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SIMULTANEOUS EMISSION OF TWO LIGHT CHARGE PARTICLES IN SPONTANEOUS FISSION OF  $^{252}\text{Cf}$

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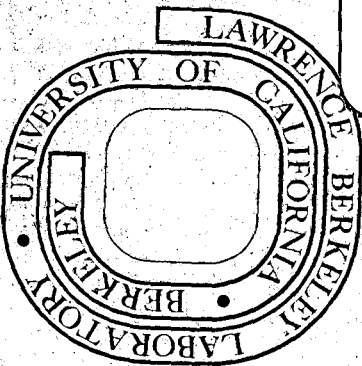
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SIMULTANEOUS EMISSION OF TWO LIGHT CHARGED PARTICLES IN  
SPONTANEOUS FISSION OF  $^{252}\text{Cf}$ \*

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The emission of two light charged particles in the spontaneous fission of  $^{252}\text{Cf}$  has been observed and studied. Two particle telescopes were placed on the opposite sides of a strong  $^{252}\text{Cf}$  source covered on both sides by platinum foils. The coincidences between the two telescopes were mainly of the type  $^4\text{He} - ^4\text{He}$  although  $^4\text{He} - ^3\text{H}$  events were also observed. The energy spectra of the detected particles--alphas and tritons, are similar in shape to the spectra of normal long range particles, but the mean energies are lower by about 2 MeV. The mean energy of one alpha particle in the  $^4\text{He} - ^4\text{He}$  coincidences does not change as a function of the energy of the other particle. The angular correlation between these particles has also been measured in another experiment (at five angles  $180^\circ$ ,  $90^\circ$ ,  $66^\circ$ ,  $45^\circ$ ,  $35^\circ$ ) without particle identification. The overall emission rate of the coincident particles is approximately two per million binary fission events. The hypothesis of

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independent emission of these two particles seems to be consistent with most of the observed data. Trajectory calculations are being performed to deduce the condition of the fissioning nucleus at scission configuration.

## INTRODUCTION

The process of nuclear fission accompanied by a third light-charged particle (LCP) has been the subject of many investigations.<sup>1</sup> The prime reason for these studies is that the light-charged particles appear to be coming from the region between the larger fragments at a time close to scission. Therefore, the study of this process is expected to yield information on the fissioning nucleus at the scission point. Recently, Kapoor *et al.*<sup>2</sup> found evidence for the simultaneous emission of two LCP in coincidence with fission fragments in the thermal neutron-induced fission of  $U^{235}$ . In the present work we have observed the occurrence of the simultaneous emission of two LCP in the spontaneous fission of  $Cf^{252}$ , we have identified the coincident LCP and obtained their energy distributions, and we have measured the angular correlation between the two particles. Various correlations between the two coincident LCP have also been studied.

The experiment has been performed in two parts. In the first part two particle telescopes were used to identify the coincident LCP and to obtain the energy distribution of each type of particle observed. In the second part two surface barrier detectors were used to obtain the yield of the coincident LCP events without any particle identifications for various angles between the two detectors.

## EXPERIMENTAL PROCEDURE

### Part 1. The Identification of the LCP in Quaternary Fission

Two semiconductor particle telescopes were placed on the opposite sides of a strong  $Cf^{252}$  source as shown in Fig. 1a. Each telescope was at a

distance of 1.2 cm from the source. The source strength was  $0.6 \times 10^7$  fissions per minute. The source was covered on both sides by two absorber foils 12.5 mg/cm<sup>2</sup> of platinum and 3.6 mg/cm<sup>2</sup> of aluminum. The thicknesses of the foils were selected in such a way as to prevent the fission fragments and the 6.18 MeV alpha particles from reaching the particle telescope. Two measurements were made using different thicknesses for the  $\Delta E$  counters in the particle telescopes. The first measurement was performed with a 50  $\mu$   $\Delta E_1$  and a 48  $\mu$   $\Delta E_2$  counter. The second measurement was carried out using a 37  $\mu$   $\Delta E_1$  and a 24  $\mu$   $\Delta E_2$  counter to study the low energy part of the alpha particle distributions in quaternary fission.

A four parameter data acquisition system was used to record the information from the coincidence events. The four parameter system was triggered by the occurrence of a fast coincidence between the two  $\Delta E$  counters. Therefore, all the two-fold events ( $\Delta E_1$ - $\Delta E_2$ ), three-fold events ( $\Delta E_1$ -TEL2 and vice-versa), and four-fold events (TEL1-TEL2) were recorded. The fast coincidences were realized by using the zero cross-over technique with a time resolution of 40 nsec. The energy calibrations for all the detectors were done twice a week during the entire measurement which lasted for two months. The timing between the two  $\Delta E$  counters was monitored using a Th<sup>228</sup> source (8.78 MeV alpha particles). Essentially, no timing or pulse-height shift was observed. The data analysis was done off-line. The particle identification was performed by using a power law approximation to the range-energy curves.<sup>3</sup>

#### Part 2. The Angular Correlation Between the Two LCP

The angular correlation experiment was carried out with a Cr<sup>252</sup> source, stronger by an order of magnitude than the one used in the Part I experiment. With the new source, the true to chance rate decreased to a ratio of 2 to 1 within the two telescope set up. Therefore, in order to have a reasonable true to chance ratio both telescopes were replaced by semiconductor detectors

and the electronic configuration was also modified. The modified configuration was not possible with the two telescope set up and the available four parameter data acquisition system. Two time pickoff units were used to obtain the timing signals which were fed to a time to amplitude converter (TAC). The two linear energy signals from the two counters and the linear time signal from the TAC were recorded event by event on the multiparameter data recording system. The time distribution obtained showed a time resolution of 3 nsec. The window on the time signal was set at 6 nsec as compared to 40 nsec in the first part. The true to chance ratio varied from 20 to 1 for the 45° measurement to 8 to 1 for the 90° measurement.

Figure 1b shows the various configurations of the two detectors which were used to obtain the angular correlation between the two LCP. These measurements were divided into three sets. In the first set A the distance between the detectors and the source was 2.0 cm for the 90° and 180° measurements and 2.2 cm for the 66° measurement. In the second set these distances were 3.2 cm for the 90° and 180° measurement and 3.5 cm for the 45° measurement. The third set includes measurements for angles 180° and 35°; the corresponding distances were 4.5 cm and 4.75 cm. For the 90° and 180° measurements the effective thickness of the absorber foils were identical for both detectors (1.4 times thickness) where as for 66°, 45°, 35° the effective thickness of the absorber foils vary (1.2-1.15 times thickness).

## RESULTS

### Part 1. Identification of LCP

The coincident events were predominantly of the type  $\text{He}^4\text{-He}^4$  but  $\text{He}^4\text{-H}^3$  and  $\text{He}^4\text{-H}^1$  events were also observed. No coincident events involving particles heavier  $Z = 2$  particles were observed. A significant part of the events involving  $\text{H}^1$  are due to the fast neutron induced (n,p) reaction in the detectors in coincidence with long range alpha particles. However, the

contribution of (n,p) reaction to the ( $H^1, He^4$ ) events where the energy of  $H^1$  is larger than 5 MeV is insignificant.<sup>4</sup> Table I shows the observed number of various types of coincidence events using thick 50  $\mu$   $\Delta E$  counter telescopes. Table II contains the relative yields of protons, tritons,  $He^4$  and  $He^6$  normalized to unity for each telescope in ternary and quaternary fission events. It can be seen that within the statistical error these relative yields in normal ternary fission events are equal to the corresponding relative yields in the fission events with two light-charged particles (quaternary fission).

