Correlated spins of complementary fragment pairs in the spontaneous fission of ^{252}Cf

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A study of the γ-ray decay of low-lying excited states in fragments produced in the spontaneous fission of ^{252}Cf has revealed a significant correlation between the angles of emission of the $2_1^+ \rightarrow 0_1^+$ transitions of complementary fragment pairs. Calculations of the amount of dealignment that is needed to reproduce the measured a_2 values, and a comparison with the results of previous fragment-γ angular distribution measurements, suggests that at scission there may be significant population of $m \neq 0$ substates associated with the projection of the fragment spin vector on the fission axis. Fragments from the spontaneous fission of ^{248}Cm emit $2_1^+ \rightarrow 0_1^+$ γ rays that show markedly reduced interfragment correlations, suggesting that either a larger role is played by the relative angular momentum of the fragments, or that the dealignment introduced by the neutron emission and statistical γ decay to the 2_1^+ state is larger in ^{248}Cm than ^{252}Cf fission.
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Spontaneous fission is a reaction mechanism that produces fragments with a mean spin of $6-8\,\hbar$. This internal angular momentum has its origin in dynamical processes that occur in the fissioning system, the so-called bending and wriggling modes [1–3], as well as in Coulomb torque immediately following scission [4]. Investigations of the properties of fission-fragment angular momentum provide one of the few means open to the experimentalist to explore the behavior of the fissioning system near the point of scission, providing a particularly crucial test in the case of spontaneous fission where the initial angular momentum of the system is well defined. For example, the spontaneous fission of an even-even nucleus results in a binary system whose intrinsic and relative angular momenta must sum to give zero. For many years it has been known that the spin of a fragment from spontaneous fission tends to be aligned in a plane perpendicular to the fragment direction, suggesting that twisting modes about the fission axis play at most a minor role in the generation of fission-fragment spins. This conclusion has been drawn from measurements of the angular correlation between the fission axis and the direction of γ rays emitted in the prompt decay of the excited fragment. Such experiments have been performed with NaI detectors [5], and also with germanium detectors, whose superior energy resolution allows fragment-γ correlations to be measured for particular γ-ray transitions from low-lying states [6]. Recent measurements of fragment-γ correlations for spontaneous fission of ^{252}Cf accompanied by light-charged-particle emission [7] have shown that the particle emission does not influence the fragment spin alignment.

In this paper we present, for the first time, measurements of angular correlations between γ rays emitted from one fragment with γ rays from the complementary fragment, for decays from low-lying excited states. We discuss the relationship between such correlations and the alignment of the spin vectors of the complementary fragments at scission.

A 120μCi ^{252}Cf source, sandwiched between two $20\ \mathrm{mg\,cm^{-2}}$ Gd foils [8], was used as a source of neutron-rich fission fragments, whose γ-ray decays were detected in the Euroball array [9] of germanium detectors. Over the course of 21 days, 9.7×10^9 events in which three or more γ-rays were detected were written to magnetic tape. These events were then sorted off-line into a hypercube [10] containing 2×10^{10} triples, for the purpose of angular correlation measurements. The first axis of the hypercube represented the energy of a γ ray (γ_0) detected at any angle. The second and third axes represented the energies of γ rays (γ_1,γ_2) detected in coincidence with γ_0. The cosine of the angle between the detection axes of γ_1 and γ_2 ($\cos\theta$) was recorded using the fourth hypercube axis in twenty bins covering the range $-1<\cos\theta<1$. Gates were applied to the first axis (isotropic) to help select the decay sequences of interest, and to the second and third axes to select pairs of γ rays that defined the angular correlations. The resulting projections of

