Angular Distributions of α Particles Emitted by Deformed Oriented Nuclei

P. Schuurmans,[1] J. Camps,[1] P. De Moor,[1] K. Freitag,[2] P. Herzog,[2] M. Huyse,[1] R. Paulsen,[2] N. Severijns,[1]
A. Van Geert,[1] P. Van Duppen,[1,3] B. Will,[2] and NICOLE and ISOLDE Collaborations
[1]*Instituut voor Kern- en Stralingsfysica, K.U. Leuven, B-3001 Leuven, Belgium*
[2]*Institut für Strahlen- und Kernphysik, Universität Bonn, D-53115 Bonn, Germany*
[3]*CERN, PPE division (ISOLDE), CH-1211 Genève 23, Switzerland*

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A detailed quantitative experimental investigation of the influence of nuclear deformation on the angular distribution of α particles emitted by oriented nuclei is reported. The favored α transitions in the decay of the deformed nuclei ^{221}Fr, ^{227}Pa, and ^{229}Pa were studied. In all three cases, very large anisotropies have been observed. The results are in good agreement with calculations based on α particle tunneling through a deformed Coulomb barrier. [S0031-9007(99)09374-6]

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Alpha decay is a textbook example of quantum mechanical tunneling of a particle through a potential barrier. The exponential energy dependence of the α decay rate is indeed well explained by the tunneling of a preformed α particle through the Coulomb barrier of atomic nuclei [1]. Hill and Wheeler [2] argued that in a nucleus with a deformed Coulomb barrier the tunneling probability becomes direction dependent, resulting in anisotropic α emission from an ensemble of oriented nuclei (i.e., nuclei with a preferential spin direction in space). A firmer theoretical framework was built later [3–6], in which the shell model—including Bardeen-Cooper-Schriefer pairing [7]—was used to compute the formation amplitude of the α particle at the nuclear surface while employing the Wentzel-Kramers-Brillouin approximation [8] to calculate tunneling through the (deformed) Coulomb barrier.

Based on the works mentioned above, the observation of anisotropic α emission from heavy nuclei has often been attributed to the tunneling of the α particles through a deformed barrier, thus relating α anisotropies to nuclear deformation [9]. This relationship, however, has not been firmly established experimentally. Indeed, the only α anisotropy experiments on nuclei known to be deformed were performed on prolate actinide nuclei more than two decades ago [10]. As predicted, a preferential emission of the α particles along the nuclear symmetry axis was observed. However, at that time, the source preparation technique and the quality of the detectors available did not allow resolution of the different α transitions in the decays investigated and no detailed conclusions could be drawn. These problems were solved for the first time when high-resolution particle detectors operating near 4.2 K were linked with ion implantation techniques for sample preparation [11]. Using this combination we have recently shown that for nuclei near the $N = 126$ and $Z = 82$ shell closures, anisotropic α emission in favored decays, i.e., in transitions which are (almost) unhindered compared to the ground-state-to-ground-state transitions in neighboring even-even nuclei, is not dominated by deformation but rather by nuclear structure effects [12]. One is thus lead to the conclusion that the assumed relation between nuclear deformation and the angular distribution of α particles is not evident. It may be noted here that only the higher order partial α waves with angular momentum $L \neq 0$ determine the α anisotropy. The α decay of unoriented nuclei is isotropic in space and hence decay rate experiments are insensitive to the different values of angular momentum involved.

