Cold (neutronless) α ternary fission of ^{252}Cf

A. V. Ramayya,[1] J. H. Hamilton,[1] J. K. Hwang,[1] L. K. Peker,[1] J. Kormicki,[1,*] B. R. S. Babu,[1,†] T. N. Ginter,[1] A. Sandulescu,[1,2,3,4] A. Florescu,[1,2,3] F. Carstoiu,[3] W. Greiner,[1,2,4] G. M. Ter-Akopian,[1,2,5] Yu. Ts. Oganessian,[5] A. V. Daniel,[1,2,5] W. C. Ma,[6] P. G. Varmette,[6] J. O. Rasmussen,[7] S. J. Asztalos,[7] S. Y. Chu,[7] K. E. Gregorich,[7] A. O. Macchiavelli,[7] R. W. Macleod,[7] J. D. Cole,[8] R. Aryaeinejad,[8] K. Butler-Moore,[8,‡] M. W. Drigert,[8] M. A. Stoyer,[9] L. A. Bernstein,[9] R. W. Lougheed,[9] K. J. Moody,[9] S. G. Prussin,[10] S. J. Zhu,[1,2,11] H. C. Griffin,[12] and R. Donangelo[13]

[1]*Physics Department, Vanderbilt University, Nashville, Tennessee 37235*
[2]*Joint Institute for Heavy Ion Research, Oak Ridge, Tennessee 37835*
[3]*Institute for Atomic Physics, Bucharest, P.O. Box MG-6, Romania*
[4]*Institute für Theoretische Physik der J.W. Goethe Universität, D-60054, Frankfurt am Main, Germany*
[5]*Joint Institute for Nuclear Research, Dubna 141980, Russia*
[6]*Department of Physics, Mississippi State University, Mississippi 39762*
[7]*Lawrence Berkeley National Laboratory, Berkeley, California 94720*
[8]*Idaho National Engineering Laboratory, Idaho Falls, Idaho 83415-2114*
[9]*Lawrence Livermore National Laboratory, Livermore, California 94550*
[10]*Nuclear Engineering Department, University of California, Berkeley, California 94720*
[11]*Physics Department, Tsinghua University, Beijing, People's Republic of China*
[12]*University of Michigan, Ann Arbor, Michigan 48104*
[13]*University Federal do Rio de Janeiro, Rio de Janeiro, CP 68528, RG Brazil*

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The phenomenon of cold (neutronless) alpha ternary fission in spontaneous fission of ^{252}Cf was experimentally observed by triple gamma coincidence technique with Gammasphere with 72 gamma-ray detectors. Correlated pairs of $_{36}$Kr-$_{60}$Nd, $_{38}$Sr-$_{58}$Ce, $_{40}$Zr-$_{56}$Ba, $_{42}$Mo-$_{54}$Xe, $_{44}$Ru-$_{52}$Te, and $_{46}$Pd-$_{50}$Sn were observed to be associated with α ternary fission of ^{252}Cf. Yields of cold α ternary fission were extracted. [S0556-2813(98)01005-X]

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I. INTRODUCTION

The spontaneous emission of light nuclei (cluster radioactivity) is now a widely observed phenomenon, starting from the emission of alpha particles to heavier clusters such as ^{34}Si [1,2]. The cold (neutronless) fission of many actinide nuclei into fragments with masses ranging from ≈ 70 to ≈ 160 has been clearly observed [3–8]. An extreme case is the bimodal fission observed for the Fm and Md isotopes [9] where two distinct fission channels were observed, one with very high total kinetic energy (TKE) corresponding to the cold fission with compact scission shape fragments and the second one at much lower TKE's with elongated fragments at scission. All these observations confirmed the theoretical predictions regarding the cold rearrangement processes of large groups of nucleons from the ground state of an initial nucleus to the ground states of the two final fragments [10,11].

On the other hand, the fragmentations involving more than two final fragments have been also observed. In thermal neutron induced fission of heavy nuclei the third fragment is usually a light charged particle (LCP), the most probable being an alpha particle [12,13]. Heavier clusters like ^{10}Be, ^{14}C, ^{20}O, ^{24}Ne, ^{28}Mg, and ^{34}Si [14] have also been detected. The two heavier final fragments have on the average 20 to 40 MeV of total excitation energy (TXE) which finally leads to the evaporation of a few neutrons in every ternary fission event. Nevertheless, ternary fragmentations characterized by much lower TXE values are sometimes observed [13,15].