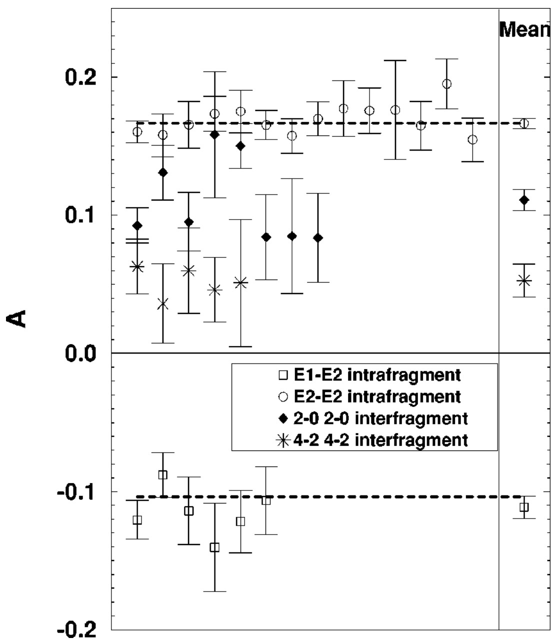

FIG. 1. Measured γ-ray anisotropies from ^{252}Cf fission. The dashed lines show theoretical values of the anisotropy for the intrafragment stretched $E2$-$E2$ and $E2$-$E1$ correlations.

the gated data on the fourth axis represented the angular correlation functions sampled at twenty points and were fitted using the method of least squares to the functional form

$$W(\theta)=a_0[1+a_2P_2(\cos\theta)+a_4P_4(\cos\theta)]. \qquad (1)$$

In each case, the anisotropy, A, could then be calculated from the deduced $W(\theta)$ as

$$A=\frac{W(180°)}{W(90°)}-1.$$

The relative detection efficiency of each angle bin on the fourth axis of the hypercube was calibrated using the eight strongest known $E2$-$E2$ correlations, namely, the $4_1\to 2_1$, $2_1\to 0_1$ pairs of γ rays in ^{144}Ba, ^{146,148}Ce, ^{100,102}Zr, ^{104}Mo, and the $6_1\to 4_1$, $4_1\to 2_1$ pairs in ^{144}Ba and ^{148}Ce. These are the first eight intrafragment $E2$-$E2$ data points shown in Fig. 1. The anisotropies of intrafragment $E2$-$E2$ transitions show no dependence on the lifetime of the intermediate state, over a wide range, from 0.70 ns [11] for the 2^+ state in ^{144}Ba to 65 ps for the 6^+ state in ^{150}Ce [12], hence ruling out spin-lattice relaxation as a major factor in dealigning the fragment-fragment spin correlation. Other $E2$-$E2$ and $E1$-$E2$ transitions were fitted as a test of the determined efficiencies; these are shown in Fig. 1 as the last six data points on the $E2$-$E2$ intrafragment plot and all the points on the $E1$-$E2$ plot, respectively. Note that the large negative anisotropies of the $E1$-$E2$ coincidences provide a good test of the reliability of the efficiencies. Interfragment γ-ray angular correlations were measured by setting an isotropic gate on the decay directly preceding γ_1 in the heavy fragment, to improve selectivity. Heavy fragment γ rays were used for this isotropic gate as they tended to produce cleaner spectra than gates on γ rays in light fragments. For a range of complementary fragment pairs, the interfragment correlation was measured between the $2_1\to 0_1$ γ ray in the heavy fragment and the $2_1\to 0_1$ γ ray in the light fragment. Measurements were also made of the interfragment angular correlations between $4_1\to 2_1$ γ rays. For complementary even-even fragments, the weighted mean interfragment anisotropy, for $2\to 0, 2\to 0$ coincidences, was found to be $A=0.101(7)$ with weighted mean values $a_2=0.057(4)$ and $a_4=0.021(5)$. The corresponding result for $4\to 2, 4\to 2$ interfragment coincidences was $A=0.05(1)$ with weighted mean values $a_2=0.035(5)$ and $a_4=-0.003(8)$.

Data from a previous experiment using a ^{248}Cm fission source [13] were reanalyzed with a method analogous to that used for ^{252}Cf. Angular correlations were again fitted using Eq. (1) and anisotropies extracted. The weighted mean interfragment γ-ray anisotropy for $2\to 0, 2\to 0$ coincidences was found to be $A=0.016(4)$ with weighted mean coefficients $a_2=0.014(2)$ and $a_4=-0.009(3)$.

