Ternary particle yields in ^{249}Cf(n_{th},f)

I. Tsekhanovich,[1] Z. Büyükmumcu,[2] M. Davi,[3] H. O. Denschlag,[3] F. Gönnenwein,[4] and S. F. Boulyga[5]
[1]*Institut Laue-Langevin, 38042 Grenoble, France*
[2]*Erciyes University, Department of Chemistry, 38039 Kayseri, Turkey*
[3]*Institute of Nuclear Chemistry, 55128 Mainz, Germany*
[4]*Institute of Physics, 72076 Tübingen, Germany*
[5]*Central Department of Analytical Chemistry, 52425 Jülich, Germany*
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An experiment measuring ternary particle yields in ^{249}Cf(n_{th},f) was carried out at the high flux reactor of the Institut Laue-Langevin using the Lohengrin recoil mass separator. Parameters of energy distributions were determined for 27 ternary particles up to ^{30}Mg and their yields were calculated. The yields of 17 further ternary particles were estimated on the basis of the systematics developed. The heaviest particles observed in the experiment are ^{37}Si and ^{37}S; their possible origin is discussed.

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I. INTRODUCTION

The nuclear fission process usually proceeds in such a way that two fragments are formed. These fragments attain about 90% of their kinetic energy within a time interval of several times 10^{-20} s. Within the same time interval, there is also a probability of three particles being formed. Theoretically, fission with the formation of three particles—ternary fission—includes the formation of a third particle, with masses ranging from scission neutrons to middle-heavy fragments when a compound nucleus splits into three parts of comparable mass. Leaving aside the scission neutrons, about 90% of ternary particles are α particles; about 7% are tritons and the rest are heavier nuclei [1,2].

Since the yield of the lightest ternary particles is comparable to that of binary fragments of given mass and charge, detailed studies could already be performed in 1960s and 1970s [3–7]. It was observed that most of the ternary α's are emitted perpendicularly to the fission axis, and only a small fraction (about 3%) are emitted from accelerated fragments (i.e., along the fission axis) [8].

As in binary fission, odd-even effects in the yields of light ternary particles were found; particles with even proton and neutron numbers (such as ^{4}He, ^{10}Be, and ^{14}C) appear more frequently in ternary fission than those with odd Z and N numbers [1].

Experimental data on the properties of particles heavier than carbon ($Z=6$) is scarce—due to the very low probabilities for their production—and quite contradictory. The earliest and probably most extensive study on heavy ternary particles was carried out by Muga *et al.* [9,10], who recorded triple fragment coincidences in solid-state detectors from ^{252}Cf(sf) and thermal-neutron-induced fission of ^{233,235}U and ^{239,241}Pu. The authors of [9,10] report having observed light particles with masses $30\leqslant A\leqslant 70$ with an emission probability of 10^{-5}–10^{-6} per binary fission, at least two orders of magnitude higher than the value given by radiochemical investigations [11]. Schall *et al.* [12] investigated ternary fission from the ^{252}Cf(sf) reaction with a powerful detection system [13]. They studied particles with masses from $A=12$ to 23, with emission probabilities down to about 10^{-6} per binary fission. The probability of symmetric tripartition ($70\leqslant A\leqslant 95$) was estimated in Ref. [12] to be lower than 10^{-8} per binary fission.

Recently, precise ternary fission experiments were performed on the fissioning systems of ^{234}U* and ^{240}Pu* [14], ^{236}U* [15], ^{242}Pu* [16], ^{243}Am* [15,17], and ^{246}Cm* [17,18], using the Lohengrin mass separator at Institut Laue-Langevin. The most comprehensive data sets were obtained for the last three nuclei, for which yields and energy distributions have been precisely determined for several isotopes of elements from hydrogen to oxygen (^{242}Pu*) and to fluor (^{243}Am* and ^{246}Cm*). Some other, heavier, nuclei (up to ^{34,35}Si) were also observed [17].

Despite the experimental and theoretical efforts expended on understanding the ternary fission process, our insight into this phenomenon is still rather poor. It is, e.g., not clear according to which law the observed yields of the heavy ternary particles decrease and whether the upper limit for the ternary particle mass, which is reached in a symmetric tripartition of the compound nucleus, is attained in low-energy ternary fission at a yield level amenable to detection in nowadays experiments. Of particular interest is also the question of how magic shells in complementary fragments influence the ternary particle yields. In consequence, every measurement on yields and energies of heavy ternary particles is of importance.

In this study, yields of radioactive neutron-rich ternary particles were determined for the compound nucleus ^{250}Cf*, with special emphasis to the heaviest ternary nuclei. ^{250}Cf* is the heaviest compound system studied so far in low-energy fission reactions on Lohengrin.

II. EXPERIMENT

The targets used in the experiment were prepared, by Hentzschel [19], by electrolytic deposition of Cf(OH)$_3$ on a titanium backing (ϕ42 mm and thickness of 0.05 mm) in the form of rectangles of dimensions 0.25×2 cm^2. The isotopic composition of the targets was 99.73% ^{249}Cf, 0.15% ^{250}Cf, 0.07% ^{245}Cm, and 0.05% other products relative to the α decay of ^{249,250}Cf. Two targets with masses of 22.7 and

10.15 μg of ^{249}Cf were used in the experiment. The fissile material was protected with a thin nickel foil (0.25 μm) against sputtering by fission fragments. The thermal stability of the target was ensured by dehydration of $Cf(OH)_3$ to Cf_2O_3, which has a melting point of 1750 °C.

The experiment was carried out at the high flux reactor of the Institut Laue-Langevin. Its recoil mass spectrometer for unslowed fission products, Lohengrin, is presently the only instrument allowing the precise study of very rare fission events, such as heavy ternary products [20]. A detailed description of the instrument, its experimental setup and particular features of the measuring technique for ternary fission events can be found in Refs. [21,16].