Figure 2a to 2e show the energy distribution of alphas, tritons, and protons observed in the quaternary fission. The smooth curve passing through the experimental points is a calculated curve. It is obtained by fitting to the experimental spectrum a Gaussian distribution with  $\hat{E}$ ,  $\hat{\sigma}$  as the most probable energy and variance parameter; after making corrections for the energy loss in the absorber foils. These energy loss corrections were performed using a Monte Carlo technique to take into account the finite source-detector geometry. The fitted values of  $\hat{E}$  and  $\hat{\sigma}$  for various spectra are shown in Table III. The dashed curve shown in these figures is the corresponding energy distribution observed in normal ternary fission using the same telescopes. Figure 2a shows the energy spectrum of alpha particles in coincidence with alpha particles obtained with the 50  $\mu$   $\Delta E$  counter telescope in the first measurement. Figure 2b shows the same energy spectrum obtained in the second measurement with the 24  $\mu$   $\Delta E$  counter telescope. The second measurement was needed to obtain reliable values of  $\hat{E}$  and  $\hat{\sigma}$  for the alpha particles whereas the first measurement gives reliable information on tritons in coincidence with alpha particles. The energy distribution of alpha particles in coincidence with tritons is shown in Fig. 2c. From these figures it is concluded that the energy distributions of the alpha particles



in quaternary fission is lower as compared to normal ternary fission by about 2 MeV as given in Table III. It can also be concluded that the alpha energy distribution in quaternary fission does not depend on the charge of the coincident light particle. The mean energy of the alpha particles does not depend on the energy of the coincident alpha particle as shown in Fig. 3. Figure 2d shows the energy distribution of tritons in coincidence with alpha particles. The energy spectrum in quaternary fission is seen to be lower as compared to the corresponding distribution in the normal ternary fission. The difference in most probable energy  $\hat{E}$  for tritons in quaternary and ternary fission is about 2.0 MeV. The energy distribution of the protons in coincidence with alpha particles is shown in Fig. 2e. The low energy peak at 4.0 MeV is due to the fast neutron induced reactions in the Si detector. The statistical uncertainties were too large to obtain any information on the most probable energy.

## Part 2. Angular Correlation of LCP

The experimental results of the eight measurements are given in Table IV. The lower energy cutoff used in the off-line analysis of the data was 5.0 MeV in the two counters to reduce the contribution of the fast neutron induced reactions in the detector material which are in coincidence with the normal ternary fission events. The coincidence rate varied from 3 events per hour to 0.1 events per hour. The measurements were carried out for a period of about six months to obtain good statistics. During the entire measurement no deterioration in the detector performance was observed. Table V summarizes the experimental information on the energy spectra of the coincident LCP and various other quantities of interest. The yields are given in terms of the constant P defined as follows:

$$N_1(\theta) = N \cdot P_3 \Omega_1(\theta) \epsilon_1$$

$$N_2(\theta) = N.P_3 \Omega_2^2(\theta) \epsilon_2$$

$$N_{12}(\theta) = N.P_4 \cdot (\theta) \Omega_1(\theta) \Omega_2(\theta) \epsilon_1 \cdot \epsilon_2$$

where

N is the source strength fissions/hr

$P_3$  and  $P_4$  are the probabilities of ternary and quaternary fission per binary fission

$\Omega_1$  and  $\Omega_2$  are the solid angles for the two detectors at angle  $\theta$ .  $\epsilon_1$ ,  $\epsilon_2$ ,

$\epsilon_1'$ ,  $\epsilon_2'$  are the correction factors due to the absorber foils

and the lower energy cutoffs

$N_1(\theta)$  and  $N_2(\theta)$  are the no. of ternary events in one measurement

$N_{12}(\theta)$  is the no. of quaternary events in one measurement of duration

H. hours.

$$P = \frac{N_{12}(\theta).H.}{N_1(\theta).N_2(\theta)}$$

$$= \frac{N.P_4 \cdot (\theta) \Omega_1(\theta) \Omega_2(\theta) \cdot \epsilon_1 \epsilon_2}{N^2 P_3^2 \Omega_1(\theta) \Omega_2(\theta) \epsilon_1 \epsilon_2} = \frac{P_4(\theta) \epsilon_1 \epsilon_2}{N.P_3^2 \epsilon_1 \epsilon_2} \quad (1)$$

The probability of quaternary fission  $P_4$  can be obtained from P, using the above relation. In Fig. 4, the measured value P for eight measurements is plotted against the angle  $\theta$  between the two detectors. Figure 5a shows the plot of  $P_4$  versus  $\theta$ . These values were calculated using Eq. (1) and taking into account explicitly the finite source and finite detector geometry of the various measurements. The correction factors  $\epsilon_1$ ,  $\epsilon_2$ ,  $\epsilon_1'$ ,  $\epsilon_2'$  were calculated assuming that the energy spectrum of the light particles does not depend on angle  $\theta$ . The parameters for the energy distribution involved were taken from the previous measured with the two telescopes.

The mean energies of particles detected in one counter does not depend on the energy of the particle detected in the other counter, as shown in Fig. 6 for one measurement. This conclusion holds for all the measurements. Moreover, it is observed that the most probable energy  $\hat{E}$  of the particles in quaternary fission does not depend on the angle between the two counters as shown in Table V for four measurements. The total probability obtained by integrating  $P_4(\theta)$  is  $(1.5 \pm 0.5 \cdot 10^{-6})$  per binary fission.

#### DISCUSSION

The experimental results on the energy distributions, angular correlation and the relative abundances of various particles in quaternary and ternary fission indicate that the two processes have very much in common. The relative abundances of tritons and alpha particles is identical within statistical error in the two types of fission events. The energy distribution of alpha particles in quaternary fission does not seem to depend on the nature of the coincident particle (tritons or alphas).

The energy distributions of tritons and of alpha particles in quaternary fission are very much similar to the corresponding energy distributions in the normal ternary fission except for the fact that mean energies for these particles in quaternary fission are lower than in ternary fission. The angular correlation experiment shows that the yield at  $90^\circ$  is smaller than at  $30^\circ$  or  $180^\circ$ , although the yield at  $30^\circ$  is higher than the yield at  $180^\circ$ . It seems that the emission mechanism of light particles in quaternary fission is similar to the emission mechanism of light particles in ternary fission. If one assumes that the light particles in ternary fission are produced at scission from one of the two fragments when the interaction between the two fragments has vanished, then the quaternary fission events represent the cases where each fragment has contributed one LCP. Under the assumption that each fragment can emit only one LCP with probability  $P_i$ , which does not

depend on the other fragment, the total probability of quaternary fission is  $P_4 = P_1 P_2$ . The probability of ternary fission will be equal to  $P_1 + P_2$ . Taking the experimental value of  $P_1 + P_2 = 1/300$  and assuming  $P_1 = P_2$ ,  $P_4$  is equal to  $2.6 \times 10^{-6}$  per fission, which is in good agreement with the experimental value of  $P_4$ . This hypothesis is also consistent with the constancy of relative abundances of tritons and  $\text{He}^4$  in two types of fission events.

Under the assumption that each LCP in the quaternary fission has a sharply peaked angular distribution with respect to the motion of the fission fragment as in normal ternary fission and assuming that the two LCP are emitted statistically independent of each other, there is an angular correlation between the two LCP as shown in Fig. 5b. The calculated curve is symmetrical around  $90^\circ$ . The magnitude of  $W(180^\circ)/W(90^\circ)$  quantity is of the order observed in the experiment. However, the experimental correlation is not symmetrical around  $90^\circ$  as calculated under the hypothesis.

. Trajectory calculations are being performed in an attempt to explain the lower mean energies of particles in quaternary fission as compared to normal ternary fission and the observed asymmetry in the angular correlation data.

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Table I. The total number of events for various types of coincidences between the two light-charged particles (LCP) emitted in the spontaneous fission of  $\text{Cf}^{252}$  observed in the experiment using 50  $\mu$   $\Delta E$  telescope systems p - protons, t - tritons and  $\alpha$  - alpha particles.

Telescope - 1	-	Telescope - 2	No. of events
p	-	p	10
p	-	t	0
p	-	$\alpha$	16
t	-	p	5
t	-	t	1
t	-	$\alpha$	66
$\alpha$	-	p	12
$\alpha$	-	t	82
$\alpha$	-	$\alpha$	551

Table II. The relative abundances of protons-tritons and alpha particles normalized to unity, observed in each telescope for quaternary and ternary fission events with 50  $\mu$   $\Delta E$  telescopes in the experiment set 1.

