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X-RAYS FROM FISSION

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## I. INTRODUCTION

This is a report on the activities pursued during the second year of the contract "X-rays from Fission." The main effort during this part of the contract has been to analyze and write-up the research work performed concerning spontaneous fission of  $^{252}\text{Cf}$ . This work involved (X-ray, X-ray) coincidence measurements, (X-ray,  $\gamma$ -ray) coincidence measurements, ( $\gamma$ -ray,  $\gamma$ -ray) coincidence measurements, and the setting up of a fission fragment time-of-flight experiment for determining fission mass yields. These experiments have resulted in new findings and have been published in The Physical Review. The results found in the (X-ray, X-ray) work was in some ways unexpected and we have conjectured that the high correlation of self X-ray coincidences occur due to internal conversion cascading. Furthermore, these internal conversion cascades are highly correlated and are not in agreement with a random or statistical model for the fission process. With this result in mind, one would expect these same measurements to vary with the internal energy of the fissioning nucleus. Thus one should measure X-ray self coincidence as a function of neutron energy for neutron induced fission. Furthermore, this result should vary with respect to the fissionable nucleus. Thus several different targets should be used. If these expectations hold, we believe that we will have a method for determining a quantity, important in determining source characteristics of the nuclide producing fission. At present, ours is the only data available.

Thus, we believe this avenue of exploration should be further investigated.

The (X-ray,  $\gamma$ -ray) work has pinned down a large number of  $\gamma$ -ray transitions as to their origin, that is, the specific isotope from which the transition arises. These  $\gamma$ -ray transitions are the clearest signature by which particular isotopic yields can be identified. Further work will be done since a nearly complete tabulation of gamma-ray energies and their transition properties needs to be tabulated to be confident in making yield determinations.

Since most measurements take approximately one month to run, careful thought needs to be given each experiment before initial data taking is begun. We believe, if no major set-backs develop, that the coming year should produce fruitful results.

Most of the work accomplished during the first six months of this year is summarized in the Semi-Annual Report. During the second six-month period of this year's contract, much attention has been given to setting up a time-of-flight experiment to measure mass yields directly. This experiment is discussed in detail in II.B. Initial results are promising.

## II. EXPERIMENTAL PROGRAMS

### A. Introduction

In recent years technological advances have provided new and more direct experimental methods (as compared to the traditional radio-chemical and mass spectrometric methods) of probing the phenomenon of nuclear fission. These methods, besides being able to provide an independent check on the results of the earlier "chemical" analyses, appear to be faster, more accurate, and capable of providing a wealth of other information on the details of fission. These new experimental procedures are due, in particular, to (1) the development of solid state particle detectors with intrinsically good energy resolution, (2) the availability of photon spectrometers of unprecedented resolution, and (3) subnanosecond time pick-off possibilities for these two types of detectors and a vast assortment of fast timing electronics from which subnanosecond timing measurements may be made. From these basic "building blocks" many types of experiments exploring the fission process may be designed and performed, the actual experiment being dictated by the particular aspect of fission which concerns the experimenter. Obviously, no one experiment will give all the answers.

The distribution of mass in nuclear fission after the emission of prompt neutrons may be directly determined by measuring the kinetic energies and the velocities of the fragments. The energy determination involves the use of the high resolution heavy particle detectors and the velocity determination involves fast timing techniques. The mass distribution before the prompt neutron emission is obtained by measuring

either the kinetic energies or the velocities of the post neutron complementary fission fragments and applying momentum conservation to the system. Of course in this case certain assumptions are implicitly made in such determinations. For the double velocity experiment it is sufficient to assume that the prompt neutron emission is isotropic in the center of mass system of the fragments. The double energy method requires other considerations based on the details of the neutron emission as a function of fragment mass.

A rather unique signature of the atomic number of a fission fragment is clearly the observation of one of its characteristic atomic X-rays. Since state-of-the-art photon spectrometers have the resolution needed to easily resolve these lines, the ability to directly detect  $Z$  for the fragment at the time of fission is at hand.

On the other hand, gamma rays emitted from excited fission fragments provide a signature of a nuclear type for identifying fragments and investigating the nuclear properties of fission products. From gamma decay studies nuclear decay schemes may be determined, which, besides being of great interest to the nuclear physicist, may help in the identification and monitoring of fission fragment yields.

It is now known that internal conversion is a very important method of prompt de-excitation of post neutron emission fission fragments. Hence, the detection of internal conversion electrons provides yet another signature as to identification and decay modes of excited fission fragments.

Experiments of the multiparameter coincidence type, in which information is stored event by event in a computer or on magnetic tape

are by far the best types of experiments from which correlations between the many fission parameters may be extracted. As an example, an experiment in which the energies of complementary fission fragments and the time-of-flight of one fragment are measured enables the determination of primary neutron masses, post neutron masses, pre-neutron kinetic energies, and prompt neutron emission as a function of mass and total kinetic energy.

The discussion in the above introduction is intended to indicate that with "state of the art" experimental equipment and procedures, a vast amount of correlated information concerning nuclear fission may be directly accumulated. Obviously, there are many types of experiments to perform, each having its own degree of complexity and information to contribute.

The experiments discussed in the following pages of the work performed at this laboratory on fission will help further the understanding of this important nuclear process.



## B. Time of Flight - Energy - X-ray Coincidence Measurements for the Determination of Isotopic Fission Yields

In order to obtain isotopic fission yields both charge and mass must be determined simultaneously. Direct physical techniques usually involve measuring in coincidence, at the time of fission, parameters from which Z and A can be determined.

Two general techniques can be used to determine the mass distribution. The pre-neutron mass distribution (for binary fission) can be extracted by measuring the kinetic energies of both fission fragments and applying conservation of energy and linear momentum. Post-neutron mass distributions (for binary or tertiary fission) can be determined by measuring the time of flight of the fragment over some distance and its kinetic energy. In the former technique three quantities determine the experimental mass resolution: (1) the influence of the angular distribution of the emitted neutrons on fragment recoil velocity, (2) the energy resolution of the fragment detectors, and (3) the energy loss of fragments in the target material. The dominant contribution originates from term (1). Typical quoted values are about  $4\mu$  (FWHM) for such experiments. In the case of the time of flight technique the experimental mass resolution is a function of (1) the energy resolution of the fragment detectors, (2) the timing resolution of the electronics, and (3) the angular resolution of the time of flight measurement. Fig. 1 shows how these quantities enter in the calculation of the mass resolution. Fig. 2 shows the contributions to the mass resolution for the reasonable

expected uncertainties  $\Delta T = 1 \text{ ns}$ ,  $\Delta E = 1 \text{ MeV}$ , and with  $\Delta\theta = 2^\circ$ ,  
 $E = 100 \text{ MeV}$ ,  $D = 10 \text{ m}$ ,  $M = 100 \text{ u}$ . The large solid angle for such a  
long flight path will be discussed later. The expected mass resolution  
 $\langle \Delta M/M \rangle \approx 1 \text{ u (FWHM)}$  which is due almost entirely to the energy uncertainty  
indicates that with this time of flight technique the integral nuclear  
mass distribution (i.e., quantized A yields) may be unfolded from the  
experimental mass spectrum with a great degree of uncertainty. Of  
course, the long flight path of 10 m implies a vanishingly small solid  
angle ( $4 \times 10^{-2} \text{ msr}$  for a  $300 \text{ mm}^2$  detector). Hence the experiment seems  
impossible because of count rate considerations. However, by using an  
electrostatic particle guide, consisting of a wire at a negative high  
voltage running down the center of a cylinder the length of the flight  
path, ions may be confined to trajectories around this wire as they  
traverse the guide. A tremendous increase in solid angle may be gained  
by using such a device. Fig. 3 shows the calculated electrostatic guide  
efficiency for both a point source a radial distance r from the center of  
the guide and an extended uniform source over the entire guide cross  
section. The collection efficiency in the case of the point source is  
proportional to the ionic charge and the voltage on the wire and inversely  
proportional to the fragment energy, as expected. As is seen from fig. 3  
an increase in solid angle of several orders of magnitude is expected  
over the case of no particle focusing. We feel that the time of flight  
technique using such a guide is a very practical means of extracting  
fission fragment integral (A values) mass yields. Such a guide has now

been constructed at the University of Texas Center for Nuclear Studies. A 10 m long stainless steel tube with an inside diameter of 5.05 cm has a 5 mil wire running its length. The high voltage is provided via a 50 keV high voltage feedthrough to which a voltage from 0 to 50 keV may be applied.

A preliminary experiment was performed to investigate the relative collection efficiency of this guide for fragments from spontaneous fission of  $^{252}\text{Cf}$ . A small  $^{252}\text{Cf}$  source (approximately  $10^6$  fissions/minute) was placed on axis about 2 inches from one end of the guide. A  $300\text{ mm}^2$  lithium drifted silicon detector with a depletion depth of  $100\ \mu\text{m}$  was placed centered on axis about 2 inches from the other end of the guide. At a pressure of  $1 \times 15^5$  torr the voltage on the wire was varied from 0 to 20 keV, and the number of fission fragments detected by the particle detector in 100 seconds was observed at each voltage setting. The results of these measurements are shown in fig. 4. Indeed, these measurements indicate that such an electrostatic particle guide increases the "effective" solid angle over the 10 m flight path by many orders of magnitude as suggested by the calculations. Therefore, the determination of the mass yields using the time of flight-energy-technique is now straightforward.