Due to the presence of the ^{250}Cf isotope in the target material, with its very high neutron capture cross section (2000 b) [22], the actual use of targets in the neutron beam was limited to only nine days for each, in order to keep the fission rate from the ^{251}Cf isotope, which is bred from ^{250}Cf by neutron capture, to a negligible level (less that 10% of total value). The results obtained in this study are, therefore, to be attributed almost exclusively to the ^{250}Cf* compound system.

III. DATA EVALUATION

The Lohengrin mass spectrometer provides separation of particles with ratios A/q and E/q with A, E, and q being particle mass, kinetic energy, and ionic charge state, respectively. The ratios A/q and E/q are selected by the settings for the magnetic and electric fields of the separator. Frequently different fission products fulfill the same conditions. In consequence, they will not be separated in Lohengrin, hence forming multiplets. The components of such multiplets were identified by the difference in their kinetic energy, using an ionization chamber as a detecting device, placed in the focus of the separator. In the case of ternary fission, using a split anode (ΔE-E_R technique, with ΔE and E_R being the signals from the first and second parts of the anode of the ionization chamber), the nuclear charge of the ternary particles could be directly determined from their specific energy losses. The separation of isobars is perfect for ternary particles and allows clean and background-free measurements. An example of typical ternary spectra measured with the ionization chamber with a split anode is shown in Fig. 1.

In the experiment, fission rates decrease with irradiation time, since the quantity of the fissile material decreases as a result of (n,f) and (n,γ) reactions. The change in target activity (burnup) was monitored for each target by daily measurements of the ternary ^{10}Be yield. For a given target with known isotopic composition, the half-life in the neutron beam can be calculated with a decay function, using the neutron flux of the reactor and the data on fission and capture cross sections. The half-life calculated for our target composition amounted to 7.18 days; experimental values extracted from the burn-up data for two targets used were 7.23 and 7.29 days, i.e., very close to the theoretical value. This signifies that practically no other losses of the target material, apart from "natural" burnup in the neutron flux, occurred during the experiment. Hence, the data corrected for the burnup can be compared to each other in a reliable way; their features will reveal those of the ternary fission process.

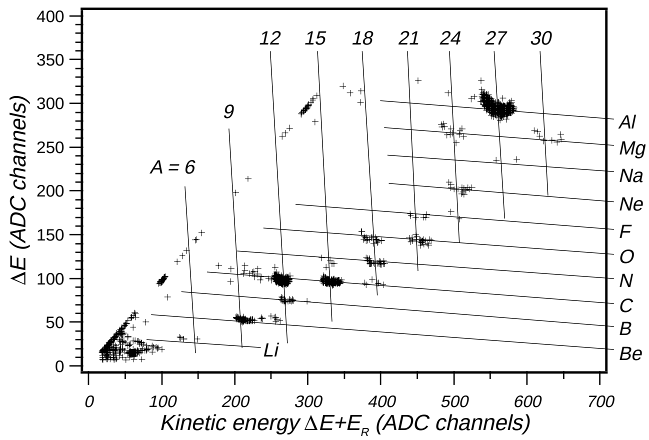

FIG. 1. Ternary particle scatter plot ($\Delta E, \Delta E + E_R$) taken with the $\Delta E - E_R$ ionization chamber at separator settings $A/q = 3$ and $E/q = 4.5$. Since the ΔE signal is proportional to the atomic number, spots located along (roughly) horizontal lines correspond to the same chemical element. Spots on (roughly) vertical lines (approximately constant kinetic energy) belong to isobaric chains. The events in the lower left corner are tritons. Those on the diagonal and close to it are ternary particles scattered on the entrance window of the ionization chamber and stopped in central collisions with the separating grid placed between the ΔE and E_R anode sections (for details, see Ref. [23]).

The fragments' energies measured in the experiment are those of fragments having passed through the target material and the nickel foil. The loss of energy in these substances was estimated for each measured ternary particle with the TRIM code [24]. Additional energy losses can occur when part of the particle kinetic energy is taken away by the emission of neutrons or other particles. In our study, we assumed zero probability for the ternary particles' decay.

Another process to be considered is the ionization of ternary particles due to their interactions with atoms of the target and nickel foil. This ionization is a statistical process, so the ternary particles may end up in different ionic charge states q at the entrance of the separator. Hence, to get correct values for the fission product yield at Lohengrin, the ionic charge distribution of each particle needs in principle to be measured completely. For most particles this would often require very long measuring times and is, therefore, not possible in practice. In addition, for technical reasons there is an upper limit to the separator field strength. Therefore, some assumptions about ionic charge distribution have to be made.

Since the characteristic kinetic energy for fragments from a fission reaction is about 1 MeV/nucleon, according to Shima [25] a Gaussian distribution can be used for the description of ionic charge state probabilities. The parameters of a Gaussian function (mean ionic charge and width) depend on the particle kinetic energy, mass, and atomic number. To obtain their values in cases where they could not be determined from the experimental data a simple prescription given by Nikolaev and Dmitriev [26] was used. To this purpose, the set of constants in the equations from Ref. [26]

characterizing the ionic charge was adapted to our experimental data. The particle-yield correction with respect to q could then be performed with average values for ionic charge and standard deviation, which were either measured or calculated with formulas from Ref. [26], and taken as parameters of Gaussian functions.

The particles' energy is a unique observable, showing whether these particles originate from fission or were knocked out from the construction material of the mass spectrometer. Only particles emanating from a ternary decay process will exhibit a characteristic spectrum with a Gaussian shape, while for recoil particles the Rutherford scattering law shows that scattering probability increases with decreasing recoil energy. In our experiment, the shape of the ternary particle-energy distribution was well reproduced by a Gaussian function in all those cases where a comprehensive distribution was measured (see Table I). The total relative yields of the isotopes were then calculated by integration over their yield at different kinetic energies. Although some stable particles can emanate directly from the ternary fission process, in the present study only yields of unstable particles were considered, due to possible contamination of the former from the material used in the construction of the mass separator (a striking example is ^{27}Al in Fig. 1).

