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Ternary fission of ^{252}Cf: 3368 keV γ radiation from ^{10}Be fragments

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Ternary fission of ^{252}Cf was studied at Gammasphere with eight $\Delta E \times E$ particle telescopes. After Doppler correction, the 3368 keV $2^+ \to 0^+$ transition in ^{10}Be was found in coincidence with the ^{10}Be fragments with no evidence for any γ rays not Doppler shifted for ^{10}Be with >21 MeV kinetic energy. The ratio of the first 2^+ to ground-state population probabilities was estimated as $N(2^+)/N(0^+)=0.160\pm0.025$. The angular distribution for 3368 keV γ rays indicated the spin alignment of excited ^{10}Be nuclei preferentially is parallel or antiparallel to their momentum.

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Nuclear fission accompanied by light charged particle (LCP) emission (often called ternary fission) has been widely studied; see Refs. [1–3] and references cited therein. A major interest to this process is related to the possibility of obtaining additional data about the dynamics of the fission process. Relative emission probabilities measured for various LCPs were used by different authors as an important test for theoretical models destined to clarify the picture of ternary fission. Since the LCP yields depend on a poorly defined quantity, the "energy cost" for the production of various particles [1], uncertainty is inherent to these models. Comparative data giving the yields of LCPs obtained in their ground and low-lying excited states would be more relevant to the case.

Existing data on the population probabilities of excited states in ternary fission LCPs are scarce. Only recently the population of a particle unstable level in ^{8}Li lying at $E^*=2.26$ MeV was reported for the spontaneous ternary fission (STF) of ^{252}Cf [4]. Besides the large statistical error in the yield value reported in this paper, the value of this result is marginal because the population ratios of particle unstable states suffer from ambiguity. The detection of γ rays emitted from excited states of $Z>2$ LCPs would be more pertinent to the case.

Among the heavy $Z>2$ LCPs in STF, ^{10}Be has the largest yield and is suitable for detection of its $2^+ \to 0^+ \gamma$ transition with energy 3368 keV. The observation of γ rays emitted by ^{10}Be in the ^{252}Cf STF was reported in Refs. [5,6], where evidence was also presented that some of the γ rays of ^{10}Be were seen without Doppler broadening. Taking into consideration the lifetime of 125 fs for the 2^+ level in ^{10}Be the authors concluded that either ^{10}Be nuclei are at rest for an unusually long time or the γ rays are emitted predominantly in the orthogonal direction to the ^{10}Be momentum [5,6]. The result was open to question because of the poor energy resolution of the NaI detectors used in this experiment. To test this idea a γ-γ-γ coincidence experiment was made with Gammasphere with a high-energy resolution [7]. The data gave support to that result, but with limited statistics and no direct LCP identification. The possibility that the ^{10}Be nucleus may stay between two fission fragments for a long time ($\sim 10^{-13}$ s) to create a so-called triple nuclear molecule would open up exciting opportunities discussed in Ref. [8].

Thus, the observation of γ rays emitted by LCPs is important both for the deeper understanding into the dynamics of ternary fission and for examining the process for quasimolecular states appearing presumably at the scission point. To seek answers to these problems, we have made a LCPs-γ-γ coincidence study of ^{252}Cf STF. The transitions from the first excited 2^+ state in the ^{10}Be nucleus exhibited Doppler shifts expected for fully accelerated ^{10}Be fragments with $E>21$ MeV. The angular distributions indicate this γ ray is evidently preferentially in parallel or antiparallel with the ^{10}Be momentum.

The experiment was performed at the Lawrence Berkeley National Laboratory by using Gammasphere to detect γ rays and eight $\Delta E \times E$ telescopes to detect LCPs emitted in the ^{252}Cf ternary fission. Gammasphere was set to record γ rays with energy between $\sim$80 keV and $\sim$5.4 MeV. The γ ray detection efficiency varied from a maximum value of $\sim$17% typical for the $\sim$200 keV energy part of the γ ray spectrum to $\sim$4.6% at 3368 keV.

