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Quadrupole moments of the first excited states of ^{96}Ru, ^{98}Ru, ^{100}Ru, ^{102}Ru, and ^{104}Ru

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The static quadrupole moments of the first 2^+ excited states of ^{96,98,100,102,104}Ru were measured employing the reorientation effect in Coulomb excitation. For constructive interference via the second 2^+ excited states the determined quadrupole moments are (-0.13 ± 0.09) *eb* for ^{96}Ru, (-0.20 ± 0.09) *eb* for ^{98}Ru, (-0.43 ± 0.07) *eb* for ^{100}Ru, (-0.57 ± 0.07) *eb* for ^{102}Ru, and (-0.70 ± 0.08) *eb* for ^{104}Ru. These Q_{2^+} represent, respectively, 29%, 36%, 67%, 78%, and 84% of the rotational value.

[NUCLEAR REACTIONS ^{96,98,100,102,104}Ru(α, α'), $E=8.5$–9.5 MeV; ^{96,98,100,102,104}Ru$(^{16}O, ^{16}O')$, $(^{16}O, ^{16}O'\gamma)$, $E=36$–44.8 MeV; measured $\sigma(E,\theta)$, deduced Q_{2^+}, $B(E\lambda)$, $B(M1)$, J, π, $T_{1/2}$. Enriched targets.]

I. INTRODUCTION

As has been pointed out by several theoretical studies,[1-5] the neutron-rich nuclei in the mass region A around 100 may belong to a new region of deformation. Indeed many experimental studies have clearly shown that deformations occur in the neutron-rich Zr, Mo, and Ru nuclides.[6-11] However, according to some theoretical calculations[3,5] these nuclei, in contrast to those belonging to the rare-earth and actinide regions, are softer on both beta and gamma deformations. The softness in the gamma direction usually brings about such effective rotation-vibration interactions that pure rotational bands cannot occur. Thus, these nuclei should be always characterized by a more or less marked destruction of the rotational structure. Experimental studies of fission product decay[6,11] seem to confirm this feature in the neutron-rich Ru nuclei and, less markedly, also in the molybdenum ones. For instance, the value of the E_{4^+}/E_{2^+} ratio increases monotonically from ^{98}Mo $(N=56)$ $(E_{4^+}/E_{2^+}=1.92)$ to ^{106}Mo $(N=64)$ $(E_{4^+}/E_{2^+}=3.04)$, whereas the same ratio increases more slowly in the Ru isotopes reaching a constant value of 2.75 in ^{108}Ru $(N=64)$ up to ^{112}Ru $(N=68)$. Thus, it would appear that the deformation characteristics of the neutron-rich Ru nuclei taper off more rapidly than in the neighboring Mo nuclei. This trend is somewhat unexpected since in the vicinity of the $N=50$ closed shell the Ru nuclides display, apparently, a more pronounced collective nature than the corresponding Mo isotopes.[12-14]

A promising and direct way of exploring the deformation properties of nuclei is to measure the static quadrupole moments of their first 2^+ excited states.[15] This is easily realized for the Ru nuclei since several stable even-A isotopes are available over a large and important nuclear region (from $N=54$ to $N=62$). The present report can be considered as a part of a systematic study of the electromagnetic properties of nuclei around the region $A=100$ (Refs. 16–19) and it should be related particularly to a similar investigation performed on the even-A Mo nuclei.[19]

II. EXPERIMENTAL PROCEDURES AND RESULTS

The present experiment consists of two parts: gamma spectroscopy (or multiple Coulomb excitation measurements) carried out by the thick-target γ-ray yield method employing 44.8 MeV ^{16}O ions, and particle spectroscopy performed with α and ^{16}O beams. Only a brief description of the experimental and data reduction procedures will be given here since these techniques have been already described in detail elsewhere.[16-20]

A. Gamma spectroscopy

To derive the static quadrupole moment of the first 2^+ excited state of even nuclei one has to insert in the appropriate program (see below) the reduced matrix elements M_{rs} of the quadrupole operator. These matrix elements are obtained from $B(E2)$ values which are usually determined by Coulomb excitation measurements.