In the yield calculation, the width of each particle-energy distribution plays a crucial role. This parameter is often difficult to obtain experimentally, especially for the heaviest ternary species and for those on the wings of isotopic distributions where production probabilities are low. Unlike binary partition, where due to Coulomb repulsion and momentum conservation both fragments always carry a sizable part of the liberated energy, in ternary fission the third particle, being born between two main fragments, may reveal a nonzero probability of production at energies close to zero kinetic energy. This can happen in case the Coulomb repulsion between the ternary particle and fragment one and fragment two cancel each other, or nearly so. This is known experimentally for all ternary particle distributions, studied for several fission reactions with fissile actinides ranging from Th up to Cf, where comprehensive data could be taken (see, e.g., Ref. [27]). It is also borne out from trajectory calculations for emission of ternary particles from the neck joining the two nascent fragments [15]. This feature is essential in the analysis of ternary fission data and here especially for the evaluation of yields of the heaviest particles at the limit of detection where measurements can be taken only at a few energy settings, preferably at those with maximum yields. In practice, it means that one additional data point has to be added to the experimental particle-energy distributions near zero energy as a constraint to the spectral shape. Only with this artificial constraint, the shape of the distribution will conform to the systematics of ternary-particle energy distributions. Evidently the assumption here is that also the heaviest ternary particles are emitted from the neck region, i.e., they are not produced by a different mechanism. In the actual evaluation, for each particle the corresponding zero-energy probability was set to a few percent from the maximum value measured, each time with an uncertainty of $\pm 100\%$. These artificial points have practically no effect on the width in the case of comprehensively measured energy distributions (most light and middle-heavy ternary particles); they are of importance, however, where only a few points in the energy distribution could be measured (heavy particles). This is demonstrated in Fig. 2 for the examples of ^{10}Be and ^{24}Ne.

In some cases, due to the lack of measured data, a reliable fit of particle-energy distributions was impossible, even with the additional condition of a nonvanishing probability near zero energy. To obtain the yield of such particles, one of the particle-energy distribution parameters (either the width σ_E or the average kinetic energy $\bar{E}$) had to be kept fixed during the fit procedure. The corresponding σ_E and $\bar{E}$ values were deduced from the systematics developed on the basis of the present experimental data. In extreme cases, it was even necessary to fix both σ_E and $\bar{E}$ in the yield calculation for those particles measured only at one kinetic energy or for which no counts at all were observed.

In the literature, the yields of ternary particles are usually given in comparison with the yield of ^{4}He [16,27], which is the most abundant ternary particle; sometimes the normalizations with ^{10}Be or ^{14}C are used [17]. The true value of the ternary ^{4}He yield is difficult to obtain however; its energy distribution is perturbed by the α decay of the target material and the break up of unstable ^{5}He into ^{4}He and a neutron [28]. In addition, α particles may stem from (n,α) reactions on ^{59}Ni bred from stable ^{58}Ni; the latter was used in our experiment as a protecting foil. Therefore, the ^{4}He normalization could introduce a systematic error into the final data.

To avoid this problem, we used a normalization of particle intensities based on that of ^{14}C. Since the absolute yield of the latter is reliably known from Ref. [19], in this paper yields of all ternary particles are given in absolute values, i.e., as probabilities per fission event. This presentation of the data brings both binary and ternary fission to the same scale and facilitates their comparison.

IV. RESULTS AND DISCUSSION

Based on the experimental data for energy distributions extending down to zero energy as explained above, absolute yields could be accurately evaluated for 27 particles (up to ^{30}Mg). For a further 17 particles (up to ^{37}S), the yield was determined with the parameters for the energy distributions (width or/and mean kinetic energy) being obtained from systematics. For these particles, the uncertainties could not be calculated properly; in the case of the heaviest observed particles (low statistics) they may amount to 100%. Finally, for the two heaviest particles ^{39}P and ^{40}S, only upper limits could be deduced, since for them no counts were observed. The results on yields and kinetic energy distribution parameters are given in Table I and depicted graphically in Figs. 3–5.

Figure 3 shows that the average kinetic energy of ternary particles increases with their atomic number Z. This increase is estimated to be approximately 2 MeV per charge number as observed for the most abundant isotopes. This trend allows, and was used for, extrapolations towards heavy atomic numbers. The dependence of kinetic energy on the particle atomic number is expected and is mainly determined by the

TABLE I. Mean kinetic energy $\overline{E}$, width of energy distribution σ_E and absolute yield of ternary particles. Values given in *italics* are those from an "enforced" fit, as explained in the text; they should be considered as preliminary.

