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Prompt Gamma Rays Emitted from Individual Fragments in Neutron-Induced Fission

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ABSTRACT:

Prompt gamma rays associated with moving fission fragments of specific masses have been observed in thermal neutron induced fission of ^{235}U. The geometrical layout of the apparatus prevents detection of photons emitted from stopped fragment and determines the effective time resolution to about 1 n sec after fission. Spectra are measured by means of a high-resolution Ge(Li) detector with 3.5 % photopeak efficiency for ^{60}Co. An 8" Ø x 9" NaI(Tl) anti-Compton shield suppresses the Compton distribution in the spectra and the recording of events produced by fast fission neutrons. The coincident fission fragment masses are deduced from their correlated kinetic energies as measured by two Si solid-state detectors. The observed Doppler shift in gamma-ray energy allows the assignment of lines to single members of fragment pairs. Data are processed in a 256 x 256 x 2048 channel matrix. The main objective of these experiments is a study of the properties of individual neutron-rich nuclei far off the stability line.

PROMPT GAMMA RAYS EMITTED FROM INDIVIDUAL FRAGMENTS IN NEUTRON-INDUCED FISSION

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INTRODUCTION

From a comparison of the conventional chart of the nuclides with the lines of vanishing proton and neutron separation energies as derived from semi-empirical mass formulae it is evident that the number of nuclei so far not investigated considerably exceeds the number of nuclei which have already been studied satisfactorily. Therefore, extension of the methods of nuclear spectroscopy to regions far off the stability line will certainly increase the understanding of various nuclear properties. A useful means for producing very neutron-rich nuclei which are not accessible by usual nuclear reactions is provided by the nuclear fission process. The bulk of data accumulated in the past with this process has been obtained by the application of fast chemistry or other separation techniques with subsequent investigation of the radioactive decay schemes. Since a lower limit in halflives is inherent in all these methods, it is desirable to study in an on-line experiment the prompt de-excitation mechanism of individual primary fission fragments.

Such investigations are of considerable importance also for a better understanding of the fission process itself. There is increasing evidence that in asymmetric fission the properties of the nascent fragments, rather than those of the initial compound nucleus, essentially determine the characteristics of the various distributions and correlations observed for this process.

Examining the radiation emitted from fission fragments of specific mass has become possible by the rapid development of semiconductor detector technology and associated electronics during the past few years. First studies have been performed by Bowman et al. [1] and by Watson [2] using the spontaneous fission of ^{252}Cf. It is desirable to extend these measurements to thermal neutron-induced fission, both in order to compare the results with those obtained from ^{252}Cf and to cover fragment mass regions where the yield in spontanious fission is low.

Various experimental difficulties are inherent in experiments of this type at a reactor. In order to insure a negligible energy loss of the fragments in the target, the source thickness is limited to about 70 $\mu g/cm^2$. Thus for the accumulation of a sufficient number of counts in a multiple parameter experiment a neutron beam of high intensity and long measuring periods are required. The beam introduces restrictions on the geometry of the experimental setup and severe background and shielding problems arise. Electronic drifts must be controlled very carefully.

The present paper describes the measurements of prompt gamma-ray spectra associated with moving fission fragments of specific masses from fission of ^{235}U by thermal neutrons. This study is a part of a more general programme for investigating with high resolution all the prompt radiations emitted in neutron-induced fission.

