass ratio. However about 2 Mev, on the average, is used up to provide the greater number of prompt neutrons. The remaining 2 Mev may well be only partly released as kinetic energy, the rest going into nuclear excitation. It is not surprising, therefore, that no difference in energy release for the same mass ratio is observed in our measurements. The greater energy release by plutonium is apparently due only to the tendency to more symmetrical fission.

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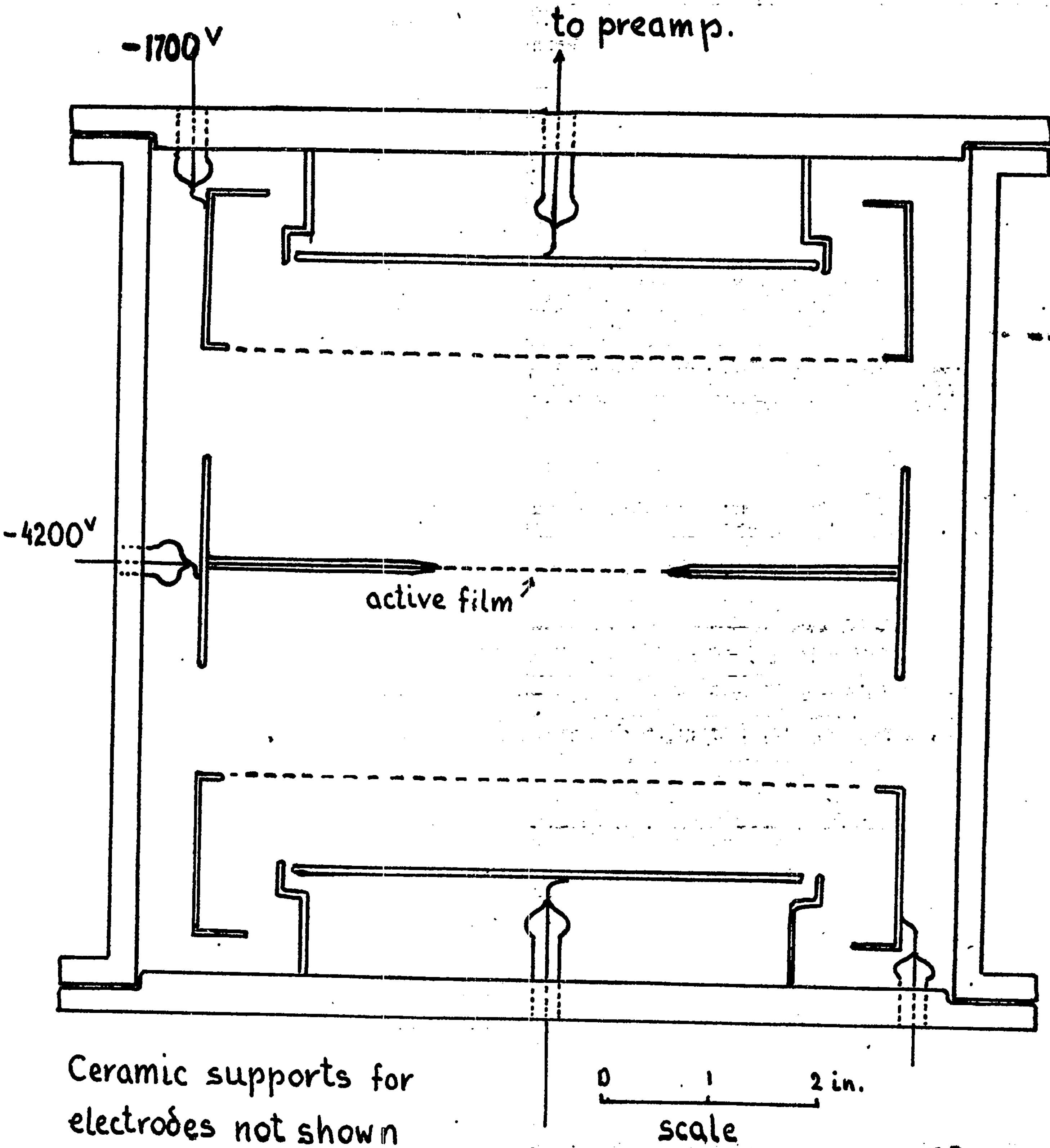
Fig. 1