	$\overline{E}$ (Mev)	σ_E (MeV)	Yield
^{8}Li	15.1±1.4	7.1±1.3	$(2.6\pm0.7)\times10^{-6}$
^{9}Li	12.5±0.9	5.5±1.0	$(3.8\pm1.0)\times10^{-6}$
^{10}Be	17.5±0.4	7.7±0.6	$(3.8\pm0.7)\times10^{-5}$
^{11}Be	16.5±1.3	7.4±0.9	$(4.7\pm1.2)\times10^{-6}$
^{12}Be	15.1±1.1	7.1±1.1	$(2.7\pm0.7)\times10^{-6}$
^{12}B	21.8±0.8	8.2±1.8	$(1.5\pm0.4)\times10^{-6}$
^{13}B	20.1±1.1	8.1±0.9	$(2.4\pm0.6)\times10^{-6}$
^{14}B	17.0±1.2	7.3±0.7	$(1.4\pm0.4)\times10^{-7}$
^{15}B	16.8±1.9	7.0±1.0	$(9.1\pm4.1)\times10^{-8}$
^{14}C	27.0±0.3	9.9±0.5	$(1.3\pm0.2)\times10^{-5}$
^{15}C	25.1±0.5	8.9±0.7	$(5.3\pm1.1)\times10^{-6}$
^{16}C	24.4±1.1	9.6±1.2	$(4.8\pm1.1)\times10^{-6}$
^{17}C	21.3±1.7	8.3±0.9	$(7.5\pm2.8)\times10^{-7}$
^{18}C	20.4±2.8	8.5±1.4	$(2.4\pm0.7)\times10^{-7}$
^{16}N	25.9±2.2	9.8±1.7	$(1.5\pm0.4)\times10^{-7}$
^{17}N	25.0±1.6	9.4±1.2	$(8.1\pm2.0)\times10^{-7}$
^{18}N	23.8±1.5	9.9±1.2	$(4.5\pm1.1)\times10^{-7}$
^{20}N	*fixed*	*7.0±0.9*	*1.3×10^{-8}*
^{21}N	*fixed*	*fixed*	*3.4×10^{-9}*
^{20}O	31.4±1.7	10.6±1.9	$(2.5\pm0.7)\times10^{-6}$
^{21}O	24.2±1.2	10.7±0.7	$(6.4\pm1.3)\times10^{-7}$
^{22}O	33.0±7.4	14.3±4.2	$(4.2\pm1.6)\times10^{-7}$
^{24}O	*fixed*	*9.5±3.2*	*5.8×10^{-8}*
^{20}F	*25.4±3.3*	*fixed*	*9.7×10^{-9}*
^{21}F	26.5±2.1	9.8±1.3	$(1.6\pm0.4)\times10^{-7}$
^{22}F	33.8±10.5	12.2±4.6	$(1.4\pm0.8)\times10^{-7}$
^{24}F	26.3±2.8	12.1±2.0	$(8.3\pm4.0)\times10^{-8}$
^{24}Ne	33.9±2.9	14.2±1.9	$(2.4\pm0.6)\times10^{-7}$
^{27}Ne	*35.9±5.9*	*fixed*	*2.0×10^{-8}*
^{28}Ne	*fixed*	*fixed*	*1.8×10^{-8}*
^{27}Na	38.4±8.2	16.3±4.5	$(8.2\pm3.2)\times10^{-8}$
^{28}Na	*fixed*	*fixed*	*1.0×10^{-7}*
^{30}Na	31.7±8.6	11.9±6.1	$(2.2\pm2.2)\times10^{-8}$
^{30}Mg	34.9±3.7	13.0±1.8	$(1.3\pm0.4)\times10^{-7}$
^{32}Mg	*fixed*	*10.8±2.7*	*3.7×10^{-8}*
^{34}Mg	*fixed*	*fixed*	*1.0×10^{-9}*
^{30}Al	*fixed*	*fixed*	*9.0×10^{-9}*
^{32}Al	*fixed*	*fixed*	*1.1×10^{-8}*
^{33}Al	*fixed*	*fixed*	*1.8×10^{-8}*
^{32}Si	*fixed*	*12.0±1.7*	*8.9×10^{-9}*
^{33}Si	*fixed*	*11.3±1.4*	*1.5×10^{-8}*
^{34}Si	*fixed*	*11.3±1.3*	*2.2×10^{-8}*
^{37}Si	*fixed*	*fixed*	*2.0×10^{-9}*
^{39}P	*fixed*	*fixed*	*$<5.6\times10^{-9}$*
^{37}S	*fixed*	*fixed*	*4.7×10^{-9}*
^{40}S	*fixed*	*fixed*	*$<3.3\times10^{-9}$*

Coulomb repulsion between all three fragments at the scission point.

The average kinetic energy is in addition expected to drop within isotopic chains. This can indeed be seen from further inspection of Fig. 3: apart from the $A=22$ isobars (where, however, error bars are very large perturbing the oxygen and fluorine data), for practically all isotopic chains, the kinetic energy decreases as a function of mass. This decrease was

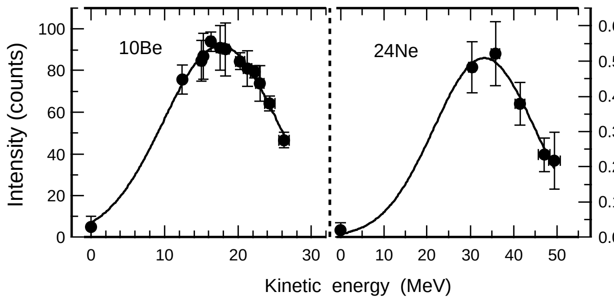

FIG. 2. Experimental energy distributions of ^{10}Be and ^{24}Ne, showing the zero kinetic-energy point as explained in the text. The drawn-out curves represent a fit of the data with a Gaussian function.

again estimated to be 2 MeV per mass unit, and this figure was used in the calculations of the corresponding particle yields.

For the same ternary particles but different compound nuclei, one should expect the kinetic energy to scale with the atomic number of the fissioning nucleus. This expectation was indeed confirmed by Baum *et al.* [15], who compared the average kinetic energies of ternary particles with masses up to $A=20(^{20}\text{O})$ from the fission of ^{236}U* and ^{243}Am*. Experimental data for some ternary particles given in Table II extend the observations made in Ref. [15] to the heavier masses and to other compound systems. Although mean energies for some particles from Table II (^{10}Be, ^{30}Mg) appear to be approximately equal, one can nevertheless conclude that the kinetic energy of ternary particles grows with increasing atomic number of the fissioning nucleus.

The observations made above on the properties of average kinetic energies of ternary particles from fission of the ^{250}Cf* compound nucleus are consistent with those known for other nuclei [14,16]. However, to develop systematics for the parameters of kinetic energy distributions, the behavior of the width σ_E also needs to be examined. From the point of view of the systematics, it appears to be more convenient to work with full width at half maximum (FWHM) values rather than with standard deviations σ_E. In Fig. 4, the FWHM data recalculated from Table I are plotted versus the corresponding average kinetic energies $\overline{E}$. A one-to-one correlation is seen in Fig. 4 between the average and the width of the energy distributions. Again, this is in a rather fair agreement with previously known experimental results [14,16,18,32]. Hence, it is obvious that, in general, a ternary particle tends to keep constant the ratio between second-to-first moments of its energy distribution.