EXPERIMENTAL PROCEDURE

Fig. 1 shows the geometric arrangement of the installation at the Karlsruhe reactor FR 2. The instrument was located at a tangential channel which passes through the heavy water of the reflector. Collimation of the neutron beam was done in such a way that the target was irradiated only by neutrons emerging from a graphite scatterer placed in the centre of the channel. A cooled bismuth single crystal filter of 20 cm length was used to reduce the gamma radiation in the beam. The thermal flux at the target position was approximately 7×10^7 n/cm^{2}sec [3]. A 50 $\mu g/cm^2$ ^{235}U fission source[1] prepared by electrospraying [4] was placed between two 600 mm^2 Si solid-state fission detectors as displayed in Fig. 2. Gamma rays coincident with fragment pairs were measured in the energy range 100 keV to 2 000 keV by means of a 28 cm^3 coaxial Ge(Li) detector surrounded by an 8" Ø × 9" NaI(Tl) anti-Compton shield [5]. This combination improved the peak-to-Compton ratio by a factor of two to four, depending on gamma-ray energy, and reduced considerably interfering gamma lines produced by inelastic scattering of fission neutrons in the germanium counter. The geometric layout of the apparatus determines the effective time resolution to about 1 nsec after fission. The gamma-ray collimator and the rounded, i.e. doughnut-shaped, fission-fragment collimators were adjusted in such a way that photons emitted later than 1 nsec time-of-flight had to penetrate several cm of lead for triggering the triple coincidence circuit. In particular, this method prevented the detection of gamma rays emitted by stopped fragments.

In order to eliminate electronic drift during the run, the gamma detector system was digitally stabilized using an ultra-stable pulse generator [6]. For stabilization of the silicon detectors the fission spectrum itself was taken as a reference. The counters were operated in the saturation region and a constant bias was maintained throughout the experiment. The energies of fission fragments were obtained from repeatedly measured single spectra with the aid of the

[1] The source was supplied by CBMN, Euratom, Geel/Belgium

mass-dependent pulse-height calibration equations given in Ref. [7]. Detectors were replaced whenever the peak-to-valley ratio became smaller than 15. The fast timing was based on the leading edge principle and the electronic time resolution was 40 nsec. The measured chance coincidence rate was well below 2 % of the true coincidence rate.

The triple pulse-height data were processed in a 256 x 256 x 2048 channel matrix via the Karlsruhe Multiple Input Data Acquisition System (MIDAS). Provisional masses μ were calculated from the correlated kinetic energies using momentum and mass conservation. Data were stored on magnetic tape in the form (μ_1, E_T, X_γ). Here μ_1 denotes the mass of the fragment moving towards the Ge(Li) detector, E_T is the total kinetic energy of both fragments and X_γ represents the gamma detector pulse-height. Fragment masses before and after neutron emission were obtained by taking into account the relationships between the average number of neutrons ν and the fragment mass [8]. The variation of ν for a specific mass with the total kinetic energy of the fragments [9] introduces an additional small mass dispersion. For the individual masses considered in the following section the average number of neutrons varies by less than $\pm$ 0.5 neutrons for total kinetic energies within plus or minus one rms width of the mean [10]. This value is small compared to the average mass resolution in this experiment which is estimated to be approximately 4 amu FWHM.

RESULTS AND DISCUSSION

Sorting of the triple data according to individual final masses of the fragments revealed a pronounced structure in the corresponding gamma-ray spectra. The pattern changed clearly for different mass ratios. Examples are shown in Fig. 3, where the results are given in the energy range from 100 keV to 1000 keV for the mass ratio values 1.26, 1.43 and 1.62. In all three cases the gamma counts were restricted to events in which the heavy fragments were moving towards the Ge(Li) counter.

Examples for the dependence of the gamma-ray energies on the fragment velocity and the direction of motion are given in Fig. 4. The upper spectrum in this figure represents the case where the light fragments (A = 96 $\pm$ 1) were travelling towards the gamma-ray detector, the lower spectrum holds for the light fragments moving away from the detector. The measured sign and magnitude of the Doppler shift were used for the identification of the emitting member of the fragment pair. The observed shift in energies was consistent with expected values derived from experimental fragment velocity distributions [11] and the geometry used in this experiment. Some weak unshifted lines which appear in the spectra have to be attributed to inelastic scattering of fission neutrons in the germanium detector. All spectra shown in Figs. 3 and 4 are unsmoothed. Only in a few cases a smoothing procedure was applied for analysis of weak lines or complex structures.

