Angular distribution of α particles from oriented ^{253,254}Es and ^{255}Fm nuclei

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The anisotropy in the angular distribution of α particles from oriented ^{253,254}Es and ^{255}Fm nuclei, which are among the strongest deformed α emitters, was measured. Large α anisotropies have been observed for all three nuclei. The results are compared with calculations based on α-particle tunneling through a deformed Coulomb barrier.

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I. INTRODUCTION

Alpha decay is often presented as a typical example of quantum-mechanical tunneling through a potential barrier. The exponential energy dependence of the α decay rate is indeed well explained by the tunneling of an already existing α particle through the Coulomb barrier [1,2]. Because of the strong dependence of the tunneling probability on barrier height and width, Hill and Wheeler [3] argued that in a nucleus with a deformed Coulomb barrier, and therefore with a nonuniform barrier height and width, the tunneling probability becomes direction dependent. Thus, α emission from an ensemble of oriented nuclei (i.e., nuclei with a preferential spin direction in space) should be anisotropic. A firmer theoretical framework was built later [4–9]. This used the shell model and Bardeen-Cooper-Schrieffer pairing [10] to calculate the formation probability of the α particle at the nuclear surface and the Wentzel-Kramers-Brillouin approximation [11] to calculate the tunneling through the (deformed) Coulomb barrier.

Based on the works just mentioned, which attribute anisotropic α emission from heavy nuclei to the tunneling of the α particle through a deformed Coulomb barrier, α anisotropies have often been related to nuclear deformation [12,13]. However, this relation has never been firmly established. The first α anisotropy measurements on deformed nuclei were performed on prolate actinide nuclei [14–16]. These, as well as later measurements [17], revealed preferential emission of the α particles along the nuclear symmetry axis, as predicted. Unfortunately, the quality of the available detectors and/or the source preparation techniques did not allow resolution of the different α transitions in the decay and so no detailed conclusions could be obtained. These problems were solved when high-resolution particle detectors operating near 4.2 K were combined with ion-implantation techniques, allowing very thin samples to be produced. This technique was first used to measure α anisotropies for near-spherical odd $^{199-211,215,217}$At and $^{205-209}$Rn nuclei [18–20] near the $N = 126$ and $Z = 82$ shell closures. This showed that for favored decays [i.e., in transitions that are (almost) unhindered with respect to ground-state-to-ground-state transitions in neighboring even-even nuclei] anisotropic α emission is not dominated by nuclear deformation but rather by nuclear structure effects [19]. For example, in the series of At isotopes that was studied the largest anisotropy was observed for ^{211}At, which is at the $N = 126$ shell closure. Measurements on the odd $^{189-193}$Bi nuclei [21] showed a similar behavior. Later, measurements were also performed on statically deformed ^{221}Fr and ^{227,229}Pa nuclei [22]. Very large anisotropies were observed, with the largest ones for the nuclei with the largest deformation. Comparison with existing theories [9,23–28] showed good agreement with the "tunneling" model calculations of Delion *et al.* [26], which use a realistic, spherical mean field and quite a large number of single particle states. This indicates that for deformed nuclei the anisotropy in α emission is dominated by the tunneling of the α particle through the deformed barrier. By combining this result with the earlier observation that for nearly spherical nuclei anisotropic α emission is dominated by nuclear structure effects, it was concluded [22] that the angular distribution of α particles emitted by heavy oriented nuclei is determined by a combined effect of nuclear deformation (tunneling) and nuclear structure (formation probability). The latter gains in (relative) importance as the deformation decreases and is dominant for nearly spherical nuclei.

Although these works have dramatically improved our understanding of the origin of α anisotropy, it should be noted that quantitative data for nuclei with a clear static deformation still exist for only three cases—^{221}Fr, ^{227}Pa, and ^{229}Pa—with deformation parameter $\beta_2 = 0.12, 0.17$, and 0.18,

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respectively. In addition, the interpretation of the data for these nuclei is complicated by the presence of a significant octupole deformation ($\beta_3 \leqslant 0.15$). To investigate the relation between α anisotropy and nuclear deformation in more detail we are therefore extending the data set now to heavy actinide nuclei with large static deformation. Here we present first results for the transuranium isotopes ^{253}Es (deformation parameter $\beta_2 = 0.236$), ^{254}Es ($\beta_2 = 0.226$), and ^{255}Fm ($\beta_2 = 0.227$). Note that all three nuclei belong to the small group of nuclei that have the largest ground-state deformation of all α emitters.