Relative Prob.	Quaternary		Ternary	
	Tel-1	Tel-2	Tel-1	Tel-2
Protons	.035 $\pm$ .01	.03 $\pm$ .01	.03 $\pm$ .01	.015 $\pm$ .01
Tritons	.097 $\pm$ .01	0.11 $\pm$ .01	.09 $\pm$ .01	.09 $\pm$ .01
Alphas	0.87 $\pm$ .01	0.85 $\pm$ 0.01	.88 $\pm$ .01	.895 $\pm$ .01
He <sup>6</sup>			.01 $\pm$ .005	.01 $\pm$ .005

Table III. Experimental and fitted parameters for energy distributions shown in Fig. 2a to 2e.  $\hat{E}$  and  $\hat{\sigma}$  are the most probable energy and the variance parameters of the Gaussian distribution. Fitted  $\bar{E}$  and  $\bar{\sigma}$  represent the mean energy and the variance of the energy distribution with ( $E_{\min} = 8$  MeV for  $\alpha$ -particles and  $E_{\min} = 4.0$  MeV for tritons) computed after making corrections for the energy loss in the absorber cover foils to the Gaussian distribution. Experimental  $\bar{E}$  and  $\bar{\sigma}$  are the mean energies and the variance calculated from the experimentally observed energy distributions.

Identification	Fitted Parameters				Exp	
	$\hat{E}$	$\hat{\sigma}$	$\bar{E}$	$\bar{\sigma}$	$\bar{E}$	$\bar{\sigma}$
Set-1. Alpha's in quaternary ( $\alpha$ - $\alpha$ ) events	13.5 $\pm$ 0.5,	4.0	12.9	, 2.8	12.9 $\pm$ 0.5,	2.9
Set-1. Alpha's in quaternary ( $\alpha$ -t) events	14.00 $\pm$ 0.8,	4.0	13.06	, 3.2	12.75 $\pm$ 0.5,	2.9
Set-1. Alpha's in ternary events	16.1 $\pm$ 0.2,	4.4	14.5	, 3.6	14.2 $\pm$ 0.2,	3.7
Set-2. Alpha's in quaternary ( $\alpha$ - $\alpha$ ) events	14.0 $\pm$ 0.6,	4.2	12.9	, 3.4	12.7 $\pm$ 0.6,	3.6
Set-2. Alpha's in ternary fission	16.2 $\pm$ 0.2,	4.4	14.2	, 4.2	14.1 $\pm$ 0.2,	4.2
Set-1. Tritons in quaternary (t- $\alpha$ ) events	6.6 $\pm$ 0.8,	2.6	7.2	, 1.9	7.3 $\pm$ 0.5,	2.1
Set-1. Tritons in ternary events	8.8 $\pm$ 0.5,	2.8	8.7	, 3.0	8.8 $\pm$ 0.5,	2.8



Table IV. Summary of angular correlation experimental data.  $\bar{E}_1$  and  $\bar{E}_2$  are the mean energies of the distribution observed in detector 1 and 2 with  $E_{\min} = 5$  MeV. Quantity P is a measure of the angular correlation as defined in the text.

Set-Det	Angle	Time hrs.	Events	P	Quaternary		Ternary	
					$\bar{E}_1$	$\bar{E}_2$	$\bar{E}_1$	$\bar{E}_2$
A.	60°	495	581	2.8±0.3	10.1±0.2	9.85±0.2	12.61±0.2	12.72±0.2
	90	220	309	1.65±0.2	10.7±0.5	10.0±0.5	12.20±0.2	11.91±0.2
	180	187	335	2.2±0.2	9.2±0.6	10.2±0.4	11.77±0.4	11.71±0.2
B.	45°	520	410	10.2±0.6	10.1±0.2	9.8±0.2	11.30±0.2	12.20±0.2
	90	520	113	1.5±0.2	11.2±0.5	9.9±0.5	13.5±0.3	12.7±0.3
	180	384	109	2.3±0.2	10.2±0.5	10.2±0.5	12.2±0.3	12.7±0.3
C.	35°	362	79	8.5±1.0	9.5±0.7	9.4±0.7	11.91±0.2	12.3±0.2
	180	691	52	2.1±0.5	9.3±0.8	9.4±0.8	12.37±0.2	11.8±0.2

Table V. Fitted parameters  $\hat{E}$  and  $\hat{\sigma}$  are the most probable energy and the variance parameters of the Gaussian distributions for alpha particles fitted to the experimental spectra observed in angular correlation experiment. The energy loss corrections used were based on assumed values of triton/alpha probability and the assumed parameters for triton energy distribution as observed in part 1 experiment. The fits were made only to four measurements.

Set	Detector	$\hat{E}$	$\sigma$	$\hat{E}$	$\sigma$	
		Quaternary		Ternary		
A	66°	i	13.15±0.8	4.20	15.3±0.5	4.72
		ii	12.70±0.8	3.65	16.30±0.5	4.90
	90°	i	14.08±0.8	4.0	15.6±0.5	4.6
		ii	15.0±0.8	4.8	16.05±0.5	4.21
	180°	i	14.22±0.8	3.63	15.0±0.5	4.70
		ii	13.5±0.8	4.3	15.5±0.5	4.45
B	45°	i	11.81±0.8	4.20	14.4±0.	4.24
		ii	12.88±0.8	3.65	15.2±0.5	4.20

## FIGURE CAPTIONS

Fig. 1a. The schematic diagram of the detector-source configuration used in Part 1 experiment. The first set of measurements were made using  $50 \mu \Delta E$  and  $48 \mu \Delta E_2$  counter telescopes. The second measurement used  $37 \mu \Delta E_1$  and  $24 \mu \Delta E_2$  counter telescopes.

Fig. 1b. The schematic diagram of the eight measurements of the Part 2 experiment. Three different distances were used for  $180^\circ$  and two distances were used for  $90^\circ$  measurements. Three measurements were made for  $66^\circ$ ,  $45^\circ$  and  $35^\circ$  angles between the two detectors.

Fig. 2. The kinetic energy distribution of LCP in quaternary fission. The dashed curve is the kinetic energy distribution of LCP in normal ternary fission. The continuous curve is the fitted curve as explained in the text.

- a)  ${}^4\text{He}$  K.E. spectrum in  ${}^4\text{He} - {}^4\text{He}$  events with  $50 \mu \Delta E$  telescope
- b)  ${}^4\text{He}$  K.E. spectrum in  ${}^4\text{He} - {}^4\text{He}$  events with  $24 \mu \Delta E$  telescope
- c)  ${}^4\text{He}$  K.E. spectrum in  ${}^4\text{He} - {}^3\text{H}$  events with  $50 \mu \Delta E$  telescope
- d)  ${}^3\text{H}$  K.E. spectrum in  ${}^4\text{He} - {}^3\text{H}$  events with  $50 \mu \Delta E$  telescope
- e)  ${}^1\text{H}$  K.E. spectrum in  ${}^4\text{He} - {}^1\text{H}$  events with  $50 \mu \Delta E$  telescope.

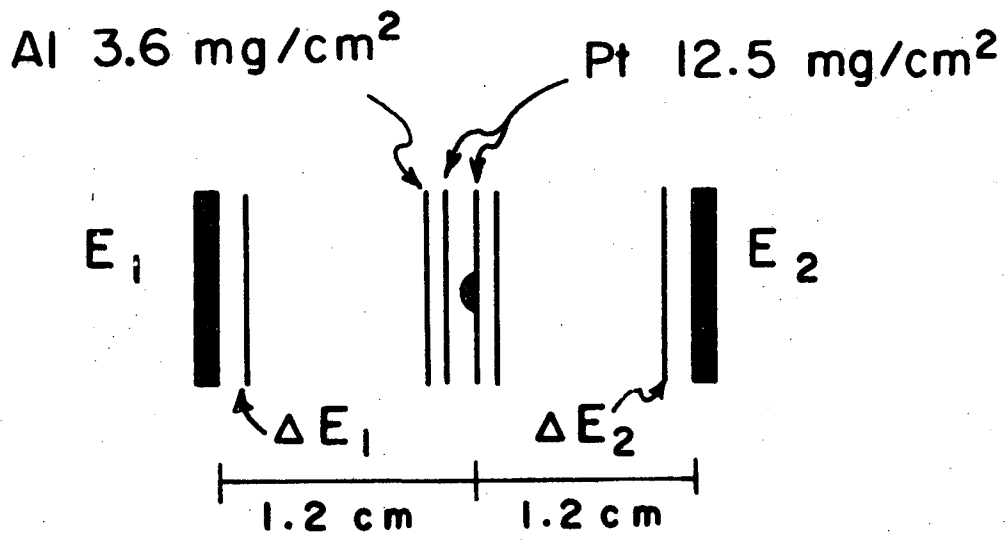
Fig. 3. The mean kinetic energy distribution of alpha particle detected in telescope 1 plotted as a function of the kinetic energy of the coincident alpha particle detected in telescope 2.