In Table I 39 gamma rays have tentatively been assigned to individual fragments. The masses were arrived at by comparing the peak intensity in adjacent mass intervals the centres of which differing in general by 2 amu. Those masses were selected where the gamma-ray lines appeared with their highest intensity. Many lines occurred distinctly only in one mass-sorted spectrum. This fact provides ample evidence for the correctness of the estimated mass dispersion and the assumed error of $\pm$ 1 amu in the mass assignment. If a gamma-ray peak which is now considered to correspond to a single gamma ray turns out to be a closely spaced doublet or a triplet, the energy listed in Table I refers only to the centroid. In some cases, a gamma line was clearly distinguished in one member of the spectrum pair whereas the Doppler shifted partner could not be located since in either possible position intense lines or complex structures occurred. Such lines were not included in the Table.

The most probable charge quoted in Table I for the given fragment masses have been derived from the Tables in Ref. [12] . For comparison, data reported by Bowman et al. [1] and by Watson [2] for spontaneous fission of ^{252}Cf have also been included in Table I. Only those transitions from these measurements have been tabulated which may be identified with gamma rays observed in the present experiment. Quite good agreement is found both in photon energy and in mass assignment. The atomic numbers derived from K-X-ray measurements coincide within experimental errors with the most probable charge, and the deviation of the Z values used for calculating the transition energy from conversion electron data is also at most one unit of charge.

It is worth-while to note that in the spectra belonging to fragment masses A $\geqslant$ 104 groups of gamma rays with energies between 250 and 320 keV and around 400 keV seem to come forward which might be identified with the regular structure observed by Johansson [13] in measurements of the delayed gamma radiation from fission fragments of ^{252}Cf. This structure occurred in the mass range 92 to 110 and suggested a rotational behaviour giving experimental evidence for the existence of a new region of stable deformation in this mass range. Unfortunately, the statistics in the present experiment above A = 102 were still too poor to locate the gamma-ray lines with confidence.

CONCLUSIONS

The experiment described has successfully demonstrated that the various difficulties can be overcome which are inherent in reactor experiments for studying the prompt radiation emitted from individual fragments in neutron-induced fission. Though the results are still too incomplete to allow any definite theoretical conclusions, they are encouraging enough to initiate systematic studies of this type, thus extending the investigation of primary fragments to mass regions which are not covered with sufficient yield in spontaneous fission. Meanwhile a new version of the experiment with increased intensity (neutron flux 1.1 x 10^9 n/cm^{2}sec) and improved system resolution has been installed at the reactor FR 2. It is reasonable to assume that such studies will reveal important information on neutron-rich nuclei far off the stability line and, via the properties of the nascent fragments, will provide a better insight into the fission process itself.

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TABLE I

TENTATIVE ASSIGNMENT OF PROMPT GAMMA RAYS TO INDIVIDUAL FRAGMENTS

Neutron-induced fission of ^{235}U This work			Spontaneous fission of ^{252}Cf Refs. [1] and [2]			
Fragment mass A	Most probable charge [a] Z_p	E_γ [keV]	Fragment mass A	Charge [b] Z	E_γ [keV] c	E_γ [keV] d
91 ± 1	36	706				
93 ± 1	37	144				
95 ± 2	38	249				
95 ± 1	38	813				
		834				
96 ± 2	39	376				
98 ± 1	40	193				
99 ± 1	40	123				
		157				
100 ± 1	40	98	100 ± 1	40[e]	97	98[f]
		210	99 ± 1	39[e]	210	210[f]
		351				
		495				
		622				
101 ± 1	41	325				
102 ± 1	41	174				
103 ± 2	41	518				
103 ± 1	41/42	136				
		296				
104 ± 2	41/42	(157)				
		556				
		581				
104 ± 1	42	191	105 ± 1	42[e]	190	191[f]
		366				
131 ± 2	51	965				
138 ± 1	53/54	585	139 ± 1	54 ± 1	586	
139 ± 1	54	283	139 ± 1			283[g]
		372				

TABLE I (continued)

