Quadrupole Moment of the 8-μs Fission Isomer in ^{239}Pu

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The quadrupole moment of the 8-μs fission isomer in ^{239}Pu has been determined with a newly developed charge-plunger technique for measuring lifetimes of states decaying by converted transitions. With a statistical accuracy of 7%, values between 34 and 39 b are obtained, depending on the unknown isomeric spin. The deformation of the second minimum described by the axis ratio of a spheroid is deduced to be $c/a = 2.0 \pm 0.1$.

The identification of a rotational band[1] based on the fission-isomeric state in ^{240}Pu with a moment of inertia more than twice as big as that of the ground-state rotational band provided the first experimental evidence that fission isomers are strongly deformed nuclear states. Since the relation between the moment of inertia and deformation is rather model-dependent, no direct value for the deformation of the second minimum could be deduced from this measurement. Besides one attempt with only limited accuracy,[2] this Letter reports the first measurement of the quadrupole moment of a fission-isomeric state.

A newly developed "charge-plunger" technique for measuring lifetimes of states decaying by converted transitions has been employed. If these transitions occur at times ≥ 0.1 ps for which recoil nuclei from a nuclear reaction have already left the (thin) target, very high ionic charge states occur as a result of fast ($< 10^{-14}$ s) Auger cascades in the atomic shells[3]; charges up to 40^+ have been observed for ^{240}Cm recoil ions.[4] These charge states can be reset to the equilibrium values (1^+ and 2^+ for recoil velocities of $v/c = 0.2\%$) by passage through a thin carbon foil. The lifetime of the level or the overall time scale of, for example, a rotational de-excitation can then be determined by measuring the intensity ratio of the high-charge and the low-charge component of the distribution as a function of the target–carbon-foil distance, since only states still excited beyond the foil contribute to the high-charge part. The method has been established[5] by measuring lifetimes of rotational levels in ^{240}Cm, which agree with experimental values for other Cm isotopes.[6]

For a cascade of 1, 2, 3, or more consecutive converted transitions, the charge distribution can, in addition, be unfolded into the individual contributions,[4] with use of their experimentally known center positions [14^+, 21^+, and 26^+ for ^{237}Np (Ref. 3)] and widths. Thus, information on the side-feeding intensities, as well as on the decay times of individual levels, becomes accessible without ever having to observe the corresponding transitions directly.

The ^{239}Pu isomer was selected as the first case because of its long half-life of 8 μs and the high cross section of the reaction ^{238}U(α, $3n$), despite the disadvantages of an even-odd nucleus. The beam of 33-MeV α particles was provided by the upgraded MP tandem accelerator at Heidelberg. The experimental arrangement is shown in Fig. 1. On the basis that the predicted value for the quadrupole moment of about $Q_0 = 38$ b[7,8] yields rotational lifetimes between 50 and 4 ps, the distance between the 20-μg/cm^2 U target and the 3-μg/cm^2 C foil was varied between 15 μm and 5 mm. The recoil ions were confined to $\pm 6°$ with respect to the beam direction. The charge distribution was measured by deflection in a magnetic field of 15 kG. After a flight time of about 100 ns, the fission isomers were stopped on the oblique sections of a Makrofol-foil detector arrangement. The horizontal disk, also covered with Makrofol foils, was completely shielded from prompt-fission events and fragments from fission in flight, thus ensuring that even at for-

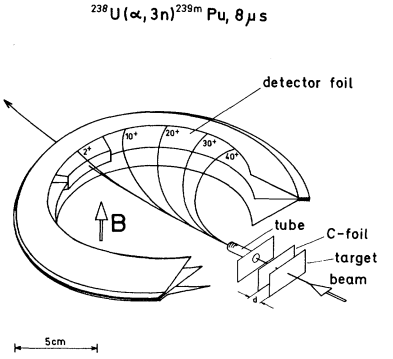

FIG. 1. Experimental setup for the measurement of charge distributions as a function of the distance between target and carbon foil.

