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Energy distribution of ternary α particles in spontaneous fission of ^{252}Cf

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The energy distribution of the ternary α particles in spontaneous fission of ^{252}Cf was measured. For the first time an energy threshold as low as 1 MeV was reached. The experiment used an array of unshielded silicon detectors measuring energy and time-of-flight (TOF) of ternary particles in coincidence with fission fragments. The TOF resolution of the system was sufficient for clear separation of ^{6}He and tritons from ^{4}He. The statistics were adequate to extract the ^{6}He/^{4}He yield ratio. For both ^{4}He and ^{6}He, an excess in the yield (as compared to a Gaussian shape) was observed at energies below 9 MeV. The measured ternary α spectrum was corrected for the distortion induced by the detection geometry covering equatorial particle emission only. The emission angle was found to affect mainly the width of the energy distribution by up to 1 MeV.

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I. INTRODUCTION

Since the discovery of ternary fission in the 1940's there have been numerous experiments devoted to the energy distribution of the ternary α particles, being by far the most frequent light charged particles (LCP) (see, e.g., reviews [1,2]). Nevertheless, surprisingly little is known about the low-energy part of these distributions, having been at issue for decades [3]. The yield at low energy is particularly important. Low-energy α particles presumably arise from low α-particle initial energies or more stretched scission configurations of the main fragments. They thus may provide important insight into both the emission mechanism of ternary particles and the scission stage of the fission process. However, particle-unstable ternary particles (e.g., ^{5}He and ^{8}Be) may also give rise to low-energy α particles in sequential processes [4] and thus mask the true ternary particle emission. To address the problem precise experimental data are needed.

Experimental studies at low energy are still scarce, and the data are not consistent. This is true also for ternary α particles from the spontaneous fission (sf) of ^{252}Cf, one of the most often studied decay processes (e.g., Refs. [3–10]). The main reasons for this situation are twofold. First the intense background from the 6.1 MeV α particles from ^{252}Cf radioactive decay has forced many researchers to use protection foils on the detectors, restricting also the ternary α-particle spectrum to energies above that value. The second is the preference given to the ΔE-E method to identify ternary particles. The method does not allow exploration of energy distributions below the penetration energy of the ΔE detector thickness ($\simeq$3 MeV for 12 μm and $\simeq$4.5 MeV for 20 μm, as examples). With ΔE-E telescopes protected by absorber foils the energy threshold is typically around 9 MeV [3]. Such rather high threshold values compared to the 16 MeV mean energy not only cut away the interesting low-energy part of the spectrum but also leave substantial ambiguity in the energy assignment of the above threshold events due to uncertainty in absorber and ΔE detector thicknesses and related energy losses. Up to now, the only experiment with unshielded energy detectors has been the TOF-E measurement by Tishchenko *et al.* [10] with the 4π Berlin Silicon Ball. Here, the detection threshold was pushed down to 2.0 to 2.5 MeV, but separation of α particles from tritons and ^{6}He was only partially achieved due to the short flight paths of 10 cm, being equal for the fission fragments (FF) and ternary particles inside the ball. In an early experiment by Loveland [6] the energy spectrum of ternary α particles in ^{252}Cf(sf) was reported to have been evaluated down to 1 MeV energy, although a ΔE-E telescope was used in the measurement. There have also been attempts to measure low-energy ternary α particles by nonelectronic methods, e.g., a mass spectroscopic measurement after using Pb catcher foils for the reaction ^{235}U($n_{\rm th}$, f) [11] and solid state nuclear track detectors (SSNTD) for ^{252}Cf(sf) [12]. While in Ref. [11] a massive "short-range" component below 7.7 MeV was stated, no intensity in excess of a Gaussian shape was found in Ref. [12] in this region.

We have remeasured the ternary α-particle spectrum from ^{252}Cf fission in a TOF-E experiment with unshielded silicon detectors at a distance of 20 cm to the source and registering the FFs with a micro channel plate detector (MCP) at a close distance of 2.5 cm. Preliminary results were presented at conferences [13,14].