To gain a better insight into the relation between nuclear shapes and the angular distributions in favored α decay, we have measured the α anisotropies of the deformed nuclei ^{229}Pa ($t_{1/2} = 1.50$ d) and ^{227}Pa ($t_{1/2} = 38.3$ min) using the KOOL on-line low temperature nuclear orientation (LTNO) setup [13] at the LISOL mass separator at Louvain-la-Neuve [14] and of ^{221}Fr ($t_{1/2} = 4.9$ min) at the NICOLE LTNO-facility [15] on-line to ISOLDE [16] (CERN). The $^{227(229)}$Pa nuclei were produced via a $^{232}\mathrm{Th}(\mathrm{p},x\mathrm{n})^{233-x}\mathrm{Pa}$ fusion-evaporation reaction at 55 MeV (38 MeV). The precursor of ^{221}Fr, ^{221}Rn, was made in a spallation reaction using 1 GeV protons on ThC. The radioactive isotopes ^{227}Pa and ^{221}Rn were mass separated and implanted at low temperatures down to 11 mK, into a magnetized high-purity iron foil mounted in a ^{3}He-^{4}He dilution refrigerator and subsequently oriented. *In situ* β^- decay of implanted ^{221}Rn yielded the ^{221}Fr nuclei [12]. The ^{229}Pa*Fe* sample was implanted at room temperature at Bonn University and thereafter top loaded into the KOOL refrigerator where the nuclear orientation data were taken. Alpha spectra were recorded with Si detectors mounted inside the refrigerators at angles $\theta_1 = 17°$, $\theta_2 = 84°$ (^{227}Pa); $\theta_1 = 71°$ (^{229}Pa) and $\theta_1 = 16°$, $\theta_2 = 50°$, $\theta_3 = 84°$ (^{221}Fr) relative to the magnetization axis. Conventional Ge detectors measured the γ spectra. The sample temperature was monitored with ^{57}Co*Fe* and ^{54}Mn*Ni* thermometers. The angular distribution function $W(\theta)$ was determined for each α transition from the ratio of the intensities $N(\theta)$ at low temperatures (i.e., $T < 100$ mK; "cold") and at ≈1.4 K (no orientation; "warm"). In order to correct for possible variations in the isotope separator beam intensity, data taken during on-line implantation are evaluated using a double ratio $W(\theta_i)/W(\theta_j)$. The angular distribution function is written as [17]

$$W(\theta) = 1 + f \sum_{k \neq 0} A_k B_k Q_k P_k(\cos\theta). \qquad (1)$$

Here, f represents the effective fraction of nuclei that is oriented by the hyperfine interaction, assuming that the rest $(1 - f)$ is not oriented at all. It is determined from the anisotropy [i.e., $W(\theta_i)/W(\theta_j)$ versus $1/T$] of γ transitions for which all other parameters in the angular distribution function are known [18]. P_k are Legendre polynomials, the Q_k account for the finite size of source and detector and the B_k are the orientation parameters. Information on the α decay is found in the directional distribution coefficients A_k. They are written as [17]

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

where F_k^α are F coefficients modified for α decay [17], 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 have $I_i = I_f = 5/2$ without parity change and α decay is parity conserving, only $L = 0, 2, 4$ are involved.

Experimental anisotropy data for the favored transitions in the decay of ^{221}Fr and ^{227,229}Pa are shown in Figs. 1 and 2. For ^{227,229}Pa α emission is preferentially along the nuclear spin while ^{221}Fr shows preferential α emission perpendicular to the nuclear spin axis. Also, the observed anisotropies are quite large as for ^{221}Fr the ratio of emission probabilities perpendicular to, and along the nuclear spin is about 1.8 while for ^{227}Pa the reverse ratio is almost 2.5. By fitting the angular distribution function to the anisotropy data the directional distribution coefficients A_2 and A_4 were deduced. Because in the case of ^{221}Fr full saturation of orientation is not reached, anisotropy data for the $5/2^- \rightarrow 9/2^-$ 6341 keV transition in ^{221}Fr were included in the data evaluation to reduce the possible correlation between the A_k and the hyperfine interaction strength parameter $\boldsymbol{\mu} \cdot \boldsymbol{B}$ For ^{227}Pa, with only two detection angles available, A_2 and A_4 were determined independently by a simultaneous fit of the data set taken during continuous implantation and data taken in the decay of the sample after interruption of the ion beam. The ^{229}Pa α transitions were observed with one detector placed at an angle of 71° where $P_4(\cos\theta)$ is almost zero. A_2 was derived here neglecting the $k = 4$ term in the expansion of $W(\theta)$. Experimental mixing ratios δ_{0L} and the fitted A_k parameters for the favored as well as for two other transitions in the decay of ^{221}Fr, ^{227}Pa, and ^{229}Pa are listed in Table I. Systematic uncertainties in the derivation of the f parameter used in the evaluation for each isotope are included in the uncertainties given in Table I. Clearly, the intensities of the $L = 2$ wave [defined as $\delta_{02}^2/(1 + \delta_{02}^2 + \delta_{04}^2)$] are quite large.