^{16}O ions (44.8 MeV) from the University of Montreal tandem Van de Graaff accelerator were used to bombard targets of ^{96,98,100,102,104}Ru whose isotopic enrichment is given in Table I. The targets were evaporated onto a thick tantalum backing and ranged in thickness from 30 to 50 mg/cm^2. The thick-target γ-ray yields were measured positioning the target at 45° with respect to the incident beam and with a 90 cm^3 Ge(Li) detector having a 2.2 keV resolution at 1.33 MeV. The detector was located at 10 or 15 cm from the target depending on the yield of the experiment and at 55° with re-

TABLE I. Isotope composition of targets in percent. All material was obtained from Oak Ridge Separated Isotopes Divisions.

Target	Isotopes 96	98	99	100	101	102	104
96	98.07(0.1)	0.12(0.03)	0.39(0.03)	0.30(0.03)	0.32(0.03)	0.49(0.03)	0.30(0.03)
98	0.56	89.34	3.61	1.70	1.71	2.17	0.90
100	<0.05	<0.05	0.54(0.05)	97.24(0.1)	1.20(0.05)	0.83(0.05)	0.19(0.05)
102	0.02(0.01)	0.01(0.01)	0.07(0.01)	0.09(0.01)	0.24(0.01)	99.35(0.03)	0.22(0.01)
104	0.02(0.01)	0.01(0.01)	0.06(0.01)	0.07(0.01)	0.13(0.01)	0.34(0.01)	99.35(0.03)

spect to the incoming beam. The γ rays which are attributed to the even-A Ru nuclei are presented in the level schemes of Fig. 1.

From the measured γ-ray yields the reduced transition probabilities were calculated by means of the first- and second-order time dependent perturbation theory of Alder *et al.*[21] The stopping power values were taken from the table of Northcliffe and Schilling.[22] The results of these measurements are summarized in Table II where the data obtained in a previous work performed with the same techniques are also shown for comparison.[23]

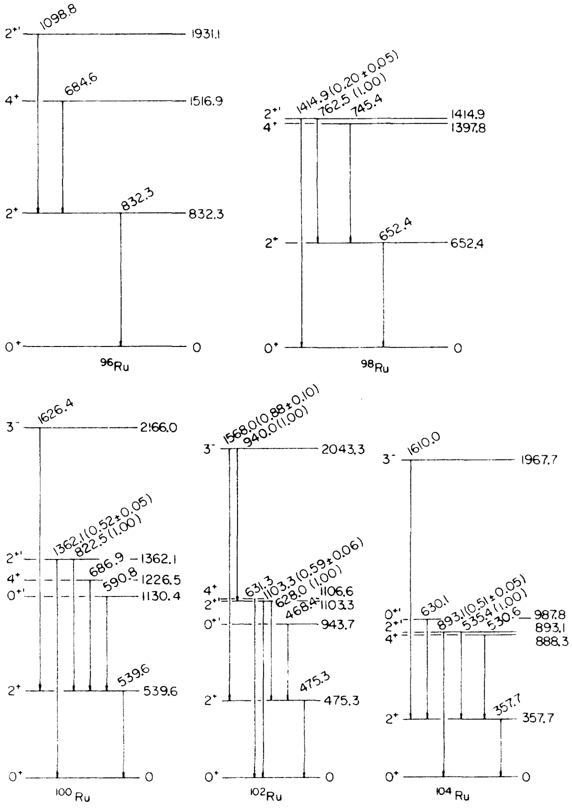

FIG. 1. Level schemes of ^{96,98,100,102,104}Ru as deduced from the present Coulomb excitation measurements. The energy and intensity values of the transitions in these nuclei have been measured in this work. The $2^+ \rightarrow 0^+$ transitions have an error of ±0.1 keV whereas the energy of the other γ rays are determined with a precision between ±0.2 and ±0.5 keV.

