Are There Signatures of Harmonic Oscillator Shells Far From Stability?—
First Spectroscopy of $^{110}$Zr


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The first measurement of the low-lying states of the neutron-rich $^{110}$Zr and $^{112}$Mo was performed via in-beam γ-ray spectroscopy after one proton removal on hydrogen at ~200 MeV/nucleon. The $2^+$ excitation energies were found at 185(11) keV in $^{110}$Zr, and 235(7) keV in $^{112}$Mo, while the $\frac{R_{42}=E(4^+_2)/E(2^+_2)}{2.7(1)}$, respectively. These results are compared to modern energy density functional based configuration mixing models using Gogny and Skyrme effective interactions. We conclude that first levels of oscillator shell stabilization scenario at $Z=40$ and $N=70$. The present data also invalidate predictions of the nuclear chart [7-10]. Despite intensive effort, the structural changes is not yet fully understood and the mechanisms that drive structural evolution differ between models. Within.

Nuclei, like atoms, manifest quantized energy states that can be interpreted in terms of an underlying shell structure—a convenient but non-observable theoretical construct [1, 2]. Within the classical picture, large gaps between adjacent shells give rise to particularly stable configurations whose proton and neutron numbers are traditionally called “magic” [3]. The magic numbers for stable nuclei were first successfully described by invoking a one-body square-well and spin-orbit potential [4, 5]; the latter was eventually replaced by a harmonic oscillator potential with $i^2$ term to obtain proper angular momentum splittings [6]. However, studies of radioactive nuclei over the past decades have shown that the magic numbers are not universal across the nuclear chart [7–10]. Despite intensive effort, the theoretical description of these structural changes is not yet fully understood and the mechanisms that drive structural evolution differ between models. Within
the shell-model picture, the tensor and central forces modify single-particle energies via interactions between valence proton and neutron orbitals according to their filling and relative spin-to-orbital orientation [3, 11–18]. In a mean-field conception, spherical shell gaps may be modified far from stability by increased surface diffuseness. Its principal effect is a quenching of the spin-orbit splittings by as much as 40% near the dripline [19, 20], but other features of shell structure are affected as well, such that shell effects might even be effaced entirely when approaching the neutron dripline [21]. Tensor interactions add additional local variations to the spin-orbit splittings [22]. In the mean-field picture, the competition between spherical and deformed shapes is illustrated by the appearance of deformed gaps in the single-particle spectrum as a function of intrinsic deformation. Large gaps may lead to shell stabilization. Qualitatively, if the spin-orbit splitting that gives rise to the N=82 shell gap is reduced by these mechanisms far from stability, the harmonic oscillator gap at N=70 may open up instead [23]. If this happens already for modest neutron-to-proton ratios, then it may be manifest at $^{110}$Zr, whose 40 protons and 70 neutrons combine two harmonic oscillator shell closures [24]. This makes $^{110}$Zr a prime benchmark for the dynamic interplay between shell structure and multipole correlations far from stability. A shell-stabilized $^{110}$Zr has potential implications for our understanding of the rapid neutron capture process. Despite recent improvements from new $\beta$-decay lifetimes [25], $r$-process calculations consistently fail to reproduce the elemental abundance distribution near mass 110. Currently this discrepancy is an entanglement of astrophysical and nuclear structure predictions, but a shell-stabilized $^{110}$Zr is one proposed solution to this anomaly [24]. Spectroscopic information near $^{110}$Zr will help delineate the N=70,82 shell evolution and constrain the structure models used in $r$-process simulations. On both the theoretical and experimental sides, $^{110}$Zr has motivated numerous studies while its structure remains unknown. A shell-stabilized $^{110}$Zr has been predicted by independent mean-field and microscopic-macroscopic approaches [26–28], which interestingly all find that the shell-stabilization coincides with a tetrahedral configuration. This exotic symmetry, hitherto unobserved, is expected to compete strongly with deformed minima and its emergence is known to be very sensitive to pairing effects [29]. If a tetrahedral configuration persists in the ground state of $^{110}$Zr however, it would manifest a unique energy spectrum [30, 31], distinguishable in a first-spectroscopy measurement. Meanwhile, most predictions of the $^{110}$Zr ground state show well deformed prolate [32, 33] or shape coexistent minima [34–39], though the exact structure is highly sensitive to the details of the effective interaction [22]. Recently, a strong deformation of $^{110}$Zr, similar to lighter Zr isotopes beyond N=60, has been predicted by Monte Carlo Shell Model (MCSM) calculations [40]. Experimentally, the weakening of the N=82 shell closure was first claimed from $\beta - \gamma$ decay spectroscopy of $^{130}$Cd and $^{130}$In where the measured $Q_\beta$ value agreed best with predictions from a shell-quenched mass model [41]. This conclusion was challenged by measurements of isomeric decays in $^{130}$Cd which showed no evidence of shell quenching [42]. More recently, mass measurements of $^{129}-^{131}$Cd show a reduction of neutron separation energy differences by 1 MeV going from $^{132}$Sn to $^{130}$Cd [43]. Thus far experiments near $^{110}$Zr, including the $\beta$-decay half-lives of $^{106-112}$Zr [25], lifetime measurements of the $2^+_1$ states in $^{104, 106}$Zr [44], and spectroscopy of the low-lying excited states of $^{108}$Zr [45], show no hint of a shell gap at N=70 and suggest that the Zr isotopes with N>60 are prolate deformed. In this Letter we present the first spectroscopic evidence of the deformed nature of $^{110}$Zr and $^{112}$Mo, and discuss the origin of their collectivity through state-of-the-art microscopic models.