To interpret our data, we used recent α anisotropy calculations by Delion, Insolia, and Liotta [19–21], by Stewart *et al.* [22], and by Berggren [23]. In their "tunneling" model, the former have adopted the same approach as in older work [3–5] but employed a much larger shell model configuration space to compute the formation probabilities and also included possible octupole deformation in the determination of the tunneling factors. In these calculations nuclear deformation is the most important factor in modeling anisotropic α decay, and it was found the angular distribution should reflect the shape of the nucleus. The theoretical A_2 coefficients [20,21] for the nuclei studied here, together with the deformation parameters used in the calculations are presented in Table II. Comparison with

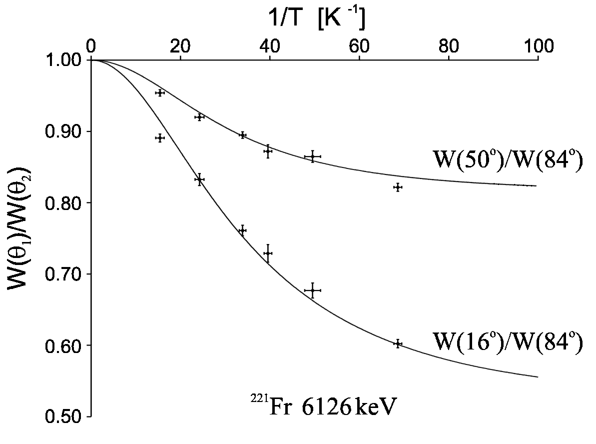

FIG. 1. Alpha anisotropy data $W(16°)/W(84°)$ and $W(50°)/W(84°)$ as a function of the inverse temperature $1/T$ for the favored 6126 keV transition in ^{221}Fr.

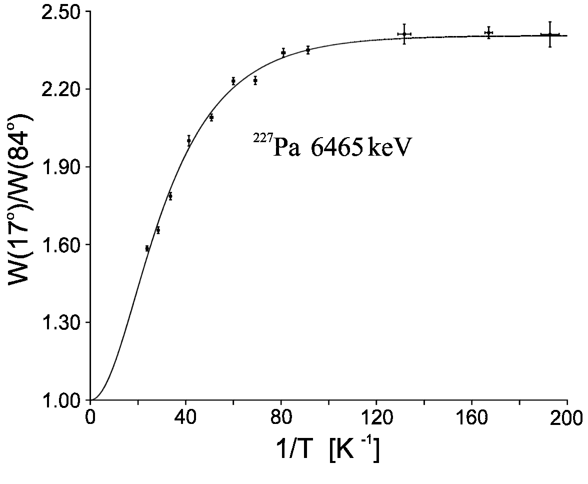

FIG. 2. Anisotropy data for the favored 6465 keV α transition in ^{227}Pa as a function of $1/T$. Data points below 10 mK were taken during the decay of the source after interruption of the beam.

TABLE I. Experimental angular distribution coefficients A_2 and A_4, the mixing ratios δ_{02} and δ_{04} ($\delta_{ij} \equiv a_j/a_i$), and the $L = 2$ intensity for transitions in the decay of ^{221}Fr, ^{227}Pa, and ^{229}Pa. The factor f is explained in the text.