Lederer *et al.*[12] in their in-beam γ-ray spectroscopy experiment proposed a second 2^+ level in ^{96}Ru at 1477 keV de-exciting to the 833 keV first excited state and to the ground state via the 644 and 1477 keV transitions. This state does not appear to be excited in the present study, casting some doubt on its existence, since if the $B(E2;\ 2^{+\prime} \rightarrow 2^+)/B(E2;\ 2^+ \rightarrow 0^+)$ had a value as large as (or even lower) that of the equivalent ratios in the other Ru nuclei, the $2^{+\prime} \rightarrow 2^+$ 644 keV photopeak should show very clearly in the singles excitation spectra. The existence of the 1477 keV level, also, was not confirmed in a recent investigation on the decay properties of the $^{96}\mathrm{Rh}^{m,g}$ isomeric pair.[24] γ rays of 644 and 1478 keV were detected in that study. These transitions, however, could be placed elsewhere in the ^{96}Ru decay scheme via coincidence measurements.

A weak 1099 keV γ ray was detected in the ^{96}Ru Coulomb excitation spectra. This photopeak was not observed in the spectra of the other even-A Ru isotopes so that it can be assigned to ^{96}Ru. An equivalent transition de-exciting a level at 1931 keV in ^{96}Ru was found by Gujrathi *et al.*[24] in the decay of $^{96}\mathrm{Rh}^m$. Those authors inferred a spin of 2^+ or 3^+ for the 1931 keV level. Since this level seems to be excited in the present Coulomb excitation experiments, we assign 2^+ to this state and consequently calculate its $B(E2;\ 2^{+\prime} \rightarrow 2^+)$ value (see Table II) [the faint possibility of a 0^+ assignment to the 1931 keV level is discarded since it would give an unrealistic $B(E2)$ value to the 1099 keV transition].

From an inspection of the results shown in Table II it can be observed that the phonon-model prediction,[25,26] i.e., $B(E2;\ J \rightarrow 2^+)/B(E2;\ 2^+ \rightarrow 0^+) = 2.0$ for $J = 4^+$, 2^+, and 0^+, is far from being satisfied in the Ru nuclei. Generally the $B(E2;$

TABLE II. Coulomb excitation results extracted from the thick-target yield measurements.