II. EXPERIMENTS AND RESULTS

The Es activity was produced by neutron irradiation of ^{252}Cf in a reactor in Dimitrovgrad (Russia), followed by radiochemical separation of the accumulated Es and a second irradiation to improve the isotopic content. The ^{252}Cf was produced by exposing a gram-scale target consisting of a mixture of Cm isotopes to a thermal neutron flux of $\cong 1.5 \times 10^{15}$ neutrons cm^{-2} s^{-1} and was then extracted by radiochemical separation. The ^{252}Cf obtained was then irradiated to accumulate Es. The production of, for example, ^{255}Es can be described with the following series of reactions:

$$^{252}\mathrm{Cf}(n,\gamma)^{253}\mathrm{Cf}(n,\gamma)^{254}\mathrm{Cf}(n,\gamma)^{255}\mathrm{Cf}(\beta^-)^{255}\mathrm{Es},$$
$$^{252}\mathrm{Cf}(n,\gamma)^{253}\mathrm{Cf}(\beta^-)^{253}\mathrm{Es}(n,\gamma)^{254g}\mathrm{Es}(n,\gamma)^{255}\mathrm{Es}.$$

To isolate Es from the irradiated targets special radiochemical procedures were developed. Ion-exchange chromatography with the Bio-Rad cation exchanger in NH_4^+ form and 0.1 mol/l solution of γ-oxy-iso-butirate as an eluent were used for separation. The efficiency of this cation-exchange system was first tested and demonstrated in laboratory experiments that were carried out with ^{91}Y, which has properties similar to Fm. The procedures that were developed can of course also be used for the production of other pure samples of Es and Fm for nuclear spectroscopy experiments or applied research needs. Finally, a batch of activity containing the isotopes ^{253}Es ($t_{1/2} =$ 20.5 d), ^{254}Es ($t_{1/2} = 275.7$ d), and ^{255}Es ($t_{1/2} = 39.8$ d) was obtained. This was loaded into the oven of a positive surface ionization ion source of the isotope separator at Bonn (Germany), mass separated, and implanted at an acceleration voltage of 160 kV into high-purity (99.99%), annealed 100-μm Fe foils at room temperature. The samples thus obtained were soldered with Woods metal onto a Cu sample holder and top-loaded into a ^{3}He-^{4}He dilution refrigerator in Leuven (Belgium) for the nuclear orientation experiments. Alpha particles were detected with Si PIN diodes mounted inside the 4.2-K shield of the refrigerator at different angles with respect to the nuclear orientation axis (the external orienting magnetic field). The energy resolution of these detectors at their operation temperature of about 10 K was about 20 keV at 6 MeV. Two large-volume HPGe detectors to measure γ spectra were installed at 0° and 90° outside the refrigerator. The sample temperature was monitored with a ^{54}MnNi nuclear orientation thermometer that was soldered to the backside of the sample holder. Count rates $N(\theta)$ were recorded for both oriented [$N(\theta)_{\mathrm{cold}}$ at $T < 300$ mK] and nonoriented

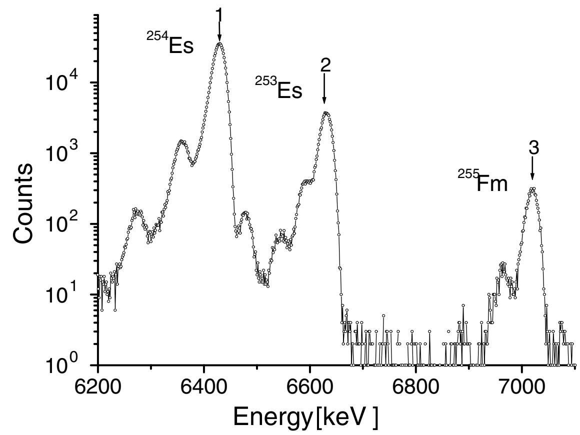

FIG. 1. Alpha spectrum observed with one of the 90° detectors. The three α lines for which anisotropies were determined are indicated with an arrow. The energies of these lines are listed in Table I.