Nucleus	E_α (keV)	$I_i^\pi \rightarrow I_f^\pi$	f	A_2	A_4	δ_{02} [δ_{24}]	δ_{04} [δ_{26}]	$L = 2$ (%)
^{221}Fr	6126	$\frac{5}{2}^- \rightarrow \frac{5}{2}^-$	0.81(2)	−0.375(14)	−0.10(5)	−0.216(9)	−0.067(30)	4.9
^{227}Pa	6465	$\frac{5}{2}^- \rightarrow \frac{5}{2}^-$	0.80(5)	0.696(44)	0.25(13)	0.364(27)	0.16(12)	9.3
^{229}Pa	5580	$\frac{5}{2}^+ \rightarrow \frac{5}{2}^+$ $(K = 5/2)$	0.68(5)	1.13(11)	⋯	0.75(16)	⋯	≈32
^{229}Pa	5670	$\frac{5}{2}^+ \rightarrow \frac{5}{2}^+$ $(K = 3/2)$	0.68(5)	0.81(11)	⋯	0.45(10)	⋯	≈17
^{221}Fr	6341	$\frac{9}{2}^- \rightarrow \frac{5}{2}^-$	0.81(2)	−0.389(10)	−0.06(6)	[−0.004(5)]	[0.037(33)]	≥99.5

the experimental A_2 data shows that for ^{227}Pa, agreement with theory is very good. Our result does not give any information on the influence of a possible octupole deformation as the experimental data overlap with both calculations excluding and including a β_3 deformation. The theoretical value (A_2^{th}) for the favored α decay in ^{229}Pa, is too small by about 30%. Agreement within the experimental uncertainty is found only with a rather improbable deformation parameter $\beta_2 \approx 0.3$. A possible explanation for this discrepancy could be that ^{229}Pa may be octupole soft such that the mean-field deformation is not described accurately with the parameters used in the calculations. Alternatively, the difference might be caused by a deficient description of the formation amplitude at the nuclear surface.

For ^{221}Fr the published theoretical prediction [20] and experiment deviate by almost a factor of 2. This difference, however, is due to the fact that Delion *et al.* assumed a nuclear spin projection $K = I = 5/2$ for the ^{221}Fr ground state with an oblate deformation $\beta_2 = -0.069$. Qualitatively, this agrees with the preferred equatorial α emission (Fig. 1). In the literature, however, the ^{221}Fr ground state is assigned $K = 1/2$ [24], with a prolate deformation. To investigate the influence of the different K assignment on the theoretical anisotropies we have recalculated the δ_{02} mixing ratio and the directional distribution coefficient A_2 for the favored α transition in ^{221}Fr. A $K = 1/2$ value was used while the deformation parameters $\beta_2 = 0.120$, $\beta_3 = 0.153$ were taken from [25]. With the α particle formation amplitudes deduced from the result published in Ref. [20], $\delta_{02} = -0.186$ is found, neglecting β_3 deformation. The influence of octupole deformation on the tunneling probability increases the magnitude of the mixing ratio δ_{02} by about 10% [21] yielding $\delta_{02} \approx -0.204$ in excellent agreement with the experimental value $\delta_{02} = -0.216(9)$. The corresponding A_2^{th} parameter is listed in Table II.

TABLE II. Theoretical A_2 coefficients calculated with deformation parameters β_2 and β_3 for the favored transitions in the decay of ^{221}Fr [20], ^{227}Pa, and ^{229}Pa [21] compared with experiment.

Nucleus	β_2	β_3	A_2^{th}	A_2^{exp}
^{221}Fr	−0.069	0.0	−0.215	−0.375(14)
	0.120	0.15	−0.373 [a]	
^{227}Pa	0.168	0.0	0.649	0.696(44)
	0.168	0.1	0.748	
^{229}Pa	0.185	0.0	0.733	1.13(11)
	0.185	0.08	0.808	

[a] Calculated with $K = 1/2$, $\delta_{02} = -0.204$, and $\delta_{04} \approx 0$.

