New results for the intensity of bimodal fission in barium channels of the spontaneous fission of ^{252}Cf

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Triple coincidence data from the fission of ^{252}Cf were used to deduce the intensity of the proposed "hot" mode in barium channels. γ-γ-γ and α-γ-γ fission data were analyzed to find the neutron multiplicity distribution for several binary and ternary charge splits. The binary channels Xe-Ru and Ba-Mo were analyzed, as well as the Ba-α-Zr, Mo-α-Xe, and Te-α-Ru ternary channels. An improved method of analysis was used to avoid many of the complexities associated with fission spectra. With this method, we were unable to confirm the second mode in the either the Ba-Mo or Ba-α-Zr splits.

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Observations of prompt γ rays produced in the spontaneous fission of ^{252}Cf have shown evidence for a "hot" fission mode in the Ba-Mo channel. The evidence for this mode is observed as a higher relative intensity for the 7–10 neutron channels [1–3]. Later analysis [4–6] did not confirm the second mode, but Refs. [5,6] did show an "irregularity" around the eight-neutron channel. In recent years, more complete data on the levels and relative intensities of transitions in barium and molybdenum isotopes has become available. There is also recent evidence that the second mode might be seen in ternary fission [7]. Because fission spectra are often complex and the events of interest are rare compared with other channels, this type of analysis is difficult and prone to errors caused by random coincidences. Therefore an improved method that avoids many of these complexities was developed to determine the relative intensity of the second mode in both the binary and ternary cases.

The data for this analysis come from two experiments using the Gammasphere detector array located at Lawrence Berkeley National Laboratory. The binary data were taken in November 2000. A 62-μCi source was placed between two iron foils to stop fission fragments. This arrangement was then put in a 7.62-cm polyethylene ball and placed in Gammasphere. The 5.7×10^{11} γ-γ-γ events were recorded. A coincidence cube was then constructed using the RADWARE software package [8]. The ternary data were taken in December 2001. A 35-μCi ^{252}Cf source was deposited as a 5-mm spot on a 1.8-μ Ti foil covered on both sides by gold foils. Eight ΔE-E detectors were placed around the source to detect light-charged particles (LCPs). The 9.0×10^5 LCP-γ-γ events were recorded [9] and a γ-γ matrix was constructed. The ternary data were analyzed using the γ-γ matrix peak fitting software written by Andrei Daniel. Earlier versions of this software were used by Refs. [1–3].

The number of prompt neutrons emitted in a fission event can be determined by finding the mass number of the fragments produced in the event. For example, if the fission fragments of ^{252}Cf are determined by some method to be ^{144}Ba and ^{103}Mo, then five neutrons must have been emitted. The relative intensity of a particular neutron channel can be found from triple coincidence data by double gating on a pair of transitions in the heavy fragment, then measuring the intensity of its partners. This must be done for each isotope of the heavy partner. The yield as a function of neutrons emitted can then be determined by summing the contributions of all possible pairs.

In practice, the yield is found by fitting peaks in double-gated spectra to find the area. For example, if a double gate is taken on ^{142}Ba, then the resulting spectra will show the 171.5-keV peak from ^{106}Mo, as well as the unresolved peaks of ^{104,108}Mo at 192.0 and 192.7 keV, respectively. The number of counts in the ^{106}Mo peak gives the relative intensity of the ^{142}Ba-^{106}Mo part of the four neutron channel. This number must be corrected for detector efficiencies and internal conversion coefficients. In principle this must be done for all possible sets of transitions to the ground state in each barium and molybdenum isotope to count all events. However, for this analysis only the strongest transitions in each barium and molybdenum isotope were measured. Because the relative intensities of other transitions are now known in the isotopes of interest, these values were used instead of attempting to measure each peak. Tables I and II show the yield matrices for the Ba-Mo and Ba-α-Zr splits calculated in this way. Note that the values given in the table are scaled and do not match the values in the following figures, which are normalized so that the total area of the Gaussian fit is equal to 1. Summing along

TABLE I. Relative yield matrix for the Ba-Mo charge split.