[$N(\theta)_{\mathrm{warm}}$ at $T \approx 1.4$ K] nuclei. The anisotropy at a temperature T is then obtained as $[N(\theta)_{\mathrm{cold}}/N(\theta)_{\mathrm{warm}}] - 1 = W(\theta) - 1$, where the angular distribution function $W(\theta)$ has the form [29]

$$W(\theta) = 1 + f \sum_{k=2,4} A_k B_k Q_k P_k \cos(\theta). \tag{1}$$

Here the implantation parameter f represents the fraction of nuclei experiencing the full orienting hyperfine interaction. It is assumed in this so-called two-site model that the rest $(1 - f)$ experiences no orienting hyperfine interaction at all. B_k are the nuclear orientation parameters, which depend on the temperature T of the sample and on the ratio $\Delta_M = \mu B / k_B I$, with μ the nuclear magnetic moment, B the total magnetic field at the site of the nucleus, k_B the Boltzmann constant, and I the spin of the oriented state. P_k are the Legendre polynomials and the Q_k solid angle correction factors account for the finite size of the source and detectors. The information on the α transition is contained in the directional distribution coefficients A_k. These are written as [29]

$$A_k = \frac{\sum_{L,L'} a_L a'_L \cos(\sigma_L - \sigma_{L'}) F_k^\alpha(L, L', I_f, I_i)}{\sum_L a_L^2}, \tag{2}$$

where F_k^α are F coefficients modified for α decay [30], and σ_L and a_L are the phase and the amplitude of the α wave with angular momentum L. The mixing ratios are defined as $\delta_{0L} \equiv a_L/a_0$. Since the favored decays studied here occur between states with the same parity, only even L values are involved. Note that in favored transitions the most intense partial α wave has $L = 0$, resulting in isotropic emission relative to the nuclear spin direction. Only the higher order partial α waves, with angular momentum $L \neq 0$, cause anisotropy in the α emission. The observed α anisotropy is thus to lowest order a direct measure of the $L = 2$ admixed amplitude. Note, finally, also that α decay of unoriented nuclei is isotropic in space such that decay-rate experiments are insensitive to the different higher order angular momenta involved.

A typical α spectrum as observed with a detector at 90° is shown in Fig. 1. Experimental anisotropy data for the

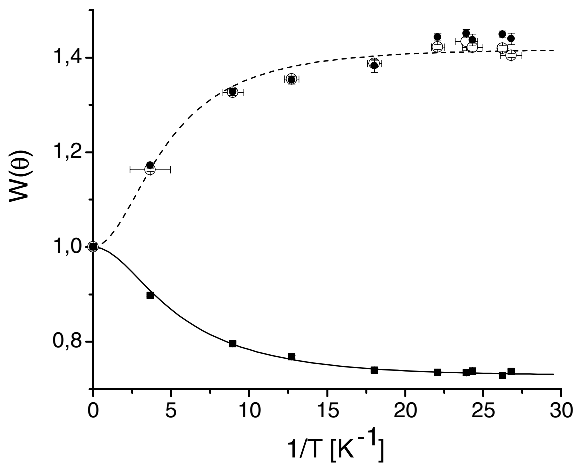

FIG. 2. Simultaneous fit of the anisotropies $W(\theta)$ for the favored $7/2^+ \rightarrow 7/2^+$ 6632 keV α transition of ^{253}Es observed with two detectors at 15° (dashed line) and one at 90° (solid line).