It is very important to establish experimentally if cold (neutronless) ternary fragmentations similar to the cold binary ones are existing in nature. This new phenomenon will be equivalent to cluster radioactivity during the fission. Such cold ternary decays will produce all three fragments with very low or even zero internal excitation energy and consequently with very high kinetic energies. Their total kinetic energy TKE $= Q_t -$ TXE will be close to the corresponding ternary decay energy Q_t or even equal to it. In order to achieve such large TKE value, the three final fragments should have very compact shapes at the scission point and deformations close to those of their ground states, similar to the case of cold binary fragmentations [16,17].

The first direct observation of cold (neutronless) binary fragmentations in the spontaneous fission of ^{252}Cf was made [6,7], by using the multiple Ge-detector Compact Ball facility at Oak Ridge National Laboratory, and more recently with the early implementation of Gammasphere [7,8]. In this paper we report the first evidence for cold (neutronless) ter-

*Also at UNISOR, ORISE, Oak Ridge, TN 37831; on leave from Institute of Nuclear Physics, Cracow, Poland.

†Present address: National Accelerator Center, P.O. Box 72, Faure, 7131 South Africa.

‡Present address: Department of Physics, Western Kentucky University, Bowling Green, KY 42104.

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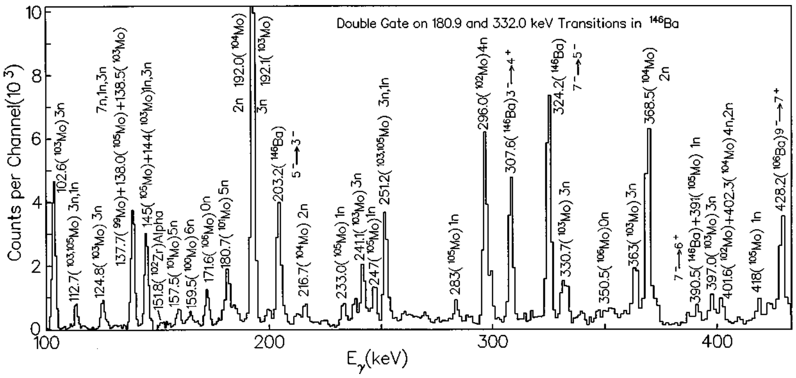

FIG. 1. Coincidence spectrum obtained by double gating on the 180.9 keV and 332.0 keV transitions in ^{146}Ba.

nary fission where the third particle is an α particle. These data were obtained in studying the spontaneous fission of ^{252}Cf with Gammasphere by using the triple coincidence technique. We were able to identify uniquely several correlated pairs with total charge and mass different from ^{252}Cf by only an α particle.

II. EXPERIMENT AND RESULTS

In order to study the fission of ^{252}Cf, a 25 μCi ^{252}Cf source was sandwiched between two Ni foils of thickness 11.3 mg/cm^2 and then sandwiched between 13.7 mg/cm^2 thick Al foils and was placed at the center of Gammasphere with 72 Compton suppressed Ge detectors at Lawrence Berkeley National Laboratory. A total of 9.8×10^9 triple or higher fold coincidence events were recorded. The Gammasphere was calibrated with ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{56}Co, and ^{57}Co sources. The fraction of ^{248}Cm in our sample was experimentally determined and found to be too small to give the observed yields. The SF rate for ^{248}Cm in our sample is 0.13 fissions/s, whereas the SF rate for ^{252}Cf is 2.81 $\times 10^4$fissions/s. A γ-γ-γ cube was built using the RADWARE software [18]. The complex γ-ray spectra obtained in SF were analyzed by the triple gamma coincidence method. In this method a double gate can be set on two γ-rays in any particular isotope or in two different isotopes which are partners of each other.

For a given fission fragment there can be several partner isotopes because 0 to $\sim$10 neutrons can be emitted from its primary partners following scission. The γ-rays emitted by the partners during deexcitation will be in coincidence with each other. If the transitions in one of the partner nuclei are known, one can identify the γ-rays belonging to other partner nuclei uniquely because one can compare the fission yields with calculated ones from Wahl's tables [19]. In the fission of ^{252}Cf, about 100 different final fragments are produced. During the fission process two primary fragments along with several neutrons and/or light clusters are emitted. The primary fragments may also emit several neutrons until the deexcitation energy is below the neutron binding energy of $\sim$6 MeV. Then the secondary fragments decay to their ground states by the emission of γ-rays. Also cold (neutronless) fragmentations are possible. The excited fragments produced in SF are too neutron-rich to emit charged particles such as protons or alpha particles which would take them farther away from β stability.

