Spin-Dependent Triaxial Deformation in Neutron-Rich Mo Isotopes

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The Eurogam-2 array has been used to study γ rays emitted following the spontaneous fission of a ^{248}Cm source. Analysis of Doppler-broadened line shapes corresponding to decays from excited rotational states in neutron-rich Sr, Zr, and Mo fission fragments has enabled the first measurements of state lifetimes in these nuclei at around $I = 10$. The results are consistent with a predicted rotation-induced change in the triaxiality of the yrast states of the Mo isotopes that contrasts with the more stable behavior observed in the Zr and Sr nuclei. [S0031-9007(96)01010-1]

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Atomic nuclei are known to exhibit a variety of shapes whose deviation from sphericity can be well described as combinations of quadrupole and higher-order multipole deformations. The shape that a particular nucleus takes up results from a delicate balance between the collective (macroscopic) and single-particle (microscopic) energies, the relative magnitudes of these energies and their dependence on deformation determining the possible shapes that the nucleus may adopt. Also, because nuclear rotation perturbs the microscopic structure of the nucleus, it may be that the shapes favored by a particular nucleus will change with spin. This threefold sensitivity of the nuclear shape to the single-particle structure, the collective behavior, and the total angular momentum provides a severe test of current theoretical approaches to low-energy nuclear structure. The neutron-rich strontium, zirconium, and molybdenum nuclei provide fertile territory for the exploration of this threefold sensitivity. It has been established that, as the neutron number is increased from the valley of stability, the onset of quadrupole deformation occurs. The transition from a spherical to a deformed shape is sudden for Sr and Zr isotopes, but is gradual for higher Z nuclei. Furthermore, increasing proton number from $Z = 40$ has the effect of softening the nuclear energy surface to triaxiality. This is evidenced firstly by recent measurements [1] that have demonstrated the existence in ^{106}Mo of a two-phonon vibrational state, which shows that ^{106}Mo is soft to triaxial deformation; and secondly by the energy levels of the neutron-rich Ru isotopes [2], which suggest the presence of a more rigid triaxial shape. It remains to explore the role of rotation in this region.

The measurement of the mean lifetimes of rotational states provides the means to probe the variation of shape with rotation, since the electric quadrupole transition matrix elements in a rotational band are simply related to the magnitude of the quadrupole deformation of the intrinsic nuclear state. To date, experimental information on the shapes of the neutron-rich nuclei near $Z = 40$ has been limited to low spins. Here we discuss the first measurements of state lifetimes in these nuclei at spins $I \sim 10$.

In this work the spontaneously fissioning isotope ^{248}Cm was used as a source of neutron-rich fission fragments whose electromagnetic decay properties were to be studied. The source consisted of about 5 mg of curium oxide (giving a fission rate of about 7×10^4 per second) embedded in a pellet of potassium chloride. The Eurogam [3] phase 2 array of Compton-suppressed germanium detectors was used to detect the γ rays emitted from the fission fragments. To obtain a spectrum that emphasized a particular even-even nucleus, the data were sorted off-line with a condition requiring the presence of the 2-0, 4-2, and 6-4 transitions in the ground-state rotational band. The γ rays belonging to events satisfying this triple-gate condition were used to increment a one-dimensional spectrum.

Partial level schemes for the neutron-rich even-even nuclei ^{98}Sr, ^{100,102,104}Zr, and ^{102,104,106,108}Mo for intermediate spins have been deduced in previous work [4]. In the course of this analysis, several new transitions have been added to the yrast bands of these nuclei and more reliable relative intensity measurements have been made, taking into account the previously unobserved broad components (see below). The γ-ray energies of the transitions relevant to the lifetime analysis (including the new transitions) and the assumed spins of the initial and final states involved in the decays are listed in Table I. The sequences of yrast γ-ray energies in these nuclei reveal gradual increases in

TABLE I. The quadrupole moments and state lifetimes deduced from our line shape analysis. Also listed are the spin and γ-ray decay energy for each state used in the decay simulation. The errors quoted for Q_{DPM} are purely statistical in origin, and a further systematic error of 5% exists due to uncertainties in the stopping powers. The DPM lifetimes are computed from Q_{DPM} and within any one band do not represent independent measurements. The first error on the lifetime represents the statistical uncertainty in the measurement, and the second is a result of a 10% uncertainty in the stopping powers. Quadrupole moments from Refs. [6] and [8] are also quoted.