	E_γ (keV)	$J_i \to J_f$	$B(E\lambda;J_i \to J_f)$ (10^{-2} e^2b^2) a	b	δ	$B(M1;J_i \to J_f)$ $(e\hbar/2Mc)^2$	$\tau_{1/2}$ (psec)	$\frac{B(E\lambda)}{B(E\lambda)_{\rm s.p.}}$ [f]	$\frac{B(E2;J_f \to 2^+)}{B(E2;2^+ \to 0^+)}$	$\beta_{J_iJ_f}$ [f]
^{96}Ru	832.5	$0^+ \to 2^+$	26.6 ± 2.6	26.8 ± 3.1			2.7 ± 0.2	20.4		0.163
	684.4	$2^+ \to 4^+$	9.8 ± 1.3				6.9 ± 0.9	20.9	1.08 ± 0.25	0.117
	1098.5	$2^+ \to 2^{+\prime}$	9.1 ± 1.3		$E2$			34.8	1.71 ± 0.41	0.150
	1931.0	$0^+ \to 2^{+\prime}$	0[c]							
^{98}Ru	652.7	$0^+ \to 2^+$	38.9 ± 3.1	39.9 ± 4.8			6.1 ± 0.6	29.0		0.194
	745.6	$2^+ \to 4^+$	19.4 ± 2.2				2.1 ± 0.3	40.2	1.41 ± 0.27	0.162
	762.0	$2^+ \to 2^{+\prime}$	14.7 ± 2.5		$E2$			54.9	1.89 ± 0.47	0.189
	1414.7	$0^+ \to 2^{+\prime}$	0.67 ± 0.11	0.74 ± 0.11			1.2 ± 0.3	0.5		0.026
^{100}Ru	539.6	$0^+ \to 2^+$	48.2 ± 2.6	51.1 ± 6.1			12.8 ± 0.7	35.0		0.213
	591.0	$2^+ \to 0^{+\prime}$	1.91 ± 0.29	<2.3			8.2 ± 1.2	34.7	0.99 ± 0.20	0.150
	686.9	$2^+ \to 4^+$	26.0 ± 2.2	26.2 ± 2.6			2.6 ± 0.2	52.5	1.5 ± 0.21	0.184
	822.5	$2^+ \to 2^{+\prime}$	8.8 ± 1.3	9.10 ± 1.55	3.2[d]	$(4.1 \pm 0.7) \times 10^{-3}$		32.0	0.91 ± 0.18	0.144
	1362.1	$0^+ \to 2^{+\prime}$	2.03 ± 0.23	1.85 ± 0.25			1.0 ± 0.1	1.5		0.044
		$0 \to 3^-$	4.32 ± 0.71					10.4		0.115
			[7.64 ± 1.40]					[18.4]		[0.152]
^{102}Ru	475.0	$0^+ \to 2^+$	65.1 ± 3.5	63.9 ± 7.7			18.0 ± 1.0	46.0		0.244
	468.5	$2^+ \to 0^{+\prime}$	1.99 ± 0.29				25.0 ± 4.0	35.2	0.76 ± 0.15	0.151
	627.9	$2^+ \to 2^{+\prime}$	11.7 ± 1.5	8.19 ± 1.19	$E2$			41.4	0.90 ± 0.15	0.164
	1103.1	$0^+ \to 2^{+\prime}$	2.1 ± 0.2	1.65 ± 0.19			3.1 ± 0.3	1.5		0.043
	631.2	$2^+ \to 4^+$	38.1 ± 4.2	36.2 ± 6.9			2.7 ± 0.3	74.9	1.6 ± 0.3	0.220
		$0^+ \to 3^-$	6.54 ± 0.97					15.1		0.138
			[7.63 ± 1.15]					[17.6]		[0.149]
^{104}Ru	357.9	$0^+ \to 2^+$	83.4 ± 4.4	81.0 ± 9.7			58.0 ± 3.0	57.4		0.273
	530.6	$2^+ \to 4^+$	43.1 ± 4.7	39.0 ± 6.9			5.6 ± 0.6	82.6	1.4 ± 0.2	0.231
	535.2	$2^+ \to 2^{+\prime}$	16.7 ± 2.0	12.3 ± 1.9	−9.0[e]	$(4.1 \pm 0.5) \times 10^{-4}$		57.5	1.0 ± 0.2	0.193
	893.0	$0^+ \to 2^{+\prime}$	3.36 ± 0.35	2.76 ± 0.30			5.0 ± 0.5	2.3		0.055
	630.1	$2^+ \to 0^{+\prime}$	1.45 ± 0.15	1.52 ± 0.15			7.9 ± 0.9	25.0	0.43 ± 0.06	0.127
		$0^+ \to 3^-$	5.79 ± 0.35					12.9		0.128
			[8.22 ± 1.07]					[18.3]		[0.152]

[a] This work. The values in brackets for the 3^- states are those calculated considering the experimentally observed transitions and upper limits of all possible γ rays de-exciting the 3^- levels. The other values are those calculated considering only the experimentally observed transitions.

[b] Reference 23.

[c] No 1931 keV γ ray was observed in the ^{96}Ru singles spectra nor in Ref. 24 (see below). However considering an upper limit of $I_{1931} = \frac{1}{50} I_{1098}$ values of 2.45×10^{-5} e^2b^2 and 4.13×10^{-3} e^2b^2 can be calculated for $B(E2;0^+ \to 2^{+\prime})$ and $B(E2;2^+ \to 2^{+\prime})$, respectively. The Q_{2^+} for ^{96}Ru shown in Table IV is not practically affected by these two possible values of $B(E2;0^+ \to 2^{+\prime})$ and $B(E2;2^+ \to 2^{+\prime})$.