However, our interest centers on isotopic yields from fission, not the determination of the A distribution alone. Clearly, with present day resolution in photon spectroscopy any characteristic X-rays emitted from fission fragments may be experimentally observed to unambiguously identify the nuclear charge of those fragments. Of course, such

observations will only identify particular fission fragment elements. No statements about elemental yields may be made unless the origin(s) of these observed X-rays are known. The evidence from K X-ray - K X-ray coincidence studies that observed K X-ray emission from fission fragments originates predominantly in most cases from nuclear deexcitation via internal conversion is conclusive. Since internal conversion probabilities are nuclear structure dependent, and since the structure of each excited fission fragment is in most cases not known, the conversion coefficients cannot be calculated from theory and included in the data analysis. Hence, it is necessary to make certain assumptions concerning K X-ray emission from fission fragments if one is to extract elemental yields, or isotopic yields if K X-rays are observed in coincidence with mass. In the literature several attempts have been made to parameterize seemingly reasonable distributions for charge probability as a function of A and K X-ray emission probability as a function of A for a given Z and fit the data by varying these parameters via least squares fits. Such techniques seem very reasonable to obtain the general trend of fission fragment mass and charge yields, but clearly, accurate isotopic yields cannot be extracted from such a technique. Nuclear properties, unlike atomic properties, do not vary smoothly in general from nucleus to nucleus.

There is another technique which exploits the observation of X-rays associated with the fission fragments to determine nuclear charge, but in this case the origin of the X-rays is well understood. This technique is to create vacancies in the electron shells of the fragments

by a known atomic process and observe the fluorescence yields (the probability that a vacancy in a given shell results in a radiative transition). Since fluorescence yields have been measured experimentally and are understood theoretically, extraction of fission charge and isotope yields should be straightforward. The electron vacancies may be created by allowing the fission fragments to pass through a thin foil (such as carbon). The X-rays resulting from electron transitions may be observed with a high resolution photon detector situated near the foil. An experiment was performed at the Center for Nuclear Studies in which fragments were detected in coincidence with X-rays from a thin carbon foil through which the fragments passed. It was observed that two groups of L X-rays (corresponding to the light and the heavy fragments) were produced. The resolution was not good enough (due to the rather poor detector used) to resolve individual lines, but the peaks were located in the energy regions where these L X-rays were expected to appear. Thus, this technique has been demonstrated to work. It seems clear, then, that the technology is at hand to determine fission isotope yields with a very reliable method.

A triple coincidence experiment using these techniques to measure fission isotopic yields has now been essentially set up. The fissioning source is placed at one end of the electrostatic guide. One fission particle detector is placed on line with the center of the guide behind the source. Another fission particle detector is placed at the other end of the guide. In front of this detector is a thin carbon foil

which is observed by a high resolution photon detector. The particle detector near the source provides the start pulse and the other particle detector provides the stop pulse for the time of flight measurement. The electronics are arranged to require a triple coincidence between the pulses in both particle detectors and the pulse in the X-ray detector. When this coincidence occurs, all three energy pulses (2 particle and 1 X-ray) plus the time of flight are stored serially on magnetic tape for off-line analysis. Fig. 5 shows the electronics set up to be used in the experiment. The fast timing part of the circuit employs complementary logic to inhibit count rate problems associated with the particle detector near the source. The particle detector timing pulses are generated by ORTEC 130 fission fragment timing units resulting in time resolution of no more than 0.5 ns with excellent (1 MeV) energy resolution.

Two extremely important points should be noted concerning the design of this experiment, besides the fact that absolute isotopic fission yields will be measured for the first time. First, since the X-ray detector is 10 m from the source, extremely high neutron fluxes ( $10^{11}/\text{sec}/\text{cm}^2$ ) can be maintained on the target without any deterioration of the X-ray detector resolution. Hence, the count rate will be increased by a factor of  $10^4$  over that which would result if the X-ray detector were in the proximity of the source (no experiments have ever been performed with any success where X-ray detectors were in fluxes greater than  $10^7/\text{sec}/\text{cm}^2$ ). Second, for the case of  $^{252}\text{Cf}$ , if the experiment is performed again with a K X-ray detector near the source, observing K X-rays originating from

internal conversion processes, the results of the two experiments can be compared to extract internal conversion probability coefficients as a function of  $Z$  and  $A$  for the fission fragments. This would greatly help in the theoretical understanding of nuclear fission, and these coefficients would allow previous K X-ray experimental data to be reanalyzed with realistic probabilities for internal conversion.

$$(1) \quad \frac{\Delta E}{E} \doteq 10^{-2} \quad (\text{typical})$$

$$(2) \quad 2 \frac{\Delta T}{T} = 2 \frac{c}{D} \sqrt{\frac{2E}{Mc^2}} \Delta T \doteq 2.5 \cdot 10^{-3}$$

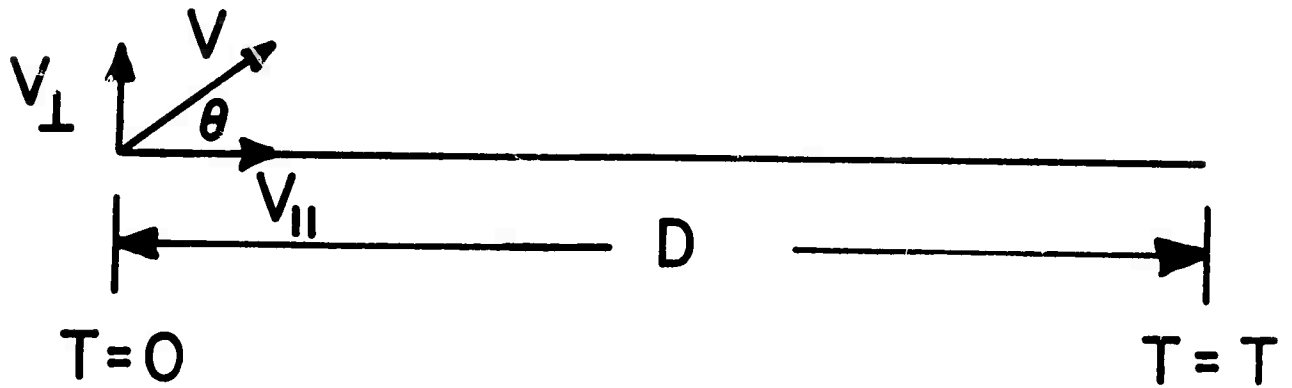
for  $E = 100 \text{ MeV}$ ,  $D = 10 \text{ m}$ ,  $M = 100 \text{ u}$ ,  
and  $\Delta T = 1 \text{ ns}$

$$(3) \quad 2 (\Delta \theta)^2 \doteq 2.5 \cdot 10^{-3}$$

for  $\Delta \Omega = 0.005 \text{ sr}$  ( $\Delta \theta \doteq 2^\circ$ )

$$\left\langle \frac{\Delta M}{M} \right\rangle_{\text{exp}} \doteq 1.06 \cdot 10^{-2}$$





$$V_{||} = \frac{D}{T}, \quad V_{\perp} = V \sin \theta = V \theta$$

$$M = \frac{2E}{V^2} = \frac{2E}{V_{\perp}^2 + V_{||}^2} = \frac{2E}{V^2 \theta^2 + \frac{D^2}{T^2}}$$

$$M(E, T, D, \theta) = 2E \frac{T^2}{D^2} (1 - \theta^2)$$

$$\frac{\Delta M}{M} = \frac{\Delta E}{E} + 2 \frac{\Delta T}{T} - 2 \frac{\Delta D}{D} - 2(\Delta \theta)^2$$

(1)                      (2)                      (3)

## ELECTROSTATIC PARTICLE GUIDE EFFICIENCY

point source

$$F(r) = (1/4) (q|V|/E) \left[ \ln(r/R) / \ln(s/R) \right] (1 - r^2/R^2)^{-1/2}$$

for  $r \doteq s \ll R$  ,  $F(r) \doteq (1/4)(q|V|/E)$

$E \doteq 100$  MeV ,  $q \doteq 20$  e ,  $V \doteq 30$  keV

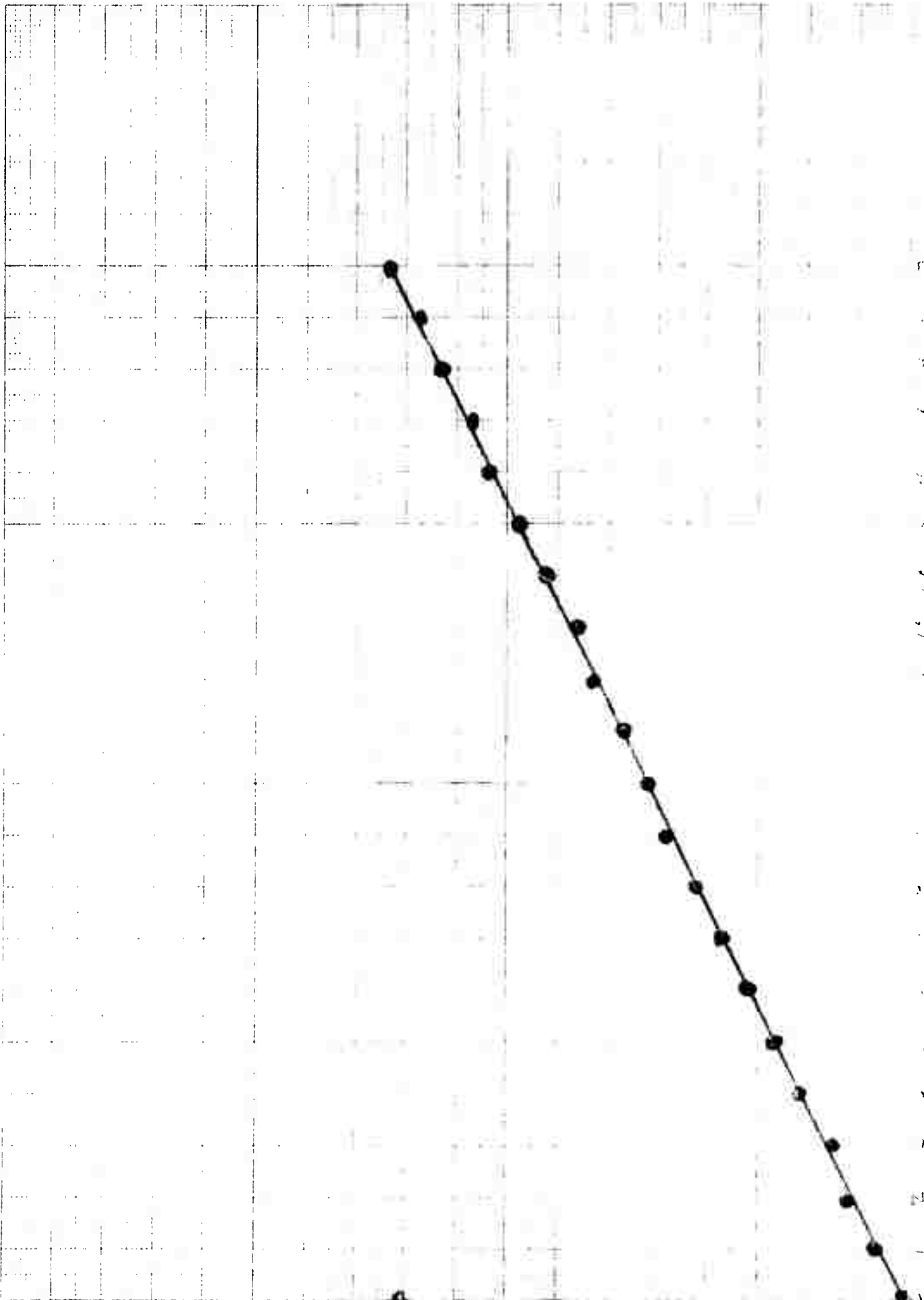
$F(r) \doteq 1.5 \cdot 10^{-3}$  or 19 msr

