Angular Anisotropy of Prompt γ Rays and Fragment Spin Alignment in Binary and Light-Charged-Particle-Accompanied Spontaneous Fission of ²⁵²Cf

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Angular correlations of prompt γ rays in binary and light-charged-particle (LCP)-accompanied spontaneous fission of ²⁵²Cf were measured with a 4π -crystal-ball NaI(Tl) γ -ray spectrometer. For ³H, ⁴He, ⁶He, Li, and Be accompanied fission, γ -ray angular distributions were deduced with respect to the motion both of the fragments and the LCPs. There is no indication for a change of the fragment spin alignment by the LCP release or for a correlation between the LCP direction and a bending motion at scission, as was argued in previous investigations. Consequences for the understanding of fragment spin formation are briefly discussed. [S0031-9007(98)08179-4]

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Fragments from nuclear fission are formed with rather high spins ($6\hbar$ to $8\hbar$, on average) which is attributed to a collective vibrational motion at scission (so-called bending or wriggling modes) [1-3], and to the Coulomb excitation (quadrupole torque interaction) in the early separation phase of the fragments [4,5]. However, recent data on fragment spin populations in cold (neutronless) fission from modern fine-grained γ -ray spectroscopy seem not to be consistent with theoretical calculations [6]. Generally, fragment spin vectors are mainly oriented perpendicular to the fission direction (i.e., aligned), in accordance both with the bending mode assumption and with postscission Coulomb excitation. This alignment of fragment spins causes the emission of the yrast γ rays to proceed anisotropically with respect to the fission axis, although fragment spin distributions are significantly influenced by neutron and statistical γ -ray emission [7,8].

Experimental access to intimate properties of the scissioning nucleus is also acquired through the rare fission mode of light-charged-particle (LCP) accompanied fission $(\approx 1/260$ of the binary fission yield, for ²⁵²Cf) [9]. It is, thus, interesting to study whether the emission of LCPs at scission influences the fragment spins and their alignment by comparing γ -ray angular correlations in ternary and binary fission, respectively. Only two experiments on γ -ray angular correlations in ternary fission were hitherto reported. They concern the angular distribution of γ -ray intensity in ²⁵²Cf spontaneous fission (sf) with respect to the fission direction, measured in coincidence with the relatively frequent ternary α particles (about 90%) of the ternary yield). Compared to binary fission, γ -ray anisotropy was found to be either removed [10] or characteristically changed [11] by the ternary α -particle release, suggesting a drastic change in the alignment of the fragment spins. On the other hand, it was argued in [12] that the results of [11] might be a signature of a mutual relation between the mechanism of ternary-particle emission and the excitation of collective modes during the saddleto-scission stage. In the present paper we present an extended study on the γ -ray angular correlation in ternary fission of 252 Cf. In addition to ternary α particles, emission of some much rarer LCP species, viz. ³H, ⁶He, Li, and Be particles, was studied, and the angular dependence of the γ -ray intensity was not only determined with respect to the direction of fragment motion, but also with respect to the emission direction of the LCPs.

In the experiment [13–15], the Darmstadt-Heidelberg crystal ball (CB) [16] serving as a homogeneous 4π γ -ray spectrometer was combined with an efficient detection system for fission fragments and ternary particles. The CB is a dense spherical package of 162 large (20 cm long) NaI(Tl) crystals of high γ -ray detection efficiency (>90%). The γ rays are separated by time-of-flight from the prompt fission neutrons which are simultaneously registered with a rather high efficiency of about 60%. The 252 Cf sample (4 × 10³ fissions/s, thin backing) and the assembly of detectors for measuring fragments and LCPs were mounted inside a CH₄-filled aluminum vessel positioned at the center of the CB. This kinematic spectrometer, described in detail elsewhere [13,17], consists of a Frisch-gridded 4π twin ionization chamber (IC) for measuring fission-fragment energies and emission angles, and of a ring of twelve ΔE -E telescopes (made from ΔE IC's and silicon *p*-*i*-*n* diodes, with a solid angle of $\pi/4$) surrounding one-half of the fragment IC for measuring the LCPs. Angular resolutions (FWHM) were $\leq 5^{\circ}$ both for the fragments and the LCPs. In four weeks of measurements we have registered, in coincidence with γ rays (and neutrons), 1.2×10^6 ternary fission events accompanied by α particles, 8×10^4 events with ³H, 2.5×10^4 events with ⁶He, and 2.5×10^3 events with the emission of Li and Be each. For LCPs heavier than the He isotopes,

experimental limitations were the relatively high energy thresholds (e.g., 17 MeV for Li, and 24 MeV for Be particles) and lack of isotopic separation. Simultaneously, 7×10^7 binary fission events were recorded, the data acquisition rate being reduced by a factor of 1/64.