The measurement was performed at the Radioactive Isotope Beam Factory operated by the RIKEN Nishina Center for Accelerator-Based Science and the Center for Nuclear Study of the University of Tokyo. A 30 pA $^{238}$U primary beam was accelerated to 345 MeV/nucleon. The radioactive isotope beams were created via in-flight fission of the primary beam on a 3-mm thick $^9$Be target positioned at the focal plane before the BigRIPS two-stage fragment separator [46]. The isotopes of interest were selected from the secondary cocktail beam using the $B_\beta$-$\Delta E$-$B_\rho$ method [47]. These isotopes, magnetically centered on $^{111}$Nb, impinged on a 99(1) mm-thick liquid hydrogen (LH$_2$) target with an incident kinetic energy of 260 MeV/nucleon. The energy loss in the hydrogen target was approximately 100 MeV/nucleon. Individual intensities for the secondary beams of interest $^{113}$Tc and $^{111}$Nb were 32 and 20 particles per second, respectively. Nuclei before and after the LH$_2$ target were unambiguously identified in the BigRIPS and ZeroDegree spectrometers, respectively, using the TOF-$B_\rho$-$\Delta E$ method [47]. $^{110}$Zr and $^{112}$Mo were created via proton removal in the LH$_2$ target. Emitted $\gamma$ rays were detected with the DALI2 gamma array, which consisted of 186 NaI(Tl) detectors covering polar angles of 12°-118° with an average angular resolution of 6° [48]. The full-energy peak detection efficiency with no addback was simulated to be 31% for 500 keV $\gamma$ rays emitted in flight [49]. DALI2 was calibrated with $^{152}$Eu, $^{59}$Co, $^{137}$Cs, $^{88}$Y, and $^{133}$Ba sources, yielding calibration peaks from 121 to 1332 keV, a calibration error of 1.5 keV, and energy resolution of 59 keV Full Width at Half Maximum (FWHM) for the 662 keV peak of $^{137}$Cs, consistent with [50]. DALI2 thresholds were set at 100 keV on average. A 300 mm long time projection chamber was placed around the LH$_2$ target in a setup
clearly identify the peaks of interest. The peaks of interest, and subtracted from the experimental spectra according to the number of nuclei incident on the LH₂ coincidence with unreacted beam particles, normalized from fast beam particles colliding with hydrogen close in energy to the Bremsstrahlung spectrum generated from a target. The Doppler-corrected γ-ray spectra of ¹¹²Mo including the total spectrum (solid black line), normalized Bremsstrahlung component (blue), and the subtracted spectrum (open circles). The fit to the subtracted spectrum is shown by the thick red line, individual simulated responses are shown by thin red lines while the exponential background is shown by the dashed line. ( Insets) Background subtracted γ-γ coincidences for the 2⁺ → 0⁺ transitions. The inset for ¹¹²Mo is restricted to γ multiplicities lower than 4 for a better signal-to-background ratio. Coincident peaks are highlighted in gray.