Interpretation of the above results requires not only consideration of the mechanisms that produce the internal angular momenta, and whether these necessarily produce correlated spin vectors, but also of the dealignment that occurs in the decay of the fragments. Following scission, the primary fragments decay by neutron evaporation and statistical γ-ray emission to yrast and near-yrast states, processes which result in a loss of alignment, as has been shown to occur in heavy-ion evaporation reactions [14]. The alignment lost in the deexcitation process has previously been calculated using statistical models which can reproduce the measured fragment-γ anisotropies [5,6], but require assumptions to be made regarding the nuclear level densities, the probability distribution of angular momentum in the initial hot fragment, and the number of statistical γ rays that are emitted.

As a means of parametrizing the degree of alignment in heavy-ion fusion-evaporation reactions, Yamazaki [15] introduced a procedure whereby the fully aligned ($m=0$) substate population is distributed over other m substates according to a Gaussian distribution of width σ. In this procedure, measurements of a_2 and a_4 coefficients for angular distributions of γ rays of given multipolarity can be translated into values of σ. Here we adopt a similar procedure for the $\gamma\gamma$ angular correlations between complementary fragment pairs. In the case of two $2\to 0$ decays, the situation is illustrated schematically in Fig. 2. The z axis is defined as the detection axis of γ_1, γ_2 being detected at an angle θ with respect to γ_1. This definition of the z axis, together with helicity arguments, means that the only possible m substates of the $I=2$ spin vector in fragment 1 are $m_1=\pm 1$. If we assume that the spin vectors of fragment 1 and fragment 2 are fully aligned, this implies that the m substate distribution for fragment 2, at $I=2$, has equal populations of $m_2=\pm 1$. Such a fully aligned case would give values of a_2 and a_4, equivalent to those found for the sequence $0\xrightarrow{E2}2\xrightarrow{E2}0$, i.e., 0.357 and 1.143, respectively. We assume that the combined statistical decay and neutron evaporation that occurs in both fragments

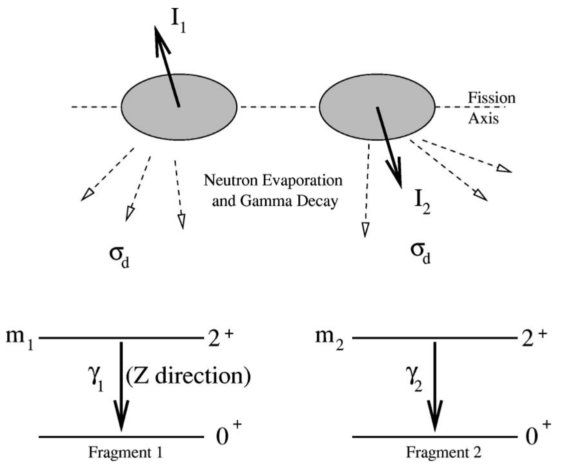

FIG. 2. A schematic illustration of the decay of the excited fission fragments. A possible alignment of fragment spins, such as might occur in a bending mode is indicated, though the spins are not necessarily aligned in a plane perpendicular to the fission axis. The detection of the γ ray from fragment 1 defines the z axis for the system, ensuring that $m_1 = \pm 1$ only. The decay paths in both fragments contribute to the dealignment of I_1 and I_2. A smearing of m substates characterized by σ_d is attributed to the neutron emission and γ decay in each fragment.