Stewart *et al.* [22] also use Fröman's method [5], but use a realistic nuclear potential to calculate the tunneling probability of the α particle through the deformed barrier, including β_2 and β_4 deformations. The formation amplitudes a_L of the partial α waves with angular momentum L at the nuclear surface, however, are extracted from the fine structure in the α decay of neighboring even-even nuclei. Hereby, all four possible choices for the relative phases between the partial α waves are considered. For one choice $[\text{sgn}(a_0, a_2, a_4) = (+, -, -)]$ the formation amplitudes turn out to be rather constant for a wide range of actinide nuclei [22]. Moreover, the a_2 amplitudes are negative for this solution. For small deformations, with little mixing of the partial α waves with different L in the barrier region, this corresponds to preferential α emission in the equatorial plane. Large prolate deformations, however, cause sufficient mixing between the different L waves to induce preferential emission along the symmetry axis. Using the values thus derived, the α anisotropies of various odd-A nuclei are computed [22]. It should be noted that in contrast with the work of Delion *et al.*, Stewart *et al.* assume that no mixing of the daughter states occurs in the decay. Moreover, the deformation parameters of the daughter, rather than of the parent nuclei are used. Theoretical predictions are given in terms of an "idealized" anisotropy ratio, i.e., $W(0°)/W(90°)$ for $T = 0$, assuming $Q_k = 1$ and $f = 1$. For ^{227}Pa and ^{221}Fr theory and experiment deviate roughly by factors 1.6 and 2, respectively. In both cases, the calculated value for $W(0°)/W(90°)$ is too close to unity. In Ref. [22] no prediction for ^{229}Pa was given. The reason for the difference between experiment and theory is not very clear. Using the (larger) parent deformation parameters improves the agreement in the case of ^{227}Pa. For ^{221}Fr, however, it would only shift the result for $W(0°)/W(90°)$towards unity, and possibly beyond, because of the increased mixing between the partial α waves.

Berggren [23] used an (α cluster + core) model in which a quadrupole (and for Pa also an octupole) type interaction between the α particle and the daughter nucleus is diagonalized. Thus several solutions are obtained, among which the best is selected on the basis of agreement between calculated and experimental branching ratios to different levels in the daughter nucleus. This particular solution then yields a prediction of the α anisotropies for all transitions involved in the decay. Although the performance of the (α + core) model is better for deformed than for nearly spherical nuclei [12], agreement with the present experimental data is not very convincing. The theoretical anisotropy ratios [23] and experiment differ by a factor of about 1.3 for the α transitions in ^{221}Fr to 1.7 for ^{227}Pa and 2.0 for ^{229}Pa.

Summarizing, our experiments constitute a detailed quantitative experimental investigation of the angular distribution of α particles emitted by deformed nuclei. Very large anisotropies were observed, the largest being found for the most deformed nuclei. Comparison with existing theories shows that both the extreme cluster model of Berggren [23] and the calculations by Stewart *et al.* [22] are in poor agreement with our findings. For the "tunneling" model calculations of Delion *et al.* [20,21], on the other hand, agreement with experiment is good. This indicates that for deformed nuclei, the anisotropy in α decay is dominated by the tunneling of the α particle through the deformed Coulomb barrier. The formation amplitude of the α particle at the nuclear surface contributes to the anisotropy but is not the most important factor here. Recently, we have shown that for nearly spherical nuclei, anisotropic α emission is dominated by nuclear structure effects [12]. Combining this with the present result we conclude that the classical idea that tunneling of the α particle through the deformed Coulomb barrier is responsible for anisotropic α emission of oriented nuclei is indeed correct only for nuclei with a strong static deformation.

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