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A B S T R A C T

A multi-step Coulomb excitation measurement with the GRETINA and CHICO2 detector arrays was carried out with a 430-MeV beam of the neutron-rich $^{110}_{\text{Ru}}$ ($t_{1/2} = 12$ s) isotope produced at the CARIBU facility. This represents the first successful measurement following the post-acceleration of an unstable isotope of a refractory element. The reduced transition probabilities obtained for levels near the ground state provide strong evidence for a triaxial shape; a conclusion confirmed by comparisons with the results of beyond-mean-field and triaxial rotor model calculations.

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The shape is a fundamental property of the atomic nucleus and, while the majority of nuclei are thought to be axially symmetric, triaxial deformation has been a subject of much recent interest in structure studies. So far, triaxial deformation has been established to occur in specific nuclei at high angular momenta [1–3] through either the wobbling motion [4] or the breaking of chiral symmetry [5] under rotation. Furthermore, its presence at or near the ground state has been studied extensively in the past, primarily via Coulomb excitation of stable nuclei [6–9], without reaching definitive conclusions for any region of the nuclear chart as the nuclei involved are more commonly viewed as “y-soft” rather than as rigidly triaxial. In addition to the general question of whether nuclei can exhibit these exotic shapes, this issue is particularly intriguing given the anticipated influence on nuclear binding energies and thus on a number of astrophysical processes [10,11]. The results of several microscopic calculations indicate that the

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neutron-rich molybdenum and ruthenium isotopes are among the nuclei with the best prospects for the observation of low-spin triaxial phenomena, mainly due to the occupation of the neutron $1f_{7/2}$ and proton $1p_{3/2}$ intruder orbitals [12–16]. However, this is a challenging issue to address as the shape degrees of freedom ($\beta$, $\gamma$, etc.) are defined in the intrinsic frame of the nucleus whereas observations are made in the laboratory, and one has to rely on the Kumar–Cline sum rules [17,18], which require knowledge of several experimental $E2$ matrix elements, to provide the bridge between the two.

The level structure of $^{110,112}$Ru has been investigated by prompt $\gamma$-ray spectroscopy of fission fragments [19–24] and in beta decay [25,26]. The excitation energy of the first $2^+$ state reaches a near constant value between $^{110}$Ru and $^{112}$Ru; i.e., close to the $N = 66$ mid-shell point, suggesting constant quadrupole deformation. However, the ratio between the excitation energies of the first $2^+$ and $4^+$ states never reaches the rotational limit for axial symmetry ($\epsilon_{22} = 10/3$), in contrast to the observations in the neighboring Sr and Zr isotopes. In addition, it has been shown that the energy of the $2^+_2$ states decreases with increasing neutron number for the Ru isotopes, thus indicating an increasing susceptibility to the triaxial (or $\gamma$) degree of freedom. In $^{110}$Ru, the $2^+_2$ level is observed to be energetically below the $4^+_1$ state, a strong indication of triaxiality according to the Rigid Triaxial Rotor Model (RTRM) of Davydov et al. [27,28]. In fact, the Mo–Ru isotopes have the lowest known $2^+_2$ states of any nucleus, with the exception of $^{192}$Os [29]. In addition, the relation $E(3^+_1) = E(2^+_1) + E(2^+_2)$, indicating a possible breaking of axial symmetry, is satisfied in $^{110–114}$Ru.

It is important to note that such empirical criteria for triaxiality are only fulfilled in one other area of the nuclear chart; i.e., in the neutron-rich Os and Pt isotopes [6], but in this case the phenomenon is limited to only a few isotopes, in contrast to a sizeable region.