extended target

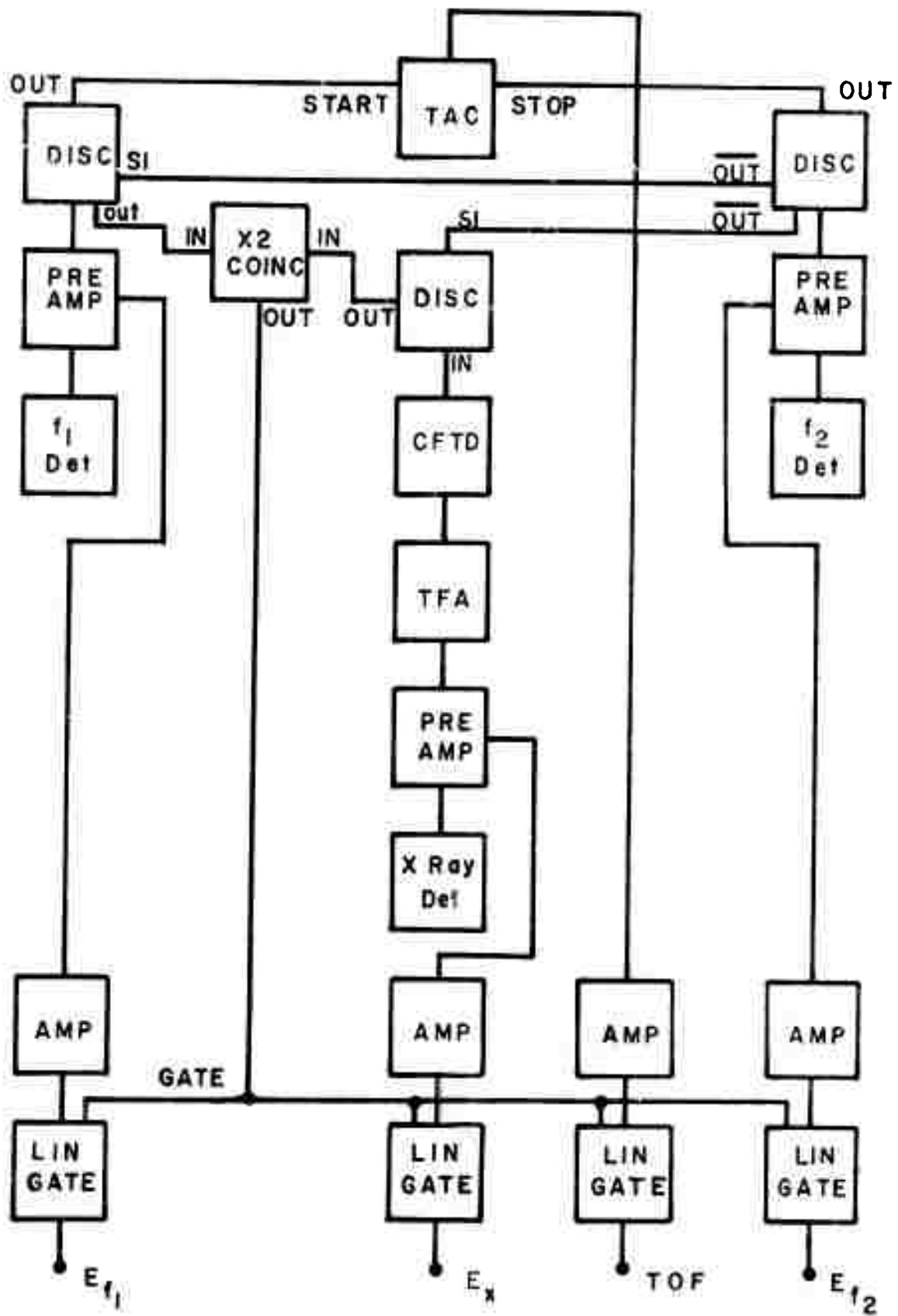
$$F \doteq 0.153 q|V| / [E \ln(R/s)]$$

for  $R = 2''$  ,  $s = 0.005''$

$F \doteq 2 \cdot 10^{-4}$  or 2.5 msr



Fragmente/100 SAC



TRIPLE COINCIDENCE CIRCUIT

### C. L,M X-ray Measurements

Signatures for the identification of specific fission product isotopes may be provided by the observation of the characteristic modes of prompt deexcitation of the excited fission fragments. Internal conversion electrons, X-rays,  $\gamma$ -rays, fragment energies and velocities, are all examples of such signatures currently being exploited to provide information concerning nuclear fission. Usually, the experiments performed are of the multiparameter coincidence type, since such techniques considerably reduce the complexity of the spectra involved.

It is well known that fission fragments may emit X-rays characteristic of the nuclear charge of the fragment nuclei. State of the art X-ray spectrometers have the necessary resolution to clearly identify the characteristic K X-ray - K X-ray coincidence experiment for  $^{252}\text{Cf}$  that the real coincidences would be predominantly between X-rays from the complementary pair, and that the assignment of elements for each complementary pair would result. Indeed, the complementary elements have been unambiguously determined. However, a very interesting and unexpected result was the occurrence of a large number of coincident events between K X-rays from the same element. This result has been interpreted as originating from multiple internal conversion processes from cascading transitions.

In order to determine the fission fragment elemental yields from the experimental observation of the characteristic X-rays emitted

by the fragments, it is necessary to know the origin of these X-rays. Only then will it be possible to make corrections on the observed yields to obtain realistic values for the absolute yields.

The question of whether or not X-rays from fission are produced as consequences of processes other than those originating from nuclear de-excitation of the fragments (e.g., internal conversion) is of considerable interest. For example, electron shell vacancies may be produced as a result of atomic collisions between the high energy fission fragment and the fissionable source material. Such processes are more likely to occur for L X-rays than K X-rays and hence the experiment which is suggested is one involving the identification of the fission fragment characteristic L X-rays. In this paper we report a K X-ray - L X-ray coincident experiment for  $^{252}\text{Cf}$  from which there is strong support for L X-ray production in fission by other than nuclear processes.

## EXPERIMENTAL PROCEDURE

### 1. Source and Detectors

A very small  $^{252}\text{Cf}$  source (approximately 500 fissions/sec) was sandwiched between a thin beryllium dish (.002 in) and a thin piece of paper (0.025 mm), and placed between the faces of two Kevex silicon X-ray detectors (active areas of  $0.09\text{ cm}^3$  and  $0.03\text{ cm}^3$ ). The dish and the paper stopped the fission fragments and prevented Doppler broadening of the X-ray spectra. The paper side of the source faced the detector used to observe the L X-ray spectrum ( $0.03\text{ cm}^3$ ).

From the K X-ray coincidence experiments it is known that the more prolific fission fragment K X-ray emitters are Y, Zr, Nb, Mo, Tc, Ru, and Rh for the lighter elements and Pr, Ce, La, Ba, Ls, Xe, and I for the heavier elements. The L X-rays for these elements are between 2-3 keV and 4-5 keV for the light and heavy elements respectively. The typical energy separation between L X-rays of adjacent elements is about 100 eV for the light fragments and 200 eV for the heavier fragments. Unfortunately, the detectors used in this experiment had suffered some neutron damage and the resolution had deteriorated to 270 eV at 6 keV. Nevertheless, it was felt that any large anomalies between K X-ray and L X-ray measurements would be discernable. It was necessary to take into consideration the attenuation of the L X-rays by the paper covering the source. Corrections were made using the normal relation  $I = I_0 \exp(-\mu x)$ . Corrections due to counter efficiency have also been made. From 2-6 keV both counters

had efficiencies ranging from 42-100% and between 10-40 keV for the K X-ray detector the efficiency varied from 100% to 24%.

## 2. Coincidence System

The usual electronics circuitry for simple X-ray - X-ray coincidence measurements was used as shown in fig. 1. It was necessary to increase the gain on the L X-ray linear amplifier by a factor of 5 over the previous experiments because of the lower energy of these X-rays, creating the problem of many long tail saturated signals originating from  $\gamma$  and K X-rays leaving this amplifier. This meant that a very weak source had to be used to stop the tails of such saturated pulses from destroying the resolution. A two parameter analysis was performed on-line using a PDP-15 computer. The gated linear signals from the L X-ray detector were sorted and stored in 1024 channel spectra according to windows set in the computer on the K X-ray peaks in the gated spectrum from the other detector.