introduces a width (σ), which determines the population distribution of m_2 substates, $P(m_2)$, according to the formula,

$$P(m_2) = \sum_{m=-I}^{I} P_a(m) \exp\left(\frac{-(m_2-m)^2}{2\sigma^2}\right) + \sum_{n=0}^{\infty} \sum_{m=-I}^{I} \times P_a(m) \exp\left(\frac{-[m_2+m+2I(1+2n)]^2}{2\sigma^2}\right) + \sum_{n=0}^{\infty} \sum_{m=-I}^{I} \times P_a(m) \exp\left(\frac{-[2I(1+2n)-m_2-m]^2}{2\sigma^2}\right),$$

where $P_a(m)$ is the fully aligned substate distribution and I is the spin of the decaying level. The second and third terms on the right-hand side of the above expression are included to account for ''reflections'' of the Gaussian smearing distribution at $m = \pm I$, with the integer n counting the number of complete cycles in m space. The new population distribution $P(m_2)$ can be easily translated into statistical tensor coefficients and thereby into theoretical values of a_2 and a_4 [16]. An analogous procedure can be used to generate theoretical fragment-γ angular distributions (for a given σ) starting with the fully aligned state in which the population is entirely $m=0$.

Figure 3 shows the effect of varying σ on the a_2 coefficient for interfragment $\gamma\gamma$ correlations, assuming two quadrupole $2 \rightarrow 0$ transitions, as well as for fragment-γ distributions with a quadrupole $2 \rightarrow 0$ γ decay. The measured value of $a_2 = 0.057(5)$ for the interfragment $\gamma\gamma$ correlations in

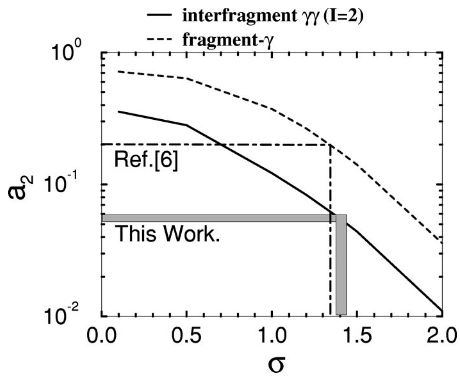

FIG. 3. Results of calculations of m substate smearing and its effect on the a_2 coefficient for fragment-γ angular distributions, as well as γ-γ correlations between the $2 \rightarrow 0$ decays of complementary fragment pairs.

^{252}Cf indicates a statistical width $\sigma_{\gamma\gamma} = 1.40(5)$. The magnitude of the corresponding theoretical $a_4(=-0.01)$ is very much attenuated at this value of sigma. The fragment-γ distributions of Wilhelmy *et al.* [6], for $2 \xrightarrow{E2} 0$ decays in ^{100,102}Zr, ^{104,106}Mo, ^{110}Ru, ^{144}Ba, and ^{148}Ce, produced in the spontaneous fission of ^{252}Cf, have a mean value of $a_2 = 0.20(4)$. As seen in Fig. 3, this translates into a statistical substate smearing with $\sigma_{f\gamma} = 1.35(5)$. The similarity in the smearing widths for interfragment $\gamma\gamma$ correlations and fragment-γ distributions in ^{252}Cf fission is somewhat surprising given that the $\gamma\gamma$ correlations suffer from substate smearing from two independent statistical processes, the deexcitation to the 2^+ states in two fragments, whereas the fragment-γ distributions suffer attenuation due to the statistical decay in one fragment only. This suggests that there is an additional statistical process which contributes to the substate smearing in fragment-γ distributions, but does not affect the interfragment $\gamma\gamma$ correlations. If we simply ascribe a common width to the statistical decay in each fragment, σ_d, and allow for no other contribution to the dealignment of the spin vectors, then, by adding the widths in quadrature, as is appropriate for independent statistical processes,

$$\sigma_{\gamma\gamma}^2 = 2\sigma_d^2.$$

If we further suppose that the fragment-γ distributions suffer from dealignment due to $m \neq 0$ components in the spin distribution at scission and attribute a dealignment parameter σ_{sc} to this effect, we obtain

$$\sigma_{f\gamma}^2 = \sigma_{sc}^2 + \sigma_d^2,$$

allowing us to extract the values $\sigma_{sc} = 0.92(8)$ and $\sigma_d = 0.99(4)$. The assertion that the statistical decays in one fragment are completely independent of those in the complementary is not strictly correct since, for a given split, the number of evaporated neutrons is a constant. Thus more neutrons out of one fragment and larger statistical smearing will imply fewer evaporated neutrons and less statistical smearing in the complementary. The detailed inclusion of this correlation may serve to increase the deduced value of σ_d slightly, but is unlikely to be a large effect since once the fragments are separated, the choice of decay paths within each fragment is made independently.