For avoiding in the coincidence time spectra any contamination in the prompt γ peak by delayed pulses due to neutrons, which were measured to yield an about 20 times stronger anisotropy than the γ rays, a rather narrow time window of ± 2 ns was set onto the prompt γ -ray coincidence peak having a width of 3.5 ns FWHM. Energy calibration of the γ -ray pulse-height spectra was performed with various γ -ray sources, following standard procedures. Because of the dominance of full energy absorption in the large crystals the measured γ -ray spectra were not unfolded for the detector response. All crystals of the CB subtend equal solid angles ($\pi/40$, each), but relative γ -ray efficiencies varied up to $\approx 20\%$ due to absorption in the charged-particle detectors mounted inside. The corresponding corrections were derived from the measured ${}^{252}Cf \gamma$ -ray spectra. Furthermore, an angular resolution correction accounting for the granularity of the CB was determined from a Monte Carlo simulation (using the GEANT code), and the various γ -ray angular distributions presented below were unfolded accordingly.



FIG. 1. Angular distributions $W'(\Theta)$ of γ rays with respect to the fragment motion for binary (left side) and α -accompanied (right side) fission of ²⁵²Cf, for different γ -ray energy intervals. The scale is correct for the lowest plots; others are shifted consecutively by 0.1 units. Solid lines are fitted curves using Eq. (1). Error bars represent statistical errors.

Prompt fission γ rays originate from a large variety of correlated pairs of fragments, the light and heavy ones moving in opposite direction with different velocities v. Therefore the measured γ -ray angular correlations are subject to the corresponding solid-angle aberrations and Doppler shifts in γ -ray energy. Corrections for these effects in first-order of v/c, justified by the low fragment velocities ($v \leq 0.05c$) and the weak anisotropy, are accomplished by averaging the γ -ray intensities $W(\Theta)$ as measured per unit solid angle at an angle Θ with respect to one fragment group and that at the respective complementary angle $\pi - \Theta$ (see [18]). Resulting symmetrized angular distributions $W'(\Theta)$ which represent the average angular correlation functions of the γ rays from the two sources in their respective rest frames are displayed in Fig. 1 for different γ -energy intervals, and for binary and α -particle accompanied fission, respectively. Solid lines are the results of least-square fits to the data assuming the multipolarity of the γ rays to be ≤ 2 , according to

$$W'(\Theta) = 1 + A_2 P_2(\cos \Theta) + A_4 P_4(\cos \Theta) \quad (1)$$

with $P_i(\cos \Theta)$ being the Legendre polynomials [7,8]. Figure 2 displays the corresponding anisotropy ratios A, defined as $A = W'(0^\circ)/W'(90^\circ) - 1$, as a function of E_γ . For binary fission, there is close agreement with previous work [8], in which the characteristic shape of $A(E_\gamma)$ was explained by the dominance of dipole over quadrupole transitions towards low energies. For α -accompanied fission, our data on $W'(\Theta)$ (Fig. 1) and $A(E_\gamma)$ (Fig. 2) do not reveal any significant difference compared to binary fission, in contrast to both former studies [10,11].

Averaged values for the anisotropy ratio A and for the angular correlation parameters A_2 and A_4 (0.1 MeV $\leq E_{\gamma} \leq 1.5$ MeV) deduced by least-square fits according to Eq. (1) are given in Table I. Our A values in binary and α -accompanied fission are in close agreement with A = 0.092(5) as obtained by [8] for binary fission, but the present result on α -accompanied fission is in obvious contradiction to the much smaller numbers published previously. We also note that, in the present experiment, we



FIG. 2. Anisotropy ratio A in binary (left side) and α -accompanied fission (right side) of ²⁵²Cf, as a function of γ -ray energy. The present results (full symbols) are compared with binary fission data from [8] (open circles).

TABLE I. Anisotropy ratio A and angular correlation parameters A_2 and A_4 for binary fission of ²⁵²Cf and ternary fission with emission of various LCPs. The γ -energy interval is from 0.1 to 1.5 MeV.

Fission mode	Anisotropy parameters		
	Α	A_2	A_4
Binary	0.0872(3) 0.092(5) ^a 0.127(3) ^b	0.0683(1)	-0.0308(2)
α -accompanied	0.0903(19) 0.015(22) ^b 0.03(2) ^c	0.0684(9)	-0.0261(12)
Other LCPs			
³ H	0.082(9)	0.062(4)	-0.023(6)
⁶ He	0.089(13)	0.068(6)	-0.027(8)
Li	0.02(4)	0.03(2)	-0.04(3)
Be	0.17(5)	0.12(2)	-0.03(3)

^aRef. [8]; ^bRef. [10]; ^cRef. [11]

have observed the γ -ray multiplicity M_{γ} in the ternary (ter) case to be lower by about 10% compared to binary (bin) fission [14], in agreement with [19] but again in conflict with [10] reporting $M_{\gamma}(\text{ter})/M_{\gamma}(\text{bin}) = 0.995(12)$. Our data in Table I reveal a slight (about 3%) increase in the *A* value for α -accompanied fission compared to the binary case, and, related to that, a somewhat lower A_4 coefficient. Such a small difference would not be surprising since the properties of fission fragments, such as the fragment mass distribution, the total excitation energy (TXE), the average number of evaporated neutrons $\overline{\nu}$, and also the proton even-odd effect in the isotopic composition of the fragments [20] slightly change upon emission of ternary α particles [9,13].