## 3. Calibration

The  $K\alpha$  X-ray spectrum was calibrated by observing the positions of 13 known  $K\alpha$  X-ray peaks. The energies of the other peaks were determined by a least squares fit to a third order polynomial function. The  $L_{\alpha,\beta}$  X-ray spectrum was calibrated with  $^{55}\text{Fe}$  and  $^{137}\text{Cs}$  sources and by observing the position of the  $M_{\alpha,\beta}$  X-ray peaks from Cm when sorting by the characteristic respective Cm L X-rays. The least squares fitting to a



third order polynomial was done as before to establish the final energy scale.

#### 4. Windows

When the peaks in the K X-ray spectrum were sufficiently resolved, 14 gates were set on the  $K\alpha$  X-rays of Y, Zr, Nb, Mo, Tc, Ru, Rh, I, Xe, Cs, Ba, La, Ce, and Pr. The coincident L X-rays were sorted and stored in 14 corresponding 1024 channel spectra for subsequent analysis.

## RESULTS

Figure 2 shows the total K X-ray coincidence spectrum for the K X-ray - L X-ray coincidence experiment. This spectrum is very similar to that obtained in the K X-ray - K X-ray coincidence experiment with the exception that very prominent L X-ray lines from  $^{248}\text{Cm}$  occur at the positions of the K X-rays from Y, Tc, and Ru. These L X-rays are in self-coincidence with  $^{248}\text{Cm}$  M X-rays in the complementary spectrum. Figs. 3-9 show some typical gated coincidence spectra. In these figures the total L X-ray coincidence spectrum is shown directly above the gated spectrum for comparison. These examples show that both fission fragment K X-ray - L X-ray and  $^{248}\text{Cm}$  L X-ray - M X-ray coincidences occur. Since  $^{252}\text{Cf}$  undergoes about 30 alpha decays for every fission, the reason for the large peaks associated with  $^{248}\text{Cm}$  is understood.

The principle of this experiment was to investigate L X-ray production in fission. Because of the nature of the experiment (i.e., L X-rays in coincidence with L X-rays) it was expected that the results would show a predominance of K - L self-coincidences for each element. One would expect for each K X-ray that a corresponding L X-ray would be emitted. In fact, because of the large solid angle of the X-ray detectors (approximately 0.5 sr) it is estimated that the number of self-coincidences should be typically a factor of 100 larger than the number of complementary coincidences. However, this is not the case.

The number of self-coincidences are in most cases comparable to the number of complementary coincidences. However, the problem is resolved when one recalls that the fluorescence yields for the elements of the fission fragments are between 0.05 and 0.15, while the total cross section for L X-ray production due to the stopping process is near unity for fission fragments traversing a piece of paper approximately 0.1 mm thick. Calculations give a theoretical self-coincidence/complementary coincidence ratio

$$R = \frac{\bar{W}_L^S}{P_L^C} + \frac{P_L^S}{P_L^C}$$

where  $\bar{W}_L^S$  = mean L X-ray fluorescent

yield for L X-rays of same element as the gate K X-ray.

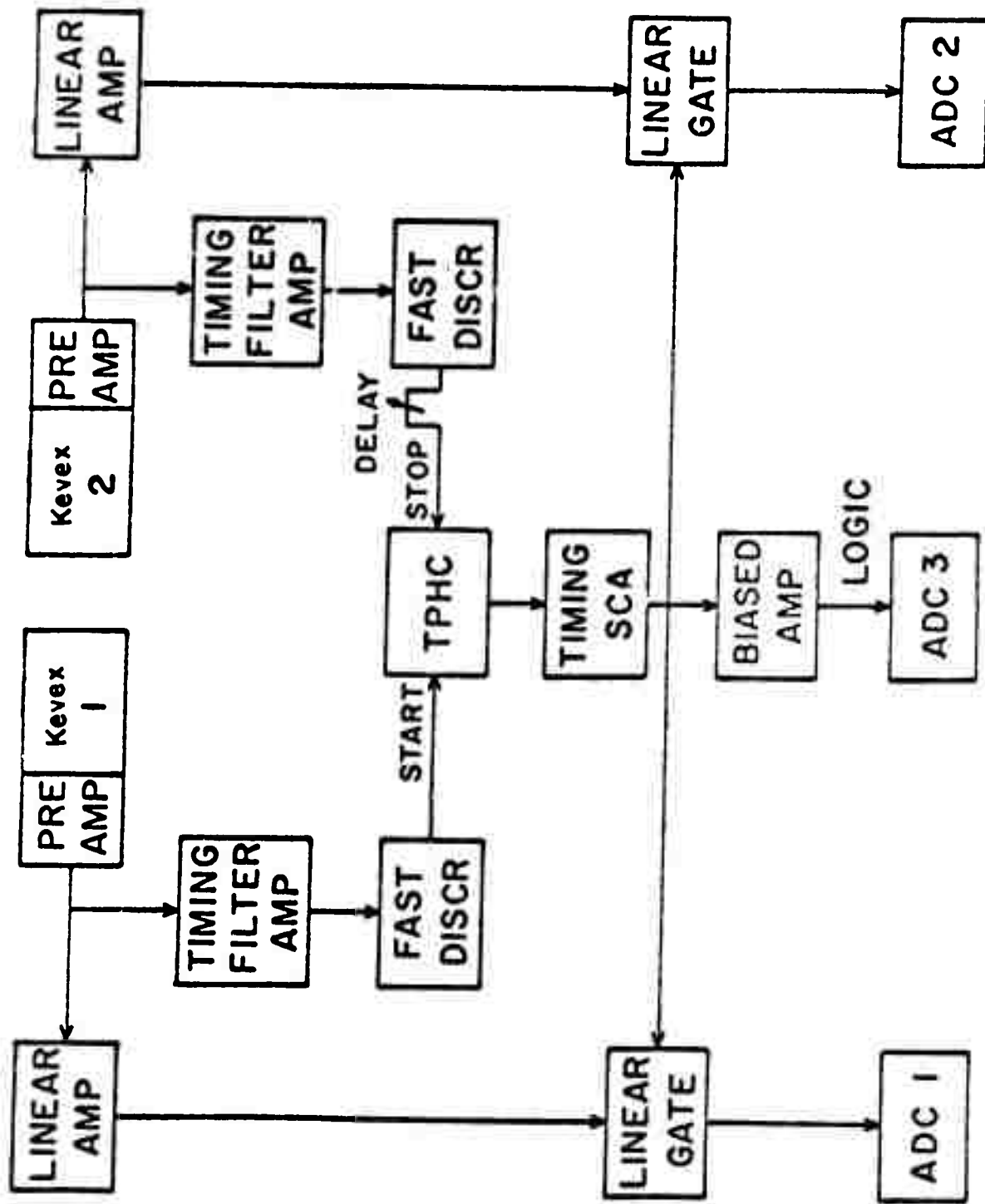
$P_L^S$  = probability of L X-ray production due to stopping process for L X-rays of same element as the gate K X-ray.

$P_L^C$  = probability of L X-ray production due to stopping process for L X-rays of complementary element of the gate.

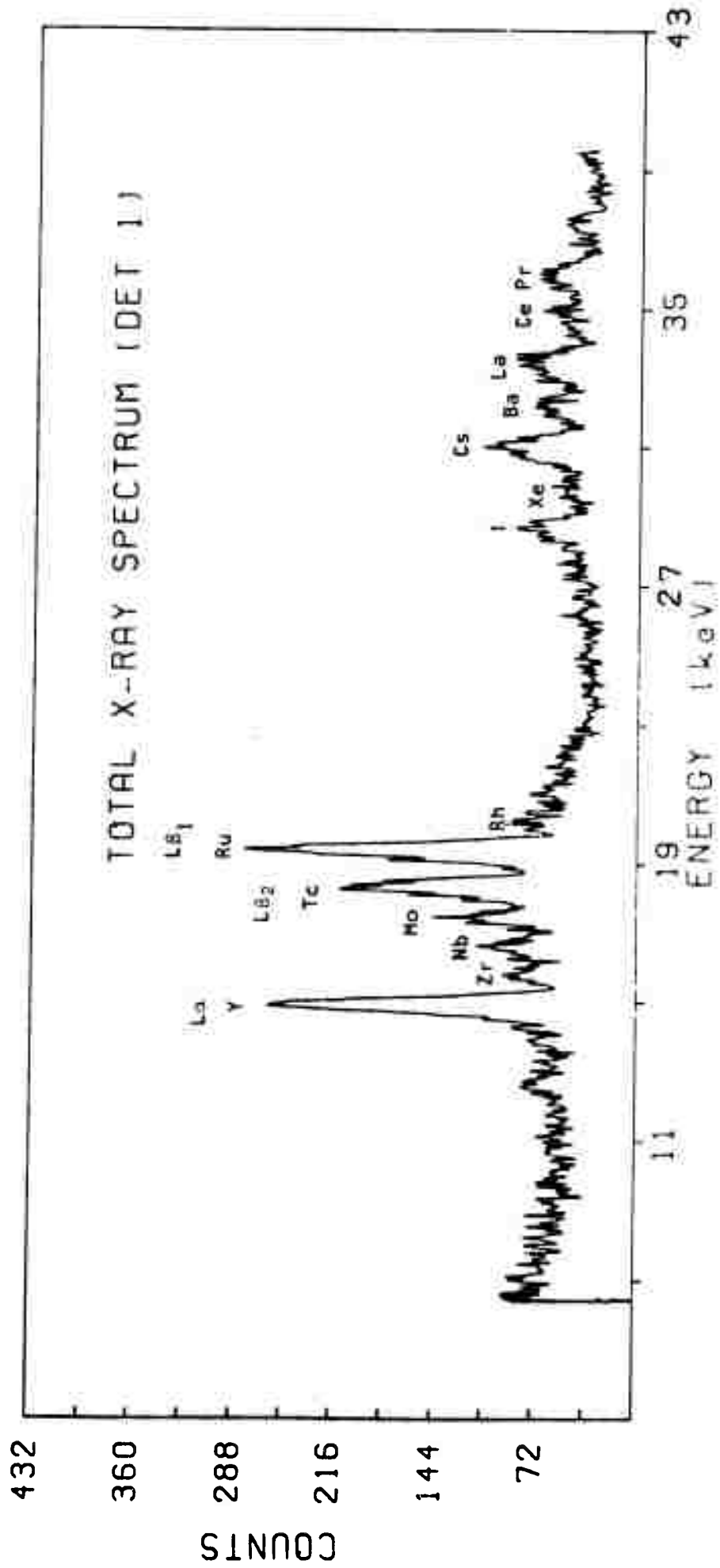
Hence, since  $\bar{W}_L^S \ll P_L^S$ ,  $R \approx \frac{P_L^S}{P_L^C}$  and the ratios are not expected to show very erratic fluxuation in moving through the complementary pair of elements.

