Coulomb excitation of the $2^{\text{+}}_{\text{ms}}$ state of 96 Ru in inverse kinematics

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We identify the $J^{\pi} = 2_3^+$ state of ⁹⁶Ru as the proton-neutron mixed-symmetry 2_{ms}^+ state of this nucleus. This identification is based on the measurement of the transition strengths $B(M1,2_3^+ \rightarrow 2_1^+)=0.78(23)$ μ_N^2 and $B(E2; 2^+_3 \rightarrow 0^+_1) = 1.6(3)$ W.u. These transition strengths were obtained from the Coulomb excitation yield for the population of the 2^+_3 state relative to the yield of the 2^+_1 state with known *E*2 excitation strength. The Coulomb excitation was done in inverse kinematics using the reaction $\text{nat}(O^{96}Ru, {}^{96}Ru^*)$ at 280 MeV. This work demonstrates the accessibility of the isovector quadrupole excitation in the valence shell using experiments with an accelerated beam of the nuclei of interest.

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The many-body atomic nucleus is one of the classic testing grounds for theories of collective quantum phenomena. The unique feature of nuclear structure physics lies in the fact that the nucleus can be described as a system built up by many bodies of two species, protons and neutrons, as the relevant degrees of freedom at low energies. This fact makes nuclear structure physics a conceptual bridge between the investigation of single-component, e.g., electronic, manybody quantum systems, and multicomponent systems, e.g., hadrons. Therefore, the exploration and thorough understanding of the impact of the proton-neutron degree of freedom on nuclear structure is one of the fundamental issues of nuclear structure physics with relevance extending to other fields of quantum physics.

Isovector excitations are particularly sensitive to the proton-neutron interaction. Extensions to collective models incorporated the isospin degree of freedom and predicted $[1,2]$ collective isovector excitations based on quadrupole deformation, albeit at higher energies than later observed.

A microscopic approach to nuclear structure is given by the interacting shell model with isospin degrees of freedom. Nuclear shell structure supports the concept of an inert core and a valence shell, which simplifies the understanding of most low-energetic excitation modes. A truncation of the valence shell provides the framework of the interacting boson model (IBM). Including the proton-neutron degree of freedom $[3]$, the IBM-2 predicts the existence of low-lying, collective, at least partly isovector, valence shell excitations, namely, the mixed-symmetry states $[4]$ with nonmaximum F -spin quantum number [5].

The relation between symmetric and mixed-symmetry states becomes particularly obvious in the *Q*-phonon scheme [6,7]. Nearly all even-even nuclei have a $J^{\pi} = 2^+$ state as the lowest-lying excited state. Its wave function can be well approximated $\lceil 8 \rceil$ by the isoscalar quadrupole excitation

$$
|2_s^+\rangle \approx \mathcal{N}_s(Q_\pi + Q_\nu)|0_1^+\rangle,\tag{1}
$$

where Q_{π} and Q_{ν} denote the proton and neutron quadrupole operators in the valence shell. Obviously, there exists an orthogonal linear combination of the collective proton and the neutron quadrupole excitations

$$
|2^+_{\text{ms}}\rangle \approx \mathcal{N}_{\text{ms}}(Q_\pi - \alpha Q_\nu)|0^+_1\rangle,\tag{2}
$$

where the parameter α ensures the orthogonality, $\langle 2^+_{ms} | 2^+_1 \rangle$ $=0$, to the symmetric one-*Q*-phonon configuration. This construct represents the fundamental mixed-symmetry quadrupole excitation in the valence shell, which is, in vibrational nuclei, the one- Q -phonon counterpart $[9]$ of the symmetric $2₁⁺$ state. Its experimental identification and investigation are of obvious importance.

The predicted $[10]$ unique signatures for the one-Q-phonon mixed-symmetry state 2^+_{ms} are a strong M1 transition to the symmetric 2^+_1 state with a large matrix element of about $1\mu_N$ in size and a weakly collective isovector *E*2 decay to the ground state. Mixed-symmetry 1^+ states were observed in many deformed nuclei [11]. Candidates for $2^{\text{+}}_{\text{ms}}$ states were suggested from the analysis of intensity ratios, e.g., [12]. An unambiguous observation of these signatures requires experimental techniques which are sensitive to the absolute size of transition matrix elements.

