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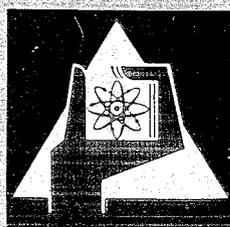
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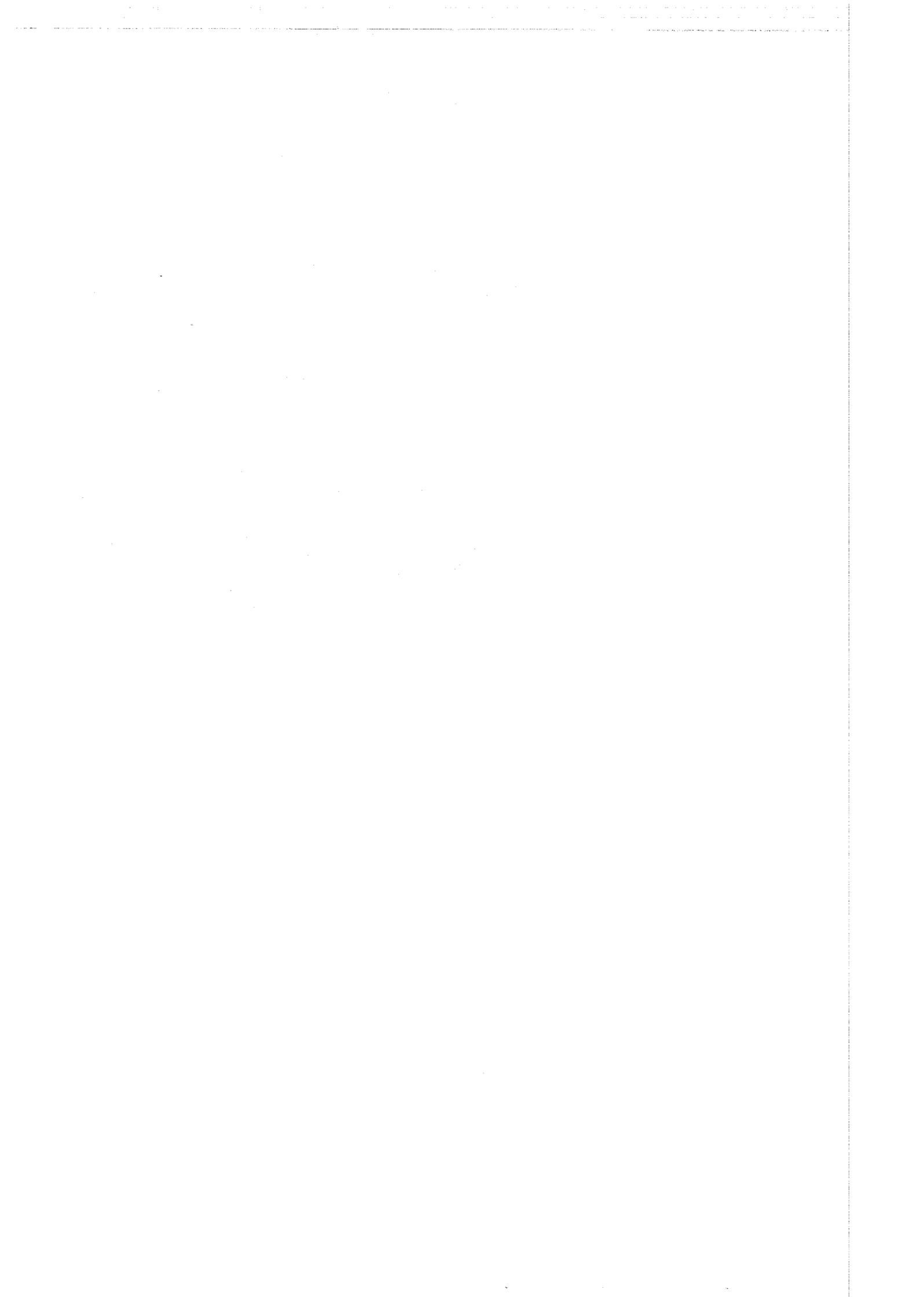
Studies of Radiative Neutron Capture and Delayed Fission Gamma-Ray
Spectra from Uranium and Plutonium as a Basis for New
Nondestructive Safeguards Techniques

P. Matussek, W. Michaelis, C. Weitkamp, H. Woda



GESELLSCHAFT FÜR KERNFORSCHUNG M. B. H.

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Studies of
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ABSTRACT

The thermal-neutron induced gamma-ray spectra from ^{235}U and ^{239}Pu samples have been studied with a Ge(Li) detector. The integral spectrum is a superposition of gamma rays from the neutron capture reaction, prompt gamma rays from fission, and delayed gamma rays emitted from fission products. In order to separate the different parts, two experimental setups were used. The prompt fission and capture gamma-ray spectra were obtained by means of a 4π plastic scintillator for the detection of prompt fission neutrons, the detector being operated in coincidence and anticoincidence with the Ge(Li) detector, respectively. A combination of three mechanical choppers covering a decay time range from 100 μs to 1 s served for the measurement of the different components of the delayed spectra.

The separation of the radiative capture, prompt fission, and delayed fission components of the total thermal neutron gamma-ray spectrum greatly facilitates the interpretation of the different gamma-ray lines observed. In particular, the structure of the capture spectra can, in many cases, be unambiguously related to hitherto known excited states and interpreted in terms of the nuclear structure of the compound nucleus. On the basis of the data measured new approaches for detection, identification, and quantitative analysis of nuclear fuel are proposed that are nondestructive, prompt, accurate, and isotope specific. The results of the measurements are discussed in view of this application. For obtaining optimum penetrability for the primary radiation the application of a filtered beam of keV neutrons in the case of the radiative capture reaction and of non-thermal neutrons or high-energy photons in the case of the delayed fission spectra is suggested.

RESÜMEE

Die Gammaspectren aus der Wechselwirkung thermischer Neutronen mit ^{235}U und ^{239}Pu wurden mit einem Ge(Li)-Detektor untersucht. Das Gesamtspektrum ist eine Überlagerung von γ -Strahlung aus der Neutroneneinfangreaktion, prompter Spaltungs-Gammastrahlung und verzögerter γ -Strahlung aus Spaltprodukten. Zur Trennung der verschiedenen Komponenten wurden zwei experimentelle Anordnungen benutzt. Die Messung des prompten Spalt- und Einfanggammaspectrums erfolgte durch Verwendung eines 4π -Plastik-Szintillators als Detektor für die prompten Neutronen aus der Spaltungsreaktion, der in Koinzidenz bzw. Antikoinzidenz mit dem Ge(Li)-Detektor betrieben wurde. Eine Kombination dreier mechanischer Chopper, die den Zeitbereich von etwa $100\ \mu\text{s}$ bis $1\ \text{s}$ überdeckt, diente zur Messung der verzögerten Komponenten.