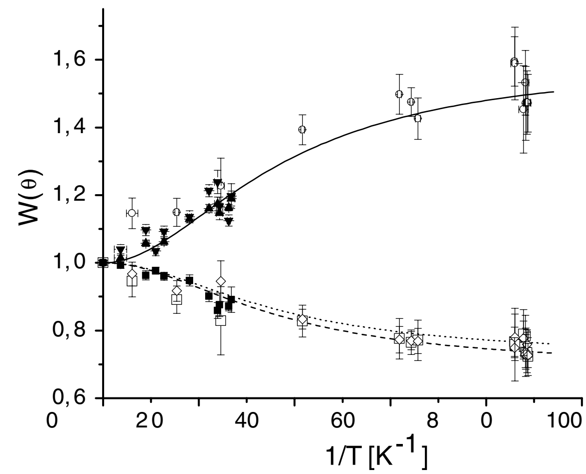

FIG. 4. Simultaneous fit of the anisotropies $W(\theta)$ for the favored $7/2^+ \rightarrow 7/2^+$ 7022 keV α transition of ^{255}Fm observed with three detectors at 15° (solid line), one at 78° (dotted line), and two at 90° (dashed line).

favored transitions in the decay of ^{253}Es, ^{254}Es, and ^{255}Fm ($t_{1/2} = 20.1$ h; the daughter isotope of ^{255}Es) are shown in Figs. 2, 3, and 4. For ^{255}Es no α anisotropies could be deduced from the data as the α lines of this isotope were hidden under the much stronger α lines of ^{254}Es. Our data show that for ^{253}Es, ^{254}Es, and ^{255}Fm α emission is preferentially along the nuclear spin direction. The observed anisotropies are quite large. At the lowest temperatures the ratio of emission probabilities along and perpendicular to the nuclear spin direction is about 1.9 for ^{253}Es, whereas for ^{254}Es and ^{255}Fm it is about 2.0. For each nucleus the anisotropies observed with the different detectors were fitted simultaneously to determine the directional distribution coefficients A_2 and A_4. The term depending on A_6 could be neglected, as was verified in the analysis. Because for ^{253}Es and ^{254}Es full saturation of orientation is reached at the lowest attained temperatures and for ^{255}Fm nearly full saturation is reached, the A_k coefficients could be obtained independent of the as-yet unknown hyperfine interaction parameters. Indeed, neither for Es nor Fm is the hyperfine magnetic field in iron host known; moreover, for ^{254}Es and ^{255}Fm the nuclear magnetic moment is not known either. It was not necessary to assume a combined magnetic and electric interaction as calculations and experimental results showed that there is no appreciable orbital moment producing an electric field gradient for Es and Fm impurities in Fe [31].

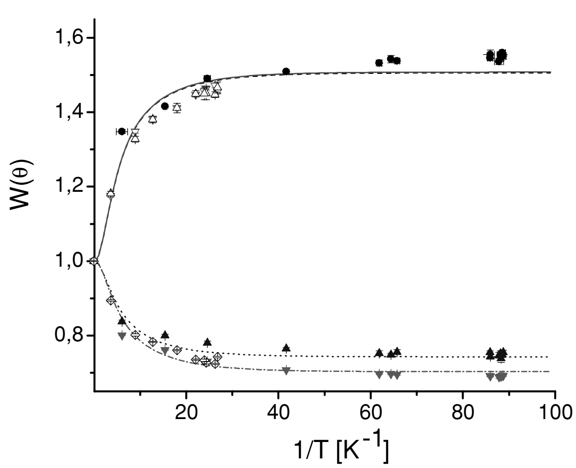

FIG. 3. Simultaneous fit of the anisotropies $W(\theta)$ for the favored $(7^+) \rightarrow (7^+)$ 6429 keV α transition of ^{254}Es observed with three detectors at 15° (dashed and solid line), one at 78° (dotted line), and two at 90° (dash-dotted line).

Fitting the α anisotropies required using the fraction $f = 0.67(10)$, which was obtained from the $W(0°)/W(90°)$ anisotropy of the 1031-keV pure E2 γ ray in the decay of ^{250}Bk, the daughter isotope of ^{254}Es. This value is similar to what was obtained earlier for Pa implanted in Fe [22]. The error of 0.03 obtained from the fit was enlarged to take into account systematic effects and the slightly different implantation conditions of Es (implanted at room temperature) and Bk (cold implanted as a decay product of the ^{254}Es α decay). The fit also yielded the amplitudes $a_0 = 0.60(4)$ and $a_1 = 0.40(4)$ for the $L = 0$ and $L = 1$ parts, respectively, in the first forbidden β decay of ^{250}Bk preceding the 1031-keV γ transition.