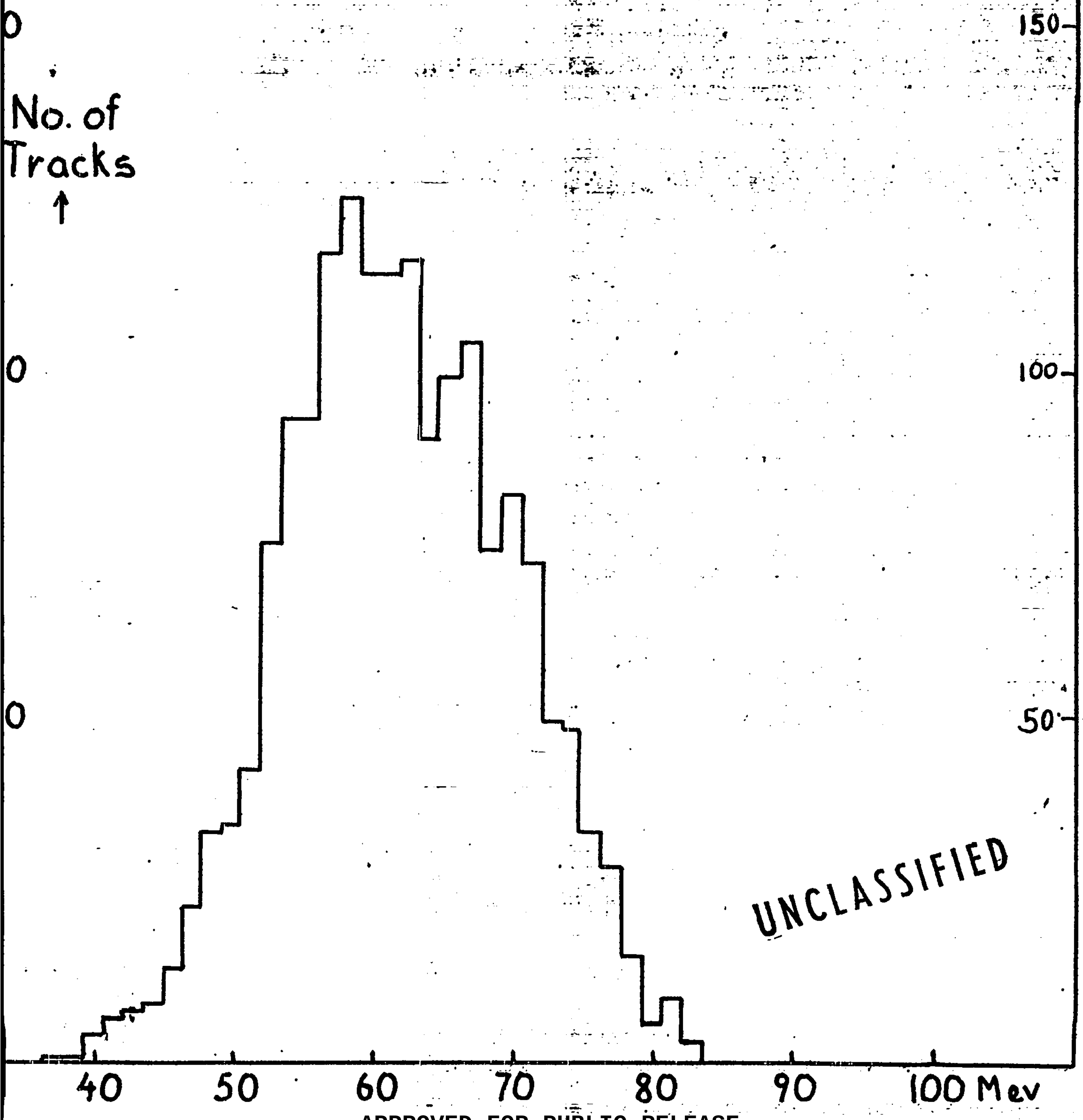


Ceramic supports for electrodes not shown

U^{235}

Energies of the Heavy Fragments

Fig 2



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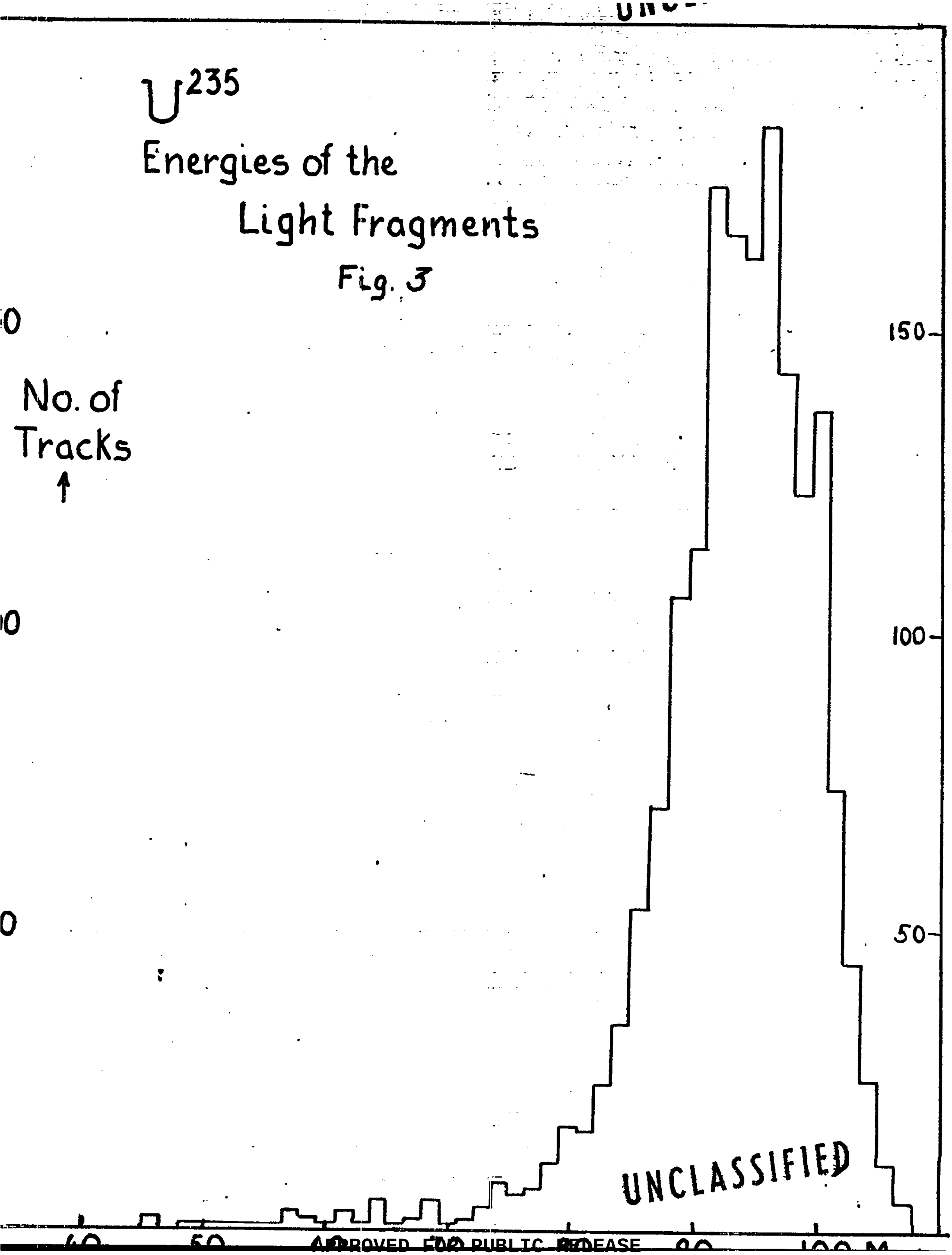
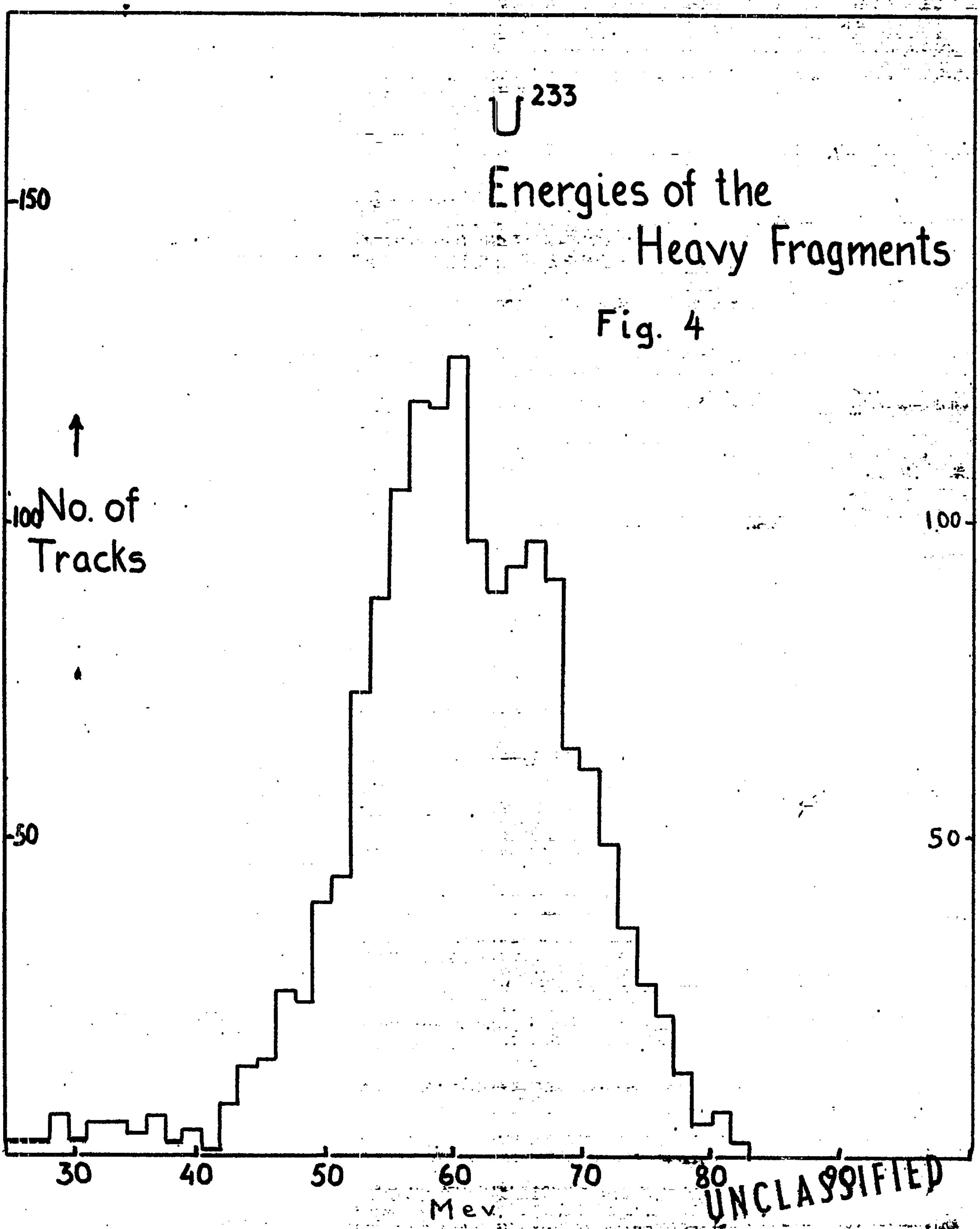
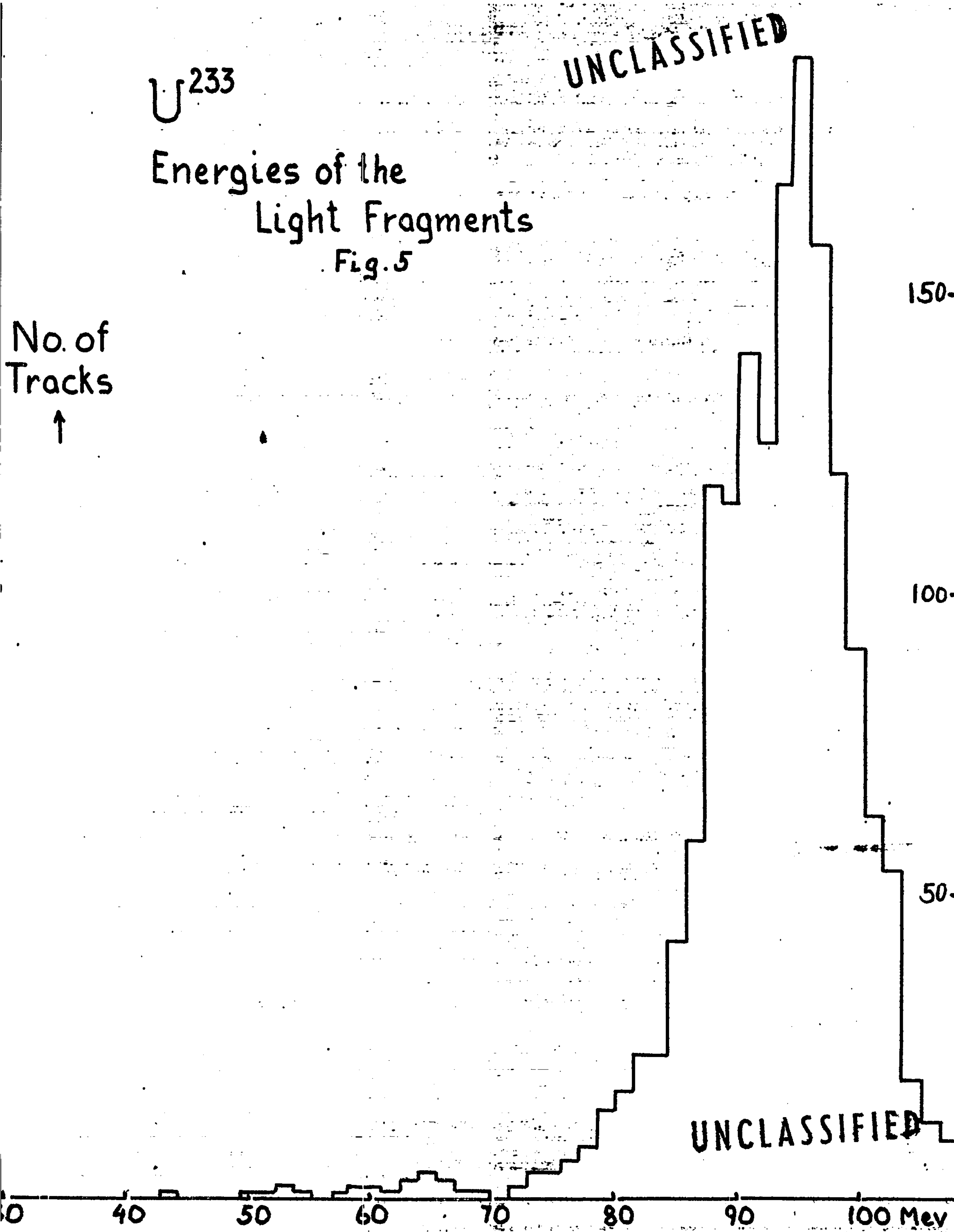


U^{235}

Energies of the Light Fragments

Fig. 3


U^{233}
Energies of the Heavy Fragments
Fig. 4
No. of Tracks
150
100
50
100
50
30
40
50
60
70
80
90
Mev.