Die Interpretation der beobachteten Gammalinien wird durch die Trennung der Komponenten aus Strahlungseinfang, Spaltung und Spaltprodukten sehr erleichtert. Insbesondere läßt sich die Zusammensetzung der Einfangspektren vielfach eindeutig bekannten angeregten Zuständen zuordnen und aus der Struktur des Restkerns erklären. Neue, auf den gemessenen Daten beruhende Verfahren zum Nachweis, zur Identifikation und zur quantitativen Analyse von Kernbrennstoff werden vorgeschlagen, die genau, zerstörungsfrei und isotopenspezifisch sind und sofort verfügbare Resultate liefern. Die Meßergebnisse werden auf diese Anwendung hin diskutiert. Zur Erzielung optimaler Transparenz für die primäre Strahlung werden im Falle des Strahlungseinfangs die Anwendung eines gefilterten Strahls Neutronen im keV-Bereich, im Fall der verzögerten γ -Strahlung nach der Kernspaltung nicht-thermische Neutronen oder hochenergetische Photonen vorgeschlagen.

STUDIES OF
RADIATIVE NEUTRON CAPTURE AND DELAYED FISSION GAMMA-RAY SPECTRA
FROM URANIUM AND PLUTONIUM
AS A BASIS FOR NEW NONDESTRUCTIVE SAFEGUARDS TECHNIQUES

P. Matussek, W. Michaelis, C. Weitkamp and H. Woda

Institut für Angewandte Kernphysik
Kernforschungszentrum Karlsruhe, Germany

1. INTRODUCTION

The observation of prompt neutron capture gamma-ray spectra has proven a powerful tool for nondestructive material analysis and is increasingly being used as an alternative or complementary method to conventional analysis procedures including activation analysis.

The application of the capture process to the assay of fissile material, however, had not been possible until an appropriate technique was proposed by Michaelis [1]. The main reason for the previous failure is the fact that spectra from capture in fissionable nuclei are usually strongly perturbed by the presence of prompt gamma rays from fission and delayed gamma rays from fission products. Capture spectra of ^{235}U have therefore never been observed, and for ^{239}Pu measurements were only recently possible in a number of resonances between 0.3 and 58.0 eV [2]. For several reasons the radiative neutron capture process is of particular interest in safeguards technology. If the high-energy part of the spectrum is utilized, the gamma-ray attenuation coefficients are sufficiently low to ensure a high penetrability. In addition the line density is low in this energy region and nuclear structure effects account for a high isotope discrimination power. Recording characteristic gamma rays in the upper part of the spectrum therefore fulfills the most important requirements of nondestructive methods.

It is the purpose of this paper to report the results of measurements done at the Karlsruhe research reactor FR 2 in order to separate the prompt fission, delayed fission, and capture parts of gamma spectra from interaction of thermal neutrons with ^{235}U and ^{239}Pu , and to discuss the results in view of an application to nuclear materials safeguards technology. The present measurements have to be considered as a first important step in developing new techniques based on radiative neutron capture. It will be shown that distinct signatures for the various fissionable species can be obtained.

2. EXPERIMENTAL PROCEDURE

For the separation of the prompt fission gamma spectrum from the capture spectrum use was made of the fact that fission of ^{235}U and ^{239}Pu is always accompanied by the emission of fast neutrons which, if detected with appropriate efficiency, can be used to separate the prompt fission gamma rays from the other components of the spectrum.

To do this an apparatus has been used that is schematically represented in Fig. 1. A collimated beam of thermal neutrons hits the fissile material target which is surrounded by a large plastic scintillator. Gamma rays are detected by means of a Ge(Li) detector operated as a double-escape spectrometer in the high-energy region. The plastic detector is used, upon registration of a neutron, to label simultaneous counts from the Ge(Li) detector as arising from fission events. As has already been pointed out in previous estimates [1], this neutron anticoincidence method should provide a sufficient suppression of the prompt fission gamma-ray spectrum at high photon energies. In addition, in this energy region the delayed spectrum is expected to be weak enough to be not a serious obstacle.

Scattered thermal neutrons from the beam are stopped by 0.55 g/cm^2 of $\text{Li}_2^{6}\text{CO}_3$ enclosed in a double-walled graphite tube. The Ge(Li) detector is shielded from fast fission neutrons by 6.0 g/cm^2 of LiH. In order to prevent cascade capture gamma-rays from interacting in the plastic detector and simulating the detection of neutrons thus reducing the number of anticoincidence events, a lead shield is inserted between the $\text{Li}_2^{6}\text{CO}_3$ and NE 102 A detector. This shield is shaped in such a way that gamma rays from the target have to penetrate 50 mm of lead before reaching the plastic scintillator. This corresponds to an attenuation by a factor of 100 for 500 keV radiation, but will not appreciably affect the detection of neutrons. Since the high-energy part of the gamma-ray spectrum consists of primary transitions proceeding from the capture state to low-lying levels, the coincidence rate caused by gamma-ray cascades is kept sufficiently low.

Because of the high count rates the plastic detector is divided into eight parts; each of the octants is viewed by three photomultipliers the signals from which are summed up and discriminated just above the noise level. The gamma-detector circuit includes leading-edge timing from the preamplifier output pulse, lower-level discrimination to cut the rate of accidental coincidences, and pileup rejection. The effective time resolution is $2\tau = 40 \text{ nsec}$. Simultaneous

acquisition of the coincidence and anticoincidence spectrum ensures that both types of spectra are taken under identical conditions and are, therefore, directly comparable.

In addition to the capture component the anticoincidence spectrum also contains contributions of delayed gamma rays from fission products. An arrangement of three mechanical choppers shown schematically in Fig. 2 was therefore set up to measure these contributions. Results have been obtained for times down to 2.5 msec after the fission process. Only one slow disk had to be spun for this purpose.

A filtered neutron beam from a through-hole of the reactor was used in the present studies. The filter consisted of a bismuth crystal cooled to liquid nitrogen temperature. The flux at the target position was approximately 2×10^7 n cm⁻² sec⁻¹.

The sample material was enclosed in polyethylene containers with 0.5 mm wall thickness.

3. RESULTS

3.1 Spectra

Due to the high neutron flux the fission neutron count rate in the plastic scintillator was very high. In order to keep the neutron and chance coincidence count rates in reasonable limits, only small quantities of sample material could be used. Table I gives a list of the size and composition of the two samples utilized. From the isotope abundances and the capture-to-fission ratios the relative cross-section contributions for radiative capture in ²³⁵U and ²³⁹Pu are calculated to 15 % and 25 %, respectively. For the samples listed in Table I the accidental coincidence rates were 22 % for the uranium target and 18 % in the case of the plutonium sample. As a consequence intense capture lines can also be identified in the coincidence spectra, though with considerably reduced intensity, while on the other hand, due to incomplete detection of fission neutrons and deadtime effects, part of the coincident counts were also registered in the anticoincidence spectrum. Background lines appeared in particular from carbon, iron, lead and aluminum due to capture in the structure materials and the sample containers. Of course all of the peaks of the delayed spectrum must be present in the anticoincidence spectrum, but the delayed spectrum also contains prompt neutron capture background lines.