A ^{252}Cf source with $\sim 4\times10^4$ spontaneous fissions per second was placed in a reaction chamber in the center of

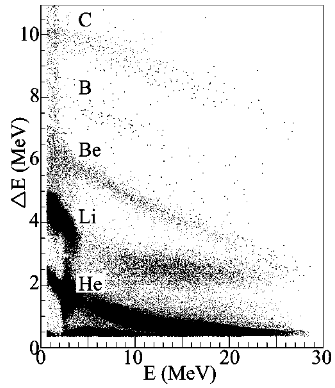

FIG. 1. $\Delta E \times E$ plot. The data were collected by the first telescope during the whole time of the experiment. The ΔE and E scales show just the values of energy deposited by the LCPs in the ΔE and E detectors.

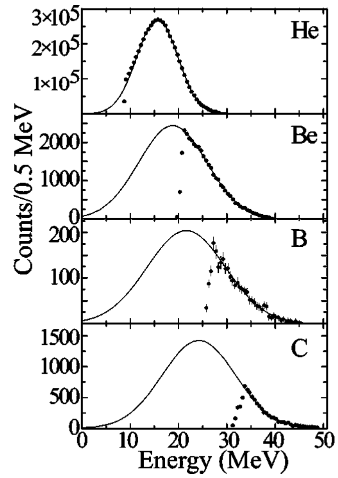

FIG. 2. Energy distributions measured for the He, Be, B, and C LCPs. Closed circles show experimental data corrected for the energy losses in the source foils taking into account the energy losses in the ΔE detectors. Solid curves are Gaussian fits (see text).

Gammasphere. The ^{252}Cf source was deposited as a 5 mm spot on a 1.8μ Ti foil covered on both sides by gold foils. These foils had the least thickness that could stop binary fission fragments. Only the light fragments emitted in binary fission with energy greater than 110 MeV could penetrate the foils.

Eight $\Delta E \times E$ Si detector telescopes were used to measure LCPs. Four telescopes were centered at the polar angle of θ=30° and φ=45°, 135°, 225°, and 315° with the other four telescopes at θ=150° and the same azimuthal angles. The telescopes were inclined so that the lines connecting the centers of two opposite ΔE detectors and going through the center of the source were orthogonal to the detector surfaces. Each ΔE detector had an area of 10×10 mm^2 and a thickness varying between 9.0μ and 10.5μ. Each E detector was 400μ thick and was 20×20 mm^2 in area. All ΔE detectors were at a distance of 27 mm from the source. The distance between the ΔE and E detectors was 13 mm in each telescope. Data acquisition was triggered by ΔE or E signals with amplitudes above the threshold values, which were set to prevent the detection of twofold pileup events of α particles emitted in the decay of ^{252}Cf. A ternary fission event was stored with a condition that at least one γ ray was detected by Gammasphere within the time interval allocated for this event. During a two week experiment $\approx 1.6 \times 10^7$ events were recorded.

The resolution of the $\Delta E \times E$ telescopes allowed us to well identify helium, beryllium, boron, and carbon nuclei in each of the eight two-dimensional plots created from the data recorded by the telescopes. One such plot is shown in Fig. 1. Only the lithium region was clouded due to the random coincidences of helium LCPs with the ^{252}Cf α particles. We separated He, Be, B, and C in Fig. 1 applying an additional condition that the energy deposited in the E detectors was greater than 5 MeV. Energy spectra obtained in this way for these LCPs are shown in Fig. 2. The lower recorded primary energies are 9, 21, 26, and 32 MeV for He, Be, B, and C LCPs, respectively. These thresholds are due to the energy losses in the source foils and in the ΔE detectors.