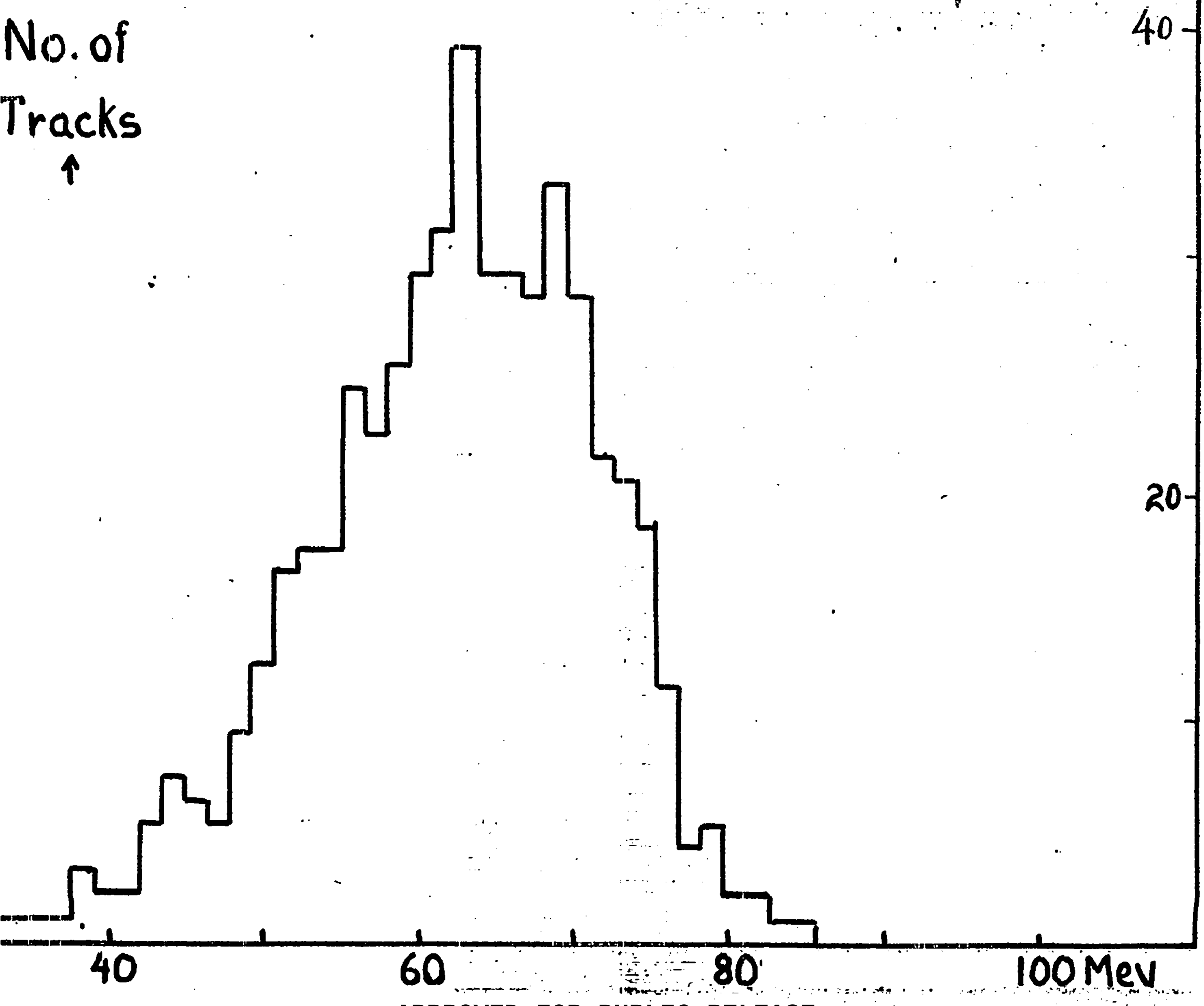
U^{233} Energies of the Light Fragments

Fig. 5

Pu^{239}

Energies of the
Heavy Fragments

Fig. 6



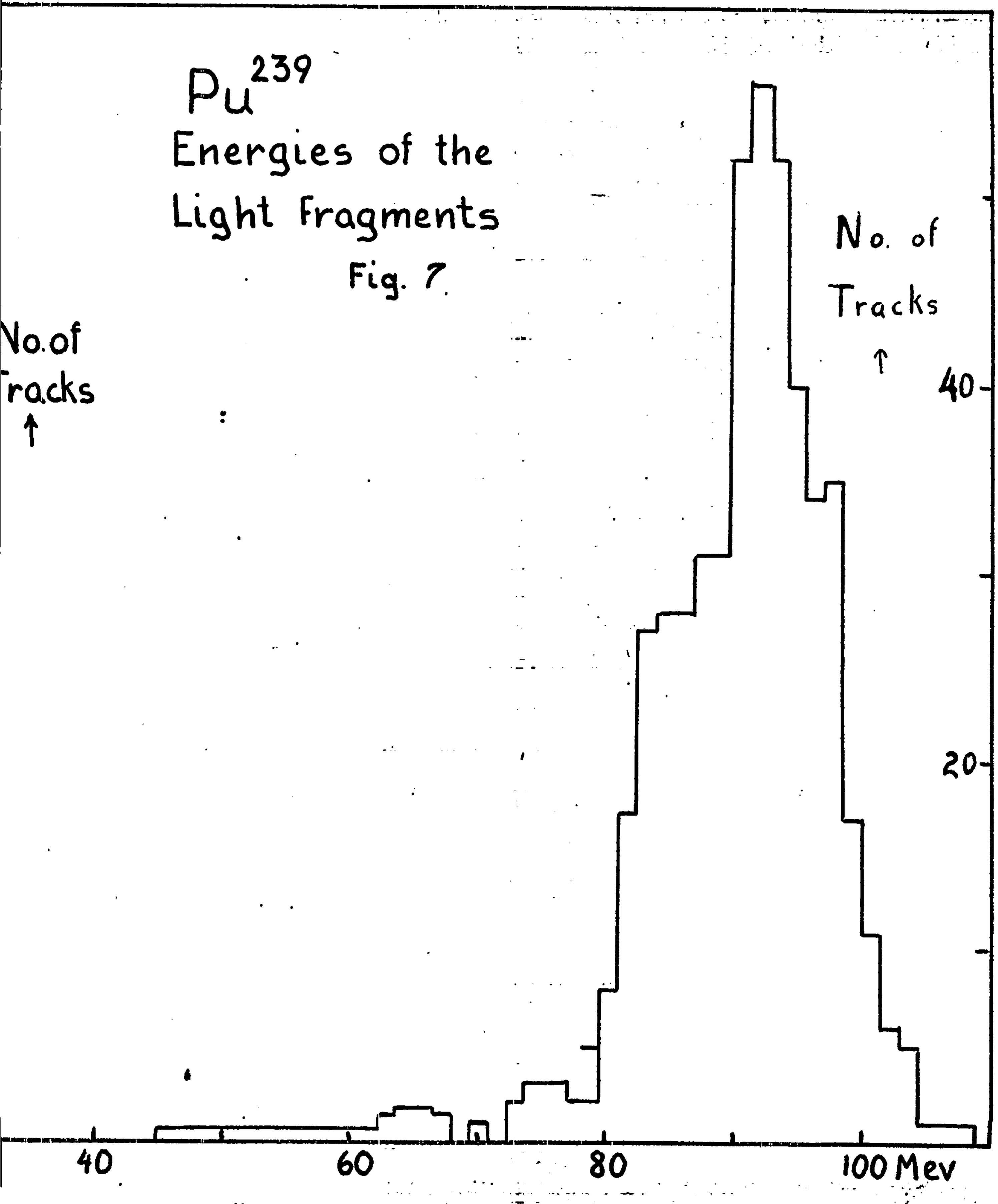
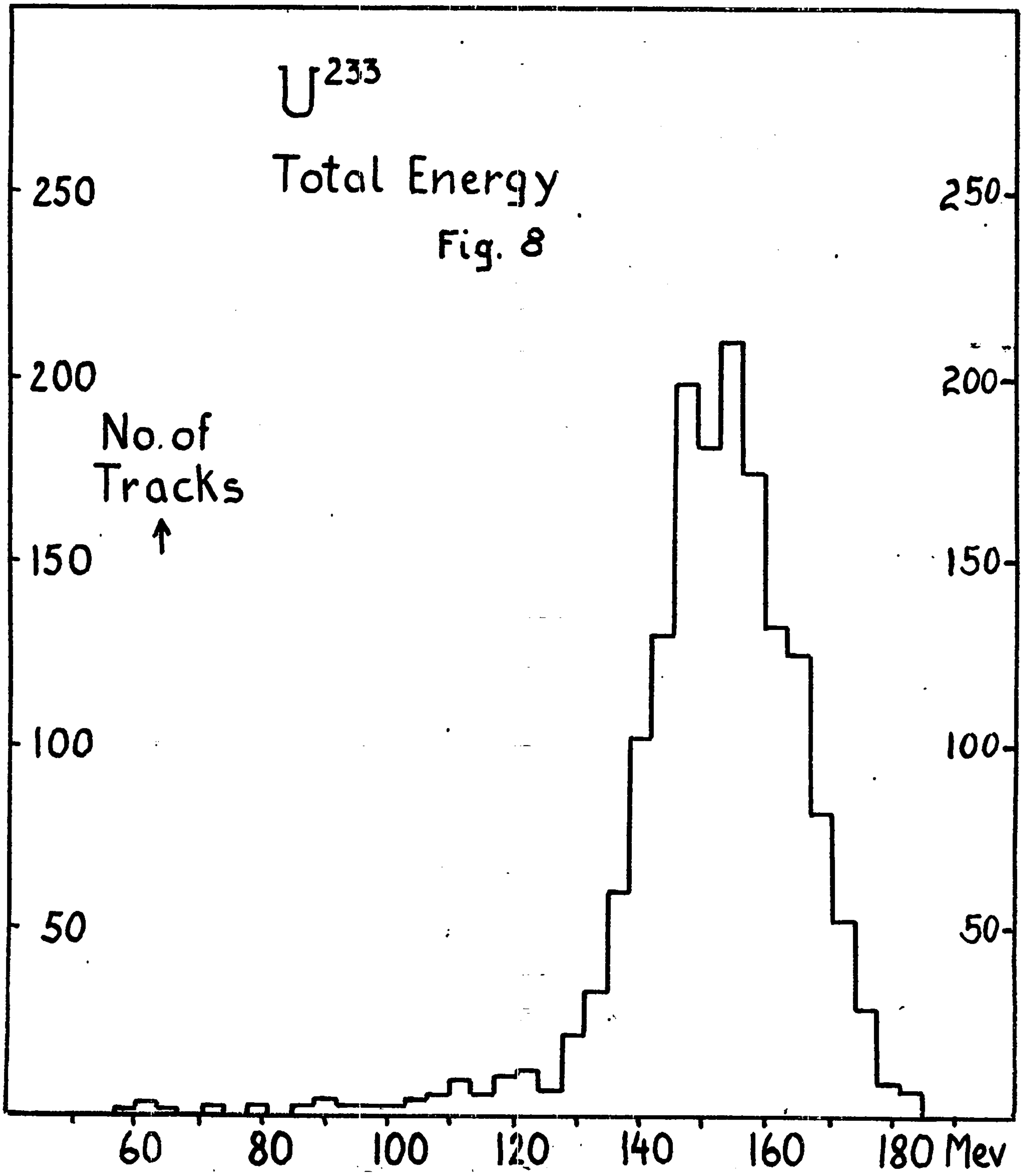

Pu^{239}
Energies of the
Light Fragments
No. of
Tracks
No. of
Tracks
40
20
40
60
80
100 Mev