	^{102}Mo	^{103}Mo	^{104}Mo	^{105}Mo	^{106}Mo	^{107}Mo	^{108}Mo	^{109}Mo
^{138}Ba				0.43(25)	0.35(11)		0.78(36)	
^{139}Ba			0.41(8)	0.36(12)	2.3(2)	2.7(6)	4.8(8)	2.5(1)
^{140}Ba		0.4(1)	0.19(12)	2.9(9)	11.8(6)	11(1)	22(5)	2.5(2)
^{141}Ba		1.2(9)	1.8(8)	13.0(7)	37(1)	26(3)	39(3)	3.2(3)
^{142}Ba	0.007(3)	6(1)	27.0(7)	45.9(9)	92(2)	41(2)	51(33)	5.5(2)
^{143}Ba	4(1)	18.6(8)	59.4(9)	93(3)	111(2)	32(3)	59(9)	2.8(8)
^{144}Ba	10.0(3)	51(1)	102(4)	99(2)	71.7(2)	10(1)		
^{145}Ba	21(1)	53(2)	79(2)	46(2)	21.9(9)			
^{146}Ba	16.7(7)	30.6(6)	39(3)	11.0(2)	4.5(2)			
^{147}Ba	4.3(2)	14.5(4)	5.1(4)	2.1(2)				
^{148}Ba	2.1(2)	1.8(2)	0.5(2)					

diagonals of this matrix gives the relative intensity for each channel. For example, the intensity of the nine-neutron channel in the Ba-Mo split would be the sum along the diagonal of the ^{141}Ba-^{102}Mo, ^{140}Ba-^{103}Mo, ^{139}Ba-^{104}Mo, and ^{138}Ba-^{105}Mo channels.

When analyzing the Ba-Mo and Xe-α-Mo splits, special care was taken in the measurement of the relative intensity of the ^{104}Mo and ^{108}Mo isotopes. With energies of 192.0 and 192.7 keV, respectively, the 2^+-to-0^+ transitions are not resolved in the double gated spectra. Instead, the intensities of the 6^+- to 4^+-519 keV transition in ^{104}Mo and the 4^+- to 4^+-414 keV transition in ^{108}Mo were measured. Because the relative intensities and detector efficiencies for these transitions are known, the individual yields of ^{104,108}Mo could be determined in this way. However, this introduces another source of error because the relative transition intensities depend on the population of the upper levels, which can depend on the neutron channel. The population of a given level can vary by as much as 30% between neutron channels in some cases.

A related problem is that only three- and-higher-fold coincidences are included in the analysis. Because the populations of upper states are low for some fragment pairs (such as the 4^+ in ^{138}Ba), this can lead to a reduction in the relative intensity of these pairs when comparing only threefold coincidences. The difficulties of measuring the intensity of the ^{103}Mo-^{138}Ba are discussed in Ref. [5].

The ternary analysis was similar to the binary analysis in that only the strongest coincidence was used to deduce the intensity of each channel. However, the ternary analysis was not done using the RADWARE software package. Instead, a two-dimensional peak fitting code was used. The code fits coincidence peaks in the γ-γ matrix and subtracts a smooth background. All the other intensities can be calculated in a manner similar to the binary analysis from the intensity of the strongest coincidence. Because ternary fission is rare (on the order of one per few hundred binary events), statistics for this analysis were much lower, producing a greater statistical error. Because α emission constitutes about 90% of ternary fission events it was sufficient to consider only α-γ-γ events, rather than all LCP-γ-γ events.

Although the authors of Refs. [1–3] derived the yields for fragment pairs by summing the observed values of all peaks corresponding to γ transitions to the ground states of odd-A fragments, the wealth of spectroscopic information now available, as well as the high statistics collected in the Gammasphere experiment conducted in 2000, make it worthwhile to revise the calculated yields of fission fragment pairs. Although taking only the 2^+-to-0^+ transitions is a good approximation for most even-even nuclei, there are some cases (^{141}Ba, for example) in which the less intense transitions make up a non-negligible percentage for the overall intensity of the channel, particularly in the 7- to 10-neutron channels.

The new method also has the advantage that most of the error from random contamination in transitions can be avoided. For example, because the 240.7-keV 2^+-to-0^+ transition in ^{110}Ru is so intense in the fission of ^{252}Cf, it will add counts to the 241.1 keV peak in ^{103}Mo, even in double-gated

TABLE II. Relative yield matrix for the Ba-α-Zr split.