All ^{4}He LCPs with energy >9 MeV are present in Fig. 2 in the He spectrum. This includes also ^{4}He nuclei originating from the ^{5}He accompanied ternary fission (see Ref. [4]). The incomplete separation of ^{6}He causes a $\leqslant$1% contribution of these LCPs in the ^{4}He spectrum. We neglected this small admixture both in fitting the spectrum with Gaussian and at the yield estimation made for the ^{4}He LCPs. A fit made for the ^{4}He spectrum by a single Gaussian is shown by a solid line in Fig. 2. The parameters $\langle E \rangle = 15.7 \pm 0.2$ MeV and full width at half maximum (FWHM)$=11.0 \pm 0.2$ are essentially the same as in Refs. [4,9]. In the case of Be, B, and C only the high-energy parts of the full energy spectra were measured. Even in the usual assumption that the LCP energy distributions are of Gaussian form, it is impossible to estimate the mean values and widths of the distributions from these data. To obtain yields, we used the mean values from an experiment in Dubna with an open ^{252}Cf source [10]. It allowed us to estimate the FWHM values by fitting the LCP energy distributions obtained with higher statistics in the Berkeley experiment. These fits, shown in Fig. 2, gave the yields of different LCPs as shown in Table I.

The histogram shown in Fig. 3 with a thin line is the energy spectrum of prompt γ rays recorded in coincidence with Be LCPs. The thick solid line in this figure is the same spectrum obtained after applying corrections for Doppler shifts. A distinct peak dominating in Fig. 3 at 3368 keV cor-

TABLE I. Parameters of the energy spectra and yields of LCPs obtained by fitting our experimental data. The yields are normalized to 10^4 He LCPs.

	Be	B	C	
$\langle E \rangle$ (MeV)	18.8(4)	21.6(1.0)	24.2(1.0)	[10]
	17.5(1.0)	21.2(1.0)	26(1)	[9]
FWHM (MeV)	16.3(7)	18.8(1.1)	17.5(9)	Present work
	18(1)	19.3(1.0)		[9]
Yield	165(10)	15.4(3.5)	106(15)	Present work
	126(30)	6.3(4.0)		[9]

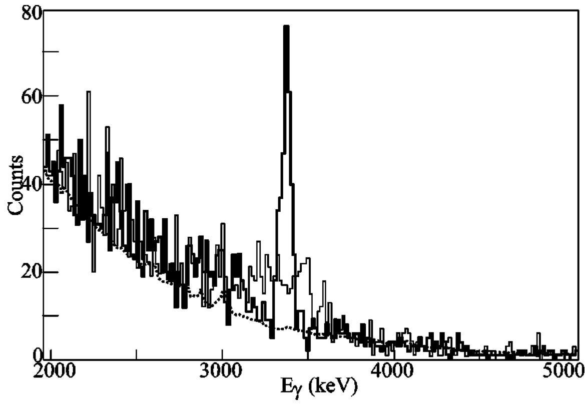

FIG. 3. Histograms drawn with the thin and thick solid lines, respectively, show the spectra of γ rays coinciding with Be LCPs obtained before and after the Doppler shift correction. The dotted line shows the spectrum of γ rays observed in coincidence with He LCP's (see text).

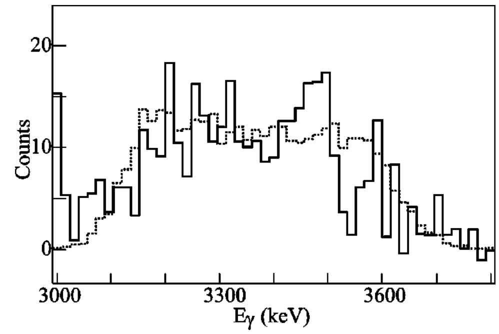

FIG. 4. Histogram drawn with a thick line shows the spectrum of γ rays coinciding with Be LCPs. It is obtained from the thin solid line histogram of Fig. 3 after the subtraction of the background shown by the dotted line in Fig. 3. The thin line histogram demonstrates the spectrum simulated under the assumption of isotropic γ emission from ^{10}Be. The second spectrum was normalized to the number of counts obtained in the first one in the energy range 3000–3800 keV.