Fig. 7



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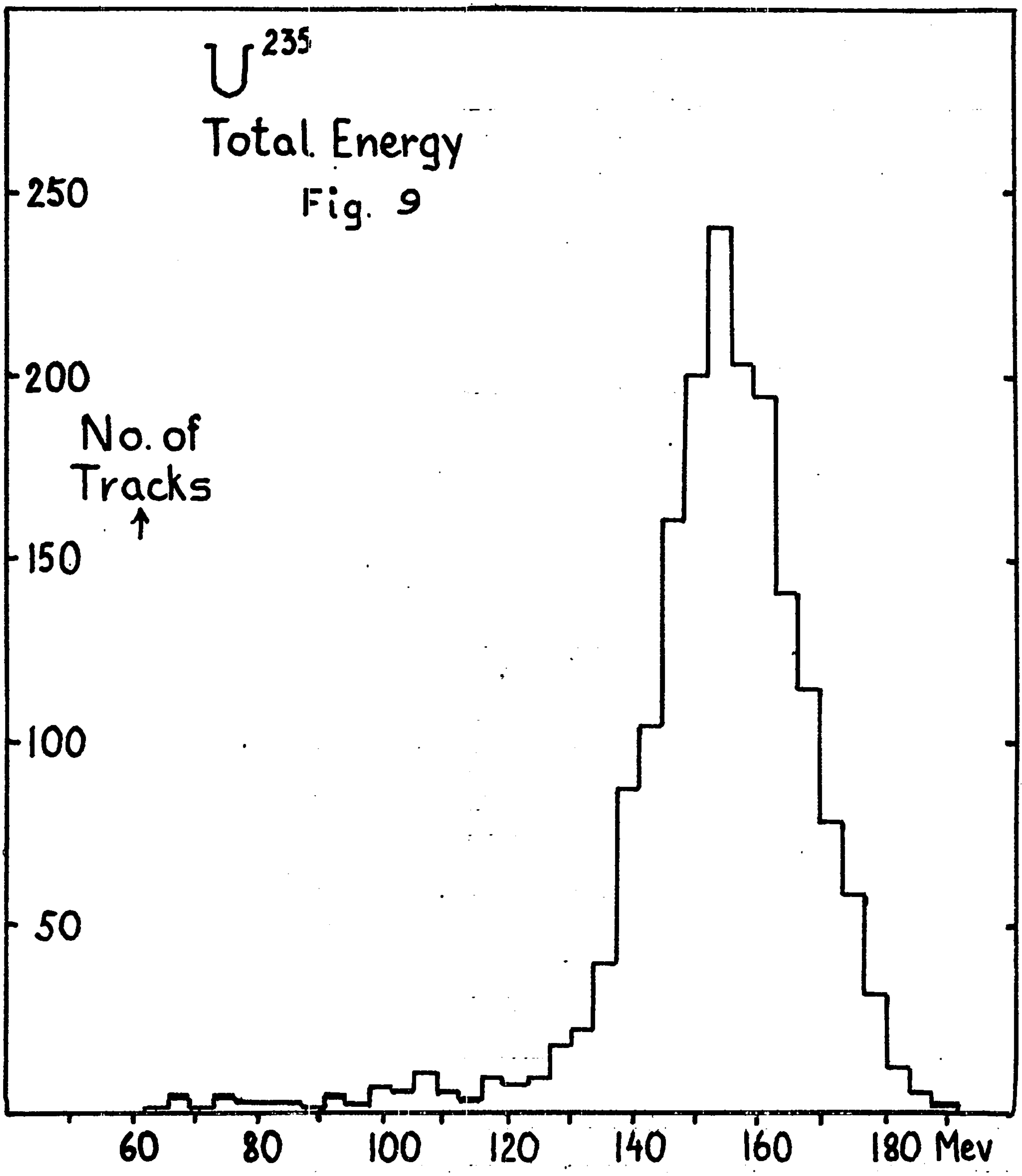

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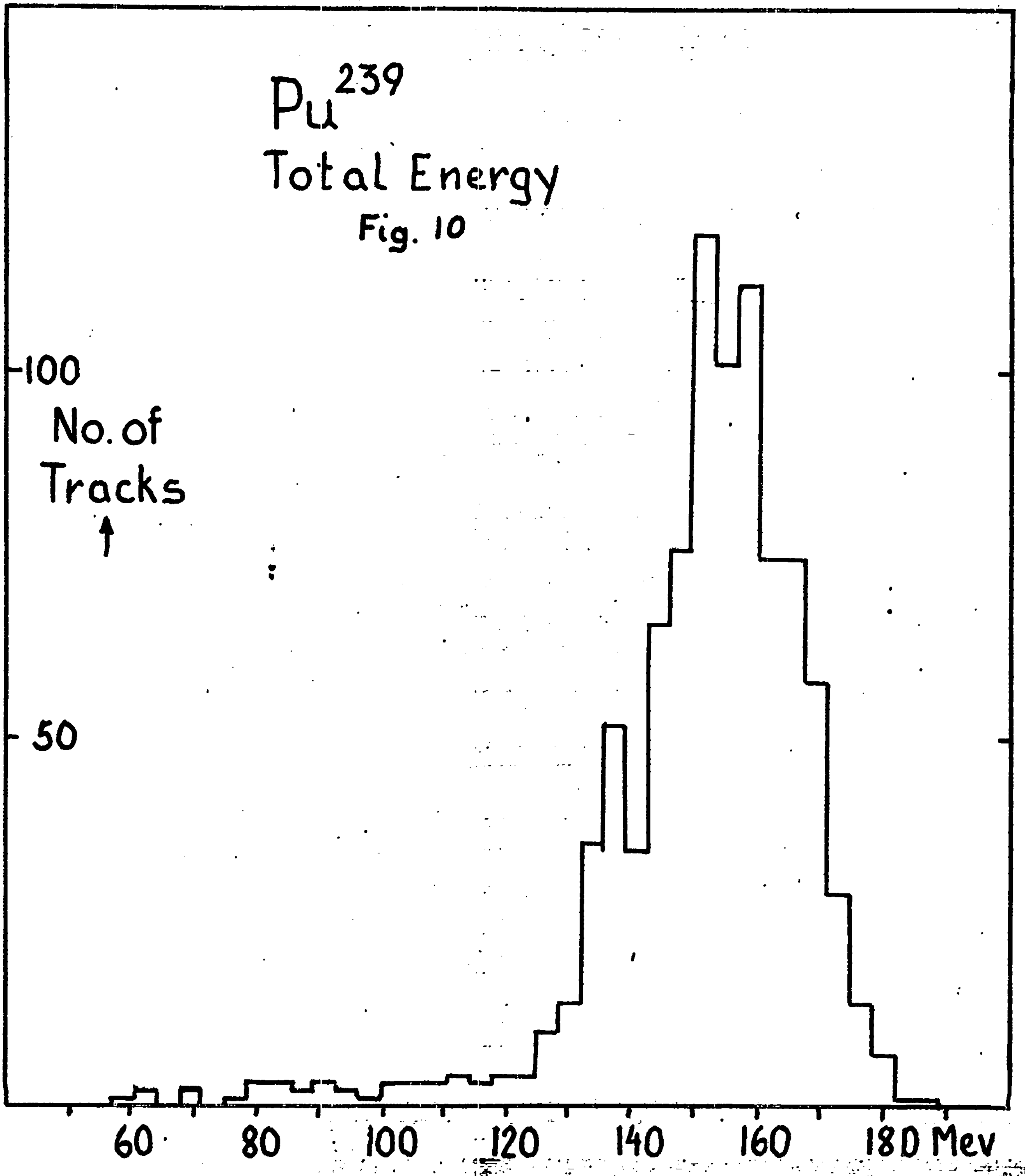

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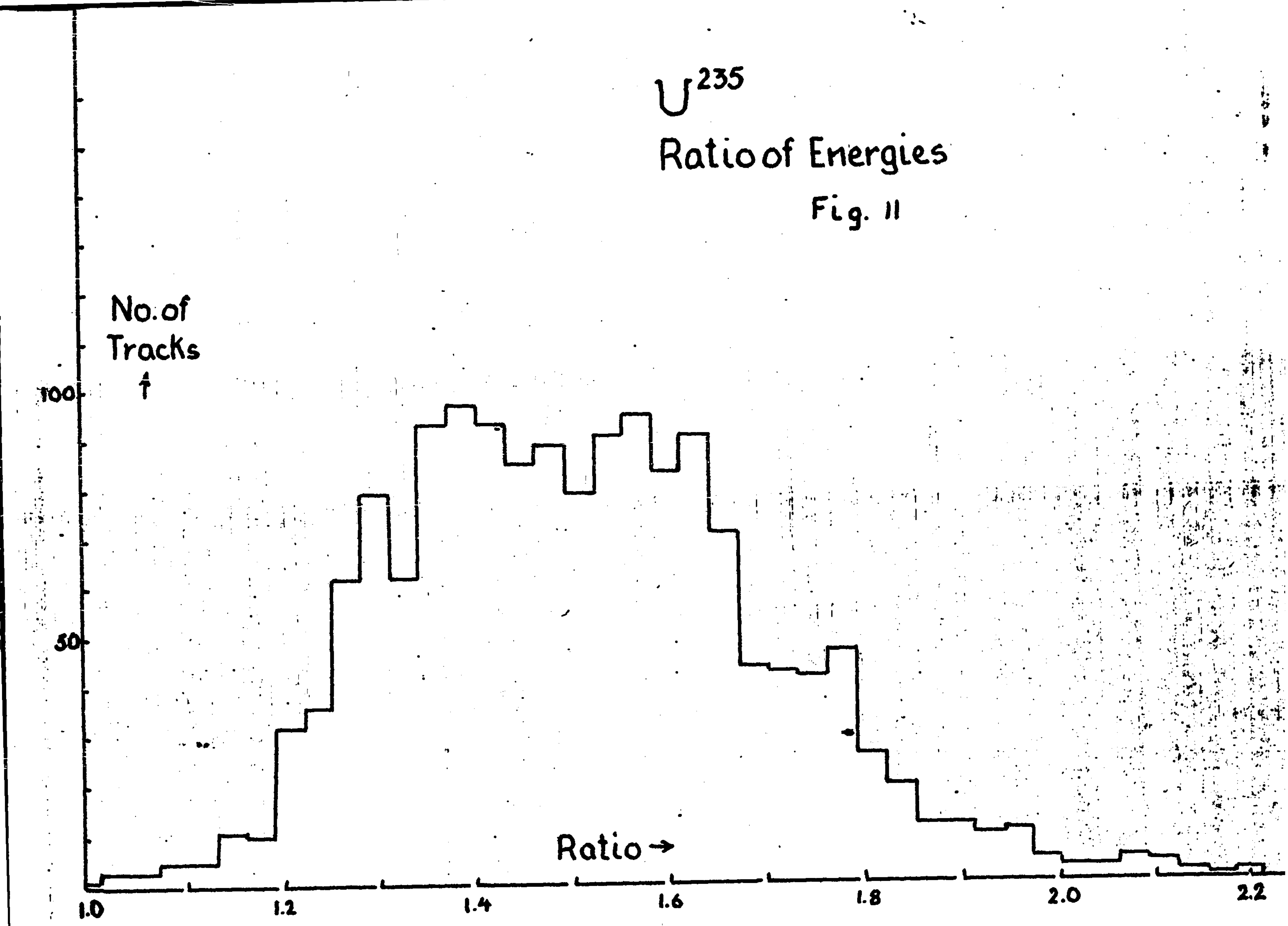