The data to obtain yields per 100 fission events were analyzed by two different methods.

Method 1. This method is described in our earlier paper [8]. In this method the cold (neutronless) binary fragmentations and 1–6 neutron channels were studied by setting double gates on $2^+\to 0^+$ and $4^+\to 2^+$ transitions in an even-even fragment (e.g., heavy or light) and then measuring the intensities of the $2^+\to 0^+$ transitions in the correlated fragments (e.g., light or heavy). The relative counts for all the pairs of correlated nuclei are determined. Correcting these relative counts for the detector efficiency of the γ-ray and for internal conversion, we obtain the relative yields for a set of particular pairs. Normalizing the total yields to the values in Wahl's table [19] or to the experimental total yields, if known, one obtains the yields per 100 fission events. If one isotope from the heavy (or light) fragment is missing, we evaluated its corresponding yield by interpolation from its neighbors with a Gaussian. A cross-check is necessary by imposing a double gate on the heavy fragment and determining the gamma intensities of the corresponding correlated light fragments. Again the sum of these yields is normalized to the Wahl tables [19] for the heavy fragment. The final isotopic yields must be consistent.

Method 2. In this method one can determine the yields by setting the first gate on the light or heavy fragment and the second gate on its correlated partner. Determining the intensities of γ transitions in both fragments and knowing the branching ratios between different transitions we can determine again the relative binary yields [7,8] which are normalized to Wahl's table [19]. We should like to mention that presently many of the spectra of the odd-Z nuclei are not known, so that we cannot determine experimentally most odd-Z isotopic yields.

In the case of cold (neutronless) alpha ternary fission of ^{252}Cf, we look at the correlation between two even-Z fragments with the sum of charges $Z=96$ and sum of masses $A=248$. For example, $^{146}_{56}$Ba and $^{102}_{40}$Zr are the partners when a ternary α-particle is involved. The Mo nuclei are partners of Ba when binary fragmentations with and without neutron emission are involved. The γ-ray spectrum observed in coincidence with the 180.9 $(2^+\to 0^+)$ and 332.0 $(4^+\to 2^+)$ keV transitions in ^{146}Ba is shown in Fig. 1. The transitions belonging to various Mo partners are shown along with the appropriate numbers of neutrons emitted during the fission. In this spectrum, one can see the $2^+\to 0^+$ transition of energy 171.6 keV of ^{106}Mo, which corresponds to the zero neutron channel. One also sees the 151.8 keV $(2^+\to 0^+)$ transition in ^{102}Zr, which corresponds to cold α-ternary fis-

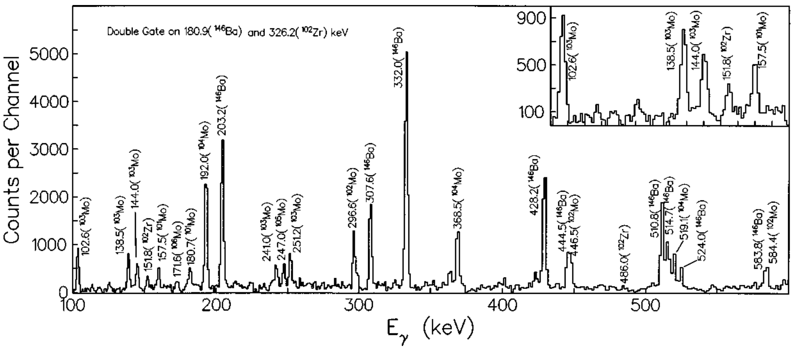

FIG. 2. The triple γ-coincidence spectrum obtained with a double gate set on the $2^+ \rightarrow 0^+$ transition at 180.9 keV in ^{146}Ba and on the $4^+ \rightarrow 2^+$ transition at 326.2 keV in ^{102}Zr. Note the clear peak at 151.8 MeV which corresponds to another transition in ^{102}Zr. The peaks at 203.2, 307.6, 332.0, 428.2, 444.5, 510.8, 514.7, 524.1, and 583.8 keV correspond to different transitions in ^{146}Ba because of the 324.2 keV ($7^- \rightarrow 5^-$) transition of ^{146}Ba in the gate.