![Gamma-ray spectra of ¹¹²Mo and ¹¹⁰Zr](image)

**FIG. 1.** (Color online) Doppler-corrected γ-ray spectra of ¹¹²Mo and ¹¹⁰Zr including the total spectrum (solid black line), normalized Bremsstrahlung component (blue), and the subtracted spectrum (open circles). The fit to the subtracted spectrum is shown by the thick red line, individual simulated responses are shown by thin red lines while the exponential background is shown by the dashed line. (Insets) Background subtracted γ-γ coincidences for the 2⁺ → 0⁺ transitions. The inset for ¹¹²Mo is restricted to γ multiplicities lower than 4 for a better signal-to-background ratio. Coincident peaks are highlighted in gray.

known as MINOS [51]. MINOS detected knocked-out protons with an efficiency to detect at least one of the two protons from (p,2p) simulated at 95%. The proton tracks were used to reconstruct the reaction vertex with a precision of 5 mm FWHM. Knowledge of the vertex allowed precise Doppler correction of the γ rays detected in DALI₂, as demonstrated in [52].

The Doppler-corrected γ-ray spectra for ¹¹²Mo and ¹¹⁰Zr are shown in Fig. 1. The transitions of interest lie close in energy to the Bremsstrahlung spectrum generated from fast beam particles colliding with hydrogen atoms in the target. This component was measured in coincidence with unreacted beam particles, normalized according to the number of nuclei incident on the LH₂ target, and subtracted from the experimental spectra to clearly identify the peaks of interest. The peaks that emerge after subtraction are visible at forward angles without subtraction. The subtraction method was validated on ⁸⁶Ge from the same experiment, where the 2⁺ → 0⁺ transition is known to be well separated from the Bremsstrahlung spectrum at 527 keV [53], and it was verified that the method did not create any artificial peaks. The subtracted spectra were then fit with response functions simulated using GEANT4 [49]. The simulation of the DALI₂ detector response functions was optimized for accuracy in the low-energy region of interest by including individual DALI₂ thresholds and accounting for absorption of the γ rays in all materials surrounding the target. The remaining background, originating from unresolved high energy transitions and particle induced background, has an unknown shape at low energy. It was taken to be an exponential cutoff with an error function, corresponding to the effect of the DALI₂ thresholds. The parameters of the exponential background were fit simultaneously with the response function intensities.

Considering the case of ¹¹²Mo, three major peaks were visible in the data, and a fourth was inferred from the shoulder on the high-energy side of the strongest transition. A matrix of response functions was simulated for each peak, corresponding to the possible combinations of transition energies and level lifetimes. The subtracted spectrum was then fit with all combinations of the four peak arrays. The Pearson’s χ² from the fitting procedure was converted to probability assuming a multi-variate gaussian probability density function, and the most probable response function and corresponding one-sigma regions of confidence in the energy-lifetime plane were extracted for each peak. The found transition energies are 235(7), 410(11), and 485(26) keV for the three major peaks. The uncertainties include the statistical region of confidence from the fitting procedure, intrinsic energy resolution, and systematic uncertainties added in quadrature. Note that the ambiguity induced by the unknown shape of the low-energy background does not allow us to determine lifetimes from the peak-shapes of the measured transitions with significant accuracy. An identical procedure was followed for ¹¹⁰Zr for the three peaks visible in the data, found at 185(11), 380(21), and 485(11) keV. In addition, ¹⁰⁸Zr (not shown) was analyzed as a reference measurement. The 2⁺ excitation energy in ¹⁰⁸Zr is found at 170(11) keV, consistent with [45], thus providing a validation of the present analysis. The γ-γ spectra allow us to construct the level schemes shown in Fig. 2. The γ-γ coincidences gated on the 2⁺ → 0⁺ transitions are shown in the insets of Fig. 1. For both ¹¹⁰Zr and ¹¹²Mo, the transition near 500 keV is not in coincidence with the other two major peaks and its intensity evolution as a function of multiplicity is consistent with a transition directly to the ground state. Therefore it is taken as the 2⁺ → 0⁺ transition. The 2⁺ → 2⁺ transition in ¹¹²Mo corresponds with the...
Assuming that the next proton shell closure below mid-shell as one departs from the Z=50 shell closure. Likewise, the systematics of 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} energies along the Zr chain are smooth as a function of neutron number with no particular increase of 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr}\ value from 108\_\text{Zr}\ to 110\_\text{Zr}\, showing no sign of any shell effect at N=70 either, and demonstrating that 110\_\text{Zr} is a well deformed nucleus with no significant structural change from lighter 100–108\_\text{Zr} isotopes.