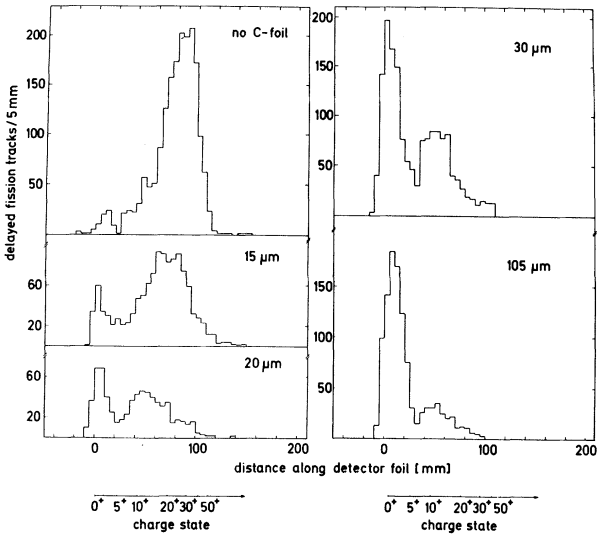

FIG. 2. Charge distributions for several distances of the carbon foil.

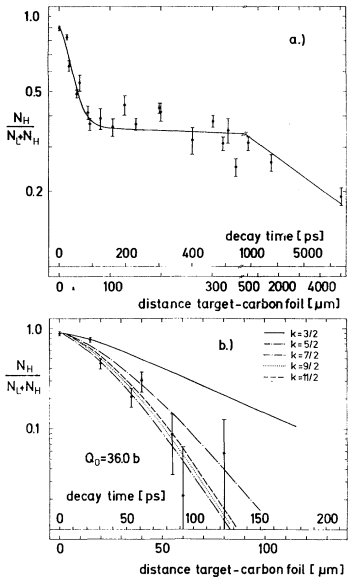

FIG. 3. (a) Fraction of highly charged recoil ions as a function of carbon-foil distance. (b) Fraction of highly charged recoil ions corrected for contribution from 12-nsec isomer. The theoretical curves represent cascade calculations for different K values of the rotational band and a quadrupole moment of 36.0 b.

ward angles only delayed-fission fragments were registered.

Figure 2 shows the charge distributions measured at several distances from the C foil to the target. The contribution of high charges reflecting excited rotational states decreases with increasing distance until it reaches a nearly constant level at about 100 μm. The yield of low charges increases accordingly. Figure 3(a) displays the percentage of highly charged ($>5^+$) recoil ions as a function of distance. The conversion into a time scale uses the known recoil velocity; the correction for energy loss in the target due to nuclear collisions is nearly negligible because of the $\pm 6^\circ$ selection. The short-lived decay is attributed to the de-excitation of the rotational band based on the fission-isomeric state. The long-lived component with a relative intensity of 36% may be explained by a hitherto unknown spin isomer in the second well of ^{239}Pu, decaying with a half-life of 12 ± 7 ns to the 8-μs fission-isomeric state; in the ^{240}Cm case[5] any background effects have been observed to be $<10\%$.

The short-lived decay is shown in Fig. 3(b) after subtraction of the long-lived component. The results of a cascade calculation for the de-excitation of the rotational band are given for comparison. This calculation is based on in-band $E2$ transition rates calculated with the rotational model for a given quadrupole moment Q_0 of 36.0 b.