[d] Nucl. Data Sheets, Vol. 11B, 279 (1974).

[e] Reference 23.

[f] For the calculation of $B(E\lambda)_{\rm s.p.}$ and $\beta_{J_iJ_f}$ see J. Barrette *et al.*, Nucl. Phys. A235, 154 (1974).

$4^+ \to 2^+)/B(E2;\ 2^+ \to 0^+)$ agree better with the value of 1.4 given by the rigid triaxial rotor model.[27] On the other hand, only in ^{96}Ru and ^{98}Ru the $B(E2;\ 2^{+\prime} \to 2^+)/B(E2;\ 2^+ \to 0^+)$ are in fair agreement with the phonon-model, whereas in ^{100}Ru, ^{102}Ru, and ^{104}Ru this ratio is approximately equal to 1. In these latter nuclei, also, the $B(E2;\ 0^{+\prime} \to 2^+)/B(E2;\ 2^+ \to 0^+)$ are considerably smaller than the theoretical ratio. Furthermore, they decrease rapidly with the increase of the neutron number.

3^- states have been Coulomb excited in three of the five studied isotopes. In ^{104}Ru the decay pattern of the 3^- level at 1970 keV agrees very well with that previously evinced.[28,29] These states clearly show collective features and our data are in excellent agreement with those found in (d,d') (^{102,104}Ru) and (α,α') (^{100}Ru) experiments.[30,31] It should be remarked, however, that the 3^- levels in the Ru nuclei are markedly less collective than the corresponding states of the Mo nuclei.[17]

B. Particle spectroscopy

Beams of α and ^{16}O particles were used for the inelastic scattering measurements. The various Ru targets were prepared by evaporation in vacuum of the enriched isotopic material (see Table I) on 10 or 20-μg/cm^2- thick carbon backings. Target thickness ranged from 3 to 30 μg/cm^2. The Coulomb excitation probabilities for both projectiles were determined by direct measurements of elastic and inelastic yields observed in four surface-barrier detectors placed at scattering angles of ±157.5° and ±172.5°. The spectra were obtained at bombarding energies ranging from 8 to 9.5 MeV for ^{4}He and from 36.0 to 37.2 MeV for ^{16}O. The energy resolution was approximately 30 keV for the alpha particles and varied from 100 to 130 keV for the ^{16}O ions depending on the target thickness and scattering angle. Typical alpha and ^{16}O spectra are shown in Fig. 2 (^{96}Ru) and Fig. 3 (^{104}Ru). The ratios $R_{\text{exp}} = d\sigma_{\text{inel}}/d\sigma_{\text{el}}$ were extracted from the data after the contributions from the isotopic impurities were subtracted from the spectra (see Table I). Particular care was also taken to detect possible target contaminants which at the bombarding energies used could produce elastic peaks underneath the Ru isotope inelastic peaks. These contaminants are isotopes with masses from $A = 54$ to $A = 76$ for the α data, and Mo and Zr for the ^{16}O data. To this end the various ruthenium thin targets were bombarded with 1.6 and 3 MeV proton beams and their elemental analysis was carried out by PIXE methods and techniques developed in this laboratory.[32-34] It was found that all the Ru targets were free of the contaminants which could have affected the ^{16}O data. However, relatively large amounts of Zn, Cu, Fe, and Ni were detected in all targets. The presence of these elements did not hamper the analysis of the α spectra of ^{100}Ru, ^{102}Ru, and ^{104}Ru, but it affected the data of ^{96}Ru and ^{98}Ru which could be analyzed