Lifetime measurements have provided evidence for fragments of the $2^{\text{+}}_{\text{ms}}$ state of heavy nuclei, e.g., [13]. Investigations of the nucleus 94 Mo have recently shown [14–16] that a very pure $2_{ms}⁺$ state can exist that is even able to support two-phonon structures. Some of the experimental methods used for the aforementioned observations (for instance, photon scattering or neutron scattering) required the long-term handling of several hundreds of milligrams of the nuclei under investigation. In practice, this has restricted the investigation of mixed-symmetry one- and two-phonon states to β -stable nuclides. However, having a reliable experimental method to identify mixed-symmetry quadrupole-phonon excitations in radioactive nuclides from absolute transition strengths would considerably enhance the study of this fundamental class of states. One viable way is the formation of an intense beam of the nuclei of interest in a rare isotope accelerator facility, like that proposed in the U.S. $[17]$, and to

FIG. 1. Comparison of the low-lying low-spin level scheme of the even-even $N=52$ isotones ⁹⁴Mo and ⁹⁶Ru. The 2^+_3 state of ⁹⁴Mo was recently identified [14] as the $2^{\text{+}}_{\text{ms}}$ state of this nucleus. The numbers denote relative γ intensities, absolute *M*1 matrix elements, and $B(E2)$ values. The transition strengths for ⁹⁶Ru are from this work.

measure the Coulomb excitation yield in inverse kinematics [18]. The corresponding cross sections for Coulomb excitation, along with γ -ray branching ratios and multipole mixing ratios, would give sufficient information on absolute transition strengths to identify $2_{ms}⁺$ states. We have carried out such an experiment at Yale, using a beam of the stable isotope 96Ru at the ESTU-tandem accelerator. The result of the experiment is the identification of the 2_{ms}^+ state of 96 Ru and the first measurement of an allowed *M*1 transition strength between mixed-symmetry and full-symmetry states in a ruthenium nuclide.

It is the purpose of this Rapid Communication to present this experiment and to discuss the results in order to first demonstrate the feasibility of Coulomb excitation in inverse kinematics for the investigation of mixed-symmetry states, second to broaden the database on $2_{ms}⁺ mixed-symmetry$ one-*Q*-phonon excitations, and third to contribute to the discussion about mixed-symmetry states in the $A \approx 100$ mass region.

The nucleus ⁹⁶Ru is particularly interesting with respect to the investigation of the $2_{ms}⁺$ state because it is the next heavier even-even $N=52$ isotone after ⁹⁴Mo, for which the hitherto most complete data set on transition strengths from multiphonon mixed-symmetry states has been observed. Figure 1 shows a comparison of the lowest parts of the level schemes of 94 Mo (left) and 96 Ru (right). The excitation energy and the decay branching ratios strongly suggest a similar character of the 2^+_3 states in both nuclei, which in ⁹⁴Mo was identified the 2^+_{ms} state [14].

We studied the $^{nat}C(^{96}Ru, ^{96}Ru^*)$ Coulomb excitation re-</sup> action at 280 MeV. The beam energy, $\approx 78\%$ of the Coulomb barrier, ensured safe Coulomb excitation. A beam of negatively charged Ru nuclei was extracted from a sample of natural ruthenium in the sputter source of the Yale ESTUtandem accelerator. After acceleration the beam was mass separated with respect to the isotope ⁹⁶Ru. A beam current of about 3×10^{9} ⁹⁶Ru nuclei per second was used on a 0.96 $mg/cm²$ natural carbon foil. The degraded beam, with an exit energy of about 220 MeV, was subsequently stopped in a Pb

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block mounted in the target chamber 64 mm downstream from the carbon foil. Gamma rays were observed with the efficient new SPEEDY Ge-detector array $[19]$ which was equipped with seven EURISYS clover Ge-detectors. The detectors were mounted at observation angles of 0° (one clover), 42° (three clovers), and $138^\circ = \pi - 42^\circ$ (three clovers) with respect to the beam axis. The total photopeak efficiency of the setup was 2.1% at 1.33 MeV. Singles γ spectra were taken for 2 d with beam on target. The room background was measured off-beam for 1 d.