At low energies most of the peaks in the spectra are due to prompt and delayed fission gamma rays, as expected, and capture peaks are hard to identify. In the region of interest, however, prominent peaks show up in the anticoincidence spectra which clearly originate from the radiative capture reaction.

Fig. 3 is a plot of the anticoincidence, coincidence, and delayed spectra from plutonium between 4 and 6 MeV along with a spectrum of an empty sample holder. The anticoincidence and coincidence spectra

were taken simultaneously, the background spectrum is normalized and compatibilized to the same neutron dose and energy scale. The delayed spectrum is also normalized so that the product of chopper duty cycle, analyzer duty cycle, and measuring time becomes equal to the measuring time of the anticoincidence-coincidence spectra. This would yield comparable conditions if all photons detected were due to neutrons from the chopped beam. Because this is obviously not so, the delayed spectrum is plotted on a reduced ordinate scale. The poor statistics required smoothing of the data by the parabola method.

For the assignment of a line to the radiative capture spectrum consideration was given to the following criteria:

- strong occurrence in the anticoincidence spectrum, with appropriate reduction in the coincidence spectrum;
- absence in the delayed spectra of both uranium and plutonium;
- absence in the background spectrum.

Several gamma-ray lines can unambiguously be identified as originating from radiative capture in ^{239}Pu . The peaks appear in the anticoincidence spectrum with remarkable intensity.

Fig. 4 shows the anticoincidence, coincidence, delayed ($t > 2.5$ msec) and background spectra as obtained from the uranium sample. At a gamma-ray energy of 6396 keV a strong line appears in the anticoincidence spectrum that is clearly absent in the other spectra. It can be seen, however, that up to 5 MeV the anticoincidence spectrum is dominated by lines which are also present in the delayed spectrum and therefore must be attributed to fission products.

3.2 Discussion

Table II gives a list of the high-energy capture gamma rays from $^{239}\text{Pu}(n,\gamma)$. The level diagram of ^{240}Pu is shown in Fig. 5. Previously known level energies and low-energy transitions as obtained from α and β decay studies were taken from Refs. [3] and [4]. Assuming that the 6490.3 keV transition reaches the 2^+ level at 42.8 keV, the neutron binding energy is calculated to be (6533.1 ± 1.0) keV. This value is in good agreement with the separation energy of (6490 ± 50) keV deduced from (d,p) data [5], and the gamma rays at 5936 and 5674 keV can then consistently be explained in terms of the hitherto known level structure. The most intense capture lines should be E1 transitions to negative parity states with the spin values 0, 1 and 2. Therefore we have assigned spin and parity 1^- to the level at 958 keV [6] excluding the spin values 0 and 2 on the basis of the systematic behaviour of octupole vibrational excitations in the heavy element region [7,8,9]. This conclusion has very recently been confirmed by the results of Chrien et al. [2] for the 0.3 eV resonance. Most probably this level corresponds to the $K=1$ octupole vibration which is predicted by Blocki and Kurcewicz [9] at about 750 keV. Amazingly enough, only a very weak 5936 keV gamma ray could be seen to the other

low-lying 1^- state at 597 keV which probably is the $K=0$ octupole state and which is fed intensely by capture in many resonances [2] including the one at 0.3 eV. On the other hand, transitions to positive-parity states at 859 keV (0^+) and 43 keV (2^+) are also observed. Because the E2 competition to primary M1 transitions is known to be very weak, the feeding of the 43 keV 2^+ level indicates that a substantial part of thermal neutron capture occurs into a $J=1$ compound state; since a large contribution to thermal capture is due to the resonance at 0.3 eV, this is further evidence for Bollinger's original spin 1 assignment [10] to that compound state. The spin 1 has been questioned by Vogt [11] and Kirpichnikov [12], but recently supported by Weinstein and Block [13] and by Chrien et al. [2]. Further lines were observed in the present study which, if assigned to primary transitions, lead to levels at 1240 and 1410 keV. None of these lines were seen by Chrien et al. who, on the other hand, found lines at 5597 and 5095 keV not seen in the present work.

The attempt of identification of low-energy lines from capture has so far not been successful, so no statements can be made on the properties of the higher-lying levels.

It follows from the level scheme of ^{236}U (Fig. 6) and the neutron binding energy derived from mass differences [14] that the 6396 keV gamma ray from Fig. 4 corresponds to a transition from the 4^- capturing state [15] to the 4^+ member of the ground state rotational band. This transition, being the only possible electric dipole transition to known low-energy levels, must indeed be the only gamma-ray visible in this energy region.

In summary, we can conclude that the identification of the observed primary transitions is consistent with the expectations from the nuclear structure of the product nuclei.

4. APPLICATION TO SAFEGUARDS TECHNOLOGY

It has been shown that the gamma-ray spectra from interaction of thermal neutrons with both ^{235}U and ^{239}Pu show intense lines from the capture reaction that lend themselves as a signature for use in a suitably designed apparatus. In an actual application the procedure will, of course, differ in several respects from that used in the present studies:

- (i) The design of the instrument will be much simpler.
- (ii) The ratio of target to container and structure material will be more favourable.
- (iii) The contribution of the delayed gamma-ray component is expected to be much smaller.
- (iv) The transparency for the incident neutrons has to be improved considerably.

Since practically no information was available before about the radiative capture process in ^{235}U and ^{239}Pu , the apparatus described in section 2 had to be designed as a basic physical instrument. Once the nature of the observed gamma rays is known, there is no longer any need for the complete separation of the different kinds of radiation. The experience acquired during our studies indicates that the anticoincidence system can be considerably simplified and that the response of the gamma-ray detector can be improved by more than one order of magnitude.

Moreover the possibility exists that the problem can be solved by singles spectroscopy without the anticoincidence concept when choosing incident neutrons of an appropriate energy between thermal energies and a few keV where the capture-to-fission ratio is much higher than for thermal neutrons.

The quality of the spectra measured with the present apparatus is, in terms of the peak areas from fissile materials, good enough for use in quantitative analytic work; for routine measurements, however, a better peak-to-background ratio will, particularly in the case of uranium, simplify the analysis and reduce statistical errors. Because the background under the high-energy peaks of the uranium and plutonium lines, as can be seen from Figs. 3 and 4, is to a large extent due to the Compton continua of higher-energy gamma rays from other elements, this is automatically achieved by an increase of the ratio of capture in fissile material to that in other materials when using actual fuel pins containing roughly 1000 times the quantity of fissile material used in the present experiment.