Fig. 11

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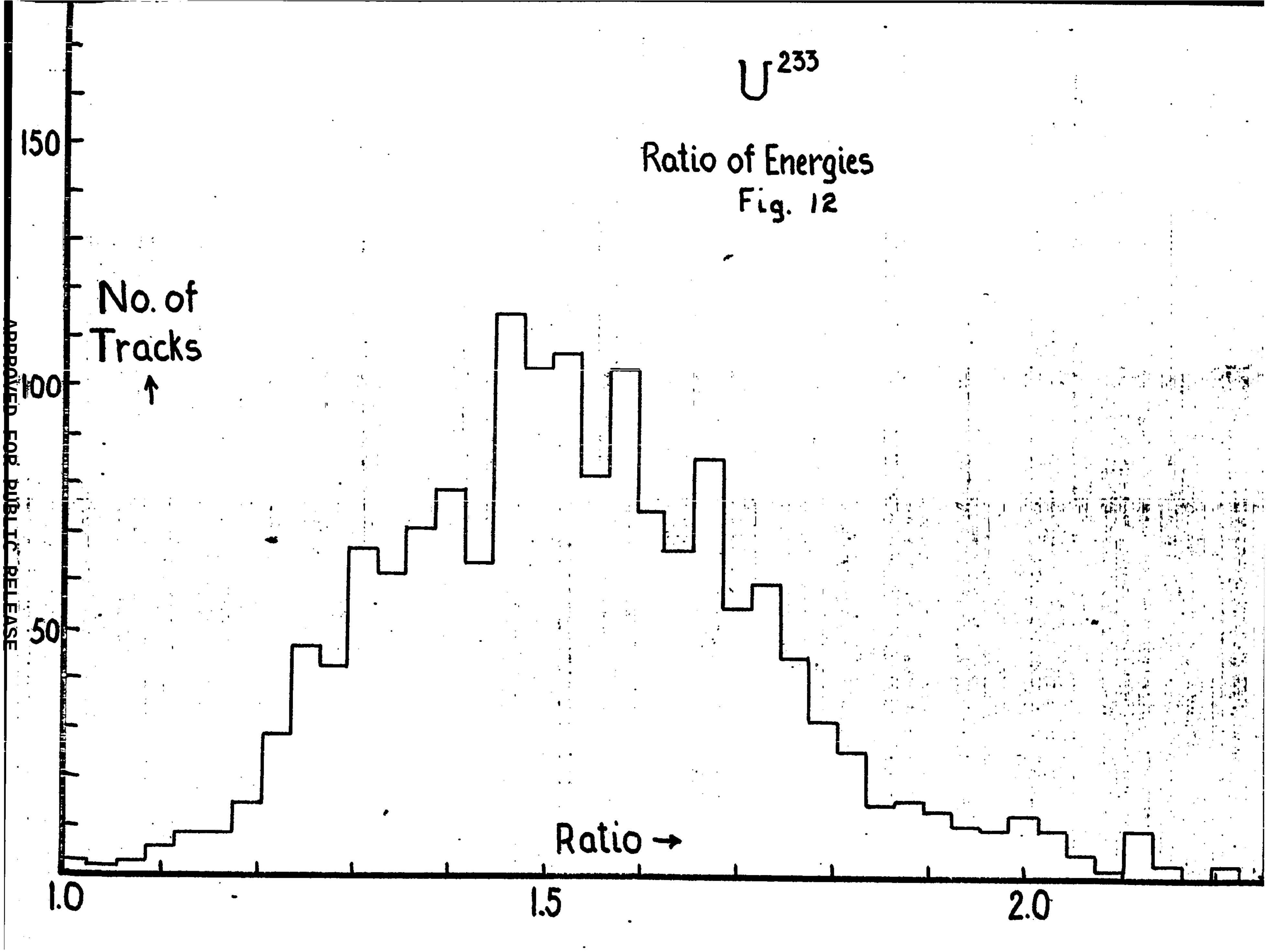

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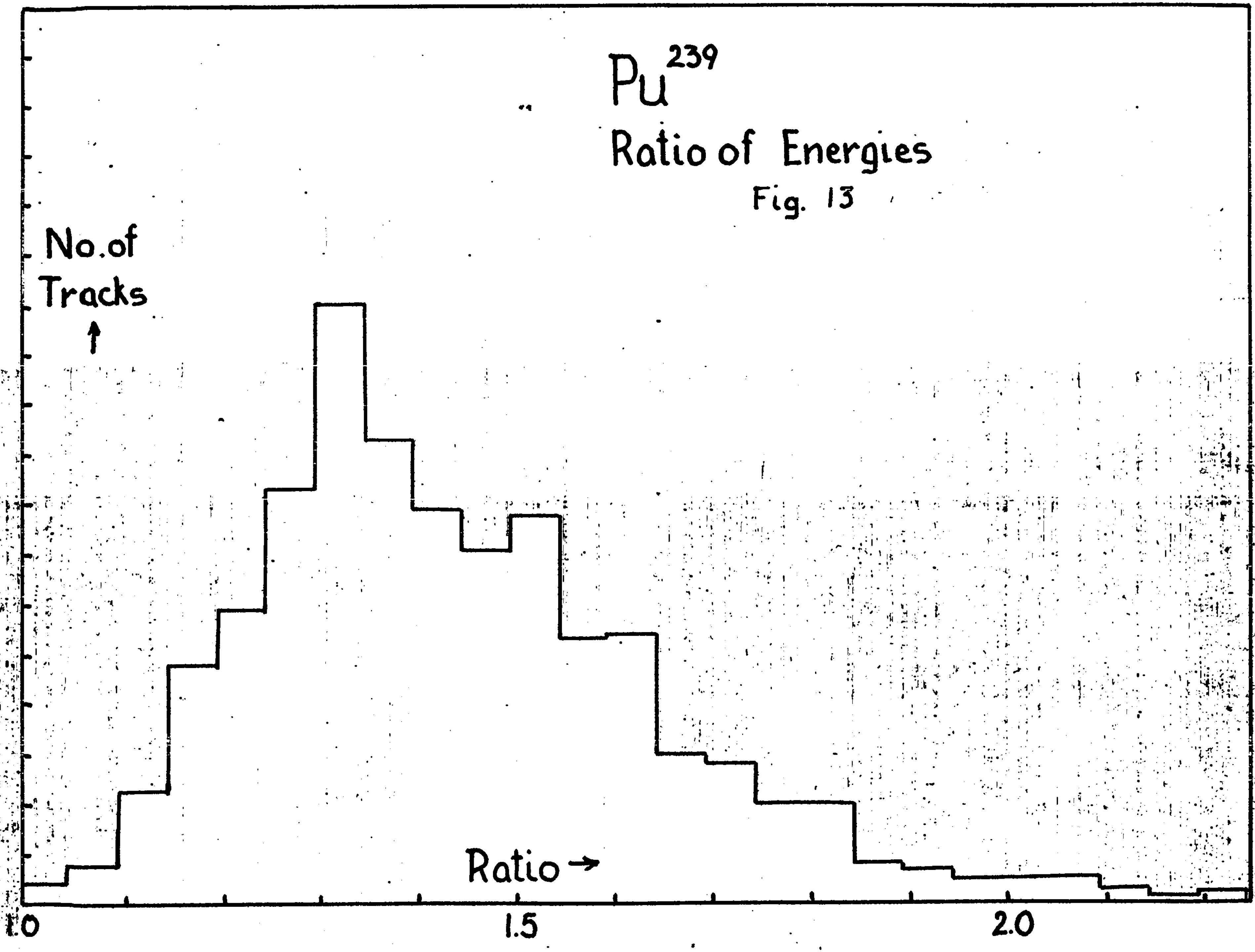
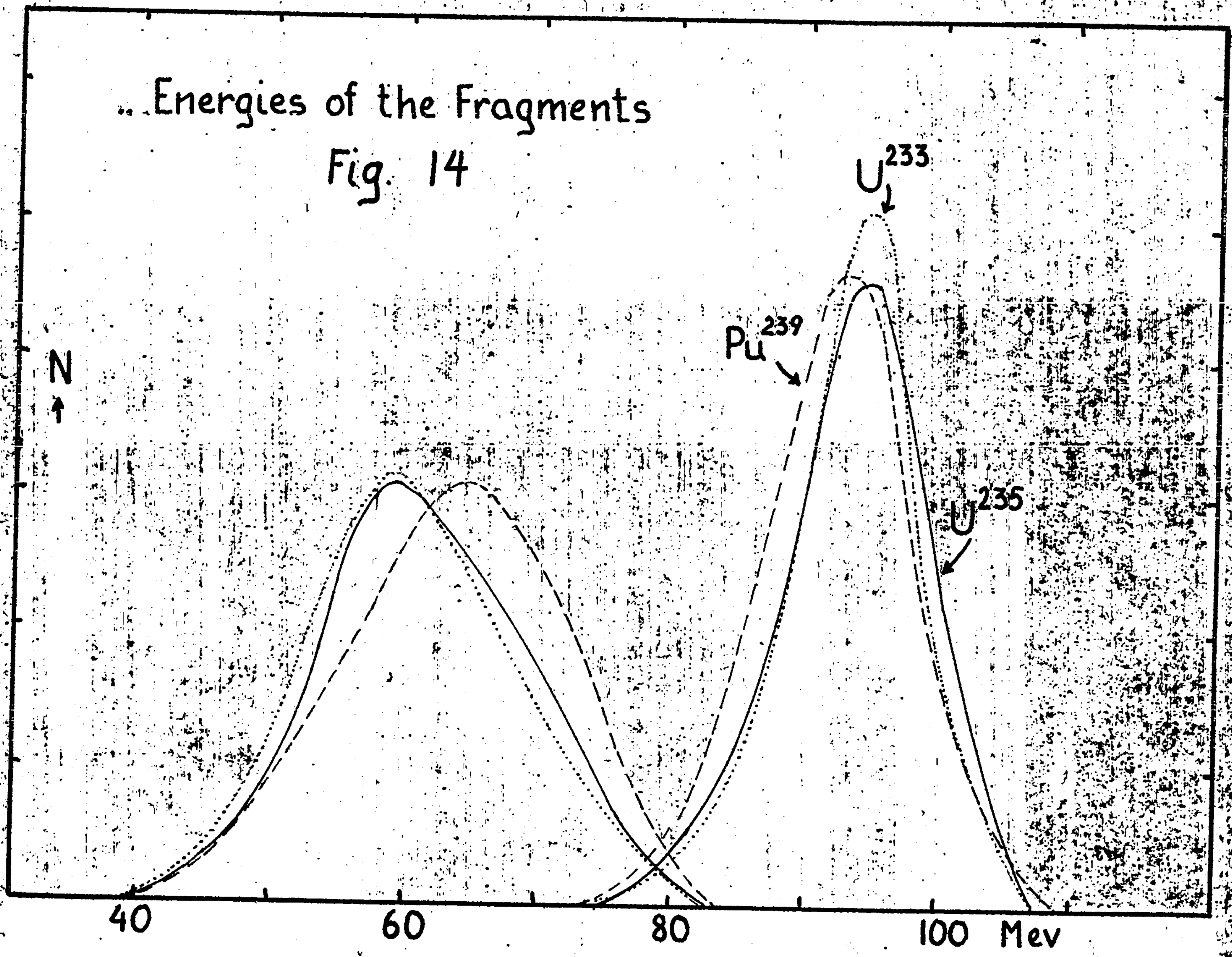
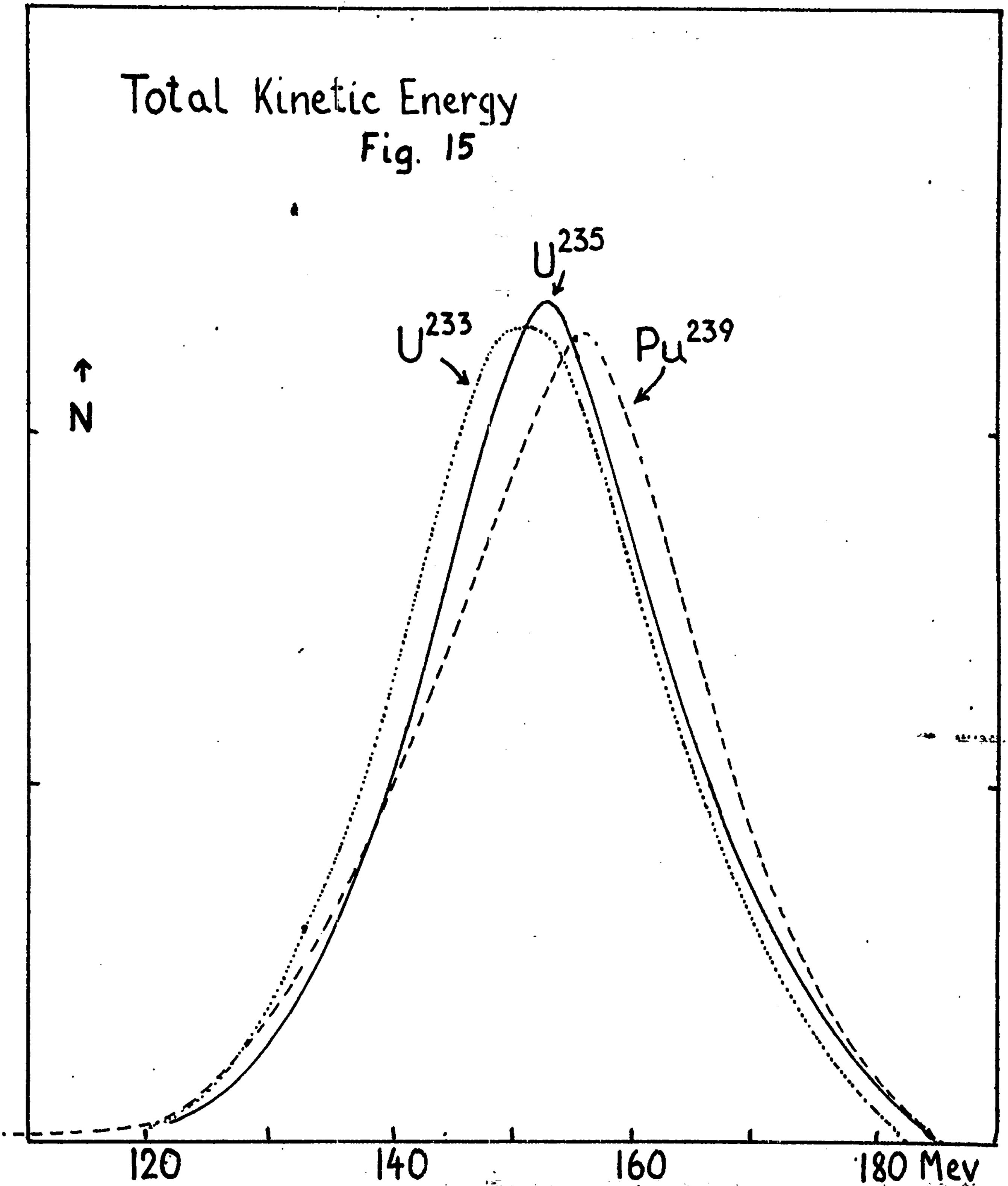