	^{98}Zr	^{99}Zr	^{100}Zr	^{101}Zr	^{102}Zr	^{103}Zr	^{104}Zr
^{140}Ba			2.2(7)		2(1)		4(1)
^{141}Ba					7(1)	4(2)	3(1)
^{142}Ba	3(1)	5(2)	1.9(5)	5(2)	7.2(8)	2.5(9)	2.0(6)
^{143}Ba	2.4(1.0)	5.2(2.6)	3(1)	7.5(3.2)	7.2(1.4)		
^{144}Ba	2.4(7)	3(1)	13.6(8)	4.1(1.4)	10.7(7)		
^{145}Ba	1.3(8)	7(3)	2(1)	11.5(3.3)	5(1)	6(1.7)	
^{146}Ba	4.2(8)		4.4(7)	4(1)			
^{147}Ba	3.2(1.5)	12(3.5)	3.9(1.3)				

spectra. This effect becomes especially problematic in channels with very low statistics such as the ^{103}Mo-^{139}Ba 10-neutron channel. Such random contaminations can be avoided almost completely in this analysis.

To extract a relative intensity from coincidence spectra, it is necessary to normalize the measured intensity of the channel to the absolute yield of one of the isotopes of the pair. For the binary analysis, the values given by Ref. [10] for the heavy fragment were used. However, no absolute yields have been calculated for ternary fission. It was therefore necessary to use an *ad hoc* normalization factor for the ternary cases. The *ad hoc* factor was calculated from the binary distributions for each neutron channel by averaging the relative increase in each channel because of normalization. The calculated ternary normalization factors were -11% for even neutron channels and $+20\%$ for odd neutron channels. Because each neutron channel has contributions from several pairs of isotopes, the net effect of normalization should be roughly the same in the binary and ternary cases even though the individual yields of each isotope may be different. The primary drawback to this method is that this type of normalization might mask the appearance of a second mode in the ternary cases. This can be checked by comparing the normalized and unnormalized spectra to see if there is enhanced intensity in the 7- to 10-neutron channels.

The Xe-Ru binary channel was the first to be analyzed. The resulting neutron distribution is fit well by a single Gaussian; there is no second mode. Furthermore,the second mode is not seen in the Ba-Mo case, as shown in Fig. 1. However, there is still a higher-than-expected yield for the nine-neutron channel. This could be evidence for a hot mode, but it is difficult to interpret this result in the usual way because this distribution cannot be fit by a double Gaussian. If there is indeed a second mode, this analysis suggests that the intensity is below 0.5% of the primary mode.

For the ternary analysis, the Te-α-Ru, Ba-α-Zr, and Mo-α-Xe splits were analyzed. The resulting normalized distribution for the Ba-α-Zr split is shown in Fig. 2. No second mode is observed in any case. Furthermore, fitting the unnormalized

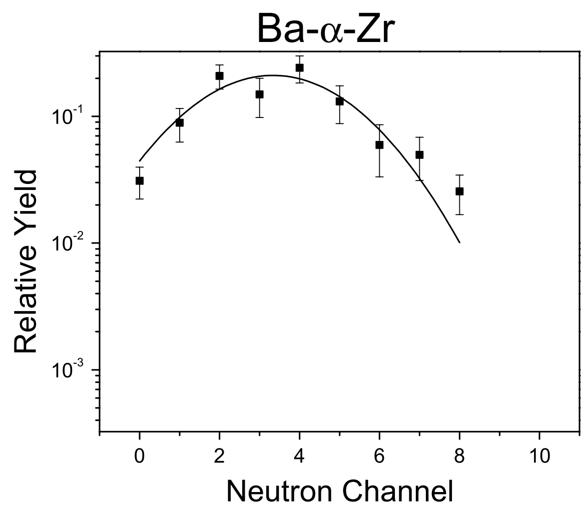

FIG. 2. Neutron distribution for the Ba-α-Zr split.

spectra in the ternary cases showed no evidence for a second mode. This contradicts our previous result [10], which was found by taking gates and peak fitting with RADWARE rather than fitting in the 2D γ-γ matrix, but is in general agreement with the results given by Refs. [11,12].