With a neutron flux of $2 \times 10^7 \text{ cm}^{-2} \text{ sec}^{-1}$, a target-to-detector distance of 30 cm, and the small samples specified in Table I the spectra shown could be taken in about 100 hours using a 35 cm^3 detector. Even if the product of flux and cross-section is reduced by a factor of 1000, this means that with improved gamma detector response a bundle of about 20 pins can still be measured with sufficient statistical accuracy within a few minutes. As for the delayed γ activity from fission products only a small amount can build up in such a short time. In addition moving of the fuel element leads to further reduction of the delayed component.

In order to ensure better penetration of the sample neutrons with higher energy, i.e. smaller total cross section, must be used. This is particularly important for the measurement of bundles of pins. As a source of non-thermal neutrons different devices are being considered. By far the simplest, from an operational point of view, is a radioactive (α, n) or (γ, n) source. Problems arise in both cases from the difficult shielding of the gamma-ray detector against fast neutrons or gamma rays from the source. Another possibility is a scandium or iron filtered neutron beam from a low-power reactor; both 2 and 25 keV neutrons provide excellent transparency. Dynamically monochromated (p, n) neutrons from a Van de Graaff generator and neutrons from an electron linear accelerator filtered by uranium and plutonium of appropriate thickness are other possibilities. Each of these has its particular appeal, but also its problems: using linac neutrons, e.g., requires additional time-of-flight discrimination of events originating

from neutrons with energies > 5 keV, and the problems associated with the gamma flash deserve special consideration.

The determination of the neutron source best suited for the present purpose is now under investigation; it is the second step towards the technical realization of an assay system based on the neutron capture reaction. The first and very encouraging step was the observation of gamma rays from neutron capture in ^{235}U and ^{239}Pu as described in this paper.

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CAPTIONS

Schematic view of anticoincidence-coincidence spectrometer.

Chopper arrangement. Principle of operation.

Gamma rays from interaction of thermal neutrons with ^{239}Pu . Top to bottom: anticoincidence, coincidence, delayed, and background spectrum. Single and double primes indicate single and double escape peaks, respectively. Note different zero suppression.

Gamma rays from interaction of thermal neutrons with ^{235}U . Top to bottom: anticoincidence, coincidence, delayed, and background spectrum. Single and double primes indicate single and double escape peaks, respectively. Note different zero suppression.

Level diagram of ^{240}Pu . Primary transitions from present work only. Band heads are indicated by heavy lines.

Level diagram of ^{236}U . Heavy lines indicate band heads.

TABLE I

Samples.

Compound	Isotope	Abundance	Quantity of heavy element	Contribution to capture fission		Capture-to-fission ratio
UO ₂	235	19.85 %	135.4 mg	91.1 %	100.0 %	0.174
	238	80.15 %	547.2 mg	9.9 %	-	-
PuO ₂	239	91.2 %	83.4 mg	91.1 %	98.8 %	0.358
	240	8.0 %	7.3 mg	7.8 %	-	-
	241	0.8 %	0.7 mg	1.1 %	1.2 %	0.382
	242	0.004 %	-	0.03 %	-	-

TABLE II

High-energy gamma rays from thermal-neutron capture in ^{239}Pu .

Energy, keV	Relative intensity
6490.3 ± 0.9	0.14 ± 0.03
5936.0 ± 1.5	0.10 ± 0.08
5674.1 ± 0.8	0.27 ± 0.04
5575.3 ± 0.8	0.96 ± 0.04
5292.8 ± 0.7	0.50 ± 0.15
5123.2 ± 0.4	1.00

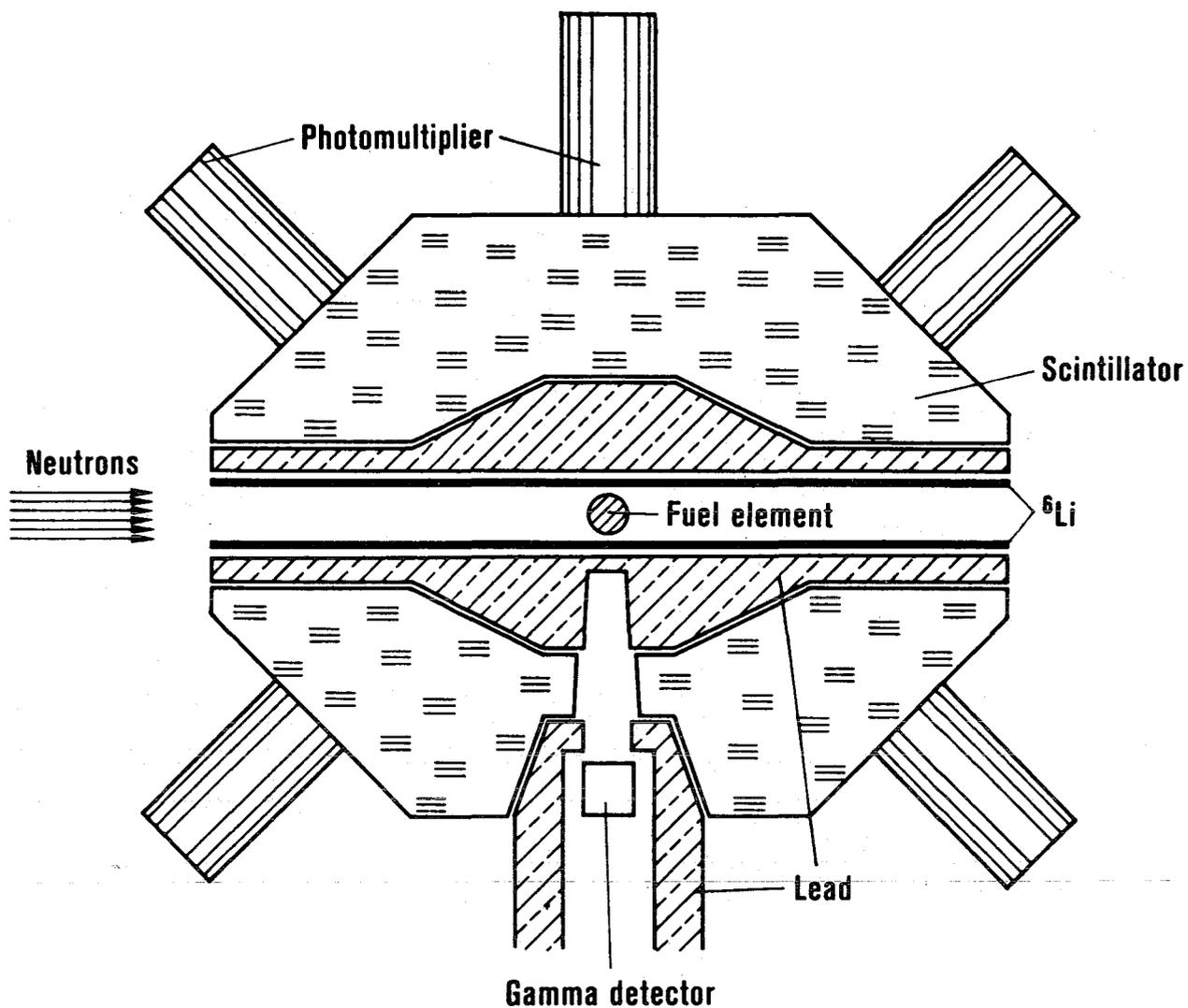


Fig.1.