Fig. 4. Plot of the quantity P which is a measure of the angular correlation as defined in text for the eight measurements. The abscissa are the respective angles between the two lines joining the centers of the detectors to the source. No corrections were made in this plot.

Fig. 5a. Plot of the angular correlation between the two light-charged particles corrected for the energy loss in the absorber foils and the finite geometry of the detector-source geometry.

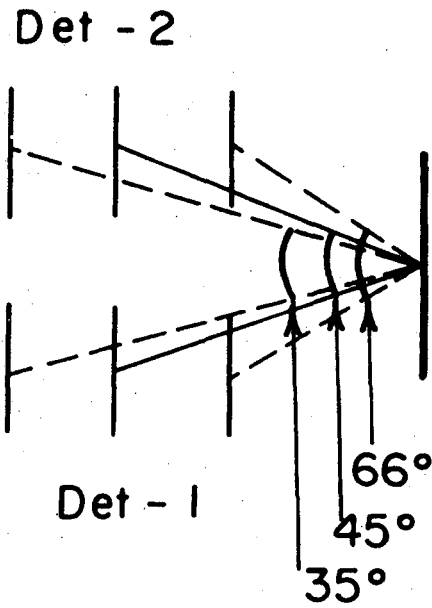
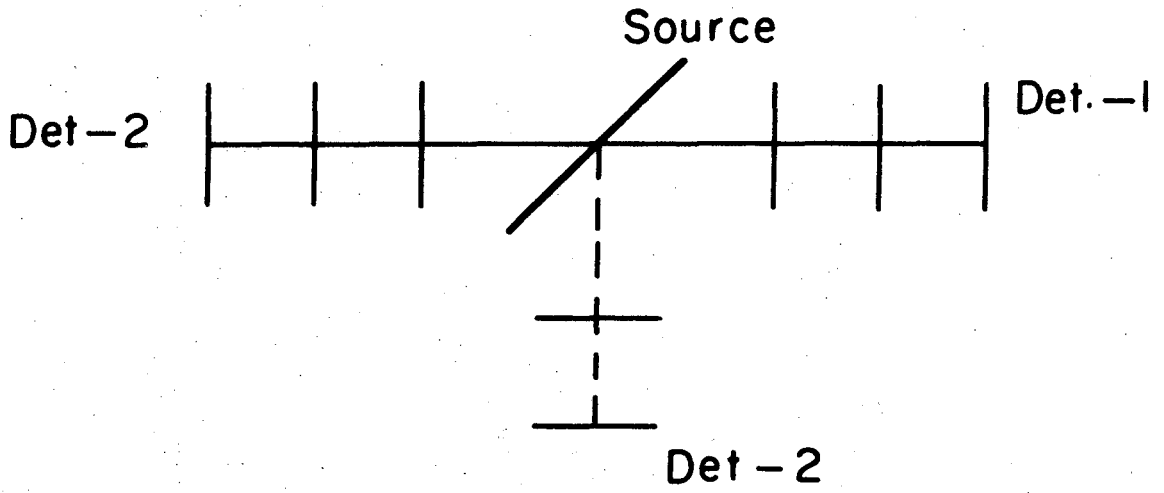
Fig. 5b. The angular correlation expected for quaternary fission based on the angular distribution of light-charged particles in ternary fission.

Fig. 6. The mean energies of light-charged particles in detector 1 in quaternary fission as a function of the energy of the second light-charged particle for set A-3.



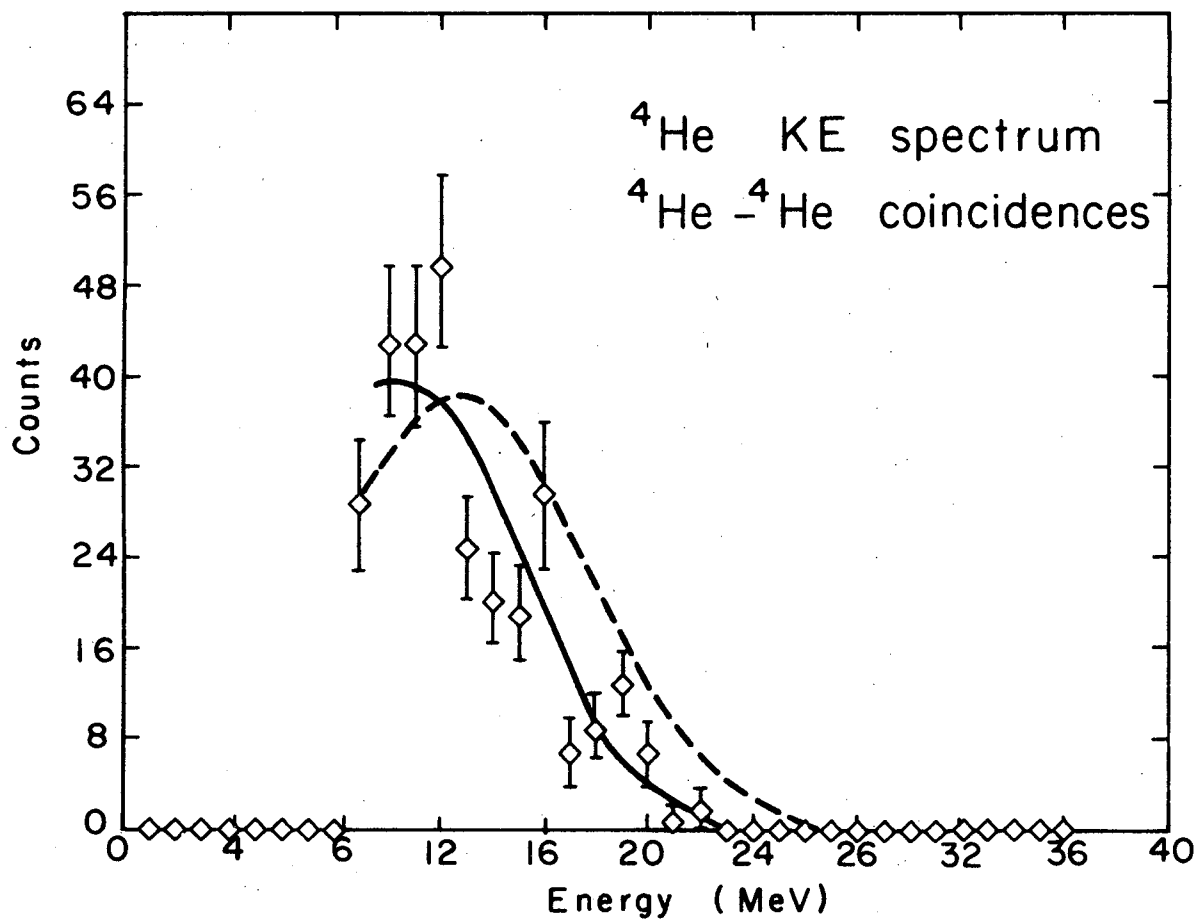
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Fig. 1a