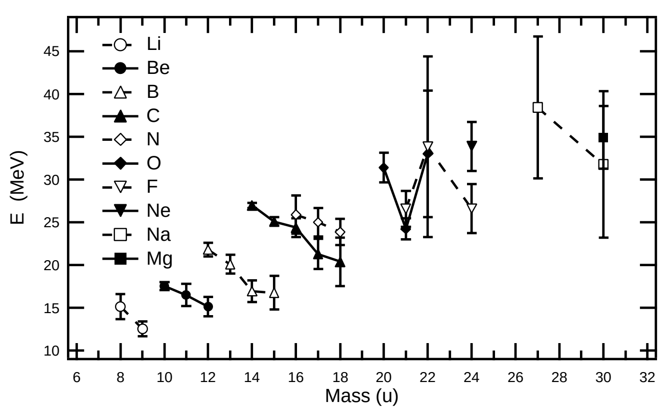

FIG. 3. Mean kinetic energy of ternary particles as a function of their mass.

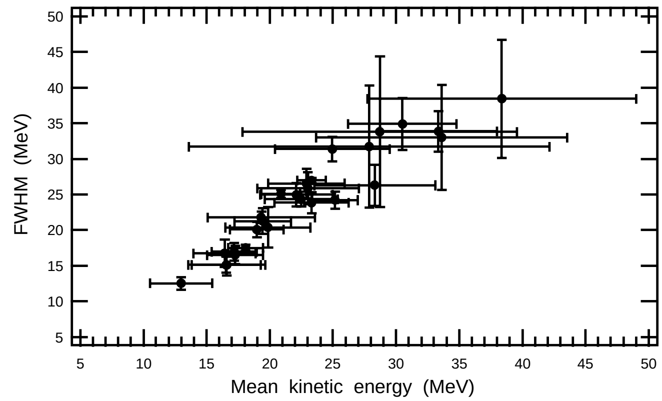

FIG. 4. Correlation between parameters (FWHM and $\overline{E}$) of the ternary particles' kinetic-energy distributions.

This property of the ternary particles' energy distribution strongly differs from that of binary fragments: for the binary fission process the σ_E parameter is known to be practically independent of the fragment mass number (if not disturbed by mixing of different fission modes). This difference in σ_E behavior could perhaps be understood as follows: in binary fission the variance of fragment energies is determined for all fragments alike by the range of compact-to-deformed scission configurations. By contrast, in ternary fission the mean energy $\overline{E}$ of the ternary particles increases with their charge number, but since all distributions are allowed to extend down to zero energies the variance is tied from the low-energy side and, therefore, has to increase with the charge number, too.

A prominent feature of the ternary particle yields is their exponential decrease with particle mass number (Fig. 5), similar to the binary fragment yields on the wings of the double-humped mass yield curve. An inspection of the isotopic distributions in Fig. 5 shows in addition the presence of some fine structure in the particle yields. For each element a staggering is observed, due to shell and odd-even effects of neutrons, as a certain deviation of the isotopic yields from a smooth yield pattern: even neutron numbers appear to be favored though in several cases it is difficult to differentiate between a genuine odd-even effect and data points with low yields in the wings of a distribution. An additional difficulty arises because yields for isotopes on the stability line have not been measured. More conspicuous is that elements with an even charge number have a larger formation probability compared to their odd-charge neighbors. In Fig. 5, odd-Z fragments are shown as open data points and even-Z fragments as full symbols. Systematically the yields of even-Z fragments are higher. The effect is strongest for fragments with low values of Z and decreases with increasing atomic number of the fragments. However, the difference in yield is

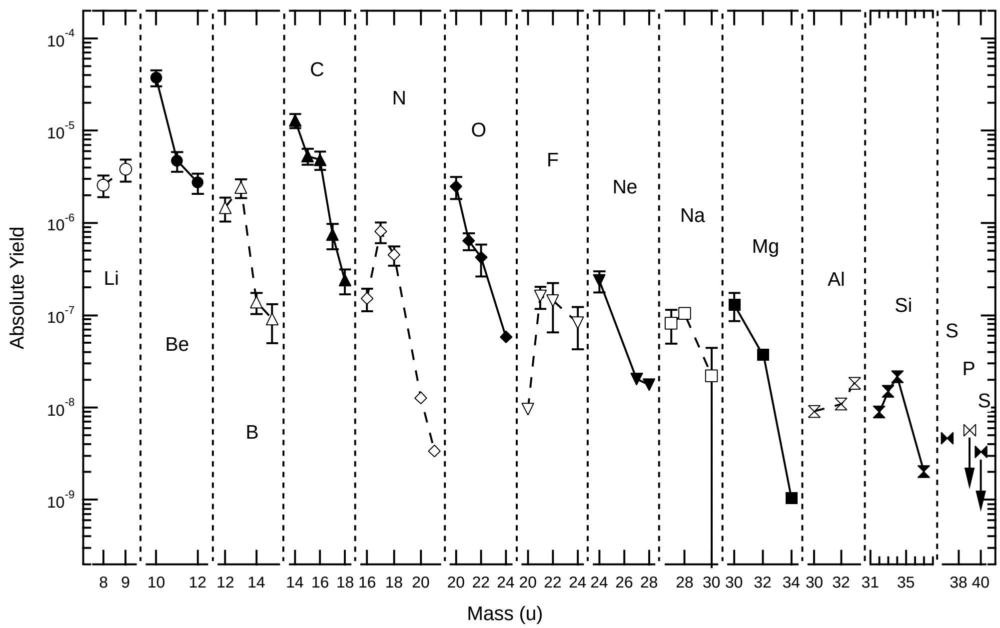

FIG. 5. Yield (probability per fission event) of ternary particles from ^{249}Cf(n_{th},f). Full symbols and solid lines are used for even-Z nuclei; those with odd-Z are represented by open symbols and dotted lines. The lines with an arrow indicate that only upper limits for the yields of ^{39}P and ^{40}S were found.

largest for isobars: The yield of even-even nuclei sometimes exceeds that of odd-odd ones by two orders of magnitude (compare semimagic ^{20}O to ^{20}N and ^{20}F). From the above observations, one concludes that, as far as the size of the odd-even staggering is concerned, neutrons and protons behave differently in the fission process. The yield dependence on the number and type of nucleons is, therefore, of importance and has to be taken into account when searching for the heaviest ternary species at the limit of observability.