Schematic view of anticoincidence-coincidence spectrometer.

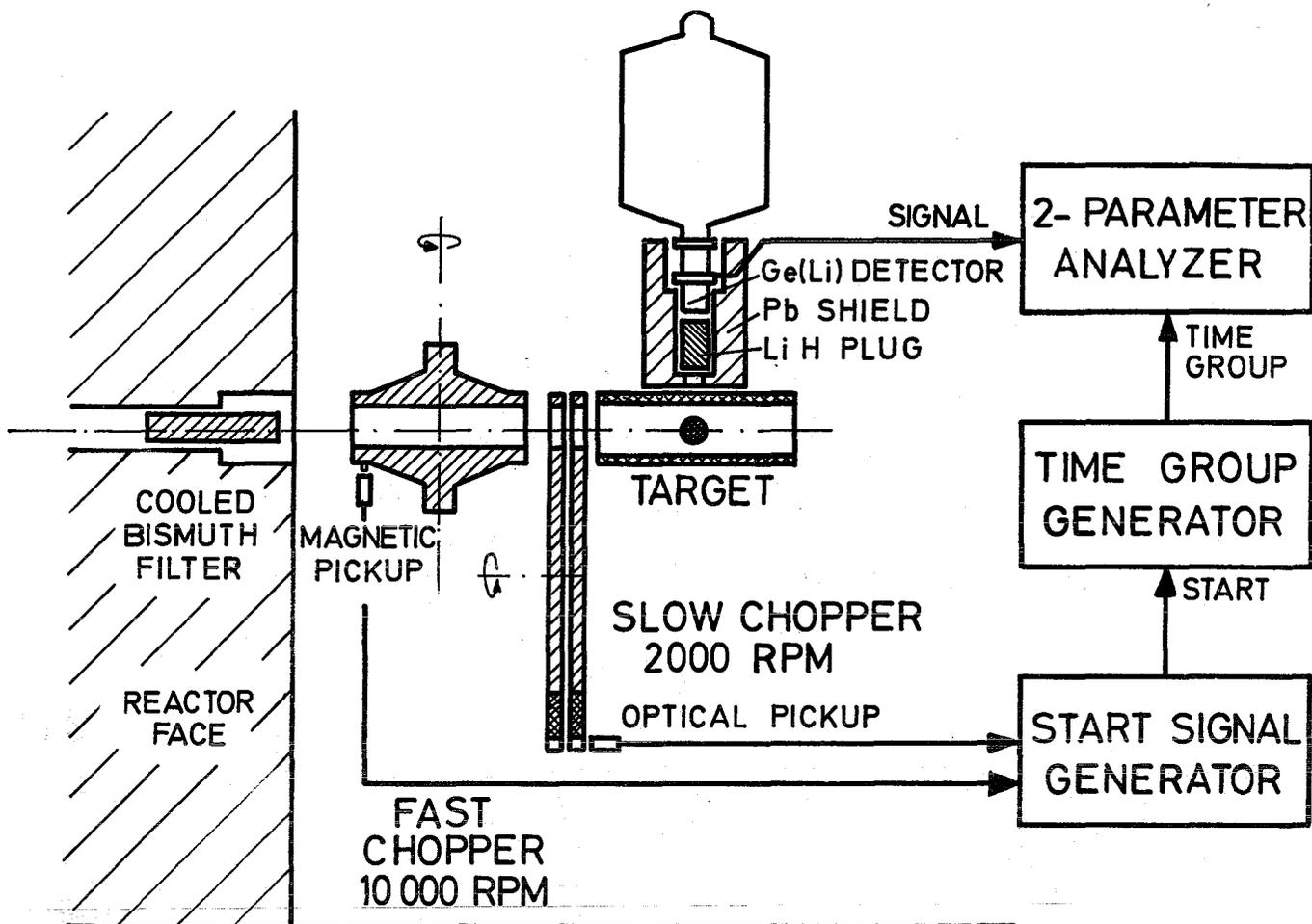


Fig. 2.
Chopper arrangement. Principle of operation.