II. EXPERIMENT

The measurements were made using a thin, double-sided ^{252}Cf source. The source, produced at the Radium Institute at St. Petersburg, had an activity of 500 fissions/s. It was prepared

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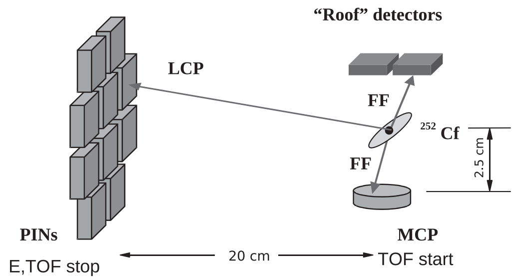

FIG. 1. Experimental setup for measuring the ternary α-particle spectrum from ^{252}Cf fission by the TOF-E method. The assembly of ^{252}Cf source, channel plate start detector, and fragment energy detectors (roof detectors) is seen on the right-hand side. Ternary particle detectors facing the open side of the sample are placed at a 20 cm distance.

by the self-transfer method onto a 22 μg/cm^2 aluminum oxide support backing with a 10 μg/cm^2 layer of gold. A schematic drawing of the setup is shown in Fig. 1.

The source was at the distance of about 20 cm from 10 silicon p-i-n diode detectors (380 μm in thickness and 30 × 30 mm^2 area) for the detection of LCPs. Proper diaphragms shielded the guard ring structure of the p-i-n diodes. The source side on which Cf was deposited was facing the array and was tilted at 45° to allow unobstructed detection of the LCPs in coincidence with the FF registered in the MCP detector. The later gave a very good start signal but no energy information. The timing extracted from the array was inferior to the MCP but fully sufficient for particle separation based on the E vs TOF analysis. The distance from the source to the converting foil of the MCP was 2.5 cm. The active part of the conversion foil had a diameter of 3 cm. Selection of fission fragment start signals in the presence of the 30 times more frequent 6.1 MeV α particles was achieved by coincident registration of the complementary fragments in 2 silicon p-i-n diodes of 20 × 20 mm^2 area (roof detectors) mounted at a distance of 6 cm to the sample, opposite the MCP.

With dedicated preamplifiers and low-noise timing filter amplifiers in the timing channels the energy threshold could safely be reduced to $\leqslant$0.5 MeV. This is the lowest cutoff value ever achieved in a ternary fission experiment. Data were collected over a period of about 6 weeks, with no significant deterioration of detector performance due to radiation damage. Energy calibration of the silicon detectors was performed with α lines from a spectroscopic thin ^{226}Ra source and a BNC PB5 precision pulse generator, to better than 50 keV (FWHM). As is stated above, in the present experiment there is no material between the open side of the ^{252}Cf source and the surface of the detectors. The nominal thickness of the aluminium layer on the entrance of the silicon detectors is 140 nm. The corresponding effective dead-layer was determined with angular dependent α spectroscopy to be 369(11) nm of silicon equivalent [15], which results in an energy loss of 110 keV for 1 MeV α particles. At 10 MeV the corresponding value is 30 keV. These small corrections were disregarded in our analysis.

III. DATA ANALYSIS

Only complete events were accepted for further analysis. In particular, we have required that both FF pairs associated with the ternary event are registered: one in the MCP and the complementary fragment in the roof detectors. The measured difference in the fragment flight times to MCP and roof detectors, respectively, vs fission fragment energy $E_{\rm FF}$ registered in the roof detectors was used to correct the measured TOF spectra of ternary particles for the difference in flight time between heavy and light fragment masses from source to MCP. The resulting TOF vs E pattern for ternary particles is shown in Fig. 2. The intense bunch in the center corresponds to ternary α particles, and the weaker bunches of the neighboring isotopes ^{3}H and ^{6}He below and above the α-particle distribution are nicely separated from it. The three bunches in the upper left corner are identified as ^{27}Al, ^{16}O, and ^{12}C scattered off from the source backing or the roof detector surface by fission fragments. Between these groups and the ternary ^{6}He particles a few events from heavier LCPs, mainly ^{8}He and ^{10}Be, are visible. It must be noted that the TOF-E pattern in Fig. 2 is particularly background free, although FFs are hitting the detectors with ≈300 times higher rates than ternary particles. The vertical line at 6.1 MeV represents random coincidences with the 10^4 times more frequent α particles from ^{252}Cf radioactive decay. The random rate is sufficiently low to permit safe subtraction of random events in the time window of the ternary α-particle distribution. It is interesting to see also a small 6.1 MeV peak at 2 ns above the pattern for the ternary α particles that is attributed to start signals from x-rays or conversion electrons in the MCP when the ^{252}Cf radioactive decay proceeds through the excited state of ^{248}Cm. This peak falls accidentally into the TOF-E pattern