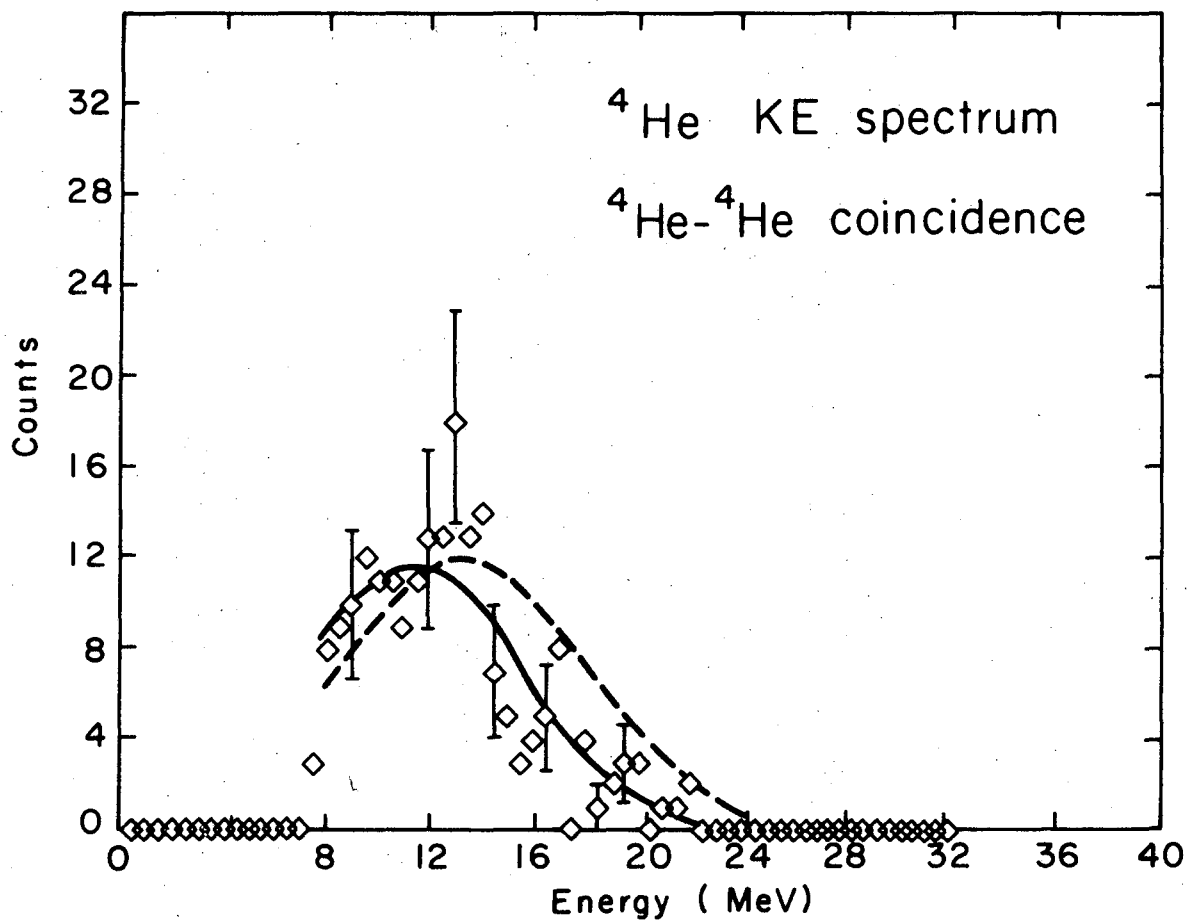
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Fig. 1b



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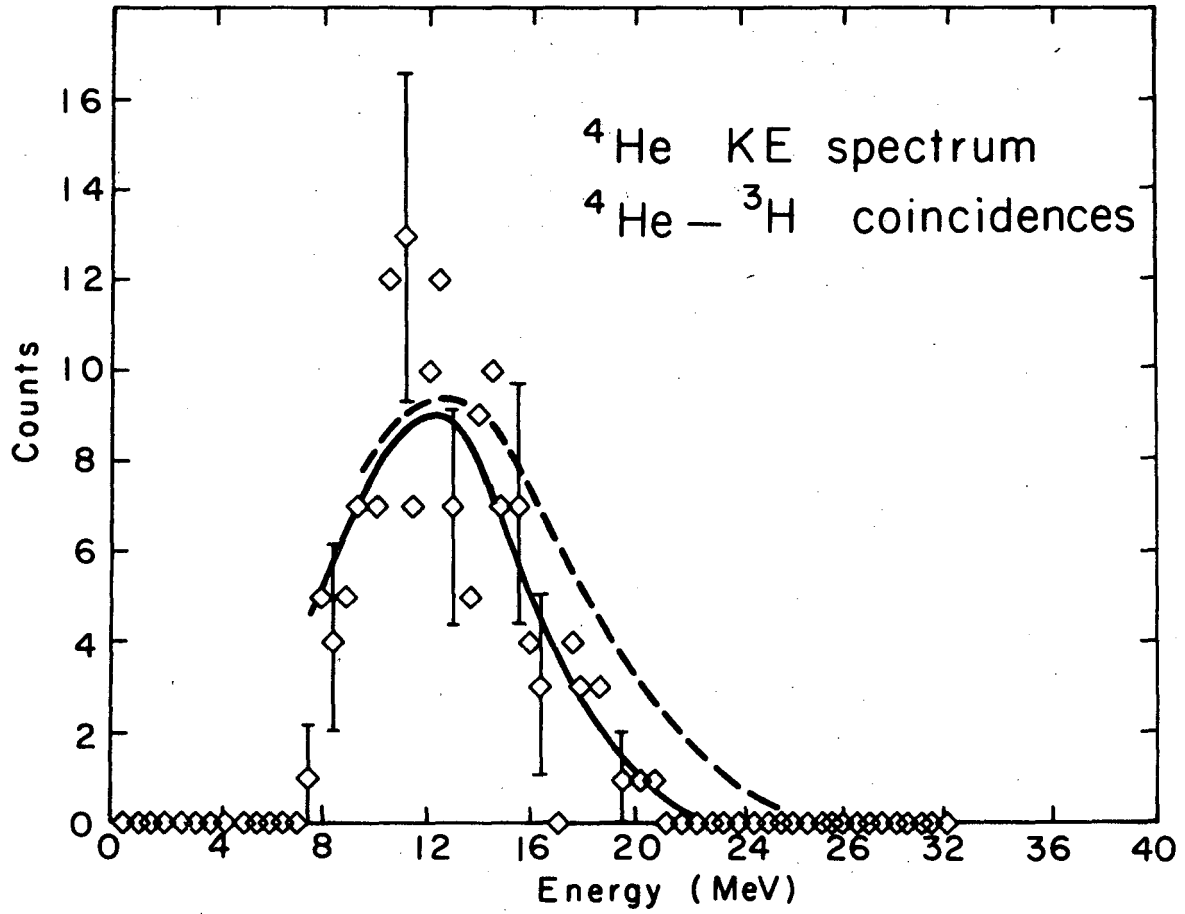
Fig. 2a



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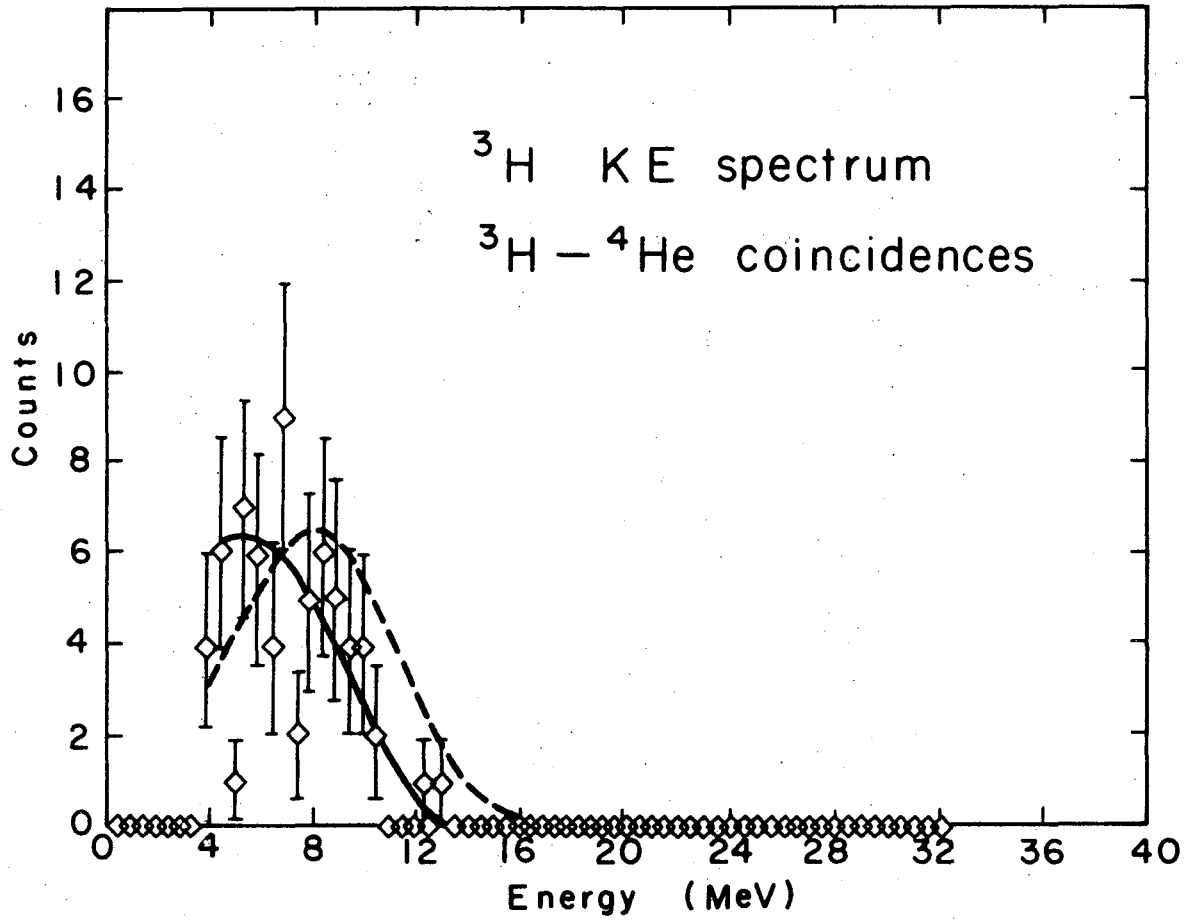
Fig. 2b