The odd-even staggering of yields, known as the odd-even effect, is one of the general properties of low-energy fission. The odd-even effect is well known in binary fission for the yields of fission fragments and serves as a sensitive probe for the excitation energy of the nucleus right at scission. Although in ternary fission the odd-even effect for the yields of the ternary particles is much more pronounced than in binary fission [16,34], no theory or model has been proposed so far to evaluate the excitation energy of the fissioning nucleus from these data. From experiments on ternary fission at higher excitation energies of the compound nucleus it is, however, evident that the odd-even effect depends on excitation [35]. At larger excitation the odd-even effect in the yields of ternary particles is indeed smoothed and eventually disappears. This may be understood from the fact that, at low excitation energies, the structure in the yields of ternary particles is strongly correlated with the corresponding Q values; this even allows models to be established to make quantitative predictions for ternary yields. In contrast, at higher excitation energies the influence of Q values can be expected to become negligible and the odd-even effect should fade away as observed.

Unfortunately, as stated above, at present the odd-even

TABLE II. Mean kinetic energy $\overline{E}$ of energy distribution for some ternary particles (TP's) from different fissioning systems.

	$\overline{E}$, Mev/Compound system				
TP	^{234}U Ref. [27]	^{240}Pu Ref. [29]	^{243}Am[a] Refs. [17,30–33]	^{246}Cm Refs. [17,18]	^{250}Cf [This work]
^{10}Be	17.2 ± 0.1	16.4 ± 0.1	17.0 ± 1.2	19.2 ± 0.6	17.5 ± 0.4
^{16}C	20.0 ± 0.6		21.1 ± 2.4	22.8 ± 2.7	24.4 ± 1.1
^{20}O	24.0 ± 1.0	24.6 ± 0.2	26.9 ± 2.6	23.5 ± 1.3	31.4 ± 1.7
^{24}Ne		28.8 ± 1.3	31.0 ± 2.8	33.6 ± 2.0	33.9 ± 2.9
^{27}Na			35.0 ± 3.0	35.4 ± 3.0	38.4 ± 8.3
^{30}Mg			35.4 ± 4.0	37.2 ± 4.0	34.9 ± 3.7

[a]For several of the TP's the mean kinetic energies reported by different authors are scattered; therefore, the average values carry quite large error bars. References [30–32] are for ^{10}Be, Refs. [17,32,33] for ^{16}C and ^{20}O, Refs. [17,32] for ^{24}Ne. Data for ^{27}Na and ^{30}Mg were taken from Refs. [32] and [17], respectively.

effect of ternary yields cannot be utilized to extract excitation energies of the fissioning nuclei. It is all the more remarkable that the method of double-isotope ratios introduced in the study of intermediate-mass-fragment production in heavy ion physics [36] has been shown also to be suitable for low-energy ternary fission [37]. Making use of the whole body of ternary yield data, consistent values could be deduced for the temperatures of nuclei at scission.

By simply juxtaposing the yields of ^{32}Mg, ^{33}Al, and ^{34}Si with the yields of their neighbors, one may conclude that there is an increasing influence of the $N=20$ shell on the ternary-particle formation probability when approaching for ^{34}Si the stability line. In other words, one observes a disappearance of the $N=20$ magic-shell influence when straying from the stability line. The same is also true for the $N=8$ shell: The $N=8$ magic number dominates in the yield of carbon,[1] still plays an important role in the isotopic chain of boron, and is already of minor importance for beryllium. These observations corroborate experimental findings made in nuclear reaction studies exploring nuclear structure [38–42]. It is remarkable that the yields of neutron-rich ternary particles directly reflect the much-discussed shift in magic neutron numbers when coming closer to the neutron drip line.

Another aspect is the most probable mass for the isotopic distributions of ternary particles, which can also be expressed as the proton-to-neutron ratio Z/N in these nuclei. In binary fission, fragments' masses and charges have been extensively studied with special emphasis put on the discussion of Z/N ratios [43].

In first approximation, the most probable mass for any isotopic distribution of fission products can be described by the rule of unchanged charge density (UCD). This rule postulates that, prior to neutron evaporation, the ratio Z/N in any fragment is the same as in the fissioning compound nucleus. In second approximation, for low-energy binary fission a small correction has to be made: The light fragments are slightly more proton rich and the heavy fragments are correspondingly proton deficient when compared to the compound system. This proton excess or deficiency is usually given as a displacement ΔZ of the experimental nuclear charge Z_P relative to the charge predicted by the UCD rule (Z_{UCD}). Generally $\Delta Z(=Z_P-Z_{UCD})$ amounts to about half a charge unit ($\Delta Z\approx\pm0.5$). Physically, it is due to the Coulomb repulsion between protons, that affects heavier fragments more than light ones.

In this context, the behavior of ternary particles is interesting: Will they be even more proton rich ($\Delta Z>0.5$) than light fragments due to their considerably smaller size or will their origin from a position between light and heavy fragments lead to a charge displacement intermediate between heavy and light fragments, i.e., $\Delta Z\approx0$?

[1]The dominance of ^{14}C in the isotopic chain of carbon follows from the extrapolation to ^{250}Cf* of the experimental data known for some lighter fissioning systems (^{236}U* [15], ^{243}Am* [33], ^{246}Cm* [17]). For all of them, the yield of stable carbon isotopes (^{12,13}C) was found to be considerably lower than that of ^{14}C.

TABLE III. Most probable mass A_P of ternary fragments with the atomic number Z calculated according to the UCD rule. $A_{P|en}$ are the masses closest to A_P with an even number of neutrons.