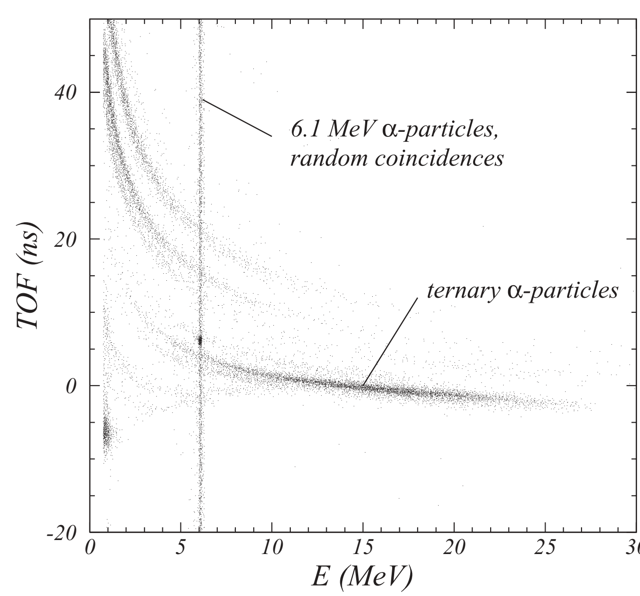

FIG. 2. Scatter plot TOF vs E of ternary particles in ^{252}Cf(sf), as measured with 10 silicon p-i-n diodes of 380 μm in thickness and 30 × 30 mm^2 in size, located at 20 cm distance from the source. Time is in ns, the scale being arbitrarily normalized to zero at the flight time of α particles with the mean energy of 16 MeV; energy E is in MeV.

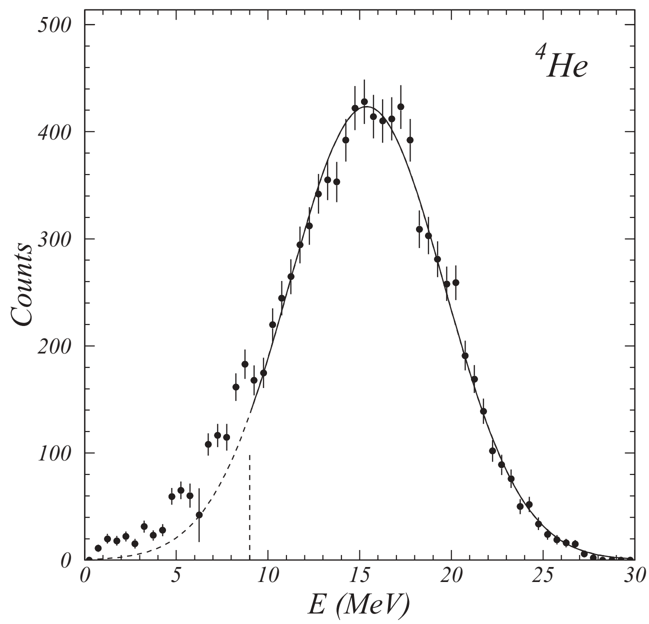

FIG. 3. Measured energy distribution of ternary α particles from ^{252}Cf fission. The solid line is a Gaussian curve fitted to the data above 9 MeV.

of ternary ^{6}He, ruling out an analysis of the ^{6}He spectrum in a small energy gap around 6 MeV. At the high-energy side the ternary α-particle spectrum is cut off at 27.5 MeV due to the limited detector thickness of 380 μm. The cutoff becomes effective at a yield level of $\approx$3% relative to the maximum yield at 16 MeV, causing only a minor distortion of the spectral shape at higher energies. For ternary ^{3}H particles the highest energy stopped in the detector is 11.5 MeV. Their TOF-E pattern bends back for higher energies (see Fig. 2) interfering with the respective pattern for the protons.