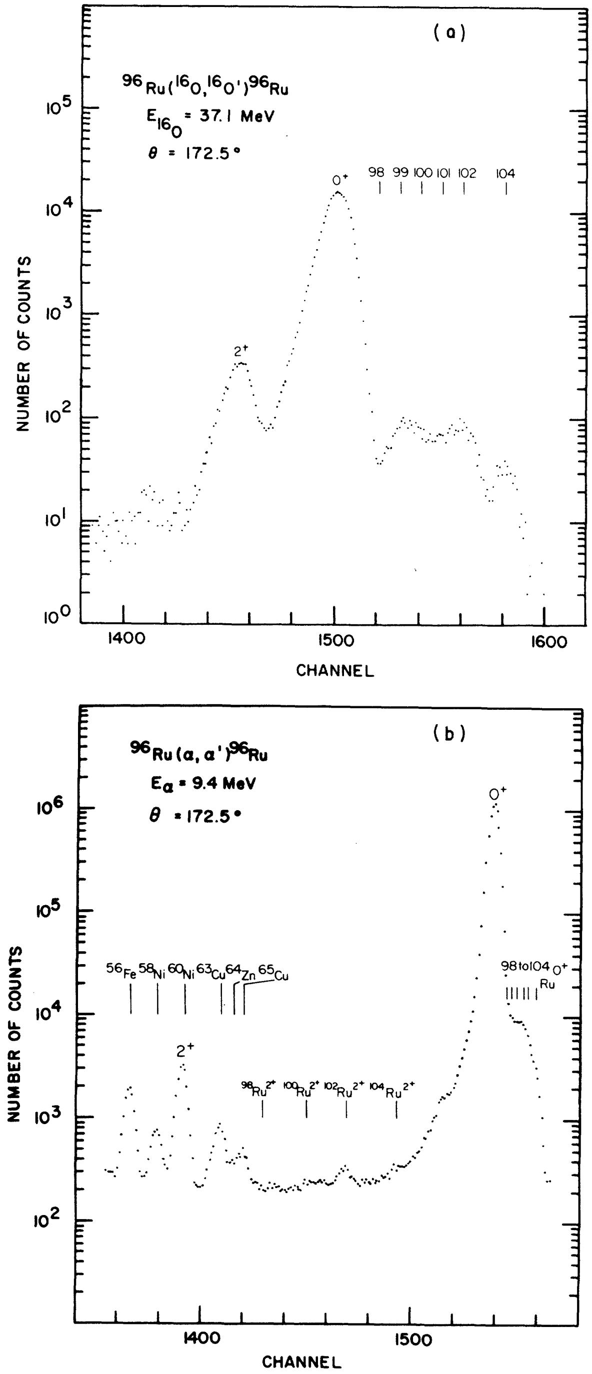

FIG. 2. (a) The ^{16}O (37.1 MeV) spectrum from ^{96}Ru at a scattering angle $\theta_{\text{lab}} = 172.5°$. (b) The α (9.4 MeV) spectrum of ^{96}Ru at a scattering angle $\theta_{\text{lab}} = 172.5°$.