Pu^{239}
Ratio of Energies
Fig. 13
No. of Tracks ↑
Ratio →
1.0
1.5
2.0

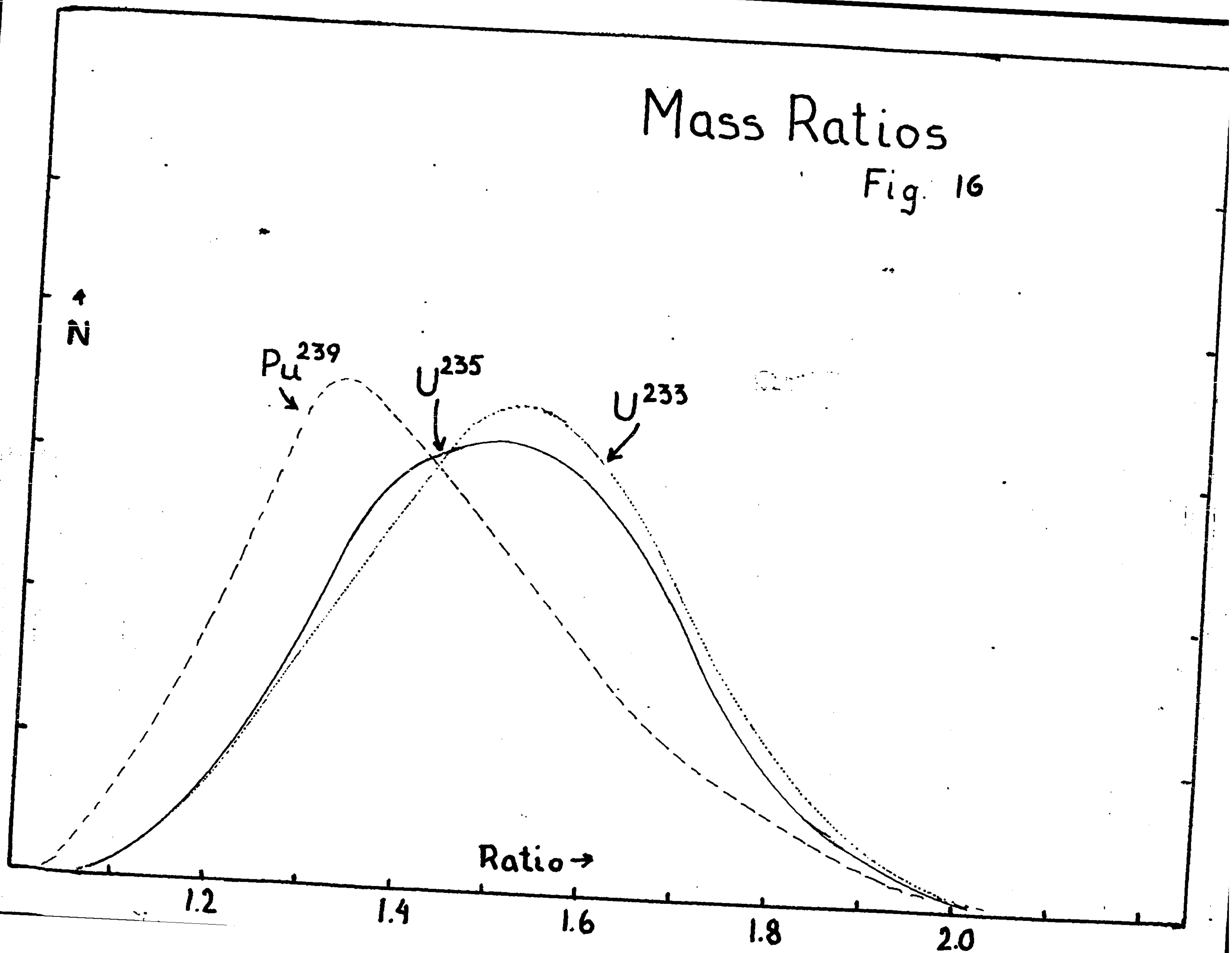


Energies of the Fragments

Fig. 14



Total Kinetic Energy
Fig. 15
U^{235}
U^{233}
Pu^{239}
N
120
140
160
180 Mev


Mass Ratios
Fig. 16
N
Pu^{239}
U^{235}
U^{233}
Ratio →
1.2
1.4
1.6
1.8
2.0

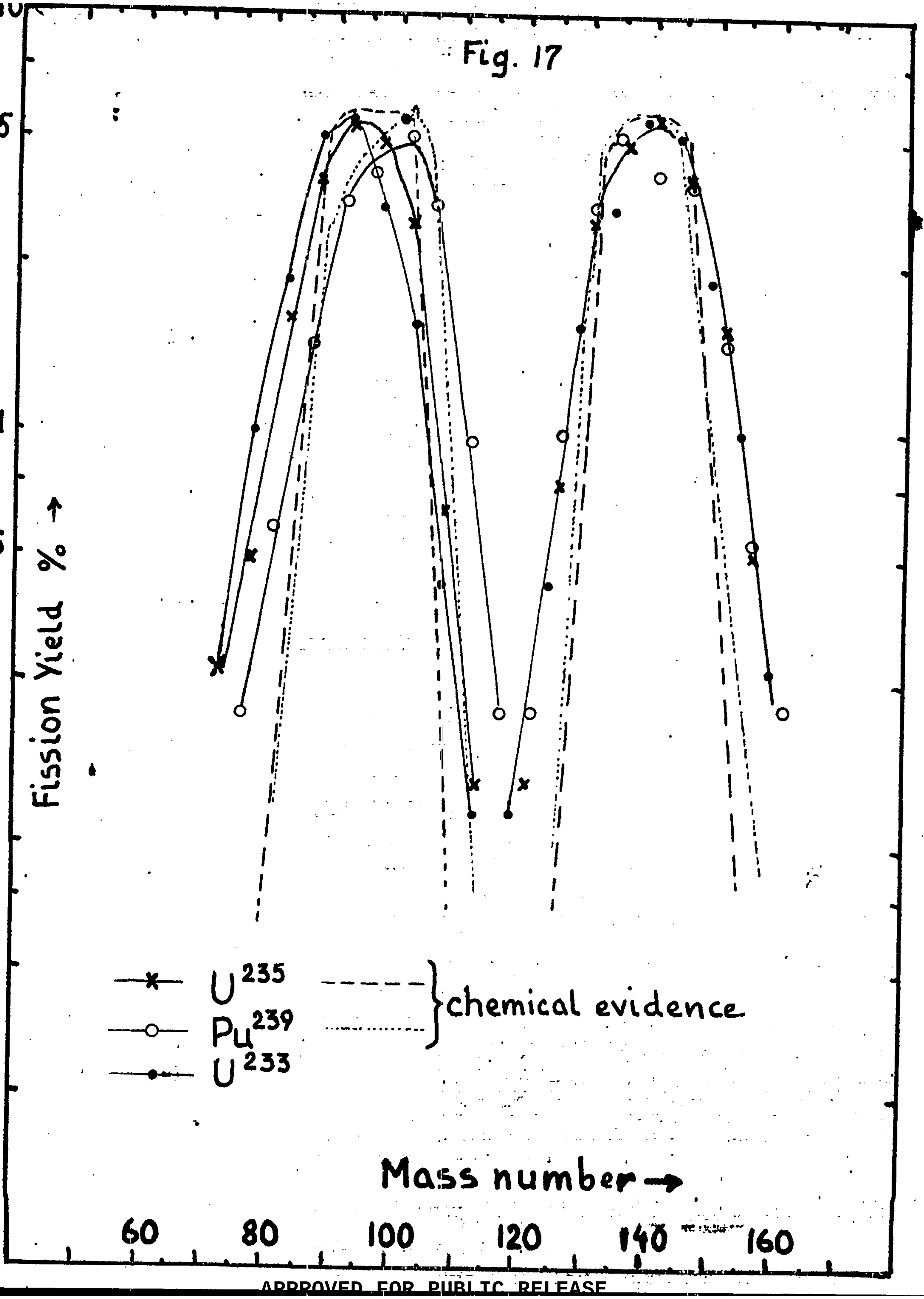
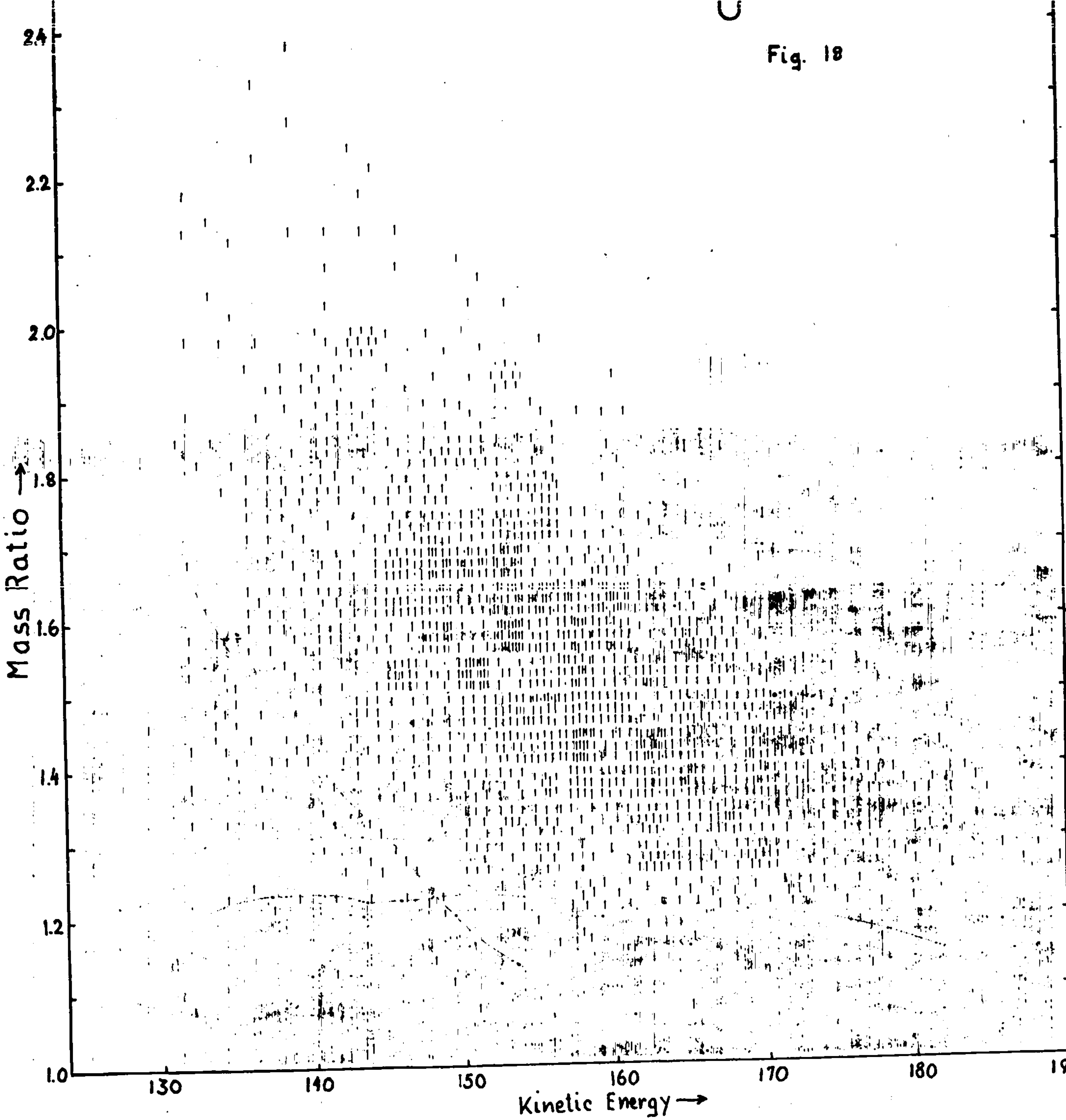


Fig. 17
10
5
1
0.5
0.1
Fission Yield % →
U^235
Pu^239
U^233
chemical evidence
Mass number →
60
80
100
120
140
160

Fig. 18



Mass Ratio →
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
130
140
150
160
170
180
190 Mev
Kinetic Energy →