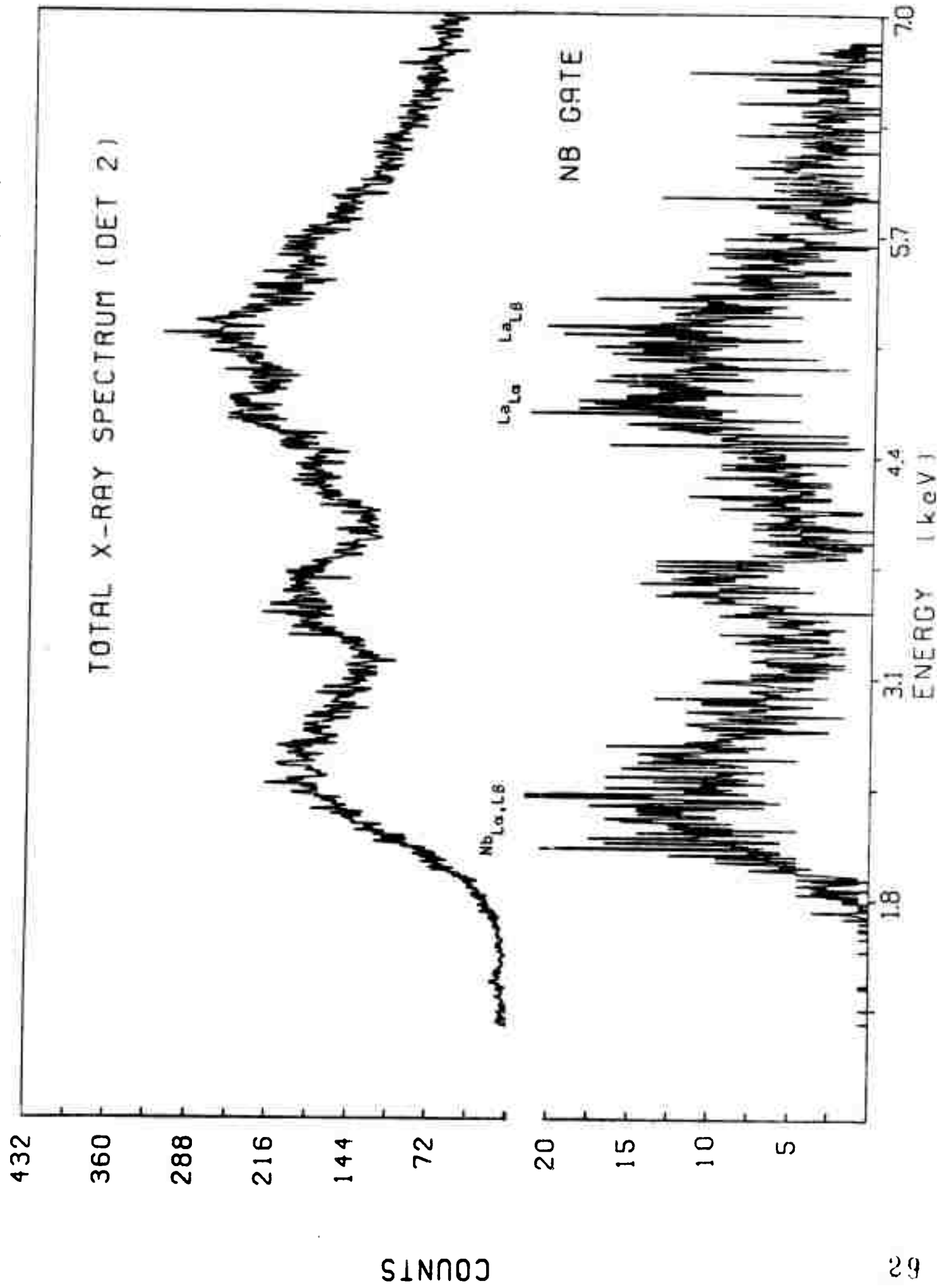
This appears to be the case from our rather poor data. Hence, we conclude that L X-rays are produced, not primarily from the fission process itself, but rather from processes involving the stopping of the fragments in matter. This latter aspect may be used in future experiments as a means of producing L X-rays for the charge identification of fission fragments.

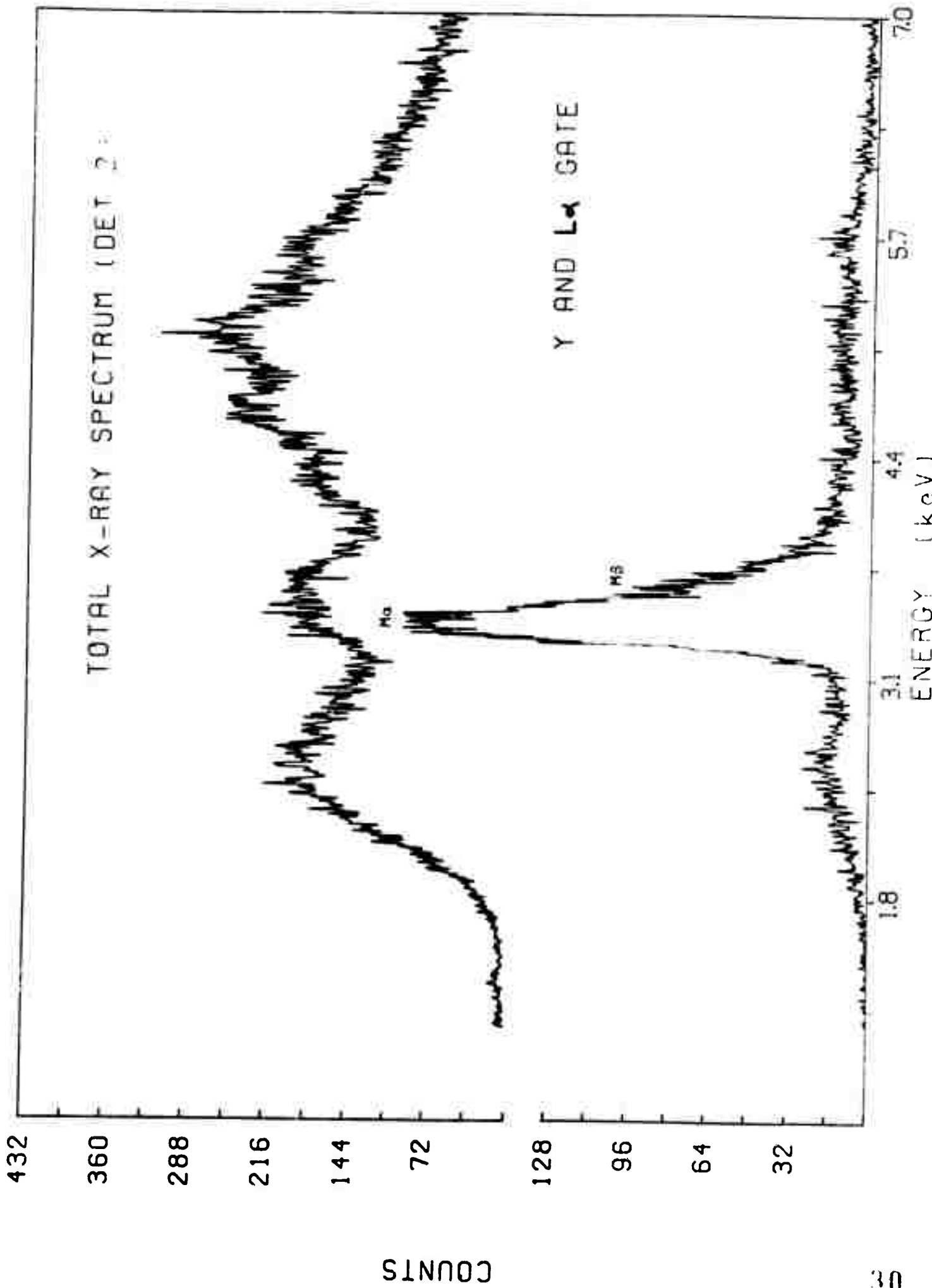
It has the advantage that (1) the electromagnetic stopping process is easy to understand and treat theoretically, and (2) the L X-ray production cross section is large.



CIRCUITRY USED IN COINCIDENCE EXPERIMENT.