Nevertheless, indisputable evidence for stable, non-axial nuclear shapes close to the ground state based solely on the analysis of energy spectra is notoriously difficult to obtain [30]. It is more instructive, therefore, to consider reduced transition probabilities in this type of analysis. Complications arise as $B(E2)$ values or transition quadrupole moments in the ground-state band are only weakly affected by gamma deformation [31], although a recent study [32] reported such moments in the $8^{-}\to 6^{-}$ bands in $^{108–112}$Ru and discussed the results in terms of $\gamma$-softness or possible triaxiality. More sensitive indications of triaxiality are the static quadrupole moments and the $B(E2)$ reduced transition probabilities between levels of an excited, $K = 2$, band and states in the ground-state band [31]. In particular, Hartree–Fock–Bogolyubov (HFB) based calculations going beyond the mean-field approach using the Generator Coordinate Method (GCM) with the Gaussian Overlap Approximation (GOA), as well as results obtained with phenomenological triaxial rotor models [27,28] predict reduced quadrupole moments and large $B(E2; J \to J)$ values between $\Delta K = 2$ bands. For the ruthenium isotopes, however, such absolute $B(E2)$ values between bands were thus far experimentally unknown beyond stable $^{104}$Ru, which was found to be characterized by a prolate-triangular shape with a degree of $\gamma$-softness [7].

In this Letter, results from the first multi-step Coulomb excitation study of $^{110}$Ru are reported with the aim of searching for evidence of a triaxial shape close to the ground state, and of here-with providing new information on shape evolution in the Ru isotopes. This unique study represents the first post-acceleration of an unstable, isotope of a refractory element and can only now be realized thanks to the CARIBU facility [33]. Despite long-standing interest, such studies have hitherto been impossible due to the physical properties of refractory metals, which prevent their release from the targets used at traditional isotope separation on-

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**Fig. 1.** (Color online.) The CHIC02 particle spectrum. The plot displays the difference in ToF between the beam and target nuclei versus scattering angle, $\theta$. The Pb recoils and various beam contaminants are labeled.

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**Fig. 2.** (Color online.) Doppler corrected $\gamma$-ray energy spectrum gated on the $A=110$ group in the CHIC02 spectrum. A number of $^{110}$Cd peaks (labeled in green) are visible in additions to the $^{110}$Ru $\gamma$ rays (red).

line (ISOL) facilities [33,34]. Furthermore, the measurement reported here also benefits from the superior Doppler reconstruction achieved by the combination of the $\gamma$-ray tracking capabilities of the GRETINA array [35] and a highly-segmented particle detector, CHIC02 [36].

The experiment was performed at the Argonne Tandem Linac Accelerator System (ATLAS). As stated above, the $^{110}$Ru beam was provided by the CARIBU facility [33,38,39], where, starting from a $\sim 1.7$Ci $^{252}$Cf source, fission fragments were efficiently thermalized and turned into a beam of $1^+$ charged ions by a gas catcher. The $^{110}$Ru$^{1+}$ ions were then selected through the isobar separator and directed to a dedicated ECR source for charge breeding (to $q = 21^+$) before being sent to the ATLAS linac for subsequent acceleration to 430 MeV. The $^{110}$Ru beam impinged on a 1.5 mg/cm$^2$–thick $^{208}$Pb target (99.9% enrichment) located at the center of the GRETINA + CHIC02 experimental apparatus. The intensity of the radioactive beam was monitored at the beam dump by measuring the yield of $\gamma$ rays associated with its beta decay (see Ref. [37] for details). On average, 2000 $^{110}$Ru ions per second hit the target. Gamma rays from multi-step Coulomb excitation were measured by the GRETINA tracking array [35] in coincidence with scattered reaction partners detected in the CHIC02 heavy-ion counter [36]. For this experiment, GRETINA consisted of 8 modules with 4 segmented HPGe detectors each. Otherwise the experimental apparatus was essentially identical to that described in Ref. [37]. With the described setup, a resolution of 4.5 keV was achieved for the 658-keV transition in $^{110}$Cd, which is shown in Fig. 2.