Neutron-induced fission of ^{235}U This work			Spontaneous fission of ^{252}Cf Refs. [1] and [2]			
Fragment mass A	Most probable charge [a] Z_p	E_γ [keV]	Fragment mass A	Charge [b] Z	E_γ [keV] c	E_γ [keV] d
		482				
		723				
141 ± 2	55	475	140 ± 1	54 ± 1	479	
141 ± 1	55	357				
142 ± 1	56	429				
143 ± 2	56	492				
143 ± 1	56	115	144 ± 1	56[e]	116	116[f]
		198	144 ± 1	55[e]	198	198[f]
		330				
144 ± 1	56	507				
147 ± 1	58	294	146 ± 1			293[g]

a Derived from the tables given in Ref. [12]

b Obtained from K-X-ray measurements.

c Directly measured gamma-ray energies.

d Calculated from conversion electron data.

e Atomic number assumed for calculating the gamma-ray energy listed in column 7.

f Calculated values with the Z assignment specified in column 5.

g Calculated using the most probable charge Z_p.

FIGURE CAPTIONS

Fig. 1 Geometric arrangement at the tangential through-hole of the reactor FR 2.

Fig. 2 Schematic view of the experimental setup.

Fig. 3 Sectional display of prompt gamma-ray spectra for various values of mass ratio with the heavy fragments moving towards the gamma-ray detector. The spectra are unsmoothed.

Fig. 4 Observed prompt gamma-ray spectra for the fragment mass ranges A = 95 - 97 and A = 136 - 138 demonstrating the dependence of gamma-ray energy on the velocity and direction of the fragment motion. The two spectra represent the cases:

(a) Light fragments moving towards the gamma-ray detector and

(b) heavy fragments moving towards the gamma-ray detector.

The letters L and H indicate some assignments to the light and heavy fragments, respectively.