432

360

288

216

144

72

48

36

24

12

COUNTS

TOTAL X-RAY SPECTRUM (DET 2)

Tc<sub>La,LS</sub>

Tc AND L $\beta$  GATE

Cs<sub>La</sub> Cs<sub>LS</sub>

1.8

3.1

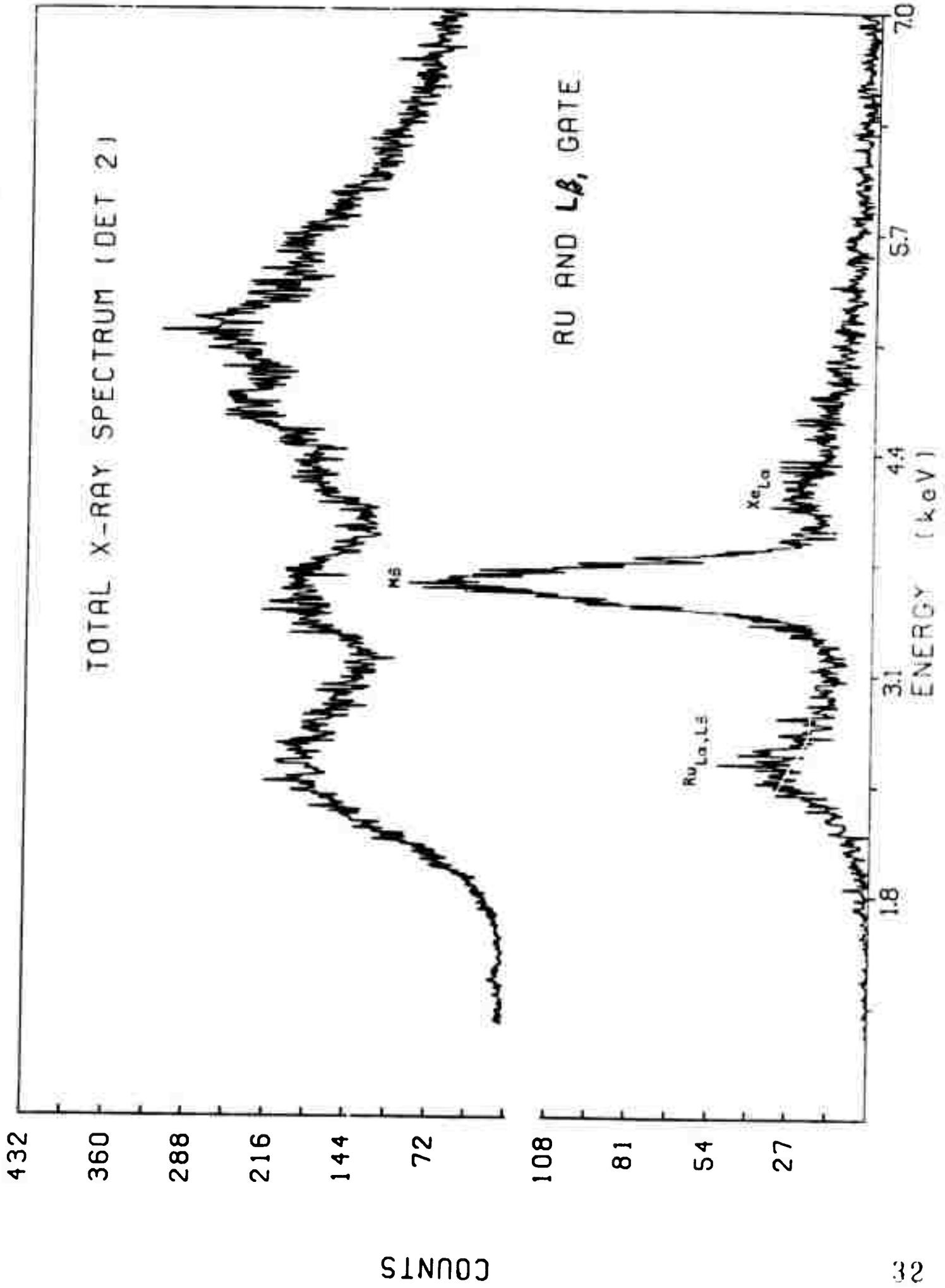
4.4

5.7

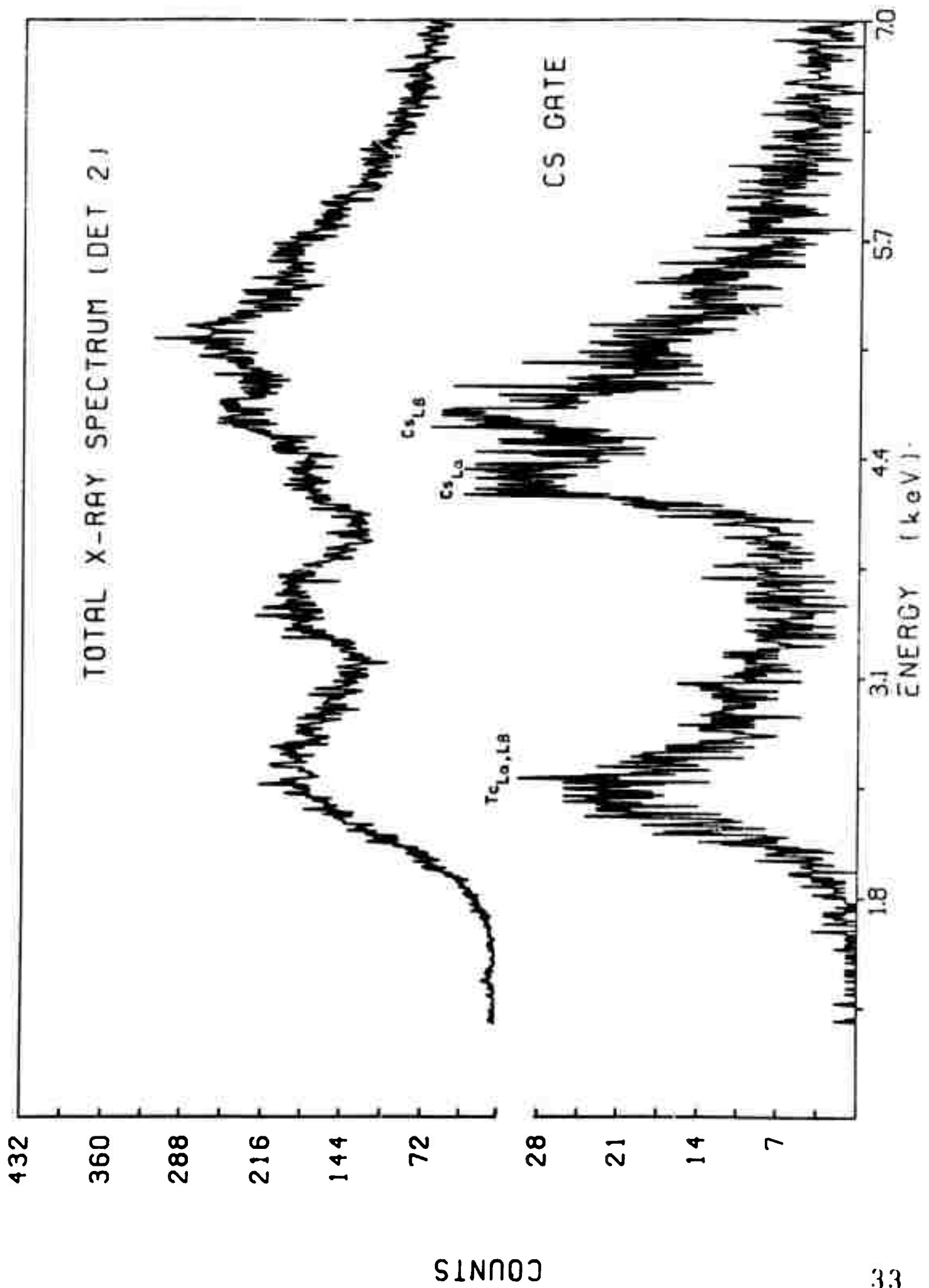
7.0

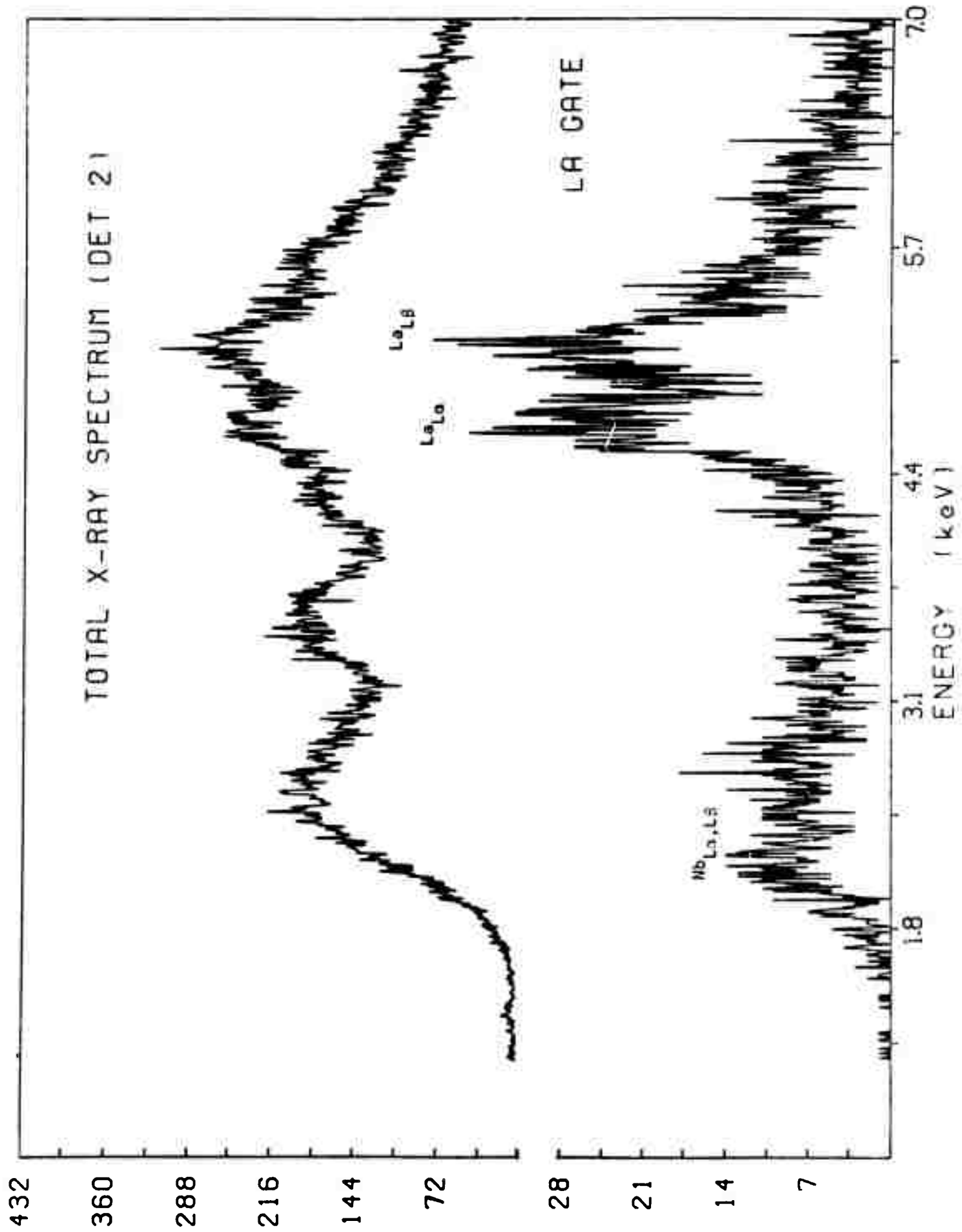
ENERGY (keV)