Nucleus	Raman	Möller	DPM results			
	Q [e b]	Q [e b]	Q_{DPM} [e b]	E_γ (keV)	$I_i \rightarrow I_f$	τ (ps)
$^{98}_{38}\mathrm{Sr}_{60}$	3.12(18)	3.14	3.17(20)	812.5	$12 \rightarrow 10$	0.67(08) (07)
				689.4	$10 \rightarrow 8$	1.55(19) (15)
				566.3	$8 \rightarrow 6$	4.28(54) (43)
$^{100}_{40}\mathrm{Zr}_{60}$	3.01(19)	3.36	3.19(10)	846.6	$12 \rightarrow 10$	0.54(03) (05)
				738.6	$10 \rightarrow 8$	1.08(06) (10)
				625.6	$8 \rightarrow 6$	2.55(17) (25)
$^{102}_{40}\mathrm{Zr}_{62}$	4.01(40)	3.51	3.52(17)	862.8	$12 \rightarrow 10$	0.40(05) (04)
				759.5	$10 \rightarrow 8$	0.77(09) (08)
				630.9	$8 \rightarrow 6$	2.01(23) (20)
$^{104}_{40}\mathrm{Zr}_{64}$		3.68	3.72(16)	898.7	$12 \rightarrow 10$	0.29(03) (03)
				765.5	$10 \rightarrow 8$	0.67(07) (07)
				624.4	$8 \rightarrow 6$	1.91(21) (20)
$^{102}_{42}\mathrm{Mo}_{60}$	3.26(19)	3.29	2.44(17)	842.0	$12 \rightarrow 10$	0.95(13) (10)
				771.5	$10 \rightarrow 8$	1.49(21) (15)
				691.0	$8 \rightarrow 6$	2.66(37) (27)
$^{104}_{42}\mathrm{Mo}_{62}$	3.29(13)	3.54	2.84(14)	733.4	$10 \rightarrow 8$	1.42(14) (14)
				641.8	$8 \rightarrow 6$	2.84(31) (28)
$^{106}_{42}\mathrm{Mo}_{64}$	3.62(10)	3.70	2.85(13)	889.9	$12 \rightarrow 10$	0.53(05) (05)
				784.3	$10 \rightarrow 8$	1.00(09) (10)
				655.0	$8 \rightarrow 6$	2.55(23) (26)
$^{108}_{42}\mathrm{Mo}_{66}$	3.68(42)	3.46	2.79(20)	867.1	$12 \rightarrow 10$	0.63(09) (06)
				776.3	$10 \rightarrow 8$	1.11(16) (11)
				662.1	$8 \rightarrow 6$	2.52(36) (25)

the moments of inertia for $\hbar\omega = 0.2-0.4$ MeV, probably a result of the crossing between the strongly interacting ground-state band and a band built on a pair of aligned $h_{11/2}$ neutrons. At spins of $I \approx 10$, symmetrically Doppler-broadened line shapes are observed in the γ-ray energy spectra of these fission fragments. The broad line shapes correspond to decays from states that have lifetimes comparable to (or faster than) the stopping time (1–2 ps) of the fission fragments in the source pellet, the broadening being due to the variable Doppler shift that is observed for the time distribution of decays from such states.

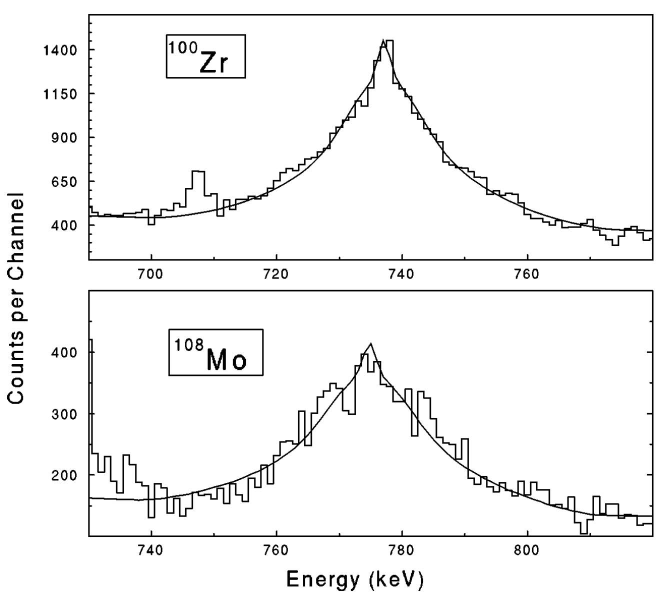
The Doppler-profile method (DPM) [5] combines a simulation of the stopping of the isotropically directed fission fragment with a simulation of the electromagnetic decay to generate a line shape that can be compared directly with the data and thereby extract state lifetimes. In modeling the electromagnetic decay of the excited fission fragment, consideration must be given to the time distribution of the intensity feeding the states whose lifetimes are to be determined. For the highest-spin state, whose decay produces an observed line shape, it is not possible to extract any information directly from the γ-ray spectrum about the feeding distribution, so the feeding of this state is simulated by a two-state feeding model in which the lifetimes of both feeding states are varied in the fit. For all other states there is some information regarding the feeding distribution contained in the line shape of the decay into the state, although a large fraction [(40–60)%] of the population of the state is provided by unobserved "side-feeding" transitions. Measurements of γ-ray multiplicities following spontaneous fission suggest that a yrast state at intermediate spin is fed predominantly by a combination of fast statistical γ rays, and the transition from the next-highest yrast state. Although slow side-feeding components have been observed [1], they constitute only a few percent of the decay intensity. The time distribution of the side feeding was simulated in the fitting procedure by a two-state model with a variable feeding time. To reduce the effect of possible contamination of the line shapes with sharp lines from the heavy fragments, the rotational band (between $I = 6$ and $I = 12$) was assumed to correspond to the rotation of a nucleus of constant intrinsic quadrupole moment (Q). Within this model, the mean lifetime (τ in ps) of a state of spin I, decaying within a rotational band by a stretched $E2$ γ ray of energy E_γ (in MeV), is given by