Figure 2 shows the relevant parts of the γ spectra obtained. Below 1 MeV, the in-beam spectrum is dominated by the 832.6 keV $2^+_1 \rightarrow 0^+_1$ transition in ⁹⁶Ru. It contains a large Doppler-shifted component and a much smaller part from emissions at rest. The shifted component originates from the population of the 2^+_1 state of ⁹⁶Ru by Coulomb excitation in the carbon target. The corresponding, Coulomb-scattering

FIG. 2. Photon spectra from the bombardment of a 1 mg/cm² carbon foil with 280 MeV ⁹⁶Ru nuclei observed with the SPEEDY array. Panel (a) displays the raw spectrum observed in-beam with the clover detector at $\theta=0^{\circ}$. Panels (b)–(d) show the in-beam spectra after subtracting the room background for the detectors at θ $=0^{\circ}$, 42°, and $\theta = 138^{\circ}$, respectively. Panel (e) displays the coincidence spectrum gated with the 832.6 keV $2^+_1 \rightarrow 0^+_1$ transition in ⁹⁶Ru. The coincidence sort was done for all detectors in the array using Doppler corrections according to the individual observation angles and a relative velocity of $v/c = 6.6\%$.

angle integrated and effective beam energy averaged, excitation cross section is $\langle \sigma \rangle (2_1^+) = 93(1)$ mb using the previously measured *E*2 matrix element $|\langle 2^+_1 || E2 || 0^+_1 \rangle|$ $=0.486(7)e b [20].$

Above the $2^+_1 \rightarrow 0^+_1$ γ line, the raw in-beam singles spectrum is dominated by γ rays originating in the room background from natural activity, e.g., the 1460.8 keV line from the decay of $40K$, and weak contaminations of the experimental hall. This fact demonstrates the extremely low background induced by the Coulomb excitation reaction in inverse kinematics which makes this method sensitive even to weak γ lines with higher energies than the dominating 2^+_1 $\rightarrow 0^+_1$ transition. We can obtain clean spectra by simply subtracting the appropriately scaled room background.

Figures $2(b)$ –(d) show the beam-induced parts of the inbeam spectra. Except for fluctuations at γ energies of strong background lines, the resulting subtracted spectra are very clean. This fact enables us to easily identify the Dopplershifted signal of the 1451.6(3) keV $2^+_3 \rightarrow 2^+_1$ transition (marked by the arrows in Fig. 2), which accounts for $93(3)\%$ of the decays of the 2^+_3 state of ⁹⁶Ru at an excitation energy of 2284.2(3) keV [23]. For a mean velocity of $v = 0.066c$ of Coulomb excited ⁹⁶Ru nuclei one expects to measure a mean Doppler-shifted energy of $E_y=1547.4$ keV at 0°, 1522.8 keV at 42°, and 1380.4 keV at 138°. The observed centers of gravity of the Doppler-shifted (and considerably broadened) $2^{+}_{3} \rightarrow 2^{+}_{1}$ transition lie at 1548(1) keV, 1521(2) keV, and $1381(2)$ keV, respectively. Figure 2(e) displays the spectrum of Doppler-corrected prompt coincidences with the shifted part of the $2^+_1 \rightarrow 0^+_1$ transition. In this coincidence spectrum we can identify two γ lines above the noise corresponding to the $4^+_1 \rightarrow 2^+_1$ and $2^+_3 \rightarrow 2^+_1$ transitions in ⁹⁶Ru with an intensity ratio (after efficiency correction) of $1.2(2)$.

The almost equal intensities of the 685.5 keV $4^+_1 \rightarrow 2^+_1$ transition with a $B(E2)$ value of 21(3) W.u. and the 2^{+}_{3} \rightarrow 2⁺ almost pure *M*1 transition with a more than 2 times larger transition energy of 1451.6 keV ensure that two-step excitation processes are negligible for the excitation yield of the 2^+_3 state in our reaction. Indeed, using¹ the $E2/M1$ multipole mixing ratio $\delta = 0.03(10)$ for the $2^+_3 \rightarrow 2^+_1$ transition [22] and the branching ratio $I_{\gamma}(2_{3}^{+}\rightarrow2_{1}^{+})/I_{\gamma}(2_{3}^{+}\rightarrow0_{1}^{+})$ $=0.075(22)$ [23], numerical calculations with the Winther–de Boer and GOSIA [24] codes yield a 99.9% dominance of one-step excitation over two-step processes for the 2_3^+ state. Consequently, the γ intensity of the $2_3^+ \rightarrow 2_1^+$ transition can be considered to be proportional to the $B(E2;0₁⁺)$ \rightarrow 2⁺₃) value. On the other hand the 2⁺₁ \rightarrow 0⁺₁⁺ transition is 3 orders of magnitude more intense than all other γ lines in our spectra, hence, the γ intensity of the $2^+_1 \rightarrow 0^+_1$ transition can be considered to be proportional to the $B(E2;0_1^+ \rightarrow 2_1^+)$ value. Therefore, we can reliably extract the $B(E2;0_1^+$

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TABLE I. Measured transition strengths.