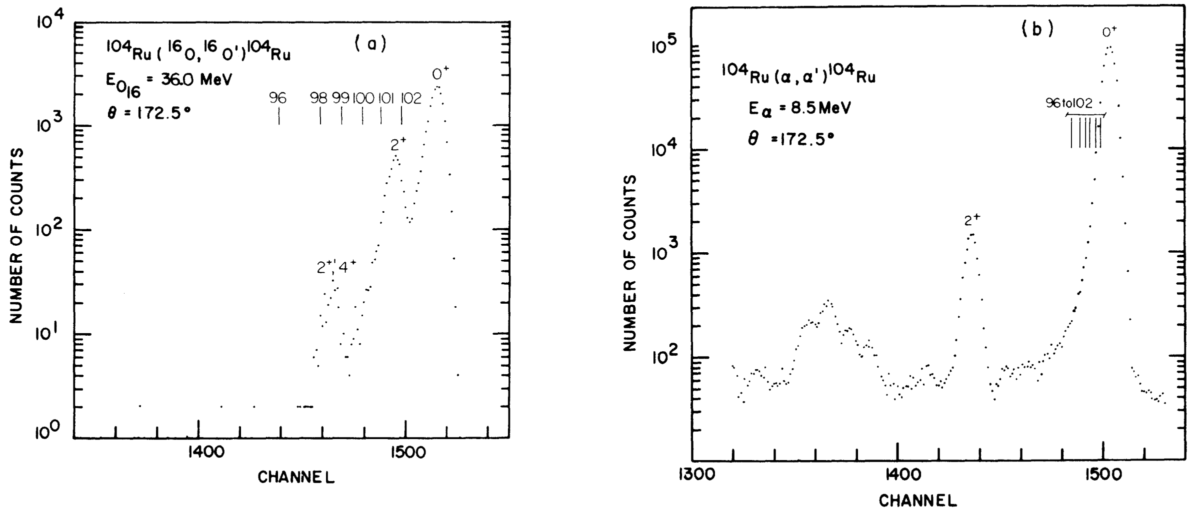

FIG. 3. (a) The ^{16}O (36.0 MeV) spectrum from ^{104}Ru at a scattering angle $\theta_{\text{lab}} = 172.5°$. (b) The α (8.5 MeV) spectrum of ^{104}Ru at a scattering angle $\theta_{\text{lab}} = 172.5°$.

only at some angles by a suitable choice of the α beam energy. Table III summarizes the R_{exp} values of all Ru nuclei. Finally the Q_{2^+} and $B(E2; 0^+ \rightarrow 2^+)$ values were assessed by the appropriate program[35] employing the reduced matrix elements M_{rs} calculated from the $B(E2)$ values given in Table II. The energy levels included in the analysis are those shown in Fig. 1 with the exception of the 3^- states. The final results are summarized in Table IV. In this table the values of the static quadrupole moments measured by other groups with a variety of methods are also presented for comparison.[36-39]

III. DISCUSSION

Only the Q_{2^+} values obtained from the constructive interference term via the second 2^+ excited states ($P_3 > 0$ in our notation) will be considered

TABLE III. Values of the experimental and least-square-fitted ratios.

Isotope	Beam energy (MeV)	Lab angle (deg.)	$R_{\text{exp}} \times 10^3$ [a]	$R_{\text{fit}} \times 10^3$ [b]
96	9.4 (^{4}He)	172.5	2.330 ± (0.84)	2.331
	37.1 (^{16}O)	157.5	20.60 ± (1.55)	20.55
		172.5	20.95 ± (1.57)	20.98
98	8.5 (^{4}He)	172.5	3.388 ± (1.33)	3.319
	9.5 (^{4}He)	172.5	5.734 ± (1.40)	5.980
	36.0 (^{16}O)	157.5	42.75 ± (1.70)	42.82
		172.5	44.20 ± (2.22)	43.95
100	8.0 (^{4}He)	157.5	4.076 ± (1.10)	4.161
		172.5	4.364 ± (1.08)	4.319
	8.5 (^{4}He)	157.5	5.604 ± (2.30)	5.656
		172.5	5.986 ± (1.88)	5.884
	36.0 (^{16}O)	157.5	72.50 ± (0.92)	72.71
		172.5	75.00 ± (1.01)	74.87
102	9.0 (^{4}He)	157.5	10.96 ± (1.17)	10.99
		172.5	11.50 ± (1.07)	11.49
	37.2 (^{16}O)	157.5	127.70 ± (1.25)	126.40
		172.5	129.25 ± (1.29)	130.60
104	8.5 (^{4}He)	157.5	14.38 ± (0.94)	14.31
		172.5	14.91 ± (0.98)	15.01
	36.0 (^{16}O)	157.5	187.65 ± (1.65)	186.70
		172.5	192.75 ± (1.69)	193.70

[a] The experimental errors for R_{exp} are statistical only and are quoted in percent.
[b] The fitted ratios are those obtained for a positive value of $P_3 = M_{02'} M_{22'} M_{02}$.

TABLE IV. Summary of the results for the $B(E2:0^+\rightarrow 2^+)$ and Q_{2^+} values obtained from the present study and from other experiments.