It is interesting to compare this value of σ_d with those required to produce the attenuation observed in γ-ray angular distributions for $2^+ \rightarrow 0^+$ decays following $(\alpha,2n)$ reactions. Such reactions tend to populate states of similar spin and excitation energy to those observed in spontaneous fission and might therefore be expected to have values of σ_d similar to that deduced here. Angular distribution measurements $2^+ \rightarrow 0^+$ decays in ^{92,94,96}Mo and ^{96,98,100,103}Ru [17] show a mean value of $a_2 = 0.30(7)$, corresponding to $\sigma_d = 1.1(2)$. Thus the attenuation in the $(\alpha,2n)$ reactions is not inconsistent with that deduced from spontaneous fission, especially since some additional attenuation in the $(\alpha,2n)$ experiments may have been a consequence of spin-lattice relaxation [17].

From our data we observe that the $2 \rightarrow 0,2 \rightarrow 0$ interfragment correlation is stronger than that for the $4 \rightarrow 2,4 \rightarrow 2$ coincidences. This result is to be expected, since the theoretical maximum correlation without any attenuation is the same as that for a $2 \xrightarrow{E2} 4 \xrightarrow{E2} 2$ decay sequence (i.e., $a_2 = 0.200$ and $a_4 = 0.092$). The decrease in the theoretical maximum a_2 values in going from the $2 \rightarrow 0,2 \rightarrow 0$ interfragment correlation to that for the $4 \rightarrow 2,4 \rightarrow 2$ coincidence is therefore roughly in line with the observed decrease of around 40%. More precisely, if we assume the same spread in the relative angles of the spin vectors at $I = 4$ as at $I = 2$, then the smearing appropriate for the interfragment angular correlation at $I = 4$ should be $\sigma_{\gamma\gamma}^{I=4} = 2 * \sigma_{\gamma\gamma}^{I=2} \approx 2.8$. According to our calculations, this value of $\sigma_{\gamma\gamma}$ gives $a_2 = 0.023$, a little lower than the measured mean value, which may reflect a tendency for $\sigma_{\gamma\gamma}/I$ to decrease with increasing spin due to increased direct feeding.

Our measurements of interfragment $2 \rightarrow 0,2 \rightarrow 0$ $\gamma\gamma$ correlations for ^{248}Cm fission have revealed a much lower a_2 $[= 0.014(2)]$ than that observed in our ^{252}Cf data. Unfortunately, the lack of data on fragment-γ correlations for ^{248}Cm makes this result difficult to interpret. The higher level of attenuation in ^{248}Cm fission could be due to an increase in the attenuation caused by the statistical decay; $\sigma_d = 1.3$ is required to reproduce the observed a_2. Such an increased σ_d would produce correspondingly more attenuated fragment-γ correlations. Alternatively, the increased attenuation could have its origin in the fission mechanism, for example, in larger contributions from wriggling modes that introduce a larger relative angular momentum between the fragments, thus smearing the angular correlation between the internal angular momenta.

This paper has discussed measurements of interfragment $\gamma\gamma$ correlations for complementary fragments produced in the spontaneous fission of ^{252}Cf and ^{248}Cm. The deduced a_2 coefficients are consistent, in ^{252}Cf fission, with a smaller attenuation than would be expected from previous measurements of fragment-γ correlations. This suggests that the internal angular momenta of the fragments at scission are well correlated with each other, but have a significant probability of being tilted with respect to the fission axis. The reduced correlation observed in the case of ^{248}Cm fission demands further experimental investigation.

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