$$\frac{1}{\tau} \approx 1.217\langle I,0,2,0|I-2,0\rangle^2 Q^2 E_\gamma^5,$$

where $\langle I,0,2,0|I-2,0\rangle$ is a Clebsch-Gordan coefficient and Q is measured in e barns. Q was used to vary the state lifetimes in a manner consistent with this assumption, the best fit to the data resulting in a solution for Q. Although Q may vary somewhat with spin, this procedure gives the most consistent treatment of the data. Nevertheless, it should be born in mind that the value of Q extracted in this analysis represents an effective quadrupole moment over the range $I = 6$ and $I = 12$.

Table I lists the quadrupole moments for eight nuclei in the neutron-rich mass-100 region determined from the line-shape analyses. In all cases the deduced mean feeding times to the highest-spin states whose decays produced observed line shapes were fast, less than 0.5 ps, as would be expected if the predominant feeding mechanism was via high energy statistical transitions. The side-feeding times to the 8^+ and 10^+ states were also found to be short, less than a third of the inband feeding time. Figure 1 shows two examples of fitted line shape spectra: those corresponding to ^{108}Mo and ^{100}Zr.

From Table I it can be seen that the results can be separated into two groups. In the first, comprising the Zr nuclei and ^{98}Sr, the DPM results agree well with measurements at lower spin, quoted in the compilation of Raman *et al.* [6]. In the second group, the Mo isotopes, the DPM results are consistently about 20% lower than those at low spin. The mean ratio of the quadrupole moment deduced from the present DPM analysis to the quadrupole moment at low spin is 0.99 ± 0.04 ± 0.05 for the Sr and Zr nuclei, and 0.80 ± 0.04 ± 0.04 for the Mo nuclei, where the first uncertainty is purely statistical and the second represents the result of including a 10% systematic error on the stopping powers [7]. In the case of ^{104}Zr, there is no determination of the quadrupole moment at low spin with which to compare our result. A comparison of the present DPM results with the ground-state quadrupole moments calculated by Möller *et al.* [8] gives very good agreement for the Sr and Zr nuclei (including ^{104}Zr), whereas the DPM results for the Mo nuclei are significantly lower than their predicted values (see Table I). Given that the band crossing ($\hbar\omega = 0.2$–0.4 MeV) is gradual, these results suggest that the Zr nuclei and ^{98}Sr hold their shape under rotation and that the Mo isotopes experience a deformation change between $I \sim 0$ and $I \sim 10$.



FIG, 1. Line shapes corresponding to the decay of the yrast $I^\pi = 10^+$ states in ^{100}Zr and ^{108}Mo. The data are represented by the histogram and the fit by the solid line.

Although a possible reason for the different behavior of the Mo isotopes could be a change in the magnitude of the quadrupole deformation (β_2) with spin, a more probable origin is connected with triaxial instability. Self-consistent mean-field calculations [9] suggest that the Mo isotopes are significantly softer in their ground-state triaxial deformation than the Zr isotopes. To examine the theoretical problem at nonzero spin, we have performed total Routhian surface (TRS) calculations [10]. The cranked Strutinsky approximation was used to obtain the shape evolution with angular momentum. At each frequency, the shape of the nucleus was minimized [10,11] with respect to the quadrupole and hexadecapole deformations and to the parameter γ, which gives the deviation from axial symmetry.