A representative time-of-flight (ToF) particle histogram is given in Fig. 1 while a $\gamma$-ray spectrum gated on mass 110 reaction prod-

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The table presents the 

\[ \begin{array}{c|cc|c|c} 
\hline 
I^+ \rightarrow I'^+ & E_\gamma (keV) & B(E2; I \rightarrow I') (e^2 fm^4) & \text{Experimental} & \text{GTRM} & \text{HFB-GCM(GOA)} \\
\hline 
2^- \rightarrow 0^- & 241 & 0.209^{+0.015}_{-0.016} & 0.209 & 0.218 \\
4^- \rightarrow 2^- & 423 & 0.293^{+0.011}_{-0.021} & 0.309 & 0.344 \\
6^- \rightarrow 4^- & 576 & 0.38^{+0.04}_{-0.04} & 0.38 & 0.45 \\
2^- \rightarrow 2^- & 372 & 0.35^{+0.13}_{-0.15} & 0.20 & 0.30 \\
2^- \rightarrow 0^- & 613 & 0.023^{+0.001}_{-0.001} & 0.023 & 0.028 \\
4^- \rightarrow 2^- & 247 & 0.31^{+0.3}_{-0.2} & 0.37 & 0.38 \\
2^- \rightarrow 4^- & 619 & 0.015^{+0.002}_{-0.002} & 0.041 & 0.003 \\
\hline 
\end{array} \]

This table contains the experimental reduced transition probabilities for low-lying states in \(^{110}\text{Ru}\), determined from the GOSIA fit to the experimental data, together with the results obtained with the two theoretical approaches discussed in the text. The bottom section of the table displays the spectroscopic quadrupole moments, \(Q_i\), determined for the \(2^-\) level in the present work (see text for details).
spectroscopic moment for the $2^+_1$ state. This provides confidence in the analysis procedures and in the results discussed below.

The reduced transition probabilities are presented in Table 1 together with their associated uncertainties. Table 1 also provides the extracted spectroscopic quadrupole moment, $Q_s$, for the $2^+_1$ state. Note that the $3^+_1 \rightarrow 2^+_2$ transition is weak and was not directly observed in the spectra. Nevertheless, a transition probability could be derived with confidence from the GOSIA fit, based on the $3^+_1 \rightarrow 2^+_2$ transition intensity measured in the present experiment and on the published branching ratio [25].

In order to aid in the interpretation of the experimental results, extensive beyond-mean-field calculations using the HFB-GCM(GOA) approach [45] with the Gogny D1S force [46,47] as well as calculations with the Generalized Triaxial Rotor Model (GTRM) [48–51] have been performed for $^{110}$Ru. The results are also presented in Table 1, while a comparison between experimental and calculated level energies, transition probabilities and the $Q_s$ moment for the $2^+_1$ state, for both approaches, is provided in Fig. 3. Note that the GTRM approach differs from the RTRM model of Davydov et al. [27,28] as it overcomes a number of limitations of the RTRM, such as irrotational moments of inertia [49], for example.