The measured energy spectrum of ternary α particles (Fig. 3) covers the wide energy range from 1 to 27.5 MeV. The total number of events collected over the 6-week period of the measurement amounts to 9447. It must be noted that having the particle detectors at right angles to the direction of emission of the fission fragments does bias the experiment to detect mainly ternary particles emitted in the neck region at the instant of scission, i.e., the so-called equatorial particles. Strictly speaking, any measured ternary α spectrum depends a little on the experimental cutoff for the emission angle $\Theta_{\alpha L}$ (angle between the ternary particle and the light group of fission fragment). The dependence is due to the well-known increase of the angular width $\Delta\Theta_{\alpha L}$ of the equatorial α-particle distribution with energy and, furthermore, the onset of the polar α particles at energies above about 20 MeV (e.g., Ref. [16]). Thus any constraint in the α-particle emission angle $\Theta_{\alpha L}$ causes the equatorial yield to be slightly suppressed with increasing energy. On the other hand, with no FF registration, as was, e.g., the case in Ref. [3], the high-energy fraction of the α-particle spectrum is slightly enhanced for two reasons, the fully covered equatorial distribution and the predominantly highly energetic polar particles.

The issue has been analyzed with the aid of a simulation calculation, using data on $\Theta_{\alpha L}$ vs E_α measured previously by Heeg *et al.* [17,18] with the double-torus ionization chamber DIOGENES. For this purpose, we determined our constraint in detection angle by Monte-Carlo simulation. Our detector geometry registered α particles with $\Theta_{\alpha L}$ angles from 55° to 130°. The efficiency function has a FWHM of 33° around a mean at $\Theta_{\alpha L} = 92°$. Folding this angular dependent registration efficiency with the α-particle angular distributions determined by P. Heeg for fine energy intervals of 1 MeV (Fig. 5.3 in Ref. [18]) we have obtained the registration efficiency in our setup vs E_α. For the discussion presented below, corrections to the measured α spectrum were made for two cases: (a) the wider than detected angular interval of equatorial particles, best defined as $50° \leqslant \Theta_{\alpha L} \leqslant 130°$ according to the DIOGENES data, and (b) the full span of angles, including polar emission, as measured in experiments without fragment registration (e.g., Ref. [3]) and in coincidence experiments with near 4π geometry (e.g., Refs. [4,10]).

IV. RESULTS AND DISCUSSION

For demonstrating the influence of the detection angle on the spectral shape, on the one hand, and providing reference ternary α spectra in ^{252}Cf for different experimental geometries, on the other hand, we are listing in Table I mean energies and widths of Gaussians curves fitted to various α spectra: (i) the measured spectrum, and the measured one after corrections were applied for (ii) the equatorial range $50° \leqslant \Theta_{\alpha L} \leqslant 130°$ and (iii) the full 4π emission angle, respectively. All spectra have been fitted for energies above the 9 MeV threshold, and the 4π emission spectrum also for above 12.5 MeV. It is obvious that taking the emission angle into account affects mainly the width of the energy distribution, by up to 1 MeV, while the mean energy changes up to 0.3 MeV only. There is also a slight but significant variation of the spectral parameters with the threshold values chosen.

The spectral parameters analyzed this way are compared with related literature data in Table I. Within rather small experimental errors, our data with the full-angle correction applied compare favorably with data from Refs. [3] and [10] with a fitting threshold of 12.5 MeV and with data from Ref. [4] with the threshold at 9 MeV. On the other hand, the somewhat narrower spectral width obtained in a coincidence experiment by Grachev *et al.* [8] is in good agreement with the present data being corrected for equatorial emission. So, the cited literature data tend to confirm the slight dependence of the spectral shape of ternary α particles on emission angle, which has been analyzed, to our knowledge, for the first time in the present work.

Figure 4 shows a decomposition of the measured ternary α spectrum, corrected for equatorial emission, into the components from true ternary α particles and the about 17% contribution of residual α particles from the decay of ternary ^{5}He, as recently measured by Kopatch *et al.* [4] with a 9 MeV threshold. Two Gaussian curves were fitted to the present data above 9 MeV, taking the relative positions, widths, and intensities from Ref. [4] as constraints in the fitting procedure. It is obvious from Fig. 4, that the ternary α spectrum shows more low-energy α yield than could be explained by the lower-energetic α particles resulting from the decay of ^{5}He,

TABLE I. Spectral parameters of the ternary α spectrum in ^{252}Cf.