The energy surfaces for ^{102}Zr and ^{102}Mo (Fig. 2) are presented as representative examples for the purposes of our discussion. A pronounced minimum at axially symmetric prolate shape is obtained for ^{102}Zr at spin zero, and the surface is practically unchanged at $\hbar\omega = 0.38$ MeV ($I \approx 9$). In contrast to ^{102}Zr, ^{102}Mo is very soft with respect to γ deformation with a minimum extending from $\gamma \approx -20°$ to $\gamma \approx +20°$. As the ^{102}Mo nucleus rotates, a pair of $h_{11/2}$ neutrons align their angular momentum with the rotation axis and cause the minimum in the surface to move to positive γ at $\hbar\omega = 0.38$ MeV. The total quadrupole moment Q has contributions from two spherical-tensor components, Q_{20} and Q_{22}. At zero spin the minimum lies at $\gamma = 0$, and the energy surface is symmetric about $\gamma = 0$, resulting in a contribution only from the Q_{20} term. However, as the nucleus rotates, the minimum moves towards positive γ, resulting in a contribution to the quadrupole moment from the (negative) Q_{22} term, and giving a reduction of Q with increasing spin. From the calculations it is difficult to determine generally whether the shift is to negative or positive γ, since the corresponding minima are close in energy. Since a shift in the nuclear mean shape towards negative γ would result in an increase in the quadrupole moment

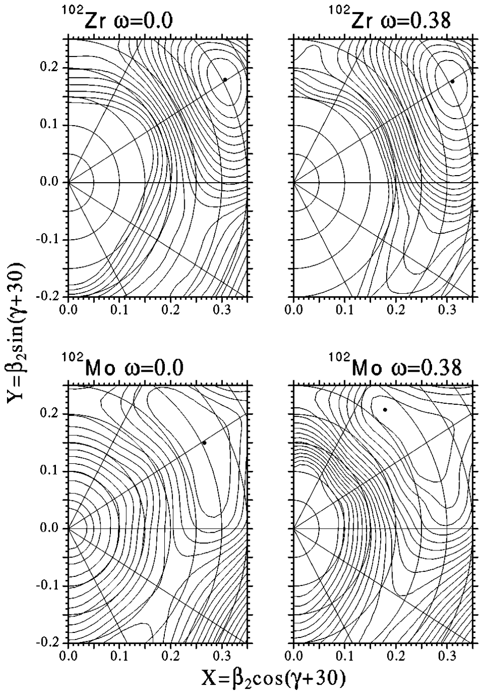


FIG. 2. Total Routhian surfaces for the isobars ^{102}Zr and ^{102}Mo calculated at $\hbar\omega = 0$ and 0.38 MeV. The energy interval between the contours is 200 keV.

with spin, our measurements suggest that the yrast states in ^{102,104,106,108}Mo near $I = 10$ have positive γ deformation. The γ softness of the Mo energy surfaces is consistent with the observation [1] of γ vibrations in ^{106}Mo.

In order to estimate the magnitude of the predicted decrease in the quadrupole moment, we calculated the expectation value of the quadrupole operator from the intrinsic wave functions at the minimum-energy deformation, at $I \sim 10$. Since the β_2 deformation parameter corresponding to the energy minimum does not change much between $I = 0$ and $I = 10$, an estimate of the effective quadrupole moment at $I = 0$ may be found by taking Q_{20} only, and at $I = 10$ by taking the sum $Q_{20} + Q_{22}$. This gives a predicted reduction in the case of ^{102}Mo of the order of 0.6 eb. This approach does not include a precise mixing calculation, nor does it include any effects of rotation-vibration coupling, and as such can only provide an estimate of the magnitude of the change in Q with spin, but nevertheless shows that changes in the triaxial deformation can produce changes in Q that are at least as large as those observed experimentally.

In summary, we have used the DPM to extract information on the quadrupole moments of neutron-rich fission fragments near $A = 100$ at $I \approx 10$. These are the first such measurements, and when compared with measurements at low spin they indicate a reduction in the intrinsic quadrupole moment of the Mo fragments between $I \approx 0$ and $I \approx 10$. The Zr isotopes and ^{98}Sr have quadrupole moments consistent with low-spin measurements. Our TRS calculations suggest that the origin of the change in the quadrupole moments of the Mo isotopes may lie in their softness to γ deformation. The aligning $h_{11/2}$ neutrons can drive the shape towards positive γ deformation and thereby reduce the effective quadrupole moment.

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