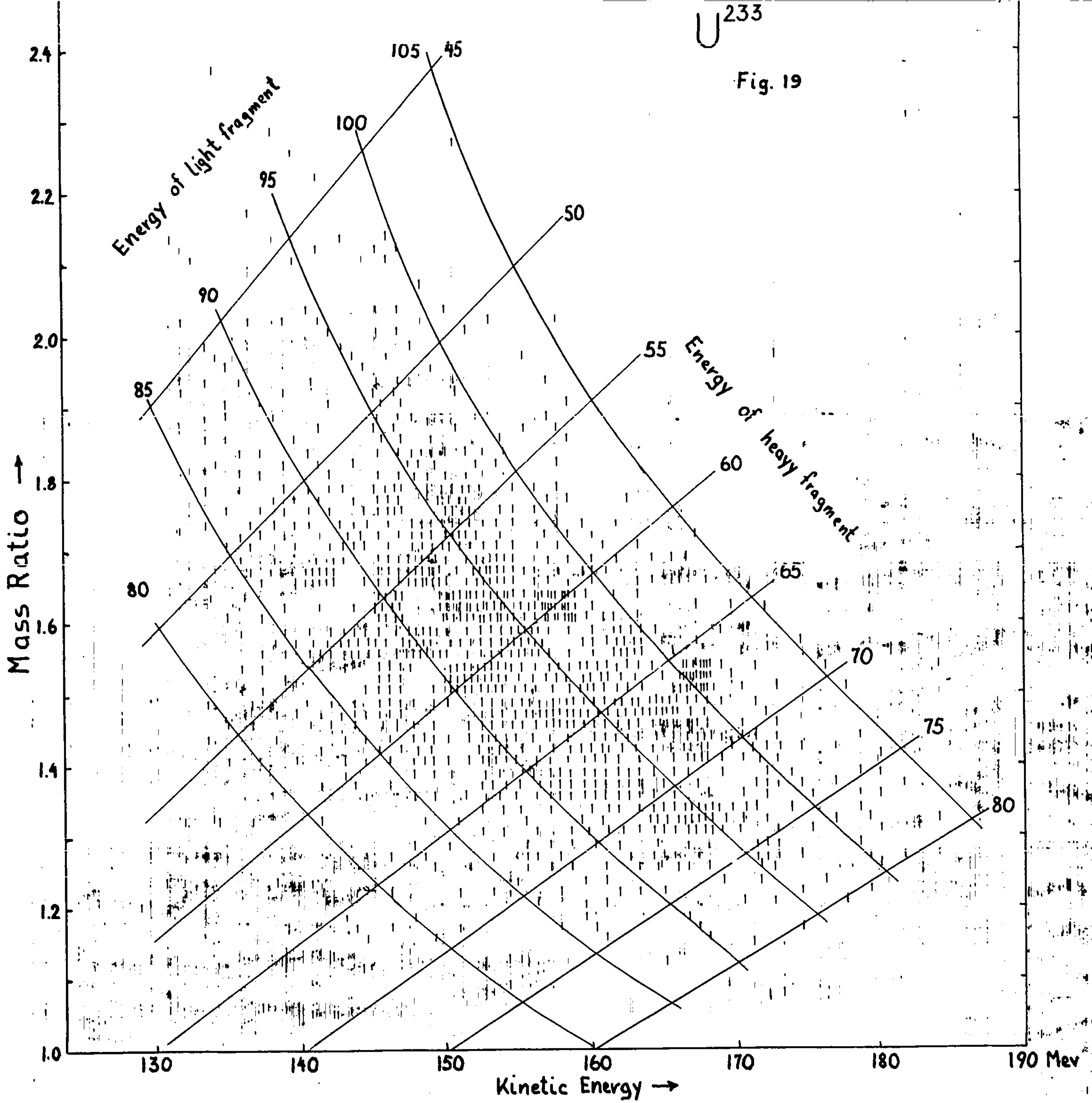


Fig. 19

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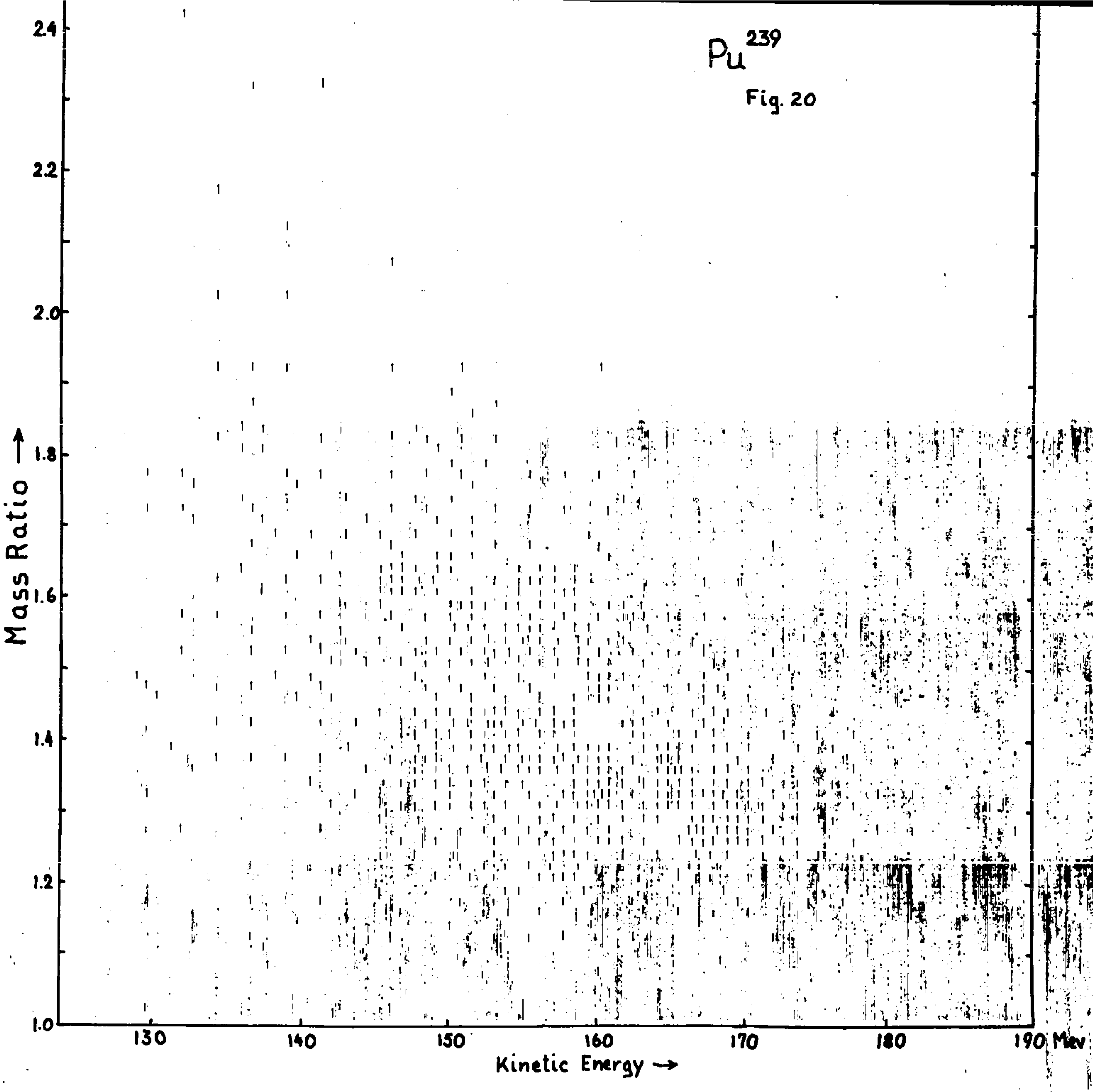
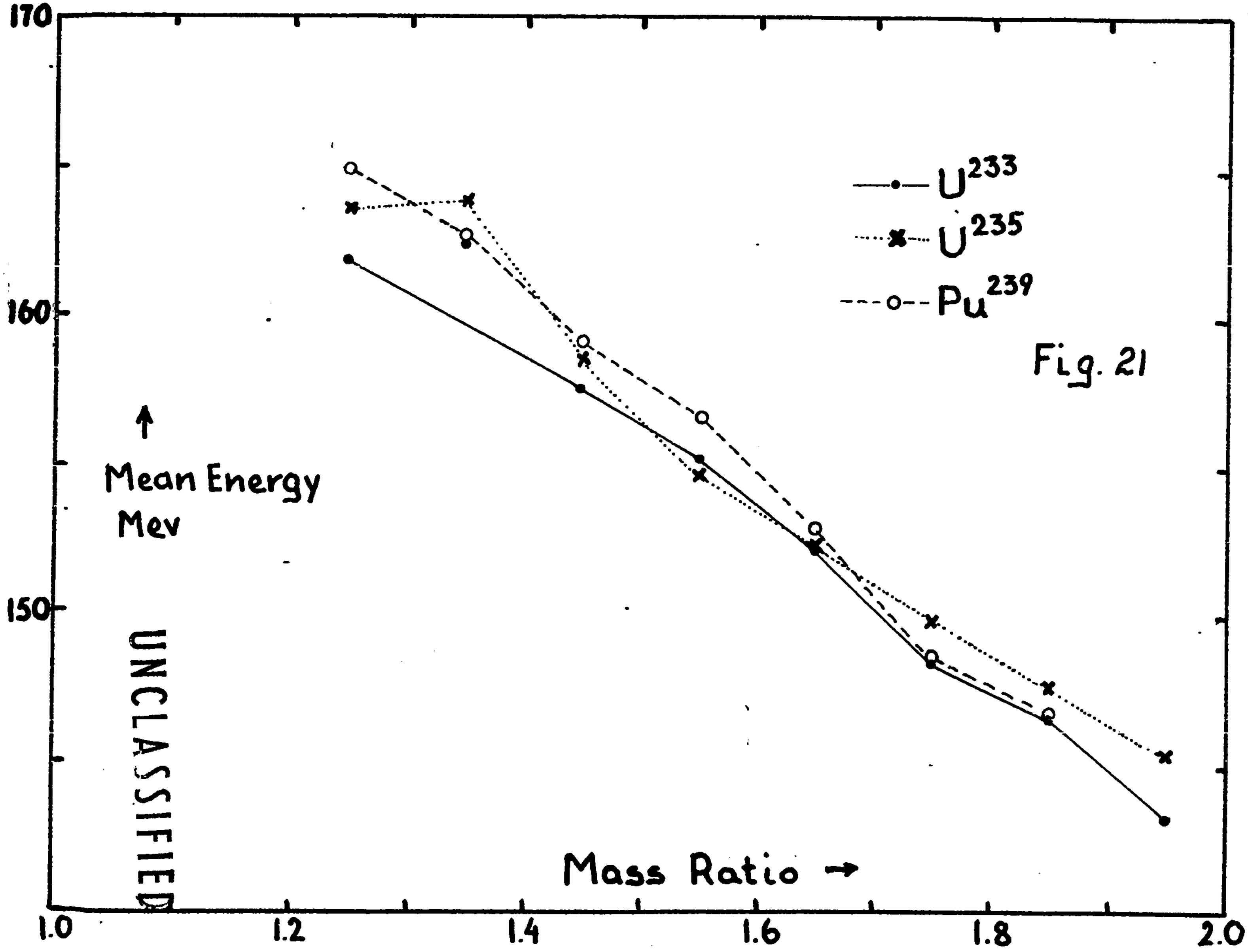


Pu^{239}
Mass Ratio →
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
130
140
150
160
170
180
190 Mev
Kinetic Energy →

