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In-beam spectroscopy reveals competing nuclear shapes in the rare isotope 62Cr

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In recent decades, rare-isotope facilities have enabled the study of short-lived, neutron-rich nuclei. Their measured properties indicate that shell structure changes in the regime of unbalanced neutron-to-proton ratios compared with that of stable nuclei. In the so-called islands of inversion in the nuclear chart-around the neutron-rich nuclei 32 Mg, 42 Si and 64 Cr, for example—the textbook shell model predicts spherical shapes due to the respective magic neutron numbers of 20, 28 and 40 of these nuclei. However, nuclei in these regions turn out to be deformed in their ground states. Another hallmark of these islands is shape coexistence, where a nucleus assumes diferent shapes with excitation energy. Here we present evidence for this phenomenon from the observation of an excited 0⁺ state in ⁶²Cr, two neutrons away from the heart of the island of inversion around neutron number *N* = 40. We use large-scale shell-model calculations to interpret the results, and we report extrapolations for the doubly magic nucleus ⁶⁰Ca.

Just like in atomic physics, the shell model has been exceptionally successful for nuclei, even though they are composed of two constituents, protons and neutrons, that interact strongly and at short range. On the nuclear chart and near stability, nuclei with magic numbers of protons or neutrons are spherical in shape and more inert than their open-shell neighbours. As a result, they often serve as immutable cores in attempts to solve the nuclear many-body problem by restricting computations to a valence configuration space. Towards the fringes of the nuclear chart, nuclei, or rare isotopes, are short-lived and undergo a sequence of radioactive decays until reaching stability. In the past decades, rare-isotope facilities have enabled the study of short-lived, neutron-rich nuclei. Their measured properties indicate that the shell structure that is well established at stability changes substantially in the regime of unbalanced neutron-to-proton ratios. Energy gaps between shells that would normally correspond to magic numbers disappear and new shell gaps emerge $1-4$. There are several regions on the nuclear chart where such a breakdown of neutron magic numbers (*N*) occurs: around ¹²Be, ³²Mg, ⁴²Si and ⁶⁴Cr with $N = 8$, 20, 28 and 40 (refs. 1,2),

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Fig. 1 | γ-ray and particle-identification spectra for 62Cr. a, Event-by-event Doppler-reconstructed γ-ray spectrum of ⁶²Cr populated by two-proton removal from 64Fe. The bins of the histogram cover 8 keV in energy. The visible peaks are labelled by their energies with uncertainties attributed to the determination of the peak position and the Doppler reconstruction. Inset: spectrum in logarithmic scale. There are many γ-ray transitions above 2,000 keV. **b**, Event-by-event

particle identification through energy loss versus time of flight of the reaction residues entering the S800 spectrograph focal plane with ⁶²Cr (indicated in red) cleanly separated. For orientation, ⁶¹Cr, ⁶³Mn and the 25⁺ charge state of ⁶⁴Fe are labelled in orange. All particles shown in the spectrum have a γ-ray detection associated with them.

respectively. At stability, *N* = 40 is a subshell gap that was originally thought to stabilize doubly magic ⁶⁸Ni ($Z = 28$, $N = 40$), just four protons north from ⁶⁴Cr on the nuclear chart. These regions of marked change on the nuclear landscape are often referred to as islands of inversion because deformed, shell-breaking configurations are energetically favoured over closed-shell ones. In fact