responds to the $2^+ \rightarrow 0^+$ transition in ^{10}Be. The peak width, FWHM$=65.7\pm4.5$ keV, was estimated by using a Gaussian fit. This width is compatible with the value FWHM $=62.1$ keV obtained by the Monte Carlo simulation of the correction procedure made for γ rays emitted from moving ^{10}Be nuclei. The simulation has been made taking into consideration the experimental energy distribution of ^{10}Be LCPs, the energy resolution of Gammasphere, and the real three-dimensional positions of Ge detectors and particle telescopes. The dotted line in Fig. 3 shows the spectrum of γ rays detected in coincidence with helium nuclei. Being normalized to the total number of counts obtained in the whole spectrum associated with Be LCPs (E_γ=90–5000 keV) this spectrum reflects to some extent the background from γ rays emitted by fission fragments.

One infers from Fig. 3 that the 3368 keV peak restored after Doppler correction (see the thick line histogram) is smeared in the raw spectrum (thin line histogram) over an energy range extending from about 3000 keV to 3700 keV. Comparison made in Fig. 4 between the spectrum of γ rays coincident with Be LCPs and the spectrum obtained by Monte Carlo simulation gives evidence that the 3368 keV γ rays are mainly emitted by moving ^{10}Be nuclei.

In the raw spectrum of γ rays recorded in coincidence with Be LCPs, we do not see any distinct peak near 3368 keV, which could be associated with γ emission from stationary ^{10}Be nuclei (see Figs. 3 and 4). Obviously, γ rays emitted by the moving ^{10}Be in directions almost orthogonal to the ^{10}Be momentum result in a broad bump centered around the maximum of the 3368 keV peak. To get rid of this bump we picked out only such events where the angles between the trajectories of γ rays and ^{10}Be nuclei were less than 45° or greater than 135°. The resulting spectrum obtained after the background subtraction is shown in Fig. 5. The energy region around 3368 keV now has a clear zero. In other words, there is the absence of any γ line which could be attributed to the emission from stationary ^{10}Be with $E(^{10}\mathrm{Be}) > 21$ MeV bound in the potential well of two fragments.

The number of counts in the 3368 keV γ line is 263 ± 22 (see the thick solid line histogram in Fig. 3). Taking into consideration the peak efficiency ($\simeq 4.6\%$), the total number of 3368 keV γ rays emitted by ^{10}Be nuclei is $I_\gamma = 5700\pm480$. After correction for the number of beryllium LCPs escaping detection because of the lack of recorded signals from γ rays, we estimated the total number of Be LCPs recorded by the $\Delta E \times E$ telescopes as $N_{\mathrm{Be}}=51800\pm5000$. The energy resolution of the ΔE detectors did not allow us to directly separate the loci of beryllium isotopes in the $\Delta E \times E$ plot (see Fig. 1). Comparing the loci obtained by Monte Carlo simulations for ^{9}Be, ^{10}Be, and ^{11}Be with the experimental $\Delta E \times E$ distributions we estimated the yield of ^{10}Be nuclei as ~80% of all beryllium LCPs emitted in the ternary fission of ^{252}Cf. This observation is in agreement with the experimental data presented in Ref. [11].