Z	A_P	$A_{P\|en}$	Z	A_P	$A_{P\|en}$
2	5.1	6	10	25.5	26
3	7.7	7	11	28.1	29
4	10.2	10	12	30.6	30
5	12.8	13	13	33.2	33
6	15.3	16	14	35.7	36
7	17.9	17	15	38.3	39
8	20.4	20	16	40.8	40
9	23.0	23			

For this purpose, the experimental yield distribution of the elements for various masses as shown in Fig. 5 will be compared with a most probable mass $[A_P(Z)]$ calculated assuming UCD, according to

$$A_P(Z)=Z\times A_F/Z_F$$

with $Z_F=98$ and $A_F=250$ the nuclear charge and mass numbers of the compound nucleus, respectively. The most probable masses A_P calculated according to this simple recipe are given in columns 2 and 5 of Table III.

For a more realistic comparison with the maximum of isotopic yield distributions (see Fig. 5), we have to take into account the strong modulation of yields due to the odd-even effect of neutrons. Columns 3 and 6 in Table III give the masses $A_{P|en}$ with an even number of neutrons which lie closest to the masses A_P. Comparing the mass numbers $A_{P|en}$ in Table III with the masses of the isotopes produced in highest yield in Fig. 5 we observe that the calculated mass numbers seem to basically agree with the present experiment for the elements with $Z=4$ (Be), $Z=5$ (B); $Z=7$ (N); $Z=8$ (O); $Z=12$ (Mg) and $Z=13$ (Al). It has to be noted, however, that for $Z=4$, 8, and 12 no experimental value below $A_{P|en}$ could be obtained so that higher yields at masses $<A_{P|en}$ cannot be ruled out completely. The distribution obtained for $Z=6$ (C) shows the highest yield at ^{14}C whereas $A_{P|en}=16$. We attribute this mainly to the shell closure at $N=8$, which has already been discussed above. Another effect of this shell closure is observed for the yield of ^{13}B. Similarly, it is known from the literature that ^{4}He dominates the distribution of ternary helium isotopes whereas the value of $A_{P|en}$ amounts to 6—certainly an effect of the double shell closure at $Z=2$ and $N=2$. Finally, the—less pronounced—effect of the shell closure at $N=20$ observed in the yields of ^{32}Mg, ^{33}Al, and ^{34}Si has also been discussed above.

Even though we are aware that both, more and more accurate data are needed, and that a systematic and critical study of the body of existing partially contradictory ternary yields of different origin available for various fission reactions would be advisable, the formulation of the following ideas will be based essentially on the present data.

In consequence, returning to the discussion of a possible charge displacement ΔZ keeping in mind the modulations

TABLE IV. Largest atomic number expected and observed for ternary particles from thermal-neutron-induced-fission reactions for U [26], Np and Pu [16,28], Am and Cm [17].

Zmax	U	Np	Pu	Am	Cm
Expected	$Z=10$ (Ne)	$Z=11$ (Na)	$Z=12$ (Mg)	$Z=13$ (Al)	$Z=14$ (Si)
Observed	^{24}Ne		^{30}Mg	^{35}Si	^{32}Mg

due to odd-even and shell effects, we observe that for the elements with $Z\geqslant 9$, particularly for F, Ne, Na, Si, and S, the maxima of the experimental distributions tend to be below $A_{P|en}$, i.e., the fragments tend to be neutron deficient with respect to the compound nucleus. The reason can be twofold as follows:

(a) As has been discussed above, the size of these ternary fragments is even smaller than that of the light binary fragments and the Coulomb energy could be at the origin of this observation.

(b) A second explanation would be that these relatively large ternary fragments are born with some deformation and will emit prompt neutrons before they are detected.

One argument against assumption (a) and in favor of assumption (b) is that in the first case ternary fragments with $Z<9$ should be even more neutron deficient. This is not observed.

On the other hand, an argument supporting assumption (a) (primary formation of neutron deficient heavier ternary fragments) is as follows: Several phenomenological models reproduce the whole body of ternary yields as a function of ternary mass and charge reasonably well. In the Halpern model, for example Ref. [44], it is argued that the yields depend on the energy it costs to pick up nucleons from the prefragments and to place a charged light particle in between the two main fragments. The general trend of the yields, ranging from the He isotopes to the heaviest ternary elements such as Si and S, is qualitatively well understood in this model. According to the model, for a given charge number Z the isotopic yields depend mainly on the Q values. It is then readily calculated from mass tables that, e.g., for silicon isotopes the favored mass number should be $A_P=34$ as observed, while for sulfur several mass numbers below (but including) $A=40$ are in competition, in agreement with observation.

However, it should also be remembered that for light nuclei the neutron drip line comes very close to the stability line. Therefore, among the lighter ternary particles, nuclei can be produced in fission, which are particle-unstable against neutron decay in their ground state (e.g., ^{5}He, ^{7}He) or in low excited states (e.g., ^{8}Li*, ^{9}Be*). The presence of these unstable nuclei in ternary fission is accounted for in all models of ternary fission. Experimentally, neutron emission in coincidence with light ternary particles has recently been studied quite extensively for fission of ^{252}Cf(sf) [45].