Mean energy (MeV)	FWHM (MeV)	Gaussian fit range (MeV)	Angular range for $\theta_{\alpha L}$	Method (detectors)	Reference
15.4± 0.1	10.0± 0.1	9–27	Experiment[a]	TOF-*E* (MCP-silicon)	Present work
15.5± 0.1	10.5± 0.1	9–27	Equatorial[b]		
15.6± 0.1	10.9± 0.2	9–27	Full		
15.7± 0.1	10.6± 0.2	12.5–27	Full		
15.7± 0.2	10.4± 0.2	⩾12.5	Full	ΔE-E (silicon-silicon)	Wagemans *et al.* [3]
15.7± 0.2	10.9± 0.1	8–28	Full	ΔE-E (gas-silicon)	Kopatch *et al.* [4]
15.7± 0.1	10.6	⩾10	Full	TOF-*E* (silicon ball)	Tishchenko *et al.* [10]
15.8± 0.1	10.2± 0.1	8–28	Equatorial	ΔE-E (gas-silicon)	Grachev *et al.* [8]

[a]For the current experimental setup.
[b]Defined as within the range $50^\circ \leqslant \Theta_{\alpha L} \leqslant 130^\circ$.

when Gaussian shapes were assumed for both partial spectra. Apparently, the spectral shape measured previously [3,4] at energies $E > 9$ MeV cannot be extrapolated meaningfully to low energies. For the explanation of the low-energy tailing, another, still quite nebulous, low-energy component was postulated in Ref. [3]. A simpler explanation could be that both the α and ^{5}He energy distributions show about the same asymmetry. This assumption might be corroborated by the apparent asymmetry of the ternary ^{6}He spectrum (see below).

A comparison of our data, corrected for full emission angle, with the spectrum measured in 4π by Tishchenko *et al.* [10] is shown in Fig. 5. There is good agreement between both sets of data concerning both the low-energy part between 2.5 and 9 MeV and the spectral shape above 9 MeV. See also Table I for the spectral parameters.

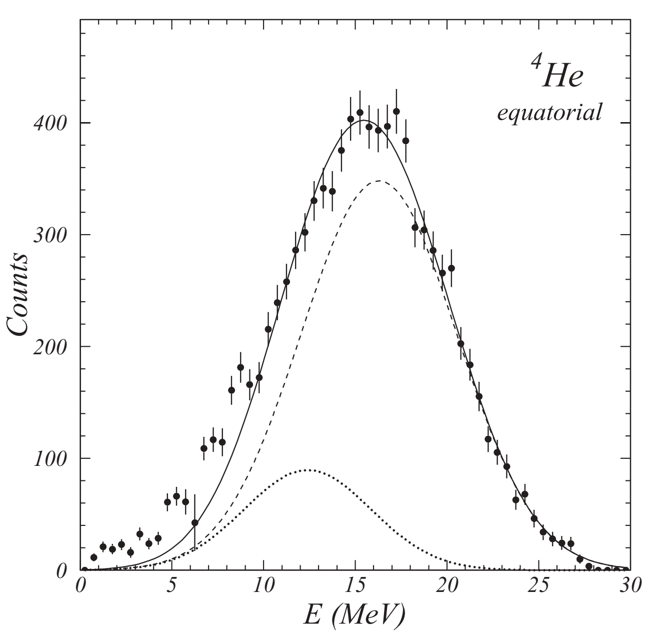

FIG. 4. Energy distribution of ternary α particles from ^{252}Cf fission, corrected for equatorial angles $50^\circ \leqslant \Theta_{\alpha L} \leqslant 130^\circ$. Two Gaussian curves were fitted to the data above 9 MeV, taking true ternary α particles (dashed curve) and residual α particles from ^{5}He decay (dotted curve) into account, according to the results of Kopatch *et al.* [4] (see text).