sion. In Fig. 1, the 151.8 keV transition is not as clear as some of the other peaks because of background fluctuations. When the double gates were set on two transitions in ^{146}Ba, many normal fission channels to Mo nuclei are open. However, restricting to one α-channel by setting the double gates on the 326.2 keV (^{102}Zr) and 180.9 (^{146}Ba) as shown in Fig. 2 one can decrease the background effect. In Fig. 2 (and also the inset) one can clearly see the 151.8 keV ($2^+ \rightarrow 0^+$) transition in ^{102}Zr. The inset in Fig. 2 shows the region around 151.8 keV (^{102}Zr) transition. This procedure does not imply relaxing the gating condition.

As an additional evidence, in Fig. 3(b) is shown the spectrum by gating on the $2^+ \rightarrow 0^+$ transitions in ^{102}Zr and ^{146}Ba. In this spectrum one can clearly see the $4^+ \rightarrow 2^+$ transitions in both these nuclei. The peak area of the 151.8 keV transition in Fig. 1 is similar to that of 332.6 keV transition in Fig. 3(b), as they should be because of the coincidence gating conditions. In Fig. 2, one observes also γ-rays corresponding to Mo isotopes because the gate 326 keV also contains the $7^- \rightarrow 5^-$ transition of energy 324.2 keV transition of ^{146}Ba. We also see the enhanced octupole band relative to the yrast band in ^{146}Ba during the α-ternary fission which may imply that the octupole shape is playing an important role. These and similar spectra, which are not shown here, clearly prove that we are observing a new phenomenon, cold α-ternary fission with the 0 neutron emission. The question now is whether the process observed is sequential binary process as opposed to the ternary fission, where one of the fragments formed in binary process emits an α-particle. The emission of an α-particle by one of the fragments, when they are in excited states is highly unlikely because these excited nuclei emit neutrons instead of an α-particle because of the Coulomb barrier. Now can the nuclei emit α-particle when they are formed near the ground states such as cold fission. In order to check for this process we studied several correlations as follows. When we gated on the $2^+ \rightarrow 0^+$ transitions in ^{146}Ba and ^{106}Mo as shown in Fig. 3(a) we observed the $4^+ \rightarrow 2^+$ transitions in both these nuclei. The evidence for ^{102}Zr is questionable which implies that we are not observing sequential binary fission. When the gates were set on the $2^+ \rightarrow 0^+$ transitions in ^{102}Zr and ^{146}Ba in Fig. 3(b), we observed clearly $4^+ \rightarrow 2^+$ transitions in ^{102}Zr and ^{146}Ba but not the transition in ^{106}Mo, which implies that there is no correlation between ^{106}Mo and ^{102}Zr. If the α-particle is emitted by ^{106}Mo, one should observe this correlation. When the gates were set on the $2^+ \rightarrow 0^+$ transitions in ^{102}Zr and ^{106}Mo in Fig. 3(c), we do not see the $4^+ \rightarrow 2^+$ transitions in ^{106}Mo or ^{102}Zr, which also implies that the α-particle is not emitted by the light fragment. Similar tests were also performed to check whether the α-particle is emitted by the heavy fragment, and found to be negative. Furthermore, the Q_α for these cases is ≈ -6 MeV. Hence the α particle is unlikely to be emitted after binary fission.

In Table I, we present for the first time the neutronless alpha ternary fission yields observed in the spontaneous fission of ^{252}Cf. The highest experimental yields are found for the Zr + Ba isotopes. Significant yields also are observed for