The two heaviest particles searched for, ^{39}P and ^{40}S, could not be observed in the experiment, despite relatively long measuring times (6.8 and 13.6 h, respectively). Upper limits for their yields are given in Table I. Longer measurements could possibly lead to a positive result. However, we cannot reject the possibility that these nuclei are not formed in ternary fission, at least at the present level of detection, which is about $10^{-9}/f$, where f stands for fission, in the case of the ^{249}Cf(n_{th},f) reaction. This supposition is supported by the observation of surprising stability in the yields of binary fragments around masses $A=132$ in the heavy and $A=80$ in the light peaks of the fission yield curve. The yield in these mass regions was found to be virtually equal for all fissioning systems [46]. This stability undoubtedly results from the structure of fragments around $A=132$ (doubly magic, $Z=50$ and $N=82$) and $A=80$ (close to doubly magic $A=78$ with $Z=28$ and $N=50$). Assuming them to be preformed at the scission point, one may put the rest of the nucleons into a neck, as suggested in Ref. [47]. Such a naive picture will then allow us to estimate the maximum atomic number and mass of the ternary particles expected to be formed from such a neck, with intact cores of the prefragments. For californium, this would be $Z=16$. In the particular case of ^{250}Cf*, the heaviest ternary particle would be ^{38}S, provided zero neutron emission takes place from fission fragments (cold fission). The nucleus ^{37}S observed in the experiment is only one neutron away from this limit.[2] Upper limits for nuclear charges of ternary particles for other fissioning systems, estimated in the same way, are given in Table IV together with the heaviest ternary particle hitherto observed.

As seen, the expected values for even-Z fissioning systems are in good agreement with the experimental findings. For odd-Z Am, the experiment shows an excess of one proton in the heaviest ternary particle. The reason could be the preferential formation of even-Z ternary particles (odd-even effect), as discussed above. Being more tightly bound than the complementary partners (i.e., light fragments; heavy ones being supposed to remain doubly magic), even-Z ternary nuclei will be preferred energetically, even in the case of odd-Z fissioning systems. In summary, at the practical limit of some $10^{-9}/f$ for the measurements of ternary yield probabilities at the Lohengrin mass separator, the masses and charges of the ternary particles virtually exhaust all neck nucleons without breaking the magic cores of both fragments. It should be most interesting to study yields of ternary particles beyond

[2]Based on the analysis of spectroscopic data, it has been recently demonstrated [48] that the ^{37}S nucleus is the last one where the $Z=16$ subshell still persists; it disappears for the heavier S isotopes. Recalling the major importance of the proton number for the ternary particles yield, it is very likely that the break up of the $Z=16$ subshell at ^{38}S results in a smaller formation probability if compared to that of ^{37}S. But this conjecture should be tested experimentally.

this limit since it is conjectured that these yields should drop much faster with the mass and/or charge of the particles than anticipated from an extrapolation, e.g., in Fig. 5.

Finally, we wish to address the question of whether all the heaviest particles observed in our experiment result from ternary fission (i.e., from the split of the compound nucleus into three parts), or whether they could be formed by other processes, such as binary cluster emission from excited compound systems and/or nuclear breeding from surrounding materials.

As to breeding reactions, only ^{37}S has to be considered. Given our experimental conditions, about 10^5 atoms of stable ^{36}S on the covering foil (=rectangle of target material) would be needed for the production of one nucleus of ^{37}S during neutron irradiation. With the high flux of fission fragments ($\sim 10^{12}$ fission/s for the target used) this nucleus could then be knocked out of the foil. The requirement for knockout at the appropriate solid angle of the separator acceptance (see Ref. [21]) increases the minimum amount of ^{36}S to 10^{10} atoms. This corresponds to 5.3×10^{13} atoms (or 2.8 ng of mass) of the most abundant isotope ^{32}S. This is already a macroscopic quantity, allowing its determination with non-nuclear methods. Since the effective Ni layer had a mass of 0.111 mg, the required sensitivity of the method to be applied should be better than 2.5×10^{-5} g/g.

We used a double-focusing sector-field inductively coupled plasma mass spectrometer (ICP-SFMS ELEMENT, Finnigan MAT, Bremen, Germany) for the determination of the sulfur concentration in the nickel foil. A piece of Ni foil identical to that used in the experiment was dissolved in high-purity nitric acid and diluted with deionized Milli-Q water. The method applied allowed us to recognize mass interferences caused by molecular ions and doubly charged matrix ions ($^{16}O_2^{+}$, $^{64}Ni^{2+}$, etc.) and to distinguish them from sulfur isotopes. The accuracy of the analytical method was tested with the NIST SRM 1160 standard. Further details on the ICP-SFMS technique can be found in Refs. [49,50]. The measurement showed the sulfur concentration to be lower than 1.0×10^{-5} g/g of solid nickel foil, leading to the conclusion that the ^{37}S observed could not originate in a breeding process.

It was shown theoretically [51,52] and found experimentally [53,54] that nuclei in their ground states may emit particles heavier than α's (so-called "clusters"). Since the energy distribution of ^{37}Si and ^{37}S was not measured in this work, we have no direct experimental proof that ternary particles were indeed observed. Some discussion is, therefore, required on whether the heavy isotopes observed could not be due to binary cluster decay.

One may first consider spontaneous cluster decay from the target nuclei ^{249}Cf. However, the amount of target material is small and the half-lives predicted for cluster emission are extremely low [52]. Taken together with the short measuring times of only a few hours, spontaneous cluster emission may safely be excluded as a source for the events observed. One might next consider induced cluster emission from ^{250}Cf* following neutron capture in ^{249}Cf. Induced α decay and exotic cluster emission from excited states of ^{238}U have been studied in Coulomb scattering of ^{238}U on ^{238}U [55] with the result that induced α decay has very low cross sections of 10^{-37} cm^2 even for excitation energies close to the Coulomb barrier. For cluster emission, the cross sections at excitation energies of nuclei having captured a thermal neutron are predicted to be even smaller and speculation on cluster emission induced by neutron capture as an observable process is explicitly rejected [55]. A further and probably even more compelling justification that the events detected in the present experiment are due to a ternary fission process is the kinetic energy at which the particles were intercepted. In a binary decay, the kinetic energies of the light partners from super-asymmetric fission with masses around $A=40$ should have energies roughly twice as large as those measured here. Such light fragments at high kinetic energies have been intensively sought on Lohengrin, with measuring times exceeding those of the present work by several times, but no positive result has been reported [17,32].

We, therefore, believe we can unequivocally attribute the observed heaviest particles to the ternary fission process. The nuclei ^{37}Si and ^{37}S are up to now the heaviest particles observed experimentally from ternary fission at low excitation energies.

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