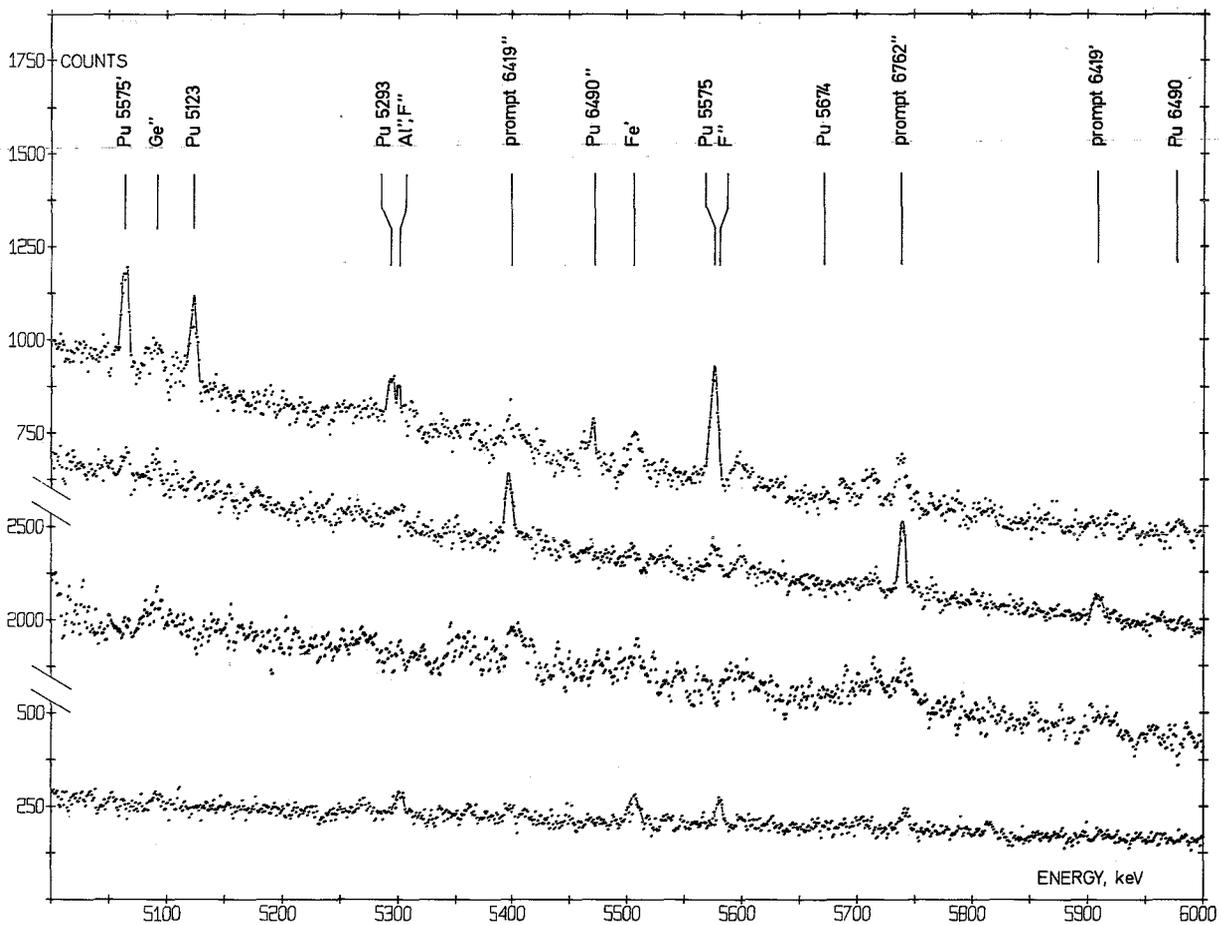
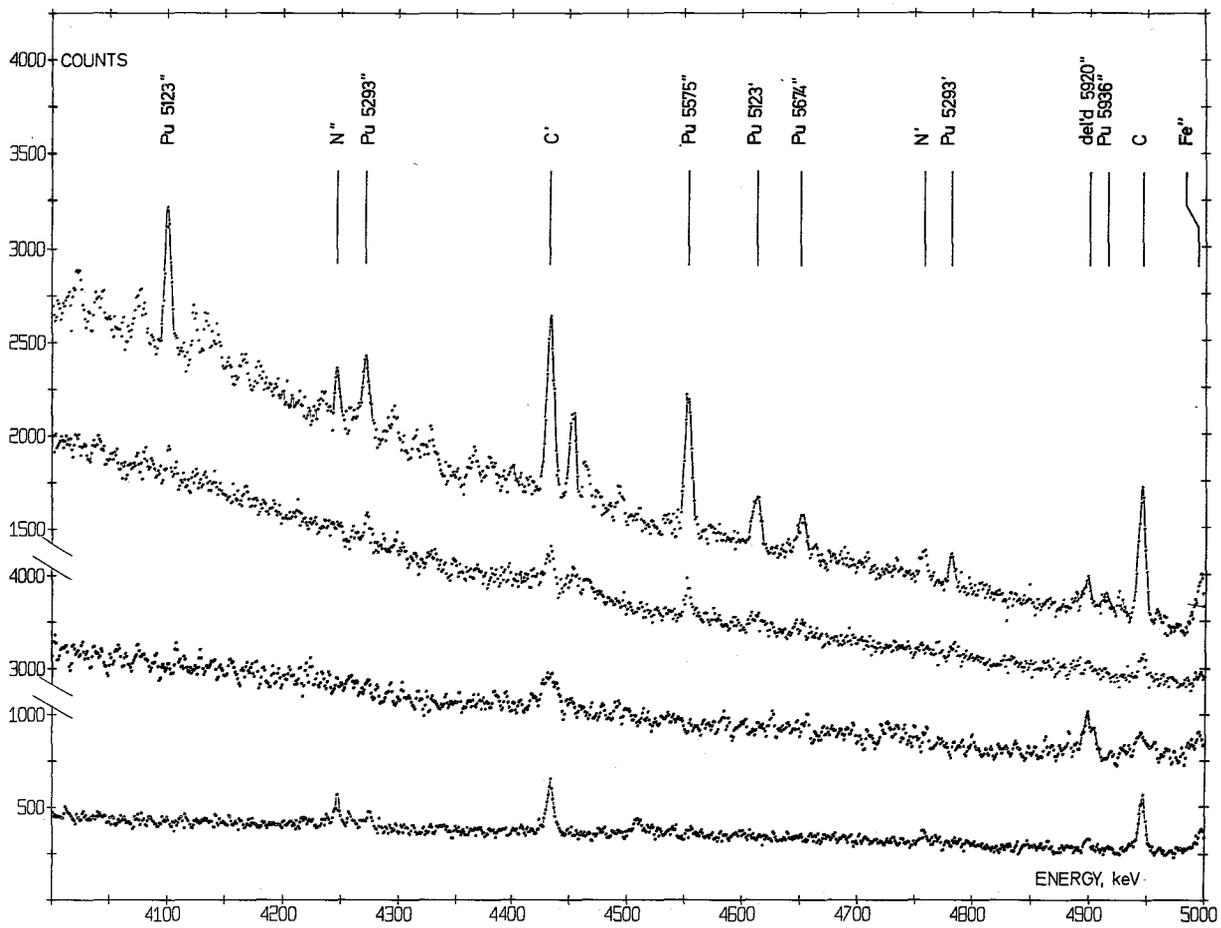


Fig. 3.

Gamma rays from interaction of thermal neutrons with ^{239}Pu . Top to bottom: anticoincidence, coincidence, delayed, and background spectrum.

Single and double primes indicate single and double escape peaks, respectively. Note different zero suppression.