We have also compared our data with the early work by Loveland [6]. Because in Ref. [6] the ternary α particles were measured in coincident with FFs in 90° geometry, we compare the spectrum published by Loveland with our data corrected for equatorial emission (see Fig. 6). The spectrum from Ref. [6] shows somewhat higher yield both, at the lowest and highest energies. It should be noted that in previous discussions on the subject, e.g., Fig. 3 in Ref. [3], the data from Ref. [6] were erroneously related to experimental spectra measured over the

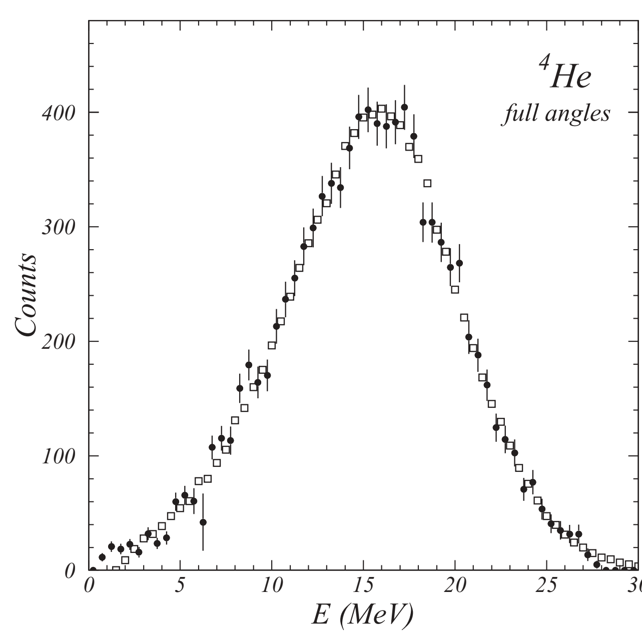

FIG. 5. Energy distribution of ternary α particles from ^{252}Cf fission, corrected for full emission angles, in comparison with data by Tishchenko *et al.* [5] (open squares). The spectra were normalized to equal integral yield.

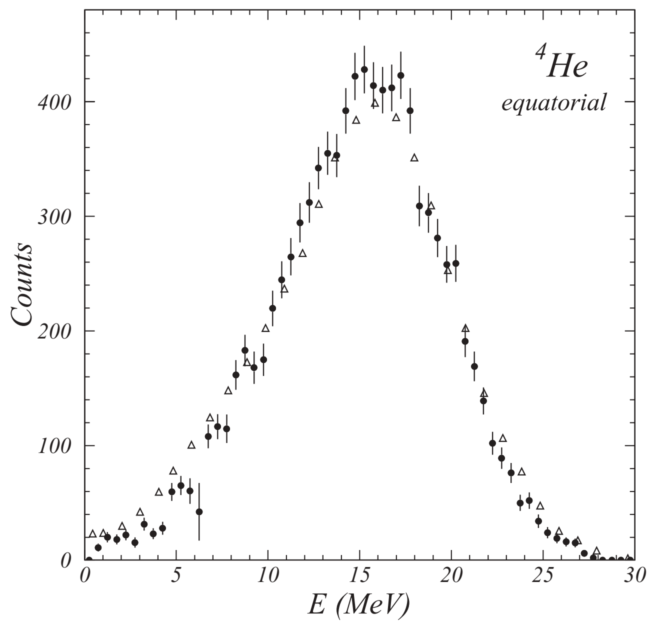

FIG. 6. Energy distribution of ternary α particles from ^{252}Cf fission, corrected for equatorial emission angles, in comparison with data by Loveland [6] (open triangles). The spectra were normalized to equal integral yield.

full emission angle, or without fragment registration. Such a comparison suggests a larger overestimation of the low-energy yield in the Loveland data.

Finally, we have also extracted the energy spectrum of ternary ^{6}He from our data shown in Fig. 2, leaving out the energy region around 6 MeV. The ternary ^{6}He spectrum is shown in Fig. 7, the total number of events collected over the 6-week period of the measurement being 468. This is a rate of about 10 events per day. To our knowledge it is the first time that ternary ^{6}He particles from ^{252}Cf(sf) were measured over their full energy range. Because of the low statistics involved, we have continued on without applying any correction for the emission angle. The spectrum turns out to be asymmetric as well, although some uncertainty remains here on the magnitude of the asymmetry because minor interference with some background in the analysis window cannot be fully excluded.