In order to further investigate the structure of 112\_\text{Mo} and 110\_\text{Zr}, we compared our results to state-of-the-art beyond-mean-field calculations [59]. The 5-Dimension Collective Hamiltonian (5DCH) with the Gogny D1S effective interaction [60, 61] reproduces well collective properties across the nuclear landscape [35]. Experimental systematics of 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} energies as well as R_{42} ratios are compared to predictions in Fig. 3. Rather constant underprediction of collectivity is found between Z=48 and 42, but 112\_\text{Mo} and 110\_\text{Zr} in particular mark a clear departure from the theoretical trend showing significantly lower 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} and higher R_{42} ratios than predictions. In 120\_\text{Sn}, the Gogny D1S interaction gives a spherical gap between the 2\[^+\]p_{1/2} and 1g_{9/2} orbitals (Z=40) of 3.2 MeV, and a gap between the 3\_s_{1/2} and 1h_{11/2} orbitals (N=70) of 1.0 MeV. The same gaps are 3.0 MeV and 2.1 MeV, respectively, for 110\_\text{Zr}, showing a sizeable neutron gap away from stability, yet a deformed minimum is energetically preferred both at the mean-field and beyond-mean-field levels.

We confirm that the 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} and R_{42} disagreement between theory and experiment for the two nuclei studied does not stem from the 5DCH approximations by comparing with predictions from projected configuration mixing (referred to as PCM in the following)\(^1\) using the same effective interaction. PCM takes into account triaxial degrees of freedom, as does the 5DCH approach, but solves exactly the Gribov-Wheeler equation in the deformation coordinates for particle-number and angular-momentum projected Bogoliubov states. Excitation energy predictions from the two models are shown in Fig. 4 and are very similar. However, D1S+PCM calculations predict a lowering of the 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} energy for 108\_\text{Sr}, in better agreement with the trend observed in the data. D1S+PCM calculations also better reproduce the R_{42} trends, but both calculations clearly underpredict the collectivity of 110\_\text{Zr}. The dependence on the effective interaction is illustrated by PCM calculations using the SLyMR0 force, a recent Skyrme parametrization [55, 63]. The systematics of 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} energies are rather flat

\[^1\] Also called Symmetry-Conserving Configuration Mixing model (SCCM) [54] or projected Generator Coordinate Method (pGCM) [62].

(FIG. 3. Experimental 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} energies (A) and R_{42} ratios (B) for the N=70 isotones compared with theory: 5DCH [35] and PCM [54] with Gogny D1S interaction, PCM with Skyrme SLyMR0 [55], and MCSM calculations [40]. Experimental data are taken from [59] and this work. (Color online))

shoulder visible to the right of the 2\[^+\]\_\text{1}\_\text{\text{p}}\_\text{Zr} peak, and is visible in the \(\gamma - \gamma\) coincidences as the highlighted peak near 300 keV, however no clear minimum in the \(\chi^2\) surface was found from the fitting procedure. The energy of the response function shown in the fit in Fig. 1 is 280 keV. With the available statistics, we do not observe the 2\[^+\] \_\text{Zr} transition in 110\_\text{Zr}. For both 112\_\text{Mo} and 110\_\text{Zr}, the non-observation of the 2\[^+\] \_\text{Zr} transition suggests that the 0\[^+\]\_\text{Zr} state lies higher, or very close in energy to the 2\[^+\]\_\text{Zr}.

The experimental values of the 2\[^+\]\_\text{p}_{3/2}\_\text{Sn} and decreases symmetrically for both larger and smaller proton numbers towards maximum collectivity at mid-shell. Towards the neutron dripline, the 2\[^+\]\_\text{p}_{3/2}\_\text{Sn} and 110\_\text{Zr} measured in this work show a steep decrease. Similarly, the experimental R_{42} = E(4_{1/2}^+)/E(2_{1/2}^+) ratios shown in the lower panel of Fig. 3 suggest increased collectivity towards 110\_\text{Zr} with a value of 3.1(2). This pattern is consistent with a transition from a harmonic vibrator to a deformed symmetric rotor towards mid-shell as one departs from the Z=50 shell closure. Assuming that the next proton shell closure below 120\_\text{Sn} (Z=50) occurs in 98\_\text{Ni} (Z=28), expected to be beyond the neutron dripline [56], the maximum of collectivity would take place at Z=39 close to 110\_\text{Zr}, consistent with our results. The present measurement shows no sign of a subshell closure at Z=40 along the N=70 isotonic chain.
from 40≤Z≤50, missing the trend towards 120Sn while approaching but still overestimating our measured the 2+ \text{\footnotesize energies} for 110Zr and 112Mo.