Experimental directional distribution coefficients A_2 and A_4 as well as the corresponding mixing ratios δ_{0L} for the favored transitions in the decay of ^{253,254}Es and ^{255}Fm are listed in Table I. As can be seen, the intensities of the $L = 2$ wave [defined as $\delta_{02}^2/(1 + \delta_{02}^2 + \delta_{04}^2)$] are quite large. Also listed are theoretical values for A_2 and A_4 and/or δ_{02} and δ_{04} for ^{253}Es and ^{255}Fm obtained from different theories. For the odd-odd nucleus ^{254}Es no theoretical expectations are available yet.

III. DISCUSSION

The theoretical predictions listed in Table I are all based on the "tunneling" model. In this model nuclear deformation is the most important factor in modeling anisotropic α decay, and it was found that the angular distribution should reflect

TABLE I. Experimental directional distribution coefficients A_k, the mixing ratios δ_{02} and δ_{04} ($\delta_{ij} \equiv a_j/a_i$), and the $L = 2$ intensity for the favored α transitions in the decay of ^{253}Es, ^{254}Es, and ^{255}Fm. These are compared to calculations we performed on the basis of the theories outlined in Refs. [4,6] and theoretical predictions from Ref. [32].

Nucleus	E_α (keV)	$I_i^\pi \rightarrow I_f^\pi$	$BR(\%)$	A_2	A_4	δ_{02}	δ_{04}	$L = 2(\%)$	Reference
^{253}Es	6633	$\frac{7}{2}^+ \rightarrow \frac{7}{2}^+$	90	0.53(8)	−0.126(20)	0.30(5)	−0.115(12)	8.2	exp., this work
						0.354	0.040		theory, [4]
						0.327	−0.031		theory, [6]
				0.960	0.226				theory, [32]
^{255}Fm	7022	$\frac{7}{2}^+ \rightarrow \frac{7}{2}^+$	93	0.65(10)	0.04(9)	0.35(7)	−0.02(7)	10.9	exp., this work
						0.338	0.052		theory, [4]
						0.353	−0.033		theory, [6]
				0.933	0.212				theory, [32]
^{254}Es	6429	$(7^+) \rightarrow (7^+)$	93	0.47(7)	−0.030(14)	0.25(4)	−0.043(8)	5.9	exp., this work

the shape of the nucleus. We performed calculations of the δ_{02} and δ_{04} mixing ratios for ^{253}Es and ^{255}Fm using the theories of Refs. [4] and [6]. As can be seen, the experimental results are in reasonable agreement with predictions from these theories, where the calculations from Ref. [6] seem to reproduce the experimental results slightly better than those from Ref. [4]. Delion *et al.* [32] have adopted in their model the same approach as in older works [4–7] but employed a much larger shell model configuration space to compute the formation probabilities. Their predictions are given in terms of "idealized" amplitudes A_L, which are equal to the factor $A_k B_k$ in Eq. (1) for $T = 0$. For comparison we have recalculated their amplitudes A_L to the corresponding values for the angular distribution coefficients A_k in Eq. (1). Their predictions for A_2 turn out to be about 50% larger than our results. The difference might be caused by a deficient description of the formation amplitude at the nuclear surface.

In summary, our experiments provide new information on the angular distribution of α particles emitted by deformed nuclei, extending the systematics now to the α emitters with the largest deformation found in nature. Large anisotropies were observed. Comparison with existing theories based on the "tunneling" model are in good to reasonable agreement with the experimental results. This confirms the earlier finding [22] that for deformed nuclei the anisotropy in α decay is dominated by the tunneling through the deformed Coulomb barrier and that the formation amplitude of the α particle at the nuclear surface, which was earlier shown to be the dominant factor for nearly spherical nuclei [18–20], is not the dominant factor for these nuclei.

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