Fig. 20



170
160
150
Mean Energy
Mev
Mass Ratio
1.0
1.2
1.4
1.6
1.8
2.0
U^{233}
U^{235}
Pu^{239}

Fig. 21

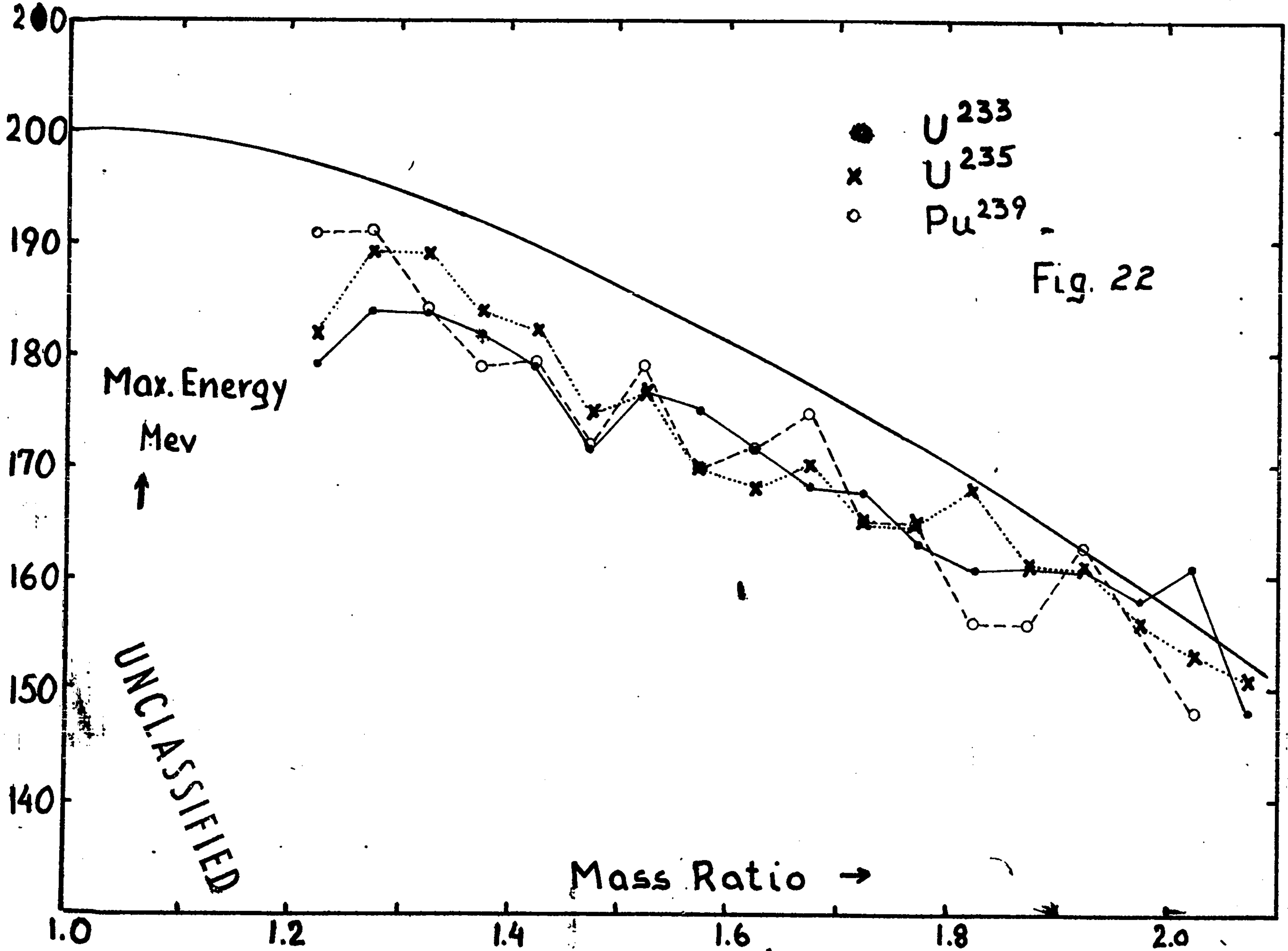

U233
U235
Pu239
Max. Energy Mev
Mass Ratio →
200
190
180
170
160
150
140
1.0
1.2
1.4
1.6
1.8
2.0

Fig. 22

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