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Fig. 2c



XBL735-3026

Fig. 2d

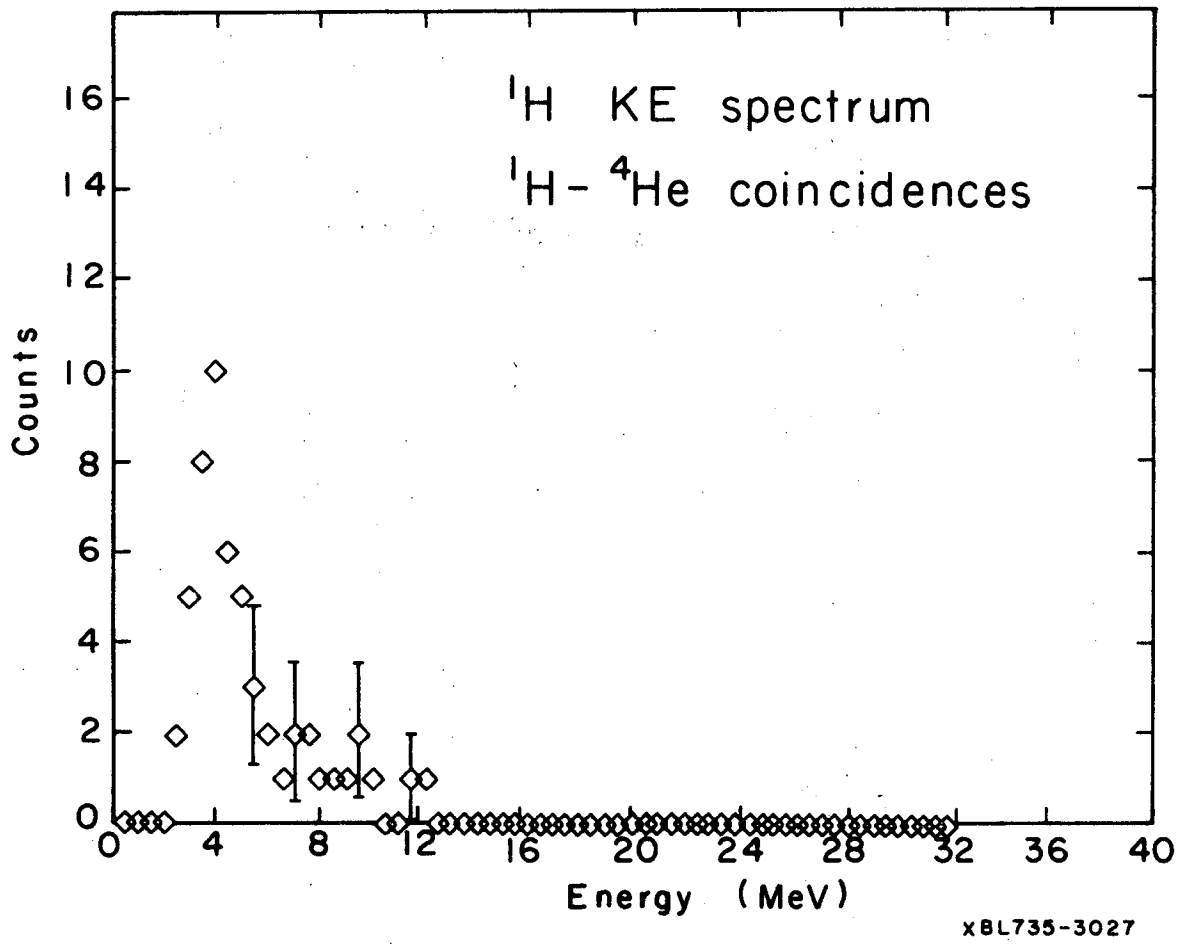