31

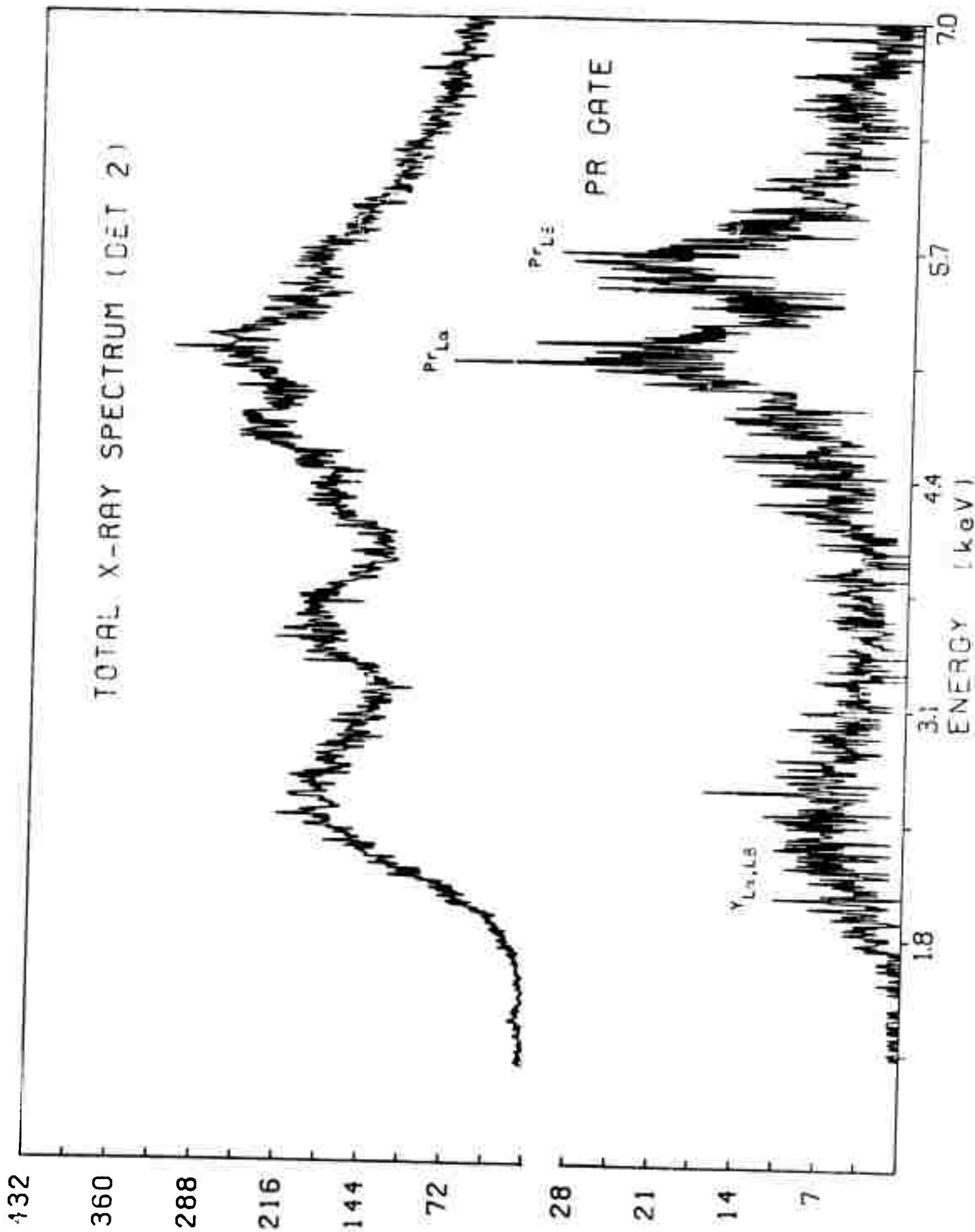


COUNTS





COUNTS



#### D. $\gamma$ - $\gamma$ Coincidence Measurements

Previous experiments<sup>1,2</sup> have placed several gamma transitions in specific fission fragment isotopes of  $^{252}\text{Cf}$ . The X- $\gamma$  coincidence technique used resulted in spectra in which only the strongest lines from the most frequently detected isotopes were of sufficient intensity to be discerned. These lines serve as a basis for a  $\gamma$ - $\gamma$  coincidence experiment in which sequential  $\gamma$  decay of particular post-fission isotopes are being investigated. Fission fragments have large kinetic energies ( $\sim 100$  MeV) and travel at velocities on the order of  $1$  cm/ns. Hence, in allowing the fragments to separate, and by stopping one of the fragments in a short time with a thin foil placed between two high resolution Ge(Li) photon spectrometers, an unambiguous  $\gamma$ - $\gamma$  coincidence measurement can be made for that fragment. A  $\gamma$ -ray -  $\gamma$ -ray coincidence experiment utilizing a small high resolution Ge(Li) detector and a large Ge(Li) detector in extended geometry has been completed after about six weeks of actual data-taking time. This data is currently being analyzed. The preliminary results will be discussed at the April meeting of the American Physical Society. An additional experiment with the detectors in close geometry will be performed in the immediate future.

The determination of the level schemes is absolutely necessary in any attempt to measure yields based on either X-ray production or  $\gamma$ -ray production, since both depend to a large extent on the energy spacings and spin differences involved. The only prompt yields which

have been measured are those for the even-even products of the spontaneous fission of  $^{252}\text{Cf}^3$  which afforded a convenient but regrettably unique group of cases where the  $\gamma$ -cascades go almost always through a  $2^+$  (1st excited state) to  $0^+$  (ground state) transition. Knowledge of this particular behavior, which is not expected to be exhibited by other nuclei, was a prerequisite to knowing the yields.  $\gamma$ -rays, X-rays, and internal conversion electrons are the only prompt radiations, outside of nondescript neutron spectra, available for detection and as such represent the main hope for most any kind of accurate measurement. It is obvious that until more is known about specific transitions within odd Z and odd A nuclei, the detailed distribution for fission of a given heavy element will remain a mystery only slightly outlined by the work to date.

- 1) F. F. Hopkins et al., Phys. Rev. C 4, 1927 (1971).
- 2) F. F. Hopkins et al., Phys. Rev. C 5, 1015 (1972).
- 3) E. Cheifetz, J. B. Wilhelmy, R. C. Jared, and S. G. Thompson, Phys. Rev. C 4, 1913 (1971).

## E. Fission Studies with a Curved Crystal Spectrometer

In the last two contract years we have used Si(Li) detectors to measure X-ray yields from fission fragments. These experiments allow the determination of the charge division following fission. The main considerations in using these detectors to study fission X-ray yields are (1) the count rate that can be obtained in an experimental situation and (2) the energy resolution compared to the energy separation of X-rays from adjacent elements as well as the separation of various components of a single element. The count rate is determined by the efficiency of the Si(Li) detector and the solid angle that it subtends. The efficiency is of the order of 100% to 30% depending on the energy of the X-ray. The solid angle however is very small since the typical area of our detectors is  $10 \text{ mm}^2$ . If this detector is placed at one inch from the fissioning source then the percent solid angle is  $100 \times (d\Omega/4\pi)\% \sim 1.2\%$ . Since all the experiments performed in this project are coincidence experiments, this is a limiting feature on the feasibility of many important researches. The X-ray  $\gamma$ -ray, X-ray X-ray and the X-ray fission fragment time-of-flight coincidences are all examples of these important types of experiments to determine relative fission yields (R-values) for all isotopes. The count rate and the resolution obtained with Si(Li) detectors are closely related since the physical size of the detector determines both the limiting energy resolution and the solid angle of the device. The smaller the area of the detector the better the resolution and of course the smaller the solid angle. The state of the art resolution that can be obtained is 160 eV at 5.9 keV.



A tremendous improvement in the energy resolution can be obtained by using crystal spectrometers instead of Si(Li) detectors. It is possible to obtain 6 eV resolution with a flat crystal in the place of the 160 eV at 5.9 keV obtained with Si(Li) detectors. The difficulty is that the efficiency with the flat crystal spectrometer is of the order of  $10^{-4}\%$  compared to 1% with Si(Li). The same resolution of 6 eV can however be obtained with a curved crystal spectrometer with an efficiency in the range of 1% to 0.1%. Such spectrometers are now commercially available.

We therefore feel that using a curved crystal spectrometer is a natural step in the direction of greatly improving on the important coincidence experiments of the type X-ray - ff; X-ray  $\gamma$ -ray, etc. If this system is indeed demonstrated to work during the next contract year, it should lead to much improved R-value determinations. Another important feature in using a high resolution curved crystal spectrometer is that it will allow us to separate low energy  $\gamma$ -rays from K X-rays in cases where the energies are very nearly equal.