Observable	Unit	Ref. [20]	This work
$B(E2;0^+_1\rightarrow 2^+_1)$	$e^2 h^2$	0.236(7)	$0.236(7)^{a}$
$Q(2^+_1)$	e b	0.13(9)	$0.13(9)^{a}$
$B(E2; 2^+_1 \rightarrow 4^+_1)$	$e^2 h^2$	0.098(13)	0.106(8)
$B(E2;0^+_1\rightarrow 2^+_3)$	$\rho^2 h^2$		0.0207(36)

^aAdopted from Ref. [20] for normalization.

 $\rightarrow 2_3^+$) value relative to the known $B(E2; 0_1^+ \rightarrow 2_1^+)$ =0.236(7) $e^2 b^2$ value from the intensity ratios $I_y(2^+_3)$ \rightarrow 2⁺ $\frac{1}{1}$ /*I*_y(2⁺ \rightarrow 0⁺₁) \approx 3×10⁻³. The numerical calculations with the GOSIA code take into account integrations over Coulomb scattering angles and effective beam energy in the target, and γ angular distributions. The observed intensities correspond to Coulomb excitation cross sections calculated with the transition strengths shown in Table I. The measured $B(E2; 2^+_1 \rightarrow 4^+_1)$ value agrees with the literature [20]. The error quoted for the $B(E2; 0_1^+ \rightarrow 2_3^+)$ transition strength includes, besides the statistical error, a 17% uncertainty of the cross section due to a possible variation of the unknown quadrupole moment $Q(2^+_3)$ from zero to the extreme rotational limit $|Q_{\text{rot}}(2_3^+)| = \sqrt{10/7B(E2)\uparrow} = 0.18e$ b.

For the $B(E2; 2^+_1 \rightarrow 4^+_1)$ value this reorientation effect due to the unknown $Q(4_1^+)$ induces an uncertainty of less than 2%. The contribution from Coulomb excitation of the 96Ru beam on heavy elements present in the setup was determined to be less than 1% for the 2^+_3 state mainly because of the much higher Coulomb barrier. Deorientation due to magnetic dipole interaction is insignificant for the 2^+_3 state because of its short lifetime.

Table II compares the measured properties of the 2^+_3 states of ⁹⁶Ru and ⁹⁴Mo. We note the similarity of the weakly-collective $B(E2; 0_1^+ \rightarrow 2_3^+)$ values in ⁹⁶Ru and ⁹⁴Mo that correspond to about 10% of the *E*2 excitation strength to the 2^+_1 states. These sizable $B(E2;0^+_1 \rightarrow 2^+_3)$ values characterize the 2^+_3 states as one-phonon excitations which ideally can be studied by Coulomb excitation. We next observe the large $B(M1; 2^+_3 \rightarrow 2^+_{1})$ value,² which is even larger in ⁹⁶Ru than in the neighbor 94 Mo. This exceptional combination of large absolute $M1$ and $E2$ transition strengths to the 2^+_1 state and to the ground state is the clear signature for a $2_{ms}⁺$ state. Therefore, we conclude the 2^+_3 state of ⁹⁶Ru to be the mixedsymmetry one-phonon excitation of that nucleus. This conclusion is supported by the predictions of the IBM $[10]$ and agrees with recent microscopic calculations $[26,27]$ for the $N = 52$ isotone ⁹⁴Mo. The similar decay properties of the 2^{+}_{3} states of 94 Mo and 96 Ru with mixed-symmetry character are shown in Fig. 1. The properties of the one-phonon 2_{ms}^+ state change little when going from one $N=52$ nuclide to the even-even isotone.

¹The literature values used here for the branching ratio and the *E*2/*M*1 multipole mixing ratio agree within the experimental uncertainties with new results obtained recently by the Cologne spectroscopy group $[21]$.

²The large error bar in the 96 Ru case is mainly due to the uncertainty in the $I_{\gamma}(2^+_3 \rightarrow 0^+_1)/I_{\gamma}(2^+_3 \rightarrow 2^+_1)$ branching ratio.