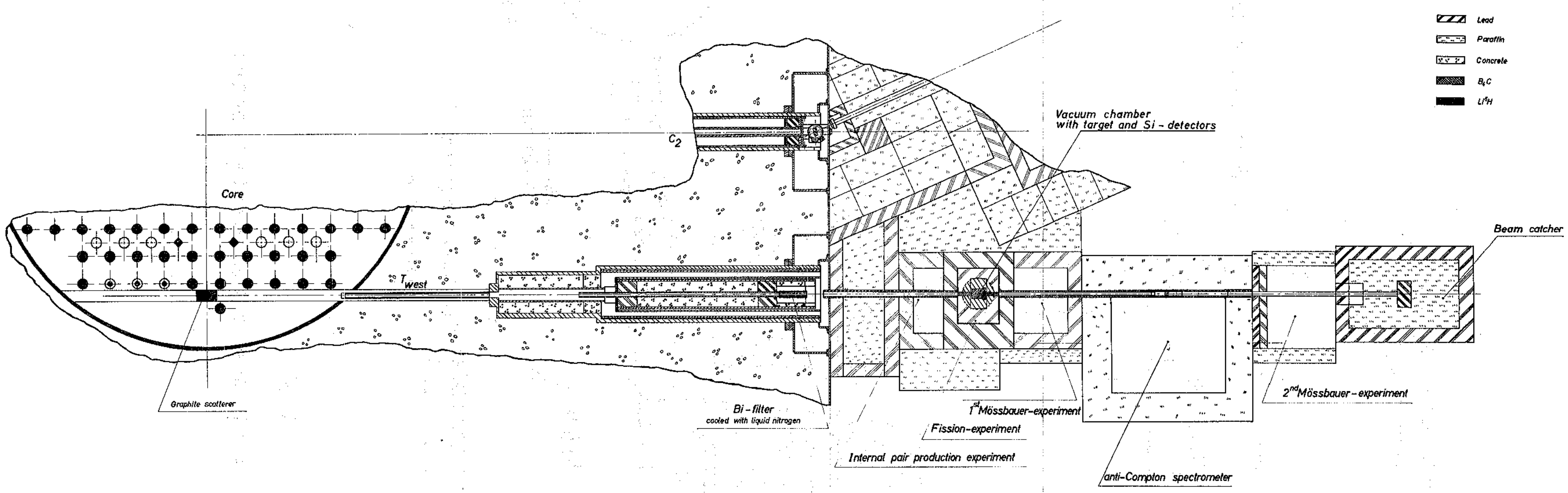


Fig. 1

Geometric arrangement at the tangential through-hole of the reactor FR 2

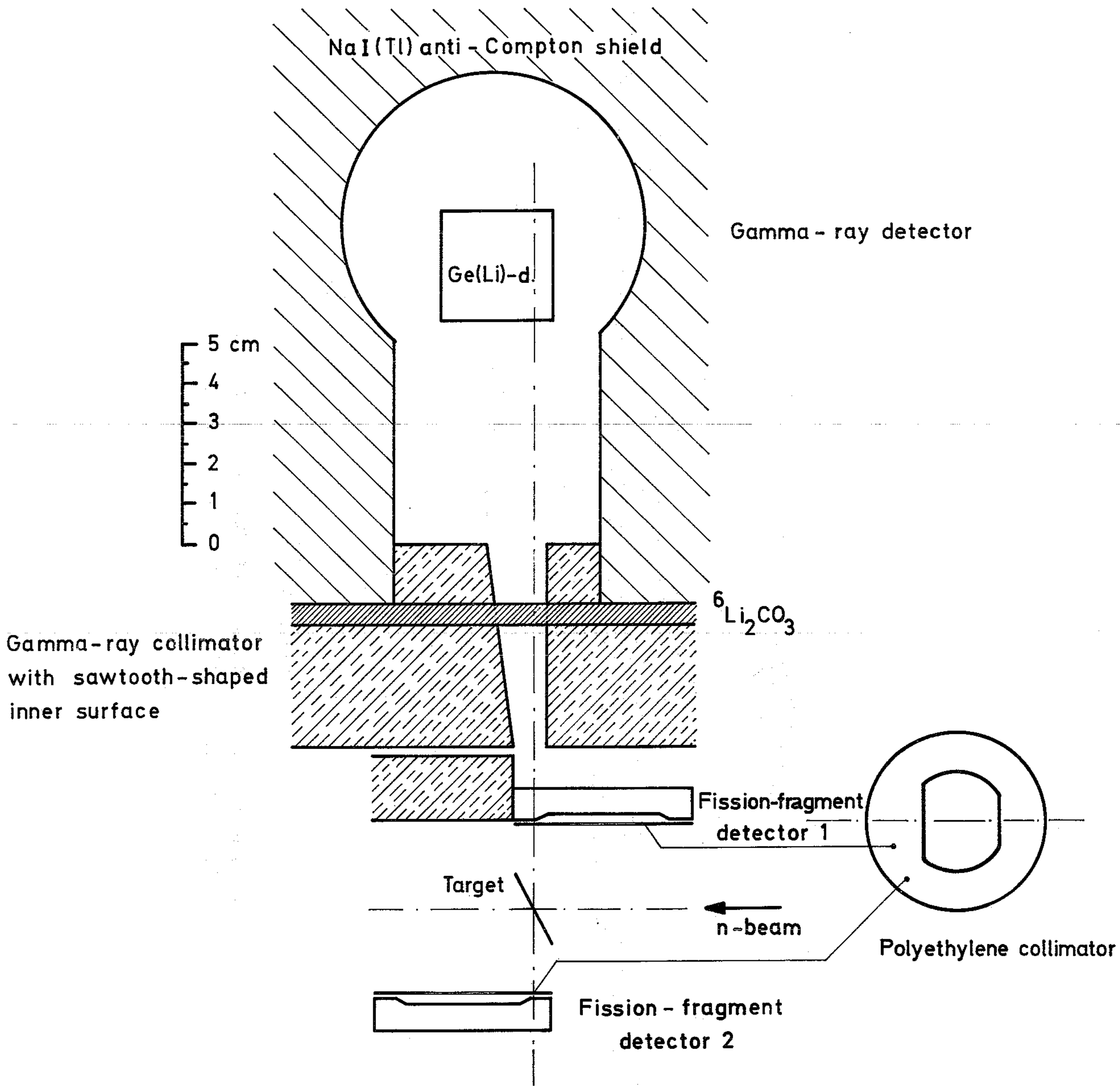


Fig. 2