The partial half-lives for the decay of a rotational state with spin I_1, K are given by

$$T_{1/2}(I_1)=\frac{8.26\times10^2 \ln 2 \text{ keV}^5\, e^2\, \text{b}^2\, \text{s}}{(\Delta E)^5 Q_0^2 \langle I_1 K 20 | I_2 K\rangle^2 (1+\alpha)}.$$

The transition energies ΔE have been calculated for a rotational constant $\hbar^2/2\theta$ lowered by 5% from the value of 3.33 keV measured[1] for ^{240}Pu to take account of an increase of the moment of inertia θ for an even-odd nucleus relative to the even-even neighbor because of blocking effects. Only for small K values, which are ruled out later, even larger moments of inertia may occur because of Coriolis mixing. The rotational constant used in the analysis is uncritical since the $(\Delta E)^{-5}$ dependence of the half-lives on the transition energies is nearly compensated by the energy dependence of the conversion coefficients α. Changes in conversion coefficients for highly stripped recoil ions due to missing electrons and altered electron densities within the nuclear volume are found to be less than 1%.[9,10] The side-feeding intensities are taken from the unfolding of the charge distribution measured without the C foil; their uncertainties, including those due to contributions from the nanosecond isomer, influence the final result by much less than the statistical error of the data.

The results of the cascade calculation depend

on the unknown K quantum number of the rotational band. For $K \geq \frac{7}{2}$, only small differences occur in the calculated decrease of the number of excited rotational states as a function of the decay time. For $K = \frac{5}{2}$ and $\frac{3}{2}$, the $\frac{7}{2} \rightarrow \frac{5}{2}$ and $\frac{5}{2} \rightarrow \frac{3}{2}$ transition can only proceed through L_{III}, $M\ldots$, and $M\ldots$ conversion, respectively; for $K = \frac{1}{2}$, the lowest transition is also restricted to $M\ldots$ conversion, independent of the decoupling parameter. Consequently, the last step of the cascade is slowed down considerably in these cases, prolonging the overall de-excitation time as shown in Fig. 3(b). An unfolding analysis[5] of the high-charge part of the distribution gives, however, nearly equal decay times of 12 ± 2, 8 ± 2, and 9 ± 2 ps, respectively, for the 1, 2, and 3 consecutive converted transitions, thus ruling out the cases with $K = \frac{1}{2}$ and $\frac{3}{2}$. For $K \geq \frac{5}{2}$, the quadrupole moments Q_0 obtained from fits of the calculated decay curves to the experimental data are listed in Table I.

Apart from the remaining ambiguity due to K, systematic errors could occur through nonzero side-feeding times. According to the unfolding analysis no significant side-feeding with times ≥ 10 ps occurs apart from the 12-ns isomer. With typical values of 5 ps for (HI, xn) reactions,[11] the quadrupole moment would increase by only about 5%. Errors could also be caused by converted transitions before reaching the rotational band. Because of the choice of an even-odd nucleus, a further problem arises from possible $M1/E2$ competition. In lack of any experimental knowledge about magnetic moments thus far, this influence has been studied in detail considering all possible g factors for each K. In the worst possible case, the quadrupole moment would be reduced by 19%.

In conclusion and independent from these uncertainties, a static quadrupole moment of the ^{239}Pu fission isomer between 34 and 39 b exceeds, by far, the values 11.0 ± 0.5 and 11.3 ± 0.5 b[12] measured for the ground states of ^{238}Pu and ^{240}Pu, respectively. It completely agrees, on the other hand, with the numbers 37.6,[7] 34.3[8] and 38.2,[7] 35.0 b,[8] respectively, calculated for the second well on the basis of the Strutinsky procedure. If the shape of the nucleus in the isomeric state is described by a prolate spheroid, an axis ratio of 2.0 ± 0.1 is deduced for a radius parameter of $r_0 = 1.16$ fm. A ratio 2:1 corresponding to the strongest shell effects is obtained theoretically by quite general symmetry arguments.[13,14]

TABLE I. Static quadrupole moments deduced by fitting calculated decay curves to the experimental data with assumption of various spins for the isomeric state. The errors quoted are purely statistical.

$K = I$	Q_0 (b)
5/2	39.4 ± 2.8
7/2	34.3 ± 2.5
9/2	36.0 ± 2.6
11/2	36.9 ± 2.7

A continuation and extension of the experiments to other fission isomers is in progress.

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