Based on these results, we estimated the population ratio of the excited-state 2^+ and the ground-state 0^+ levels in ^{10}Be as $N(2^+)/N(0^+)=0.160\pm0.025$. The upper limit for the probability that ^{10}Be emits its γ rays at rest makes only 3% of this ratio. We note, however, that these results are valid only for ^{10}Be LCPs obtained at infinity with kinetic energies greater than 21 MeV. In particular, one cannot exclude that the hypothetical triple nuclear molecule decay mainly by low kinetic energy ^{10}Be LCPs which were beyond the reach of

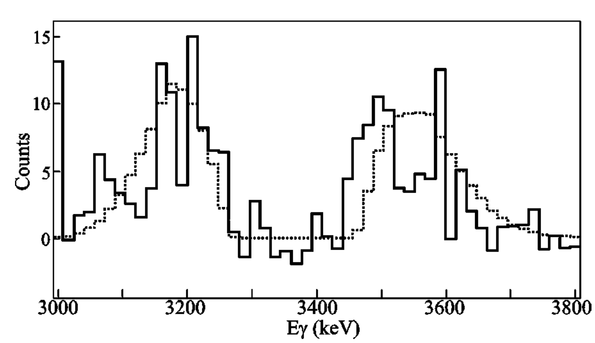

FIG. 5. Solid line histogram shows the spectrum of γ rays detected at angles $<45°$ or $>135°$ to the Be momentum. The dotted line demonstrates the spectrum simulated with the same angular limitations, assuming isotropic γ ray emission.

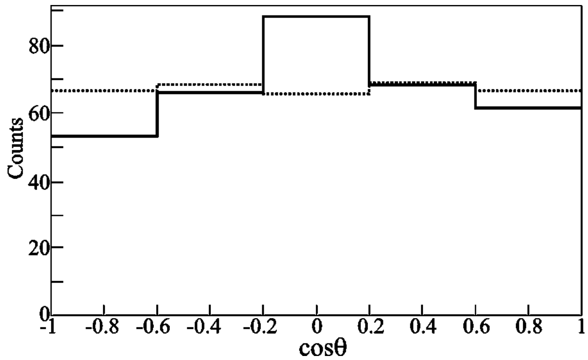

FIG. 6. Angular distribution measured in the experiment for 3368 keV γ rays emitted by ^{10}Be LCPs (solid line). θ is the angle between the ^{10}Be momentum direction and the direction of γ ray emission. Dashed line demonstrates the simulation made with the assumption of isotropic γ ray emission from moving ^{10}Be nuclei.

the present experiment. Since the earlier Gammasphere experiment [7] relied solely on the observation of γ-γ-γ coincidence events, a possibility still remains open that a narrow γ peak characteristic of the stationary ^{10}Be can be found if the low energy part of the energy distribution of ^{10}Be LCPs is also detected. At the same time, the present result excludes any possibility that an effect of triple quasi-molecular state, involving beryllium LCPs, was observed in Refs. [5,6] where the energy cutoff was 26 MeV for these clusters.

Selecting events with 3368 keV γ rays emitted from ^{10}Be we obtained the angular distribution shown in Fig. 6. A clear anisotropy of the γ ray emission probability with respect to the ^{10}Be momentum direction is evident.

In summary, He, Be, B, and C LCPs emitted in the ^{252}Cf ternary fission with kinetic energy more than 9, 21, 26, and 32 MeV, respectively, were identified and their relative yields and energy distribution parameters could be estimated. The extraordinary capability of Gammasphere in probing rare γ rays combined with $\Delta E \times E$ detector telescopes allowed us to measure for the first time the ratio of population probabilities, $N(2^+)/N(0^+)=0.160\pm0.025$, for ^{10}Be nuclei emitted in their excited (2^+) and ground (0^+) states. Assuming that a kind of statistical equilibrium is set at some stage in the ternary fission process (e.g., as discussed in the dynamical model [11]) and taking into account the statistical spin factor, this ratio suggests a value of 1.0 ± 0.1 MeV for the temperature parameter. Our data do not show any hint that these are γ rays emitted from stationary ^{10}Be nuclei as part of a triple nuclear molecule presumably formed at the scission point when the ^{10}Be energy is greater than 21 MeV. The angular distribution obtained for the first time for the 3368 keV γ rays gives a clear indication for the spin alignment of excited ^{10}Be(2^+) nuclei preferentially parallel (or antiparallel) to their momentum direction.

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