One interesting feature is the greater width of the Gaussian fit in the Ba-α-Zr case. The full width at half maximum (FWHM) of the fit is about 3.8, whereas in the other cases the width is about 2.7. This is also true in the binary case, where the width of the Ba-Mo distribution is about 2.8 and the other binary cases have a width of 2.7. The increased width of the fit may be indicative of a lower fission barrier in the potential for the barium modes or of some other unique feature of the potential.

It is also possible to fit a double Gaussian to the Ba-α-Zr distribution. If the widths of the peaks are restricted to be the same as the other cases (FWHM $= 2.7$), the distribution can be fit by two equal width Gaussians. To perform the fit shown in Fig. 3, it was also necessary to hold the peak position of the

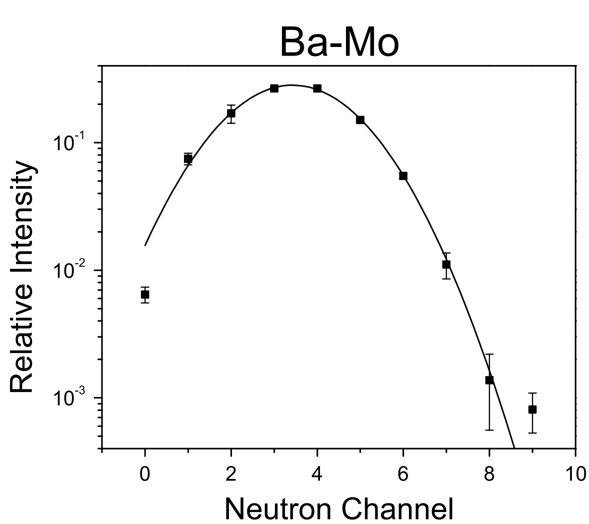

FIG. 1. Neutron distribution for the Ba-Mo split.

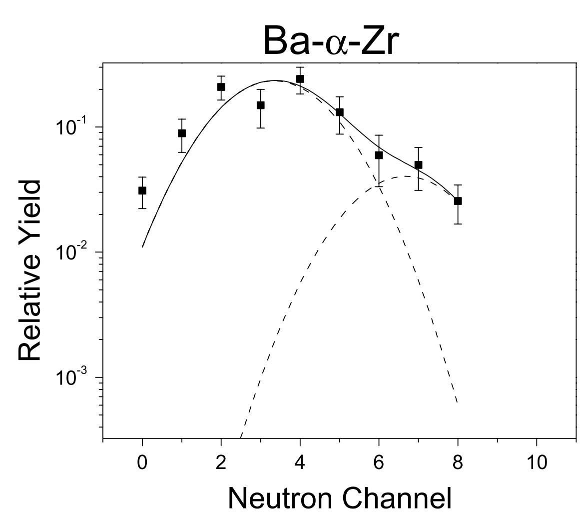

FIG. 3. Neutron distribution for the Ba-α-Zr ternary split fit to a double Gaussian distribution.

primary peak constant at 3.34 and restrict the position of the second peak to greater than five neutrons emitted. Therefore, the only variable parameters in the fit were the relative areas of the peaks and the x position of the secondary peak. The resulting distribution fits the data almost as well as the single Gaussian distribution. The relative intensity of the secondary peak is about 17% of the primary peak. This is in agreement with our previous result (Ref. [7]), but is certainly inconclusive because of the restrictive fitting parameters. However, it is interesting to note that under certain assumptions about the widths and locations of the peaks, the distribution can be fit by a double Gaussian.

In conclusion, the relative intensities as a function of neutrons emitted were determined for two binary channels (Ba-Mo and Xe-Ru) and three ternary channels (Ba-α-Zr, Xe-α-Mo, and Te-α-Ru). Using a simplified method designed to reduce error because of random coincidences and low peak-to-background ratios, no definitive hot mode was observed in any of the five cases analyzed. However, increased FWHM was observed for the two barium channels, as well as an enhanced nine-neutron emission for the binary case. Furthermore, it is possible to fit the Ba-α-Zr data to a double Gaussian distribution by restricting certain parameters, although the data are best fit by a single Gaussian with a FWHM of 3.8 neutrons.

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