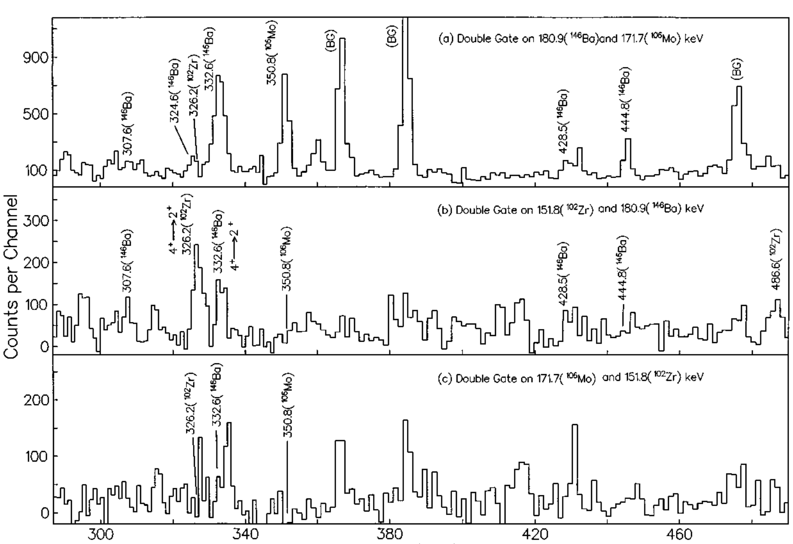

FIG. 3. Triple γ-coincidence spectra obtained with double gates set on $2^+ \rightarrow 0^+$ transitions with 180.9 (^{146}Ba) and 171.7 (^{106}Mo) keV energies (a), with 180.9 (^{146}Ba) and 151.8 (^{102}Zr) keV energies (b), and with 171.7 (^{106}Mo) and 151.8 (^{102}Zr) keV energies (c).

TABLE I. The alpha ternary isotopic yields Y_{expt} obtained per 100 fission events.

α partner nuclei	Y_{expt} (%)	α partner nuclei	Y_{expt} (%)
$^{92}_{36}$Kr-$^{156}_{60}$Nd	0.002 ± 0.001	$^{96}_{38}$Sr-$^{152}_{58}$Ce	0.008 ± 0.003
$^{98}_{38}$Sr-$^{150}_{58}$Ce	0.014 ± 0.006	$^{99}_{38}$Sr-$^{149}_{58}$Ce	0.018 ± 0.009
$^{100}_{38}$Sr-$^{148}_{58}$Ce	0.021 ± 0.010	$^{101}_{38}$Sr-$^{147}_{58}$Ce	0.014 ± 0.011
$^{100}_{40}$Zr-$^{148}_{56}$Ba	0.038 ± 0.012	$^{101}_{40}$Zr-$^{147}_{56}$Ba	0.082 ± 0.010
$^{102}_{40}$Zr-$^{146}_{56}$Ba	0.009 ± 0.004	$^{103}_{40}$Zr-$^{145}_{56}$Ba	0.084 ± 0.029
$^{104}_{40}$Zr-$^{144}_{56}$Ba	0.017 ± 0.008	$^{106}_{42}$Mo-$^{142}_{54}$Xe	0.018 ± 0.007
$^{107}_{42}$Mo-$^{141}_{54}$Xe	0.030 ± 0.014	$^{108}_{42}$Mo-$^{140}_{54}$Xe	0.007 ± 0.003
$^{112}_{44}$Ru-$^{136}_{52}$Te	0.011 ± 0.006	$^{116}_{46}$Pd-$^{132}_{50}$Sn	0.006 ± 0.003

the Mo + Xe and Sr + Ce ternary fragmentations, too. In Table II, the relative binary isotopic yields obtained by using methods 1 and 2 for Mo-Ba pairs are shown. The yields in column 2 [(a),(b)] were obtained by double gating on the $2^+\rightarrow0^+$ and $4^+\rightarrow2^+$ transitions in ^{146}Ba, extracted from transitions to the ground states and normalizing the Mo yields to the values in Wahl's table [19] and the ^{102}Zr yield extracted from its $2^+\rightarrow0^+$ transition intensity relative to the Mo transition intensities and yields. The yields in column 3 [(a),(b)] were obtained by double gating on the $2^+\rightarrow0^+$ transition in ^{146}Ba and $2^+\rightarrow0^+$ transitions in the Mo isotopes and ^{102}Zr and using the $4^+\rightarrow2^+$ transition in ^{146}Ba as a measure of the relative yields. The yields were obtained from analyzing the intensities of $4^+\rightarrow2^+$ transitions in ^{146}Ba. In Table III, the relative isotopic yields obtained by using methods 1 and 2 for Zr-Ce pairs are shown. The yields in column 2 [(a),(b)] were obtained by double gating on the $2^+\rightarrow0^+$ and $4^+\rightarrow2^+$ transitions in ^{102}Zr. The yields in column 3 [(a),(b)] were obtained by double gating on the $2^+\rightarrow0^+$ transition in ^{102}Zr and $2^+\rightarrow0^+$ transitions in Ce isotopes and ^{146}Ba. The relative yields were obtained from analyzing the intensities of the $4^+\rightarrow2^+$ transitions in ^{102}Zr and

TABLE II. (a) Binary (^{A}Mo and ^{146}Ba) yields normalized to Wahl's table [19]. (b) Cold α ternary (^{146}Ba and ^{102}Zr) fission yields of ^{252}Cf. Ratio 1 = (cold α-ternary)/binary-zero-neutron, and ratio 2 = (cold α-ternary)/total-n-channel.