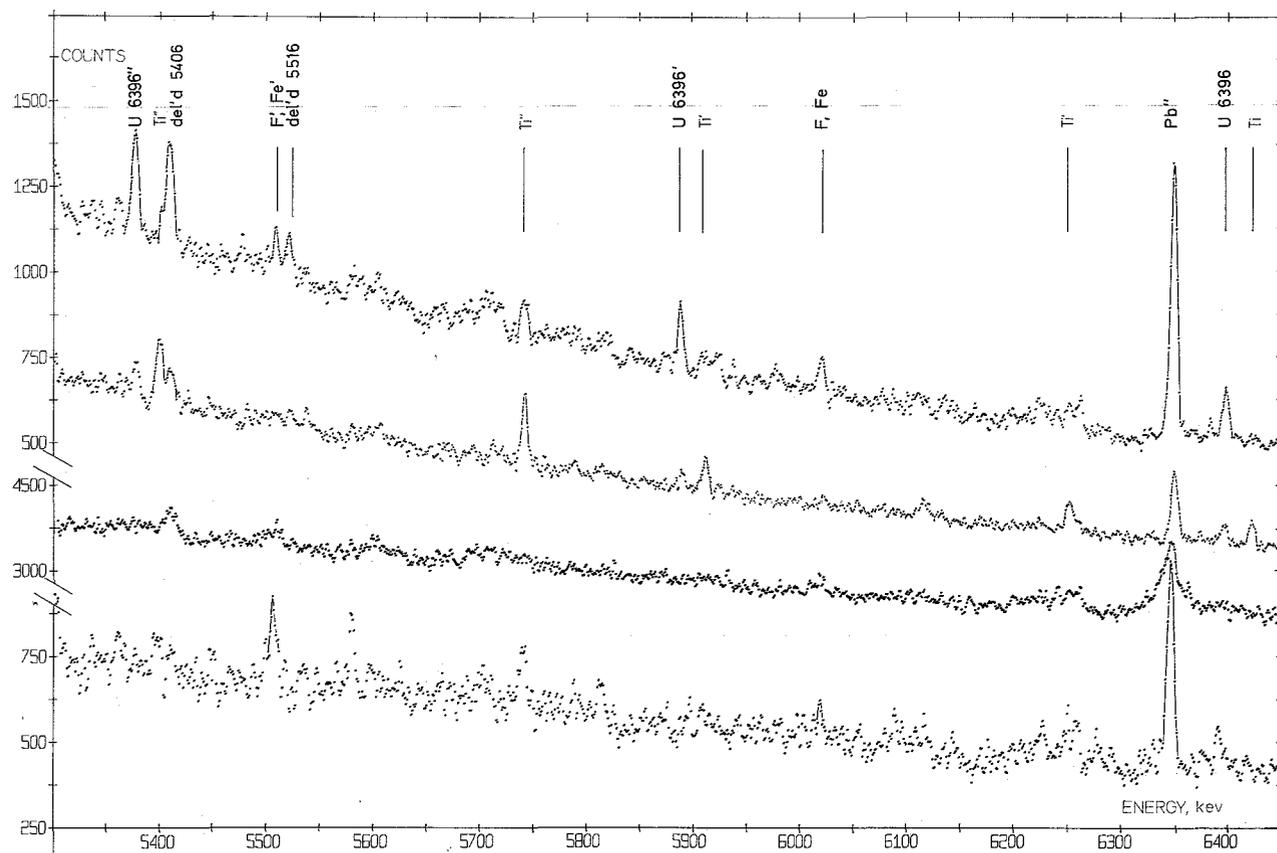
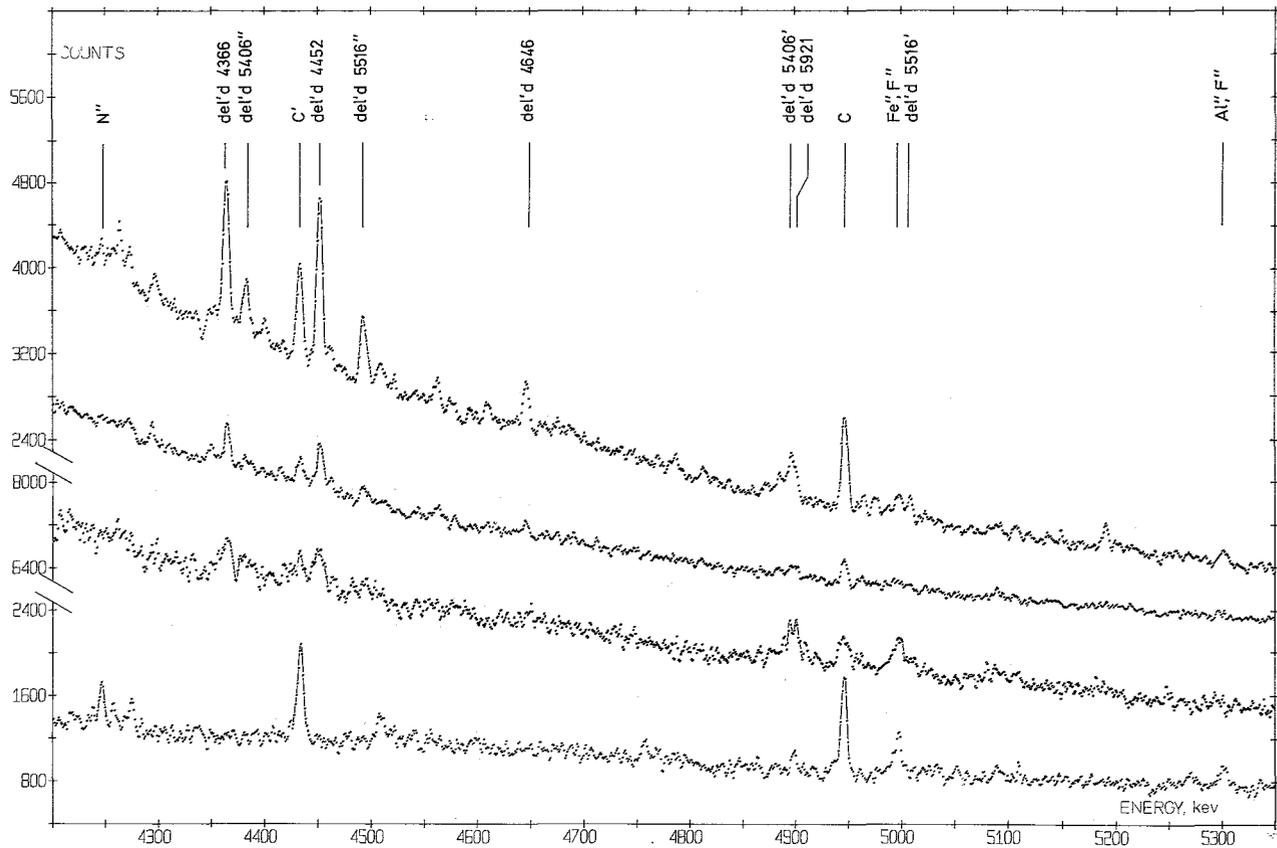


Fig. 4.

Gamma rays from interaction of thermal neutrons with ^{235}U . Top to bottom: anticoincidence, coincidence, delayed, and background spectrum.

Single and double primes indicate single and double escape peaks, respectively. Note different zero suppression.