		$B(E2:0^+\rightarrow 2^+)$ (e^2b^2)	Q_{2^+} (eb)				
Isotope	P_3	Present work	Present work	Ref. 36	Ref. 37	Ref. 38	Ref. 39
96		0.236 ± 0.007	-0.13 ± 0.09	-0.08 ± 0.19			
				$[-0.19 \pm 0.19]$[a]			
98	+	0.373 ± 0.007	-0.20 ± 0.09	-0.03 ± 0.14			
	−	0.372 ± 0.007	-0.01 ± 0.09	$[-0.17 \pm 0.14]$			
100	+	0.494 ± 0.006	-0.43 ± 0.07	-0.13 ± 0.07			
	−	0.492 ± 0.006	-0.20 ± 0.07	$[-0.30 \pm 0.07]$			
102	+	0.640 ± 0.006	-0.57 ± 0.07	-0.38 (assumed)	-0.37 ± 0.24		-0.68 ± 0.08
	−	0.640 ± 0.006	-0.35 ± 0.07	$[-0.57$ (assumed)$]$			
104	+	0.834 ± 0.007	-0.70 ± 0.08	-0.66 ± 0.05	-0.84 ± 0.21	-0.63 ± 0.20	
	−	0.835 ± 0.007	-0.35 ± 0.08	$[-0.89 \pm 0.05]$			

[a] The values of Q_{2^+} in brackets are those of Maynard *et al.*[36] renormalized to the Q_{2^+} of ^{102}Ru found in the present work.

here since these values are strongly favored from experimental and theoretical considerations.[40,41] A close examination of the results shown in Table IV shows us the following two points: (i) a prolate deformation is favored for the Ru nuclei, (ii) there is an increase in deformation with an increase of the neutron number. These two features are common to all doubly even isotopes around $N=50$ (with the exception of the Cd nuclei which do not show evidence of significant variations in Q_{2^+} and $Q_{2^+}/Q_{2^+_{\rm rot}}$ with mass number[42]) and are in agreement with the calculations of Tanaka and Tomoda,[5] Faessler *et al.*,[4] and Bucurescu *et al.*[43] Another theoretical approach predicts the same increase in deformation[1] but favors an oblate shape which is certainly not the case in point. Very recently Koo and Tassie[44] carried out a model independent energy weighted sum rule calculation to determine the Q_{2^+} for a large number of even-A isotopes. For the Ru nuclei the agreement with our experimental results is good even though the Q_{2^+} of ^{96}Ru and ^{98}Ru seem slightly overemphasized in the theoretical calculation. Furthermore, our data do not indicate the interruption of the regular increase of Q_{2^+} in ^{102}Ru shown in the calculation of Koo and Tassie.[44]

A number of nuclear models have been invoked to explain the structure properties of the even-A Ru nuclei as the rigid triaxial rotor model of Davydov and Filippov,[27] the generalized triaxial rotor which includes β vibrations,[45,46] the interacting boson approximation of Arima and Iachello,[47] and the generalized collective model of Gneuss and Greiner.[3] For ^{100}Ru, ^{102}Ru, and ^{104}Ru the experimental data seem to agree better with the predictions of the asymmetric rotor model.[48,49,29] The experimental data on ^{96}Ru and ^{98}Ru are still too scanty to have a firm comparison with theoretical predictions. It can be observed, however, that also in these two latter nuclei the $B(E2;\ 4^+\rightarrow 2^+)/B(E2;\ 2^+\rightarrow 0^+)$ value is in agreement with the triaxial rotor model expectation. Thus, it would appear that the Ru nuclei show a triaxial deformation even near the neutron closed shell $N=50$. This is in contrast to the even-A Mo isotopes whose features seem to be more classically "vibrational" at least up to ^{100}Mo.[16,17] Triaxial deformations for the Ru nuclei are, however, predicted also theoretically.[4,5,43]

As a conclusion, this work as well as other experimental studies show that the onset of deformation is not a clearcut feature at $A\sim 100$ since the isotopic chains in this region behave much less uniformly than in the traditional deformed regions as at $N=90$.

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