For the case of 110Zr, a detailed comparison between our data and the theoretical level schemes is shown in Fig. 4. All above calculations predict triaxial ground and 2+ states built on rather γ-soft potential energy surfaces [59]. The 2+ state is predicted below the 4+ \text{\footnotesize states} in qualitative agreement with the data, though the excitation energies are globally overpredicted, a trend already illustrated in Fig. 3.

Our data also suggest that the ground state band we observe is not of a tetrahedral character. According to calculations by Tagami et al. [30, 31], the lowest-lying levels of the tetrahedral band in 110Zr are predicted to be (3-, 4+, 6+). However the low energy of the transitions we measure, combined with the fact that we observe the γ decay in flight, excludes the possibility that our most intense gamma ray results from E3 and higher multipolarity transitions. The only candidates then for the γ-rays we observe would be the 4+ to 3- and 6+ to 4+ transitions, but these are predicted to have a R_{42} ratio of ~2, very far from our measured ratio of ~3. Furthermore, the spectral systematics along the Zr isotopic chain allow a direct mapping of our strongest transitions to the 2+ and 4+ levels in 108Zr, which exhibits a nearly identical ground state band [45]. For these reasons, we reject the interpretation of the ground state band as being built upon a tetrahedral deformed minimum as suggested by [27], however, we can not exclude the existence of an excited tetrahedral band, as in [30].

To further quantify the influence of the effective interaction on the above observations, we performed a local sensitivity test within the 5DCH approach by increasing the spin-orbit term of the Gogny D1S interaction from -130 to -140 MeV fm^3, with no change of the other terms of the interaction. Within this model, modification of the spin-orbit strength is an efficient, but non-unique, way to artificially increase the deformation and see the spectroscopic effects. The results show a lowering of the 2+ \text{\footnotesize excitation energies} down to 259 keV and 215 keV for 112Mo and 110Zr, respectively, in much better agreement with experimental values. The agreement for R_{42} is also improved for these two isotones, going from 2.4 to 2.8 for 110Zr, approaching the experimental value of 3. For the case of 110Zr, the collective wavefunction of the 2+ \text{\footnotesize state} shifts from a triaxial maximum at \beta = 0.25 to prolate at \beta = 0.4, concurrently increasing the g_{9/2} occupancy by 0.3 protons at the prolate minimum of the potential energy surface, consistent with increased collectivity.

Our results are finally compared to MCSM predictions for 110Zr [40], also shown in Fig. 3 and Fig. 4. Predictions of excitation energies for the 2+ \text{\footnotesize states} of 110Zr are in good agreement with our measured values. MCSM operates within the paradigm of the so-called type II shell evolution [64], wherein the low-lying spectroscopy of Zr isotopes with N≥60 is understood as coming from a modification of the neutron single particle energies driven by the deformation-triggered promotion of protons from the fp shell to the g_{9/2} orbital. This is consistent with the results of the sensitivity study, where improved agreement is obtained for a larger deformation, when more protons are promoted into the g_{9/2} orbital. However, the MCSM calculations for 110Zr predict the 0+ \text{\footnotesize state} below the 2+ \text{\footnotesize state}, at variance with the energy density functional based models. The identification of the 0+ \text{\footnotesize state}, of particular interest for a fine understanding of the structure of 110Zr, requires further experiments.

In conclusion, we performed the first spectroscopy of the neutron-rich N=70 isotones 112Mo and 110Zr. Low 2+ \text{\footnotesize excitation energies} found at 235(7) keV and 185(11) keV, respectively, as well as R_{42} values of ~3, clearly indicate that both of these nuclei are well deformed. The present study demonstrates that 110Zr experiences no stabilizing shell effect corresponding to the harmonic oscillator nucleon magic numbers Z=40 and N=70, and seems to rule out this proposed solution to the r-process anomaly at A=110. While energy density functional based predictions using the Gogny D1S interaction along the N=70 isotonic chain show an overall good agreement with the 2+ \text{\footnotesize , 4+ \text{\footnotesize systematics}} for Z≥42, a clear underprediction of collectivity is evidenced for 110Zr. The roots of this discord remain to be understood. Eventually, the spectroscopy of 60Ca with Z=20 and N=40 should further our understanding of harmonic oscillator shell effects at the neutron dripline [65–67].

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[59] See Supplementary Material at [URL will be inserted by publisher] for further details of the calculations.