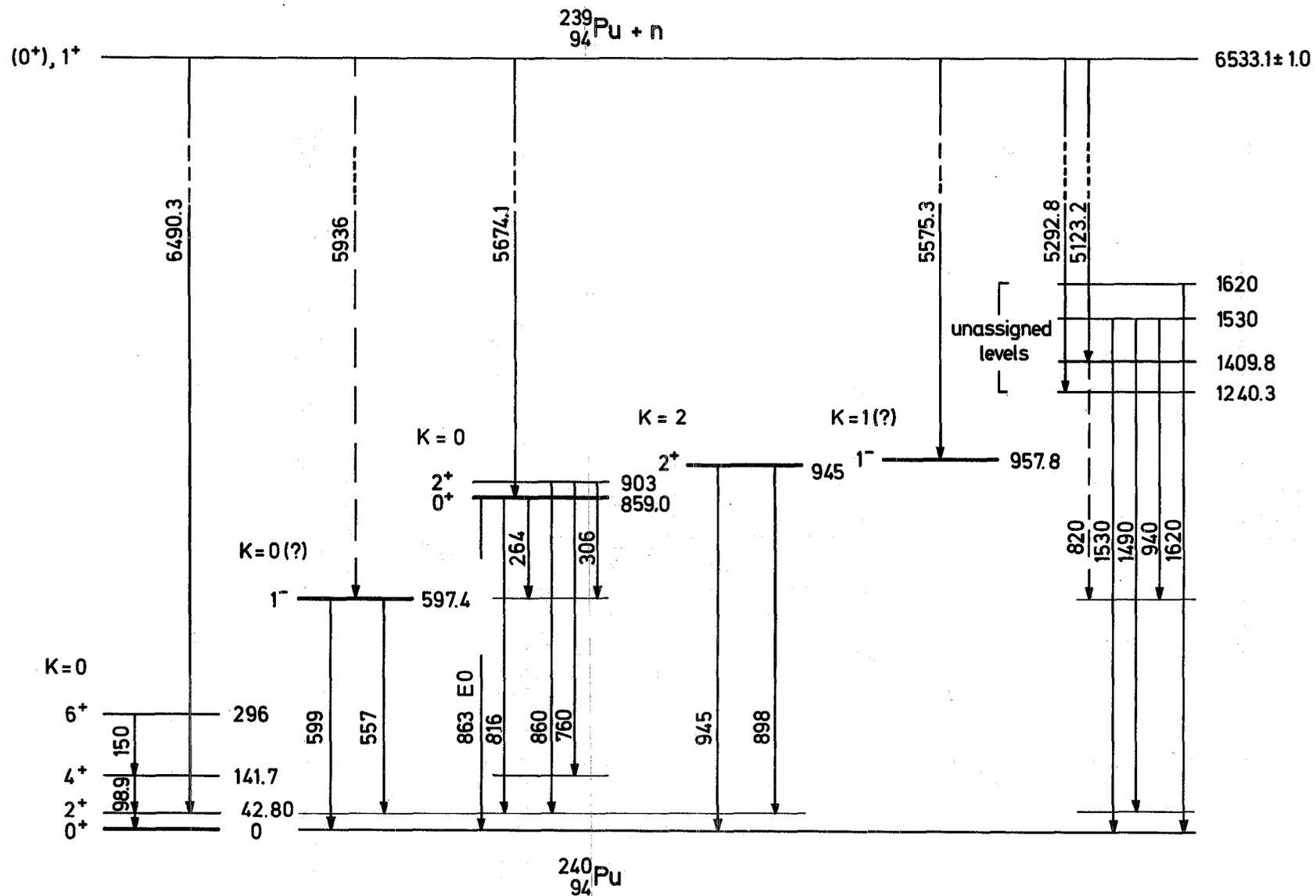


Fig. 5.
 Level diagram of ^{240}Pu . Primary transitions from present work only.
 Band heads are indicated by heavy lines.

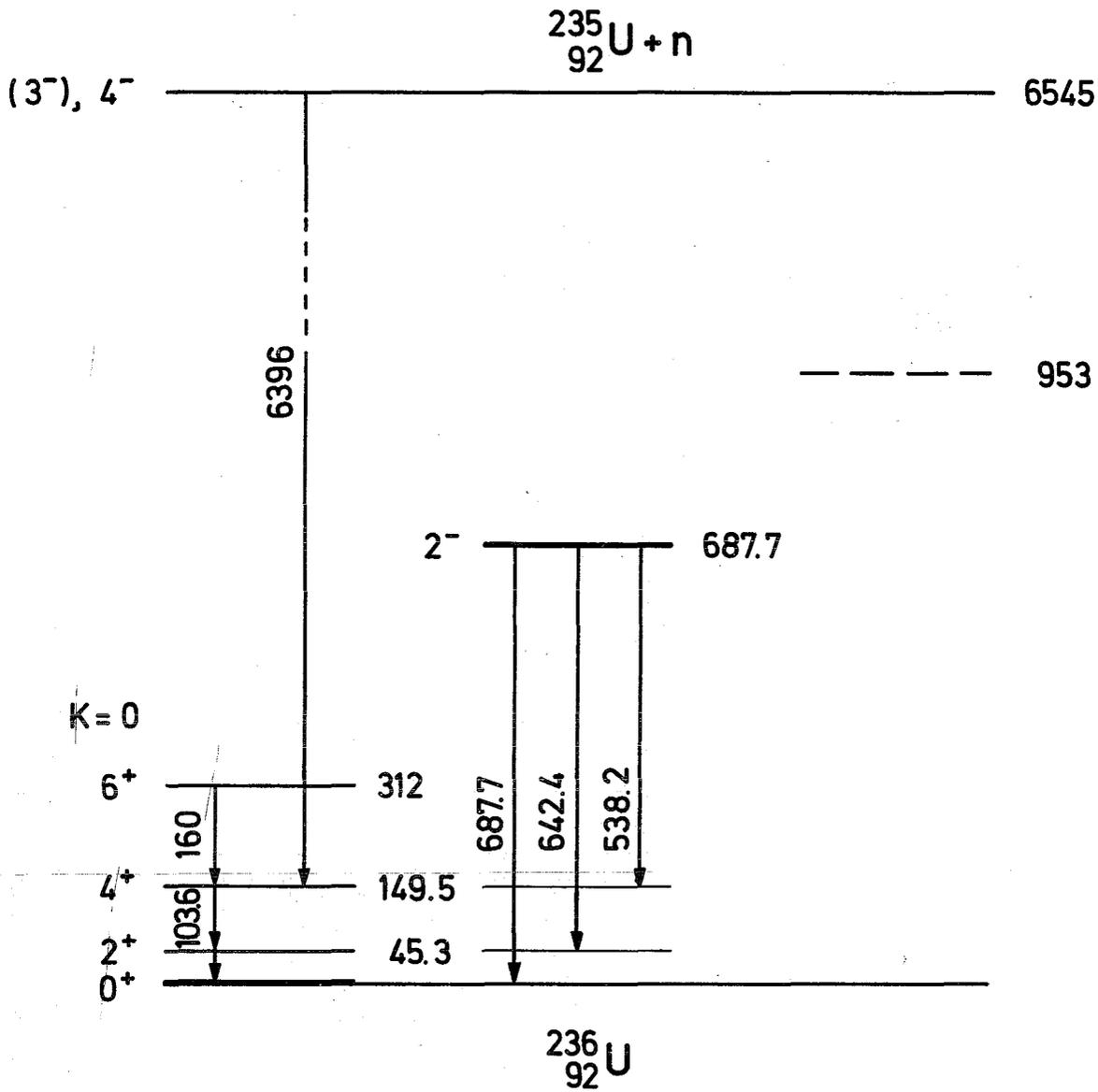


Fig. 6.

Level diagram of ^{236}U . Heavy lines indicate band heads.