In ternary fission, all LCPs (except a few so-called "polar" particles [9]) are focused into a narrow cone of angles approximately orthogonal to the direction of fragment motion. We made use of this sharp angular correlation between LCPs and fragments in the second part of our analysis by sorting the measured γ -ray emission angles into a properly chosen coordinate system xvz which allows us to deduce γ -ray angular correlations both with respect to the direction of fragment motion and the emission direction of the LCPs. In our notation, the z axis was arbitrarily chosen as the direction of fission fragments from the light mass group. The x axis is in the plane made up by the three charged reaction products (the fragment pair and the LCP) and is close to the direction of the LCP which is emitted at the mean angle $\Theta_{\alpha L} = 83^{\circ}$ (width $\approx 20^{\circ}$ FWHM) with respect to the light fragment motion [9,17]. The y axis is accordingly perpendicular to that plane making a right-handed coordinate system xyz.

For the case of α -accompanied fission and for γ -ray energies between 0.1 and 1.5 MeV, Fig. 3 displays the projections $w(\Phi_{ij})$ of the LF-LCP- γ -angular correlation on the planes (i, j) = (z, x), (z, y), (x, y), respectively, with Φ_{ij} being the respective polar angle in the plane (i, j). The projections on the zx and zy planes

containing the fragment momenta reflect the characteristic angular distribution of γ rays peaked at the directions of the fragments. The projection onto the *xy* plane, i.e., perpendicular to the fission axis, reveals an isotropic distribution of γ -ray emission with respect to the α -particle direction ($\Phi_{xy} = 0^\circ$), within rather small experimental errors. As a consequence, there is no evidence for a significant correlation between the γ -ray emission angles and the direction of motion of the ternary α particles.

Because of the high efficiency of our setup we were able to determine, for the first time, also the γ -ray angular correlations for ternary fission events accompanied by other LCPs such as ³H, ⁶He, Li, and Be particles. The resulting spectra $W'(\Theta)$ are summarized for all measured particles on the left-hand side in Fig. 4, while the right-hand side of the figure depicts the corresponding projections $w(\Phi_{xy})$ on the xy plane perpendicular to the fission axis. The results deduced for the anisotropy ratio A and for the angular correlation parameters A_2 and A_4 of $W'(\Theta)$ are included in Table I. The γ -ray angular distributions for ³H and ⁶He accompanied fission agree very well with that from binary fission. The emission



FIG. 3. Projections of the measured γ -ray angular correlation in α -accompanied fission on different planes in the coordinate system *xyz* built-up by the momenta of the two fragments and the ternary particle (see text). The emission directions of the ternary α particle and the light fission fragment are indicated by arrows.



FIG. 4. Left side: Angular distributions $W'(\Theta)$ of γ rays (0.1–1.5 MeV) for ternary fission events accompanied by ³H, ⁴He, ⁶He, Li and Be particles. Solid lines are fitted curves using Eq. (1). The dashed line corresponds to the $W'(\Theta)$ distribution for binary fission. Right side: Projections $w(\Phi_{xy})$ of the γ -ray angular correlations on the xy plane. The scales are correct for the lowest plots; others are shifted consecutively by 0.2 units.

of these particles shows similar to ⁴He no effect on the γ -ray anisotropy. However, some deviations might be observed in coincidence with Li and Be particles, but the low counting statistics involved does not permit definite conclusions.

In summary, we have performed an experimental study with high statistical accuracy on the γ -ray angular correlations in the binary and ternary fission of ²⁵²Cf, using a homogeneous setup. No significant differences in the anisotropy of γ -ray emission relative to the fission axis were observed between binary fission and various ternary fission modes. This means that the emission of ternary particles at scission does not influence (or even destroy) the alignment of fragment spins, in contradiction to what has been concluded from previous work [10,11]. In ternary fission, the γ -ray angular distribution with respect to LCP motion, investigated for the first time, reveals no correlation between the directions of the γ rays and the LCPs. The present study was thus able to clear up the experimental situation by demonstrating that the alignment of the fragment spins is not influenced by the ternary particle release at scission. Neither a previously suggested picture in terms of coupling of angular momentum between the LCP and the fragment spin [11] nor that of a correlation between the LCP emission direction and a collective bending motion at scission [12] could be confirmed. The simplest explanation for the present results could be that fragment spin formation is dominated by post-scission Coulomb excitation, i.e., after the instant of LCP release, rather than by a collective vibrational motion at scission. On the other hand, our present knowledge of the scission configuration in ternary fission is still incomplete, and the magnitude of a possible coupling of the ternary particle momentum and some collective motion of the scissioning nucleus is difficult to determine quantitatively. Evidently, the formation of fragment spins in fission is still a challenge to theory (see, e.g., [6]), and even more sophisticated experimental studies might be needed for gaining further insight into the intimate details of the nuclear dynamics at scission.

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