Nuclei	(a) ^{146}Ba/^{146}Ba	^{146}Ba/Mo
^{106}Mo($0n$)	0.029	0.029
^{105}Mo($1n$)	0.10	0.11
^{104}Mo($2n$)	0.30	0.30
^{103}Mo($3n$)	0.35	0.31
^{102}Mo($4n$)	0.15	0.16
^{101}Mo($5n$)	0.079	0.088
^{100}Mo($6n$)	0.0049	0.016
Total (Wahl)	1.017	1.017
Nuclei	(b) ^{146}Ba/^{146}Ba	^{146}Ba/^{102}Zr
^{102}Zr(α)	0.0054	0.0052
Ratio 1	18.4%	17.6%
Ratio 2	0.53%	0.51%

TABLE III. (a) Binary (^{A}Ce and ^{102}Zr) yields normalized to Wahl's table [19]. (b) Cold α ternary (^{146}Ba and ^{102}Zr) fission yields of ^{252}Cf. Ratio 1 = (cold α-ternary)/binary zero-neutron, and ratio 2 = (cold α-ternary)/total-n-channel.

Nuclei	(a) ^{102}Zr/^{102}Zr	^{102}Zr/Ce
^{150}Ce($0n$)	0.016	0.020
^{149}Ce($1n$)	0.088	0.097
^{148}Ce($2n$)	0.23	0.22
^{147}Ce($3n$)	0.46	0.46
^{146}Ce($4n$)	0.35	0.33
^{145}Ce($5n$)	0.19	0.19
^{144}Ce($6n$)	0.034	0.053
Total(Wahl)	1.367	1.367
Nuclei	(b) ^{102}Zr/^{102}Zr	^{102}Zr/^{146}Ba
^{146}Ba(α)	0.012	0.013
Ratio 1	72.05%	64.90%
Ratio 2	0.86%	0.96%

normalizing the Ce relative yields to those of Wahl's table [19]. In α ternary fission, we can set the double gates either on the heavy fragment or the light fragment. The value of 0.0054 obtained for the α ternary fission yield given in column 2 by method 1 is in agreement with the yield obtained by method 2 as shown in column 3 (b) of Table II. We calculated two ratios (cold α-ternary)/binary-zero-neutron and (cold α-ternary)/total-n-channel. These ratios are presented in Tables II and III. The results shown in Table III were obtained by setting the double gates on the light fragment. The cold α-ternary yield in Table III is a factor of two larger. This difference reflects uncertainties in intensities and difference in spin population in the light and heavy fragments. The average values obtained by these two methods are listed in Table I. A few of the cold α-ternary fragmentations presented in Table I involve odd-odd splittings. One can observe that their corresponding experimental yields are comparable or even higher than the yields for the even-even neighbors. In order to obtain the yields for odd-odd splittings, all the γ-transitions to the ground state are summed. The yields are corrected for the internal conversion and detector efficiency. The increased yields in these cases may be because of the differences in level densities in odd-odd and the even-even nuclei.

We mention here that we found enhanced experimental yields for the cold α-ternary fission in which heavier partners are Ba isotopes with masses ranging from 144 to 147. Their higher yields could be attributed to the static octupole deformation observed in this region [20], since octupole shapes at the scission configuration could significantly lower the Coulomb barrier and increase the penetrability between the final fragments. Similar enhanced yields have already been detected in the cold binary fission of ^{252}Cf [8].

We should stress here that the experimentally determined isotopic yields are integrated yields. In the spontaneous fission experiment of ^{252}Cf the majority of the binary and ternary splittings lead to highly excited final nuclei which after neutron evaporation are decaying to the lowest states by gamma cascades. Less frequently, there are also cold fragmentations which leave the final nuclei in their ground or first excited states. We define these cold fission experimental yields as integrated yields since they collect the contributions of all (neutronless) transitions over a whole range of TXE's from zero up to at least the neutron binding energy. These are the first identification and determination of yields of the particular correlated pairs associated with cold α ternary fission.

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