## F. Lifetime Measurements

As an aid in resolving the many uncertainties which will arise from the  $\gamma$ - $\gamma$  coincidence data, an experiment is about to be performed to determine nuclear lifetimes in the range from 1 to 30 ns. Previous work<sup>1,2</sup> has revealed a multitude of low energy transitions, many of which are expected to have lifetimes in this range. The determination of the level schemes of the prompt products is especially difficult since de-excitation only occurs via  $\gamma$  emission and internal conversion. In contrast, the lower masses for the same element will be populated by beta decay, a selective process which itself provides some information about spins and which leads to  $\gamma$  decay as well. The knowledge that two or more transitions are in the same nucleus and are associated with approximately the same lifetime is an indication that they may be in a cascade, i.e., one sequentially following the other. Such information will be useful in sorting out the possible  $\gamma$ -decay schemes.

The experimental method consists of a series of intensity measurements with a high resolution Ge(Li) detector placed along a well collimated parallel flux of fission fragments various distances from a  $^{252}\text{Cf}$  source, but the same distance from the particle flux.

- 1) F. F. Hopkins, et al., Phys.Rev. C 4, 1927 (1971).
- 2) F. F. Hopkins, et al., to be published.

### G. Internal Conversion Electrons from the Fission of $^{252}\text{Cf}$

The study of internal conversion electrons is a very effective way to obtain detailed information on the low energy transitions in primary post-neutron fission fragments and to identify these fragments. Previous work at this laboratory<sup>1)</sup> investigating the X-ray, X-ray coincidence spectra from the fragments arising from spontaneous fission of  $^{252}\text{Cf}$  has shown that internal conversion cascades appear to be a very prominent mode of fragment de-excitation. Observation of the internal conversion electrons will confirm this hypothesis. A large solid angle axial beta-ray spectrometer has recently been designed and constructed for observation of these electrons. Many general types of experiments are planned. A conversion electron-conversion electron coincidence experiment should help confirm the cascade nature of internal conversion suggested from the previous experiments<sup>1)</sup> and provide further insight into the decay schemes of excited fission fragments. An X-ray - internal conversion coincidence measurement will allow the identification of the conversion transition with the atomic number of the converting fragment. This experiment in conjunction with mass measurements for internal conversion transitions (see section on time of flight experiments) will help in the assignment of a particular isotope to the observed conversion electron. Hence fission isotopic yields will be determined as well as information on the nuclear excitation of the prompt fission fragment.

1) Rodrigue St-Laurent, et al., Phys. Rev. C, 4, 1948 (1971).

#### H. Neutron Induced Fission for $^{233,235,238}\text{U}$ and $^{239}\text{Pu}$ Targets

A systematic study of fission yields from neutron induced fission of  $^{233,235,238}\text{U}$  and  $^{239}\text{Pu}$  targets using the same experimental techniques as developed for spontaneous fissioning  $^{252}\text{Cf}$  is to be undertaken. However, these experiments are inherently more difficult than the previous  $^{252}\text{Cf}$  work because of the large neutron flux needed to obtain acceptable counting rates. If the only effect of the neutrons was to induce the fissioning of the target nucleus plus nucleon system, there would be no problem. However, the Si(Li) and Ge(Li) photon spectrometers as well as the fission fragment detectors must be shielded to some degree from the neutron source in order to insure continued performance for the duration of the experimental runs. A quantity of  $10^9 - 10^{11}$  neutrons is known to destroy the resolution in such detectors.

The Center for Nuclear Studies at the University of Texas acquired a Model 9400 Neutron Generator from Texas Nuclear Corporation for an extended loan period. This generator is designed to produce a positive (deuteron or proton) ion beam in excess of 1 milliamperere. The total flux produced by this machine is claimed to be about  $10^{11}$  neutrons per second.

Because the generator had not been used for some time when it was loaned to the University, it is not presently in working order. A great deal of progress has been made toward restoring the generator to good working condition, however.

The generator will be enclosed in a "Block House," the isotropic neutron source of  $10^{11}$  neutrons/sec shielded from the exterior by several meters of concrete except for a collimated opening viewing

the target. The parts of the Block House directly between the source and the high resolution detectors will be made of lead. In this way a flux of about  $10^7 - 10^8$  neutrons/cm<sup>2</sup>/sec will fall on the target, while the flux going from the source directly to the detectors will be from  $10^3 - 10^4$  neutrons/cm<sup>2</sup>/sec. Under these conditions the detectors can be expected to last about  $10^6 - 10^7$  sec. The neutron flux on the target should be sufficient to produce tolerable count rates. One advantage the neutron induced fission will have over the spontaneous fission of <sup>252</sup>Cf is that larger and thicker targets may now be used. This will greatly increase the count rates. It should be noted that the electrostatic guide used for determining the mass of the fission fragments is particularly well suited for extended targets.

The experiments to be performed are those already developed for investigating the spontaneous fission of <sup>252</sup>Cf, namely the coincidence experiments: 1) X-ray/X-ray, (2) X-ray/ $\gamma$ -ray, (3)  $\gamma$ -ray/ $\gamma$ -ray, (4) X-ray/mass, (5)  $\gamma$ -ray/mass, (6) X-ray/internal conversion electron, (7)  $\gamma$ -ray/internal conversion electron.

Hopefully, on completion of these experiments the fission process will be more fully understood and in particular, isotopic yields and specific identifiable decay schemes for fission fragments from each type of fissioning nucleus will be discovered.

### III. PUBLICATIONS

1. Rodrigue St-Laurent, Gary W. Phillips, Patrick Richard and C. Fred Moore, "Internal Conversion Cascades in Fission Products," Phys. Rev. C 4, 1948 (1971).
2. F. F. Hopkins, G. W. Phillips, J. R. White, C. Fred Moore, and P. Richard, "Low Energy Transitions from the De-Excitation of Spontaneous Fission Fragments of  $^{252}\text{Cf}$ ," Phys. Rev. C 4, 1927 (1971).
3. F. F. Hopkins, J. R. White, G. W. Phillips, C. Fred Moore, and Patrick Richard, "Further Measurements of Gamma Transitions in Spontaneous-Fission Fragments of  $^{252}\text{Cf}$ ," Phys. Rev. C 5, 1015 (1972).

#### IV. RECENT BIBLIOGRAPHY

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11. Shell Model Calculations of Fission Decay Widths and Probability in  
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## V. WORK STATEMENT FOR ARPA PROPOSAL

The contractor shall conduct research involving the measurement of prompt fission processes. This research shall include, but not necessarily be limited to the following:

- A. Studies of the spontaneous fission of  $^{252}\text{Cf}$  employing various combinations of multi-parameter coincidence experiments. Parameters may include fission fragment, x-ray, alpha particle, gamma ray, time of flight, magnetic spectrograph and time.
- B. Studies of neutron induced fission of  $^{235,238}\text{U}$  and  $^{239}\text{Pu}$  employing various combinations of multiple parameter coincidence experiments described above, less the alpha particle measurements.
- C. Studies of charged particle induced fission reactions necessary to develop neutron-induced fission measurement techniques.
- D. From an analysis of the above measurements begin a compilation of the total chain yield and the prompt isotopic yield ratios for fission of  $^{235,238}\text{U}$  and  $^{239}\text{Pu}$  as a function of neutron energy. Give priority to  $^{235}\text{U}$  and  $^{239}\text{Pu}$  fission mass distribution resulting from fission spectrum and 14MeV neutrons.



## VI. PERSONNEL

1 April 1971 to 31 March 1972

### (a) Nuclear Scientists

Patrick Richard, Associate Professor (11 Mo. <sup>*</sup> )	1 mo.
C. Fred Moore, Professor (11 Mo. <sup>*</sup> )	1 mo.
Gerald Hoffmann, Research Scientist Assoc. III	6 mo.
Gary Phillips, Research Scientist Assoc. IV	5 mo.

### (b) Pre-Doctoral Appointments (graduate students)

Forrest Hopkins, Research Assistant II	12 mo.
John R. White, Research Assistant I	12 mo.
Dee McCrary, Research Assistant I	2 mo.
Ralf-Gerald Abitz, Research Assistant I	2 mo.
Mike Picone, Research Fellow <sup>*</sup>	12 mo.
Bill Hodge, Research Assistant I	5 mo.
Casio Ore, Research Assistant II	3 mo.
Mike Senglaub, Research Assistant II	3 mo.
Rodrique St-Laurent, Research Fellow <sup>*</sup>	9 mo.

### (c) Engineering/Technical Staff

J. P. Coose, Technical Assistant II	12 mo.
Hunter Ellinger, Computer Programmer I <sup>*</sup>	12 mo.
Mary George, Accounting Clerk II <sup>*</sup>	12 mo.
A. L. Mitchell, Research Scientist Associate III <sup>*</sup>	12 mo.
Dee Munsell, Administrative Secretary <sup>*</sup>	12 mo.
John Pedracine, Tech. Staff Assistant II	2 mo.
Janice Schooler, Clerk Typist <sup>*</sup>	7 mo.
Kenric Speed, Laboratory Assistant II	12 mo.
Bonnie Wolf, Secretary <sup>*</sup>	12 mo.
Billy Yant, Instrument Maker I	2 mo.

VI. PERSONNEL

(Cont.)

(d) Laboratory Staff (undergraduate students)

Jerry Baker, Laboratory Assistant II	1 1/2 mo.
Jeffry Fitch, Laboratory Assistant III	12 mo.
Joseph Gibbs, Laboratory Assistant II	3 1/2 mo.
Gary Jacobs, Laboratory Assistant II	7 mo.
Robert Hooks, Laboratory Assistant I	6 mo.
Michael Senglaub, Lab Assistant II	2 mo.
Nat Smith, Laboratory Assistant I	2 mo.
Roger Jordon, Laboratory Assistant	6 mo.
Tom Loyd, Laboratory Assistant I	6 mo.

\* At no pay from contract