Schematic view of the experimental setup

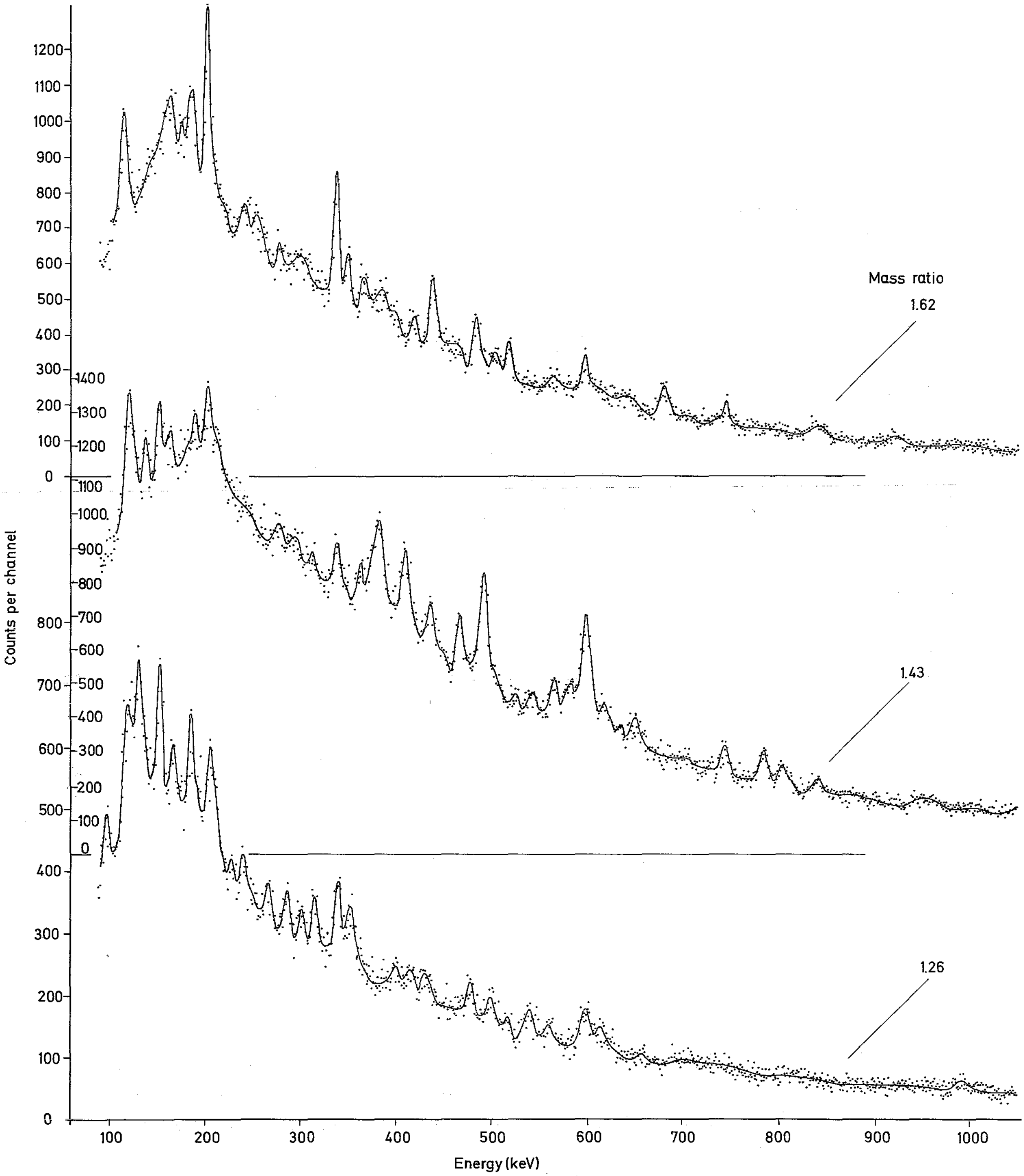
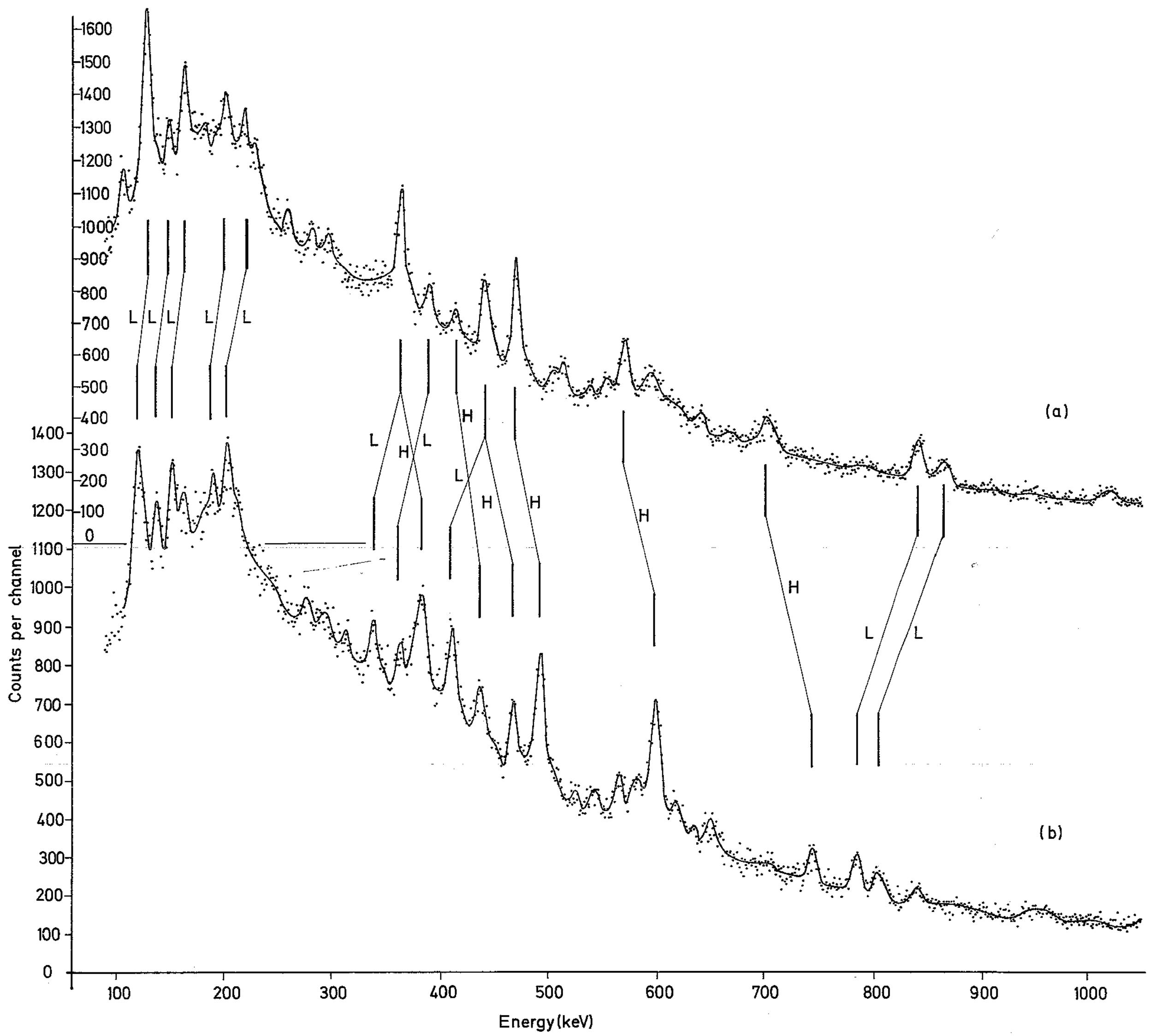


Fig. 3

Sectional display of prompt gamma-ray spectra for various values of mass ratio with the heavy fragments moving towards the gamma-ray detector. The spectra are unsmoothed.



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