Fig. 2e

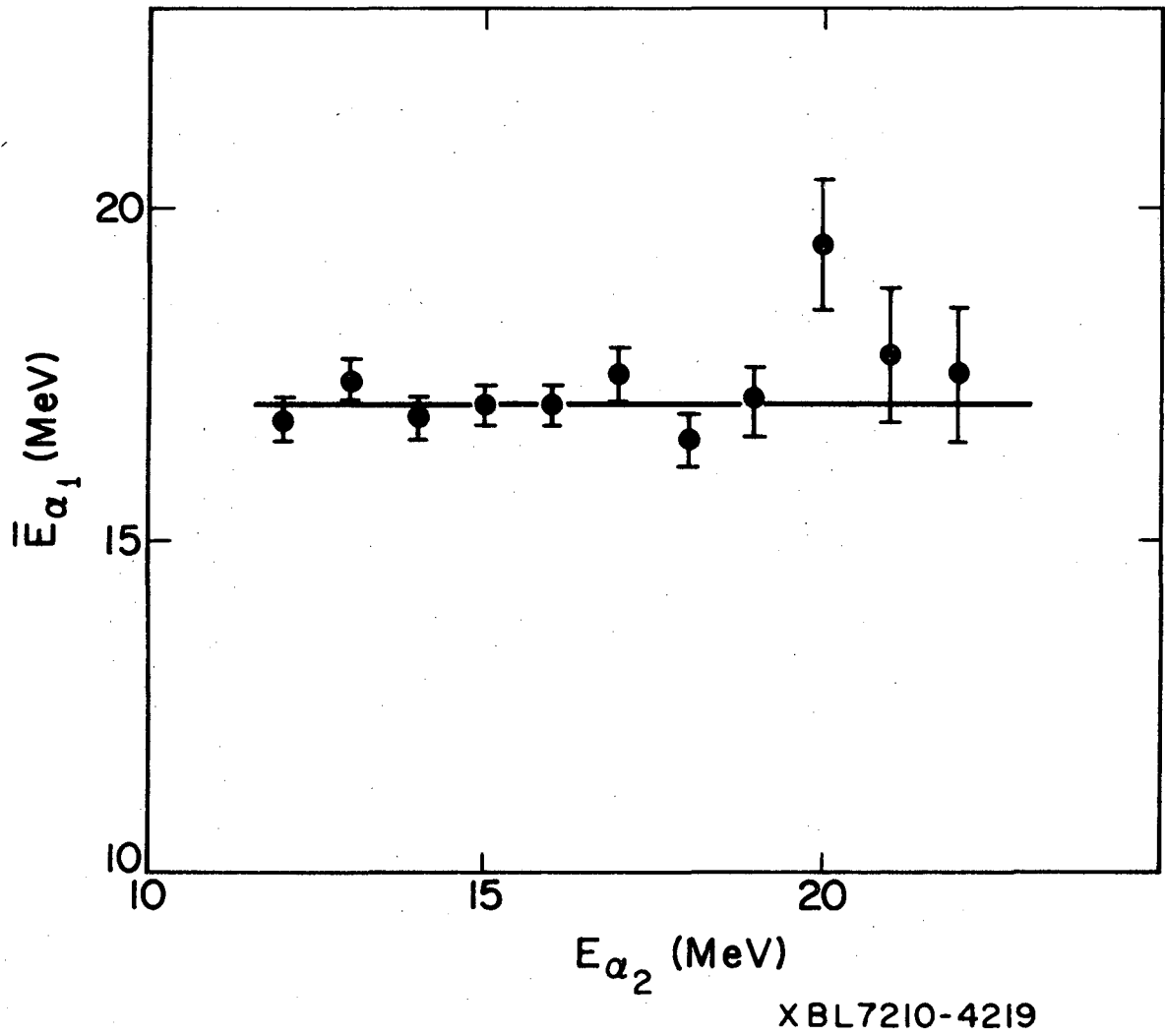
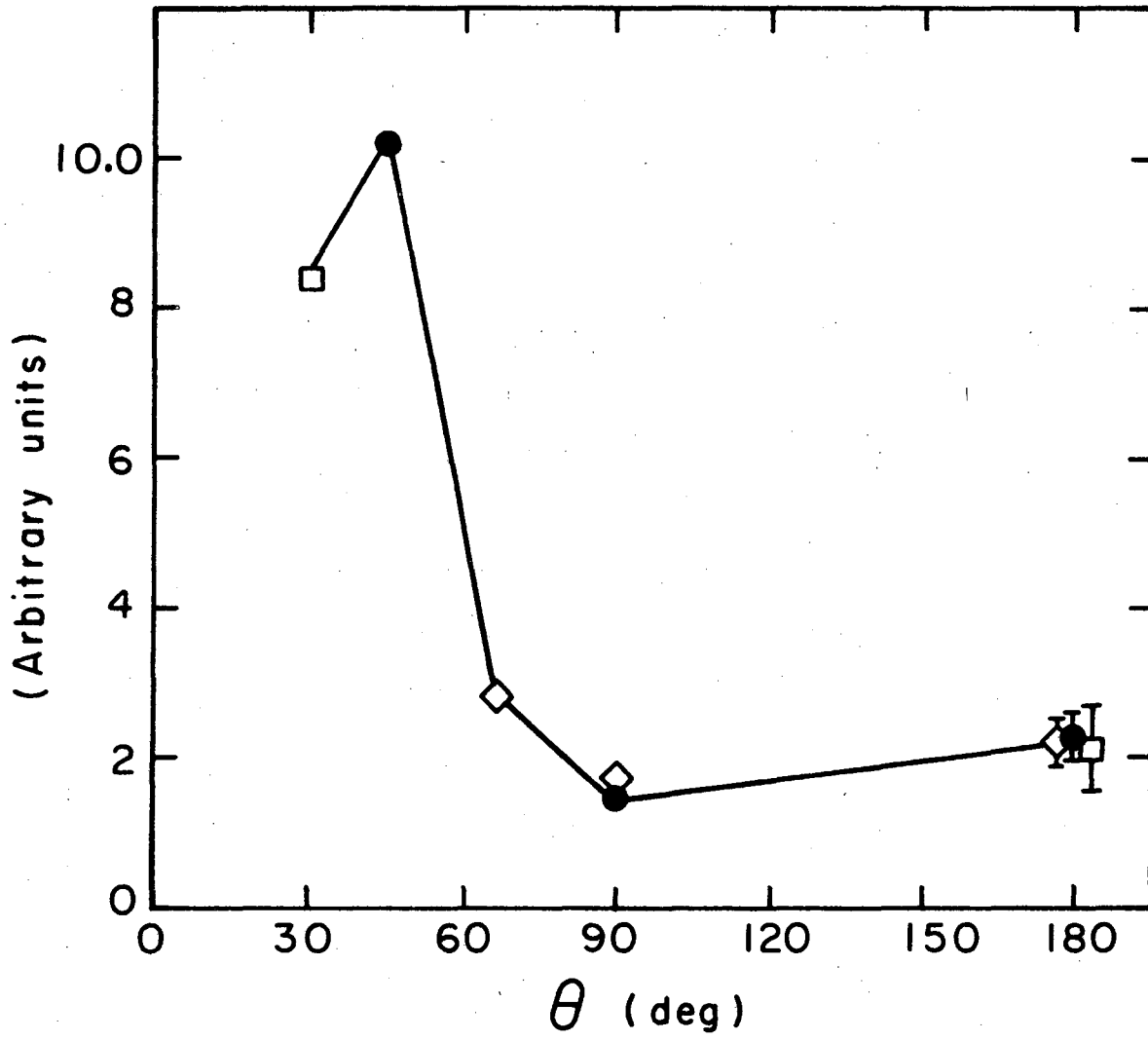
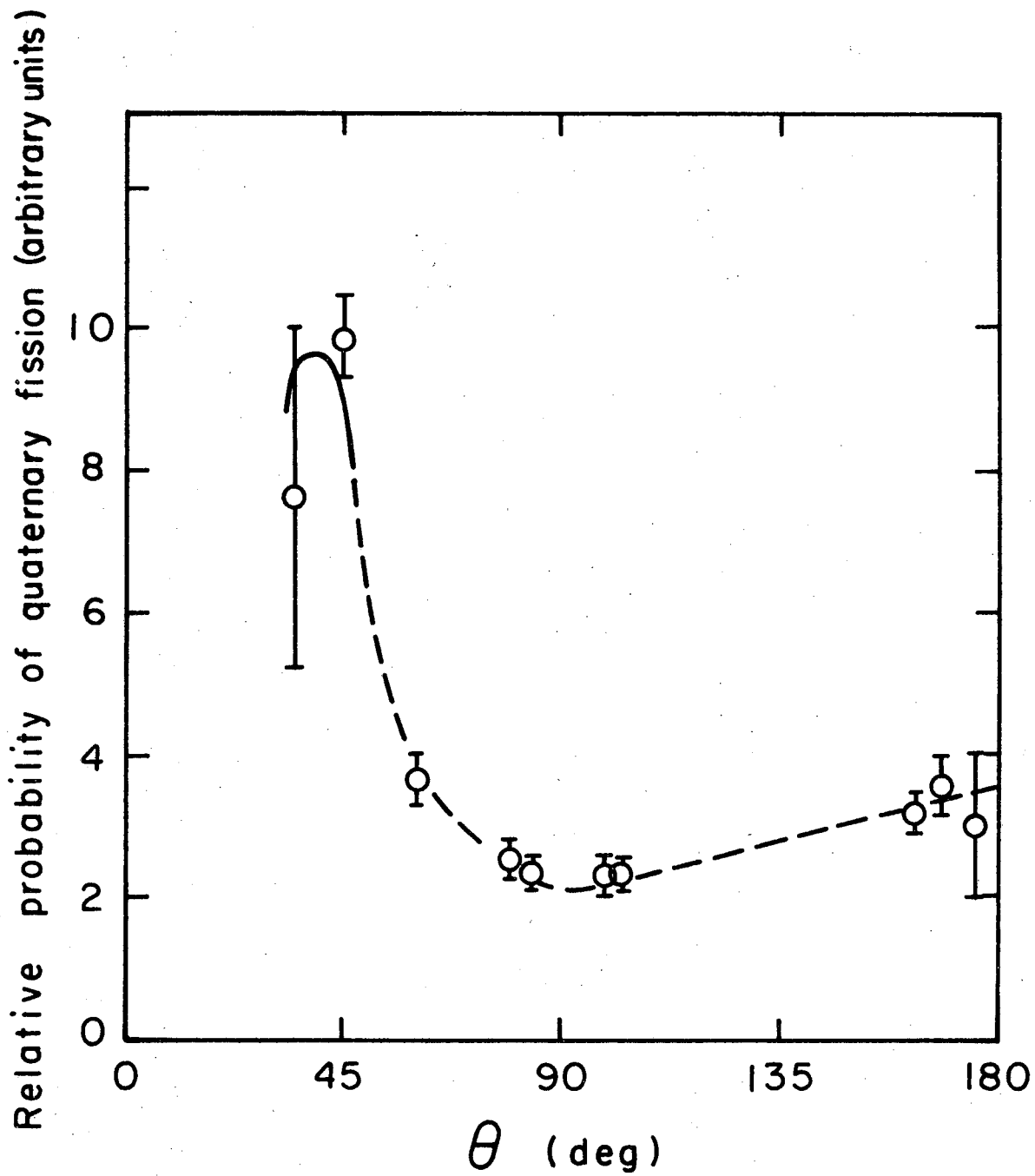