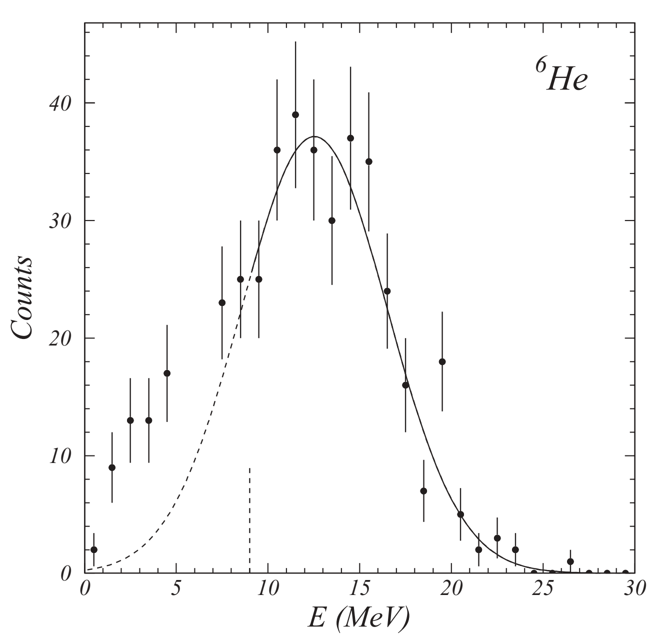

FIG. 7. Energy distribution of ternary ^{6}He particles. The solid line is a Gaussian curve fitted to the data above 9 MeV.

TABLE II. Energy parameters of ternary ^{6}He and the ratio ^{6}He/^{4}He in ^{252}Cf(sf).

Mean energy (MeV)	Width (FWHM) (MeV)	^{6}He/^{4}He ratio	Reference
12.5(5)	9.4(8)	0.041(5)[a] 0.049(3)[b]	This work
12.3(5)	9.0(5)	0.031(2)	Kopatch *et al.* [4]
11.6(2)	10.1(1)	0.040(3)	Dlouhy *et al.* [19]
11.4(3)	10.6(3)	0.039(1)	Grachev *et al.* [8]

[a]From Gaussion fits above 9 MeV threshold.
[b]Above 1 MeV experimental threshold.

Summing up the spectrum shown in Fig. 7, with interpolating the missing values around 6 MeV, and relating it to the sum of α particles shown in Fig. 3, has yielded for the ratio ^{6}He/^{4}He a value of 0.049(3). Fitting the spectrum, for energies above 9 MeV, with a single Gaussian curve yields a value of 12.5(5) MeV for the mean energy and 9.4(8) MeV FWHM for the width. Taking the area under the Gaussian as the estimate for the ^{6}He yield and relating it to the α yield fitted also above 9 MeV gives for the ^{6}He/^{4}He ratio a value of 0.041(5), which is in line with most values deduced earlier from experiments [4,8,19,20] with similar threshold energy, summarized in Table II. The analysis of the spectrum from the rare ^{6}He particles gives us confidence that any interference with background can safely be omitted in the about 25 times more intense ternary α spectrum.

V. SUMMARY

Energy spectra of ternary ^{4}He and ^{6}He particles were measured for the wide energy range from 1 to 27.5 MeV. The low detection threshold of 1 MeV was achieved by using the TOF-E method with unshielded silicon detectors. The data confirm the presence of the disputed low-energy tailing in the ternary α spectrum. For the first time, the ternary ^{6}He was determined to show similar asymmetry.

Several arguments can be invoked to explain, at least qualitatively, the observed asymmetry in the energy spectra of ternary α particles (see, e.g., Reviews [1,2]). As already mentioned above, residual α particles from ternary ^{5}He decay are presumably unable to explain the low-energy tailing quantitatively. There is reason to believe that all ternary He isotopes, including ^{5}He, are born with somewhat asymmetric energy distributions. This point of view might be corroborated by a recent trajectory calculation for ternary ^{4,5,6,7}He emission based on realistic assumptions about the ternary scission configuration [4]. This model associates the spectral shape with the distribution of particle emission points from the nascent fragments' nuclear potential.

It would be interesting to know whether the feature of low-energy tailing is unique to He isotopes or is present also in the spectra of other ternary particles. To answer this question a more sophisticated measurement is needed. For instance, particle separation could be improved by the use of pulse-shape discrimination in the silicon detectors [21]. We intend to explore such options in the future.

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