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TABLE II. Comparison of the measured properties of the isovector quadrupole excitation 2_{ms}^+ in 96 Ru and ⁹⁴Mo, to the original IBM prediction [10,25], and to recent microscopic calculations for ⁹⁴Mo [26,27].

 a From Ref. [23].

 b From Ref. [22].</sup>

^cUsing the *E*2/*M*1 mixing ratio δ =0.03(10) for the $2^+_3 \rightarrow 2^+_1$ transition from Ref. [22] and the γ -intensity branching ratio $93(3)/7(2)$ from Ref. [23] which both were recently confirmed by new data [21].

In the framework of the *sd*-IBM-2 *B*(*M*1) values are proportional to the squared difference of the boson *g* factors, $(g_{\pi}-g_{\nu})^2$. From the measured value, $B(M1;2_{\text{ms}}^+\rightarrow2_{1}^+)$ =0.78(23) μ_N^2 , we can conclude a numerical value of (g_π $(-g_{\nu})^2 \ge 1$ μ_N^2 for ⁹⁶Ru independent of any model parameters and choices of boson numbers. Moreover, a value of $(g_{\pi}-g_{\nu})^2=1$ μ_N^2 reproduces well the extensive set of *F*-vector $M1$ transition strengths measured in ⁹⁴Mo assuming a γ -soft structure [15] and it can account for the total excitation strength of the 1_{sc}^{+} mode in deformed rare-earth nuclei if one allows for breaking of the $SU(3)$ symmetry.

Previously, mixed-symmetry character has been assigned $[28,29]$ to an extensive set of low-lying nuclear states in the $A \approx 100$ mass region that could not be understood in the framework of the *sd*-IBM-1. These mixed-symmetry assignments were obtained from comparing excitation energies and electromagnetic intensity ratios to systematic *sd*-IBM-2 calculations using a strongly quenched *M*1 operator. A value of $(g_{\pi}-g_{\nu})^2=0.054~\mu_N^2$ was used to reproduce measured intensity ratios in even-mass ruthenium nuclei, $98-114$ Ru. From the satisfactory agreement with the data, dominant mixedsymmetry character was assigned to many states, because large $F \leq F_{\text{max}}$ components in the wave functions were required to generate sufficient *M*1 strength to account for the measured intensity ratios. It was concluded in Ref. [28] that for nuclei in this mass region the *B*(*M*1) values for allowed mixed-symmetry to full-symmetry $(MS \rightarrow FS)$ transitions would be of the order of 10^{-2} μ_N^2 .

The value we measured above for 96 Ru exceeds the former estimate by almost 2 orders of magnitude. This apparent contradiction could drastically change our current vision of mixed-symmetry states in this mass region: either the strengths of allowed $MS \rightarrow FS$ transitions changes by 1 to 2 orders of magnitude when going from $96Ru$ to the even-even neighbor ⁹⁸Ru, which would lead to doubts about their collectivity, or the structures discussed in Refs. $[28,29]$ contain much smaller components of mixed-symmetry character than previously thought. It would be of great importance to measure the absolute $B(M1;2^+_1 \rightarrow 2^+)$ strength distribution in ⁹⁸Ru and heavier ruthenium nuclides up to 3 MeV to resolve this problem.

To summarize, we have identified the fundamental mixedsymmetry one- Q -phonon excitation $2^{\text{+}}_{\text{ms}}$ in the near-spherical nucleus 96Ru based on absolute *E*2 and *M*1 transition strengths obtained in a Coulomb excitation experiment of ⁹⁶Ru nuclei in inverse kinematics. The new data broaden the still small data set on transition strengths from $2^{\text{+}}_{\text{ms}}$ states and add confidence in the concept suggesting the existence of more-or-less fragmented isovector valence shell excitations in all open shell nuclei. This experiment demonstrates that the technique of Coulomb excitation in inverse kinematics is capable of identifying and investigating the $2_{ms}⁺$ state. It solidifies our expectations for many-body quantum research with a powerful rare isotope accelerator and it might open up new experimental approaches to mixed-symmetry states, e.g., the measurement of *g* factors for mixed-symmetry states using transient field techniques after projectile Coulomb excitation [30]. The measured value $B(M1; 2^+_{\text{ms}} \rightarrow 2^+_1)$ $=0.78(23)$ μ_N^2 shows that allowed MS \rightarrow FS *M*1 transitions in ruthenium nuclei can reach strengths of the order of 1 μ_N^2 , about 2 orders of magnitude more than concluded in earlier work.

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