Fig. 3



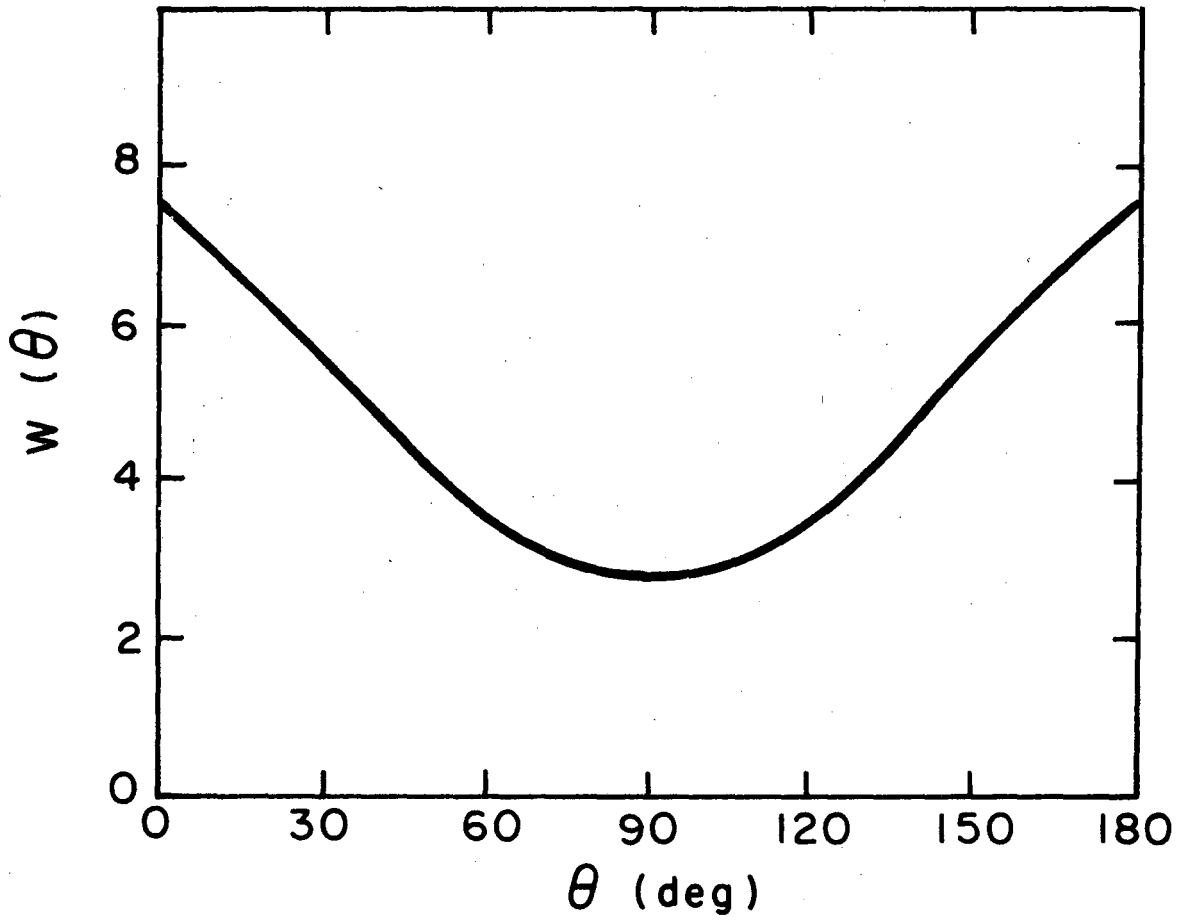
XBL 735-3032

Fig. 4



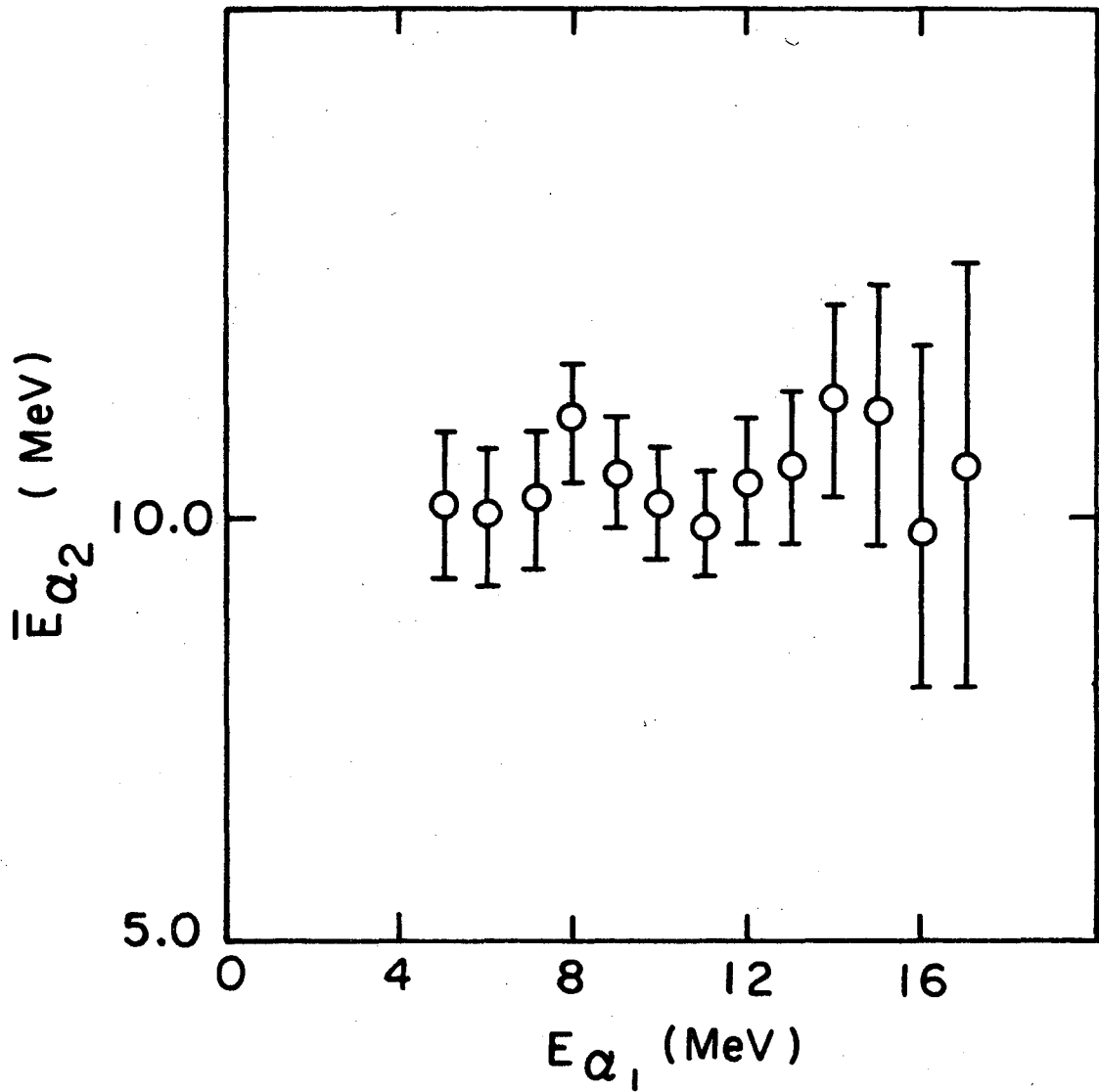
X BL735-3033

Fig. 5a



XBL 735-3034

Fig. 5b



XBL735-3035

Fig. 6



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