It is clear that the data are well reproduced. In particular, the strong coupling between $K = 2$ and $K = 0$ states, which manifests itself through a large $2^+_2 \rightarrow 2^+_1$ matrix element, is borne out by the data as are the excitation energies of the $2^+_2$ and $3^+_1$ states. These observables and, in particular, the relatively large $2^+_2 \rightarrow 2^+_1$ and small $2^+_1 \rightarrow 0^+_1$ matrix elements, are strong indications of triaxial deformation. In fact, in the GTRM calculations, the $\gamma$ deformation parameter has a value of $\gamma = 29^\circ$ and a $\beta_2$ deformation of $\beta_2 = 0.31$. The extracted gamma deformation for $^{110}$Ru is the closest to $30^\circ$ of all candidate triaxial nuclei investigated with multi-step Coulomb excitation data. Hence, the present study establishes the possibility of triaxiality near the ground state for a region other than Os–Pt and, in many regards, makes $^{110}$Ru the best candidate for triaxiality to date. The HFB-GCM(GOA) calculations yield similar deformation parameters of $\gamma = 26^\circ$ and $\beta_2 = 0.29$ with fluctuations in gamma of the order of $\sim 12^\circ$, indicating a more shallow minimum in the potential energy surface as compared to the $\gamma$-rigid assumption of the GTRM. The present data do not, however, allow for the extraction of experimental $\beta$ and $\gamma$ shape parameters as the $Q_s$ moment for the $2^+_1$ level could not be determined and no sensitivity to the relative signs of the $2^+_1 \rightarrow 0^+_1$, $2^+_1 \rightarrow 0^+_1$ and $2^+_1 \rightarrow 2^+_1$ matrix elements was found in the GOSIA fit. For the analysis, the adopted convention was that all in-band matrix elements and the one linking the $2^+_1$ level to the ground state were chosen to be positive, while ones between bands were allowed to take positive or negative values. Nevertheless, all the available experimental evidence of Table 1 and Fig. 3 is supported by the results of calculations pointing to a significant role for triaxiality in this nucleus ($\gamma \sim 30^\circ$), albeit with a degree of $\gamma$ softness. It is also worth remembering that beyond-mean-field calculations within the same framework also reproduce satisfactorily data on, for example, the neutron-rich Sr isotopes [53] and the neutron-deficient Kr [54,55] and Se isotopes [56] where shape changes and/or shape coexistence have been observed. Similarly, the GTRM model has been found to successfully reproduce the extensive set of measured $E2$ matrix elements for the candidate triaxial nuclei $^{186–192}$Os [51].

Furthermore, the spectroscopic quadrupole moment, $Q_s$, was determined for the $2^+_1$ state to be $Q_s = -83^{+35}_{-20}$ e fm$^2$. The large uncertainty is due in part to (i) the limited sensitivity in the angular range ($30^\circ$–$75^\circ$) where the analysis could be performed (due to the lack of separation between different nuclei at lower angles and the lack of ToF information at backward angles), and to (ii) insufficient precision in the determination of some other transition probabilities in $^{110}$Ru (in particular that of the $2^+_1 \rightarrow 2^+_1$ transition, which has a second-order effect on the excitation cross section of the $2^+_1$ state). Nevertheless, the sign of the $Q_s$ moment is determined and firmly establishes that $^{110}$Ru has a prolate shape near its ground state. While the $Q_s$ measurement agrees within $\sim 1.3\sigma$ with the HFM-GCM(GOA) and GTRM results described above, it is in clear disagreement with the calculations of Ref. [57] within the D1N formulation of the Gogny energy density functional [14] (without the GCM(GOA) treatment) which predict a static oblate deformation for the $^{108–112}$Ru isotopic chain. Not all models give predictions for quadrupole moments, however, it is worth also pointing to recent calculations within the Cranked Skyrme–Hartrye–Fock–Bogoliubov (CSHFB) framework [13]. In this work, $^{110}$Ru is determined to be triaxial with deformation of the same magnitude ($\beta_2 = 0.16$ and $\gamma = 25^\circ$) as those computed here. The quoted $Q_s$ moment [13] translates into a $Q_s$ value of $-100$ e fm$^2$, which is also in agreement with the measured spectroscopic quadrupole moment, $Q_s = -83^{+35}_{-20}$ e fm$^2$.

In conclusion, a number of recent developments have proved vital to the success of this measurement. First post-acceleration of a $^{110}$Ru beam is reported, which, when coupled with the enhanced performance provided by the large angular sensitivity of the CHICO2 particle detector and the $\gamma$-ray tracking capabilities of the GRETINA array, provides direct evidence for of relatively rigid triaxial deformation near the ground state in a neutron-rich Ru isotope. The success of this measurement represents a technical milestone and paves the way for future studies involving unstable, neutron-rich refractory isotopes, where non-axial shapes and other intriguing shape phenomena are predicted to occur, in particular $^{112}$Ru where triaxiality is expected to reach a maximum in the Ru isotopic chain.

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References


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1 Not every model predicts triaxiality in $^{110}$Ru: the present results contradict an analysis within the framework of the interacting boson model [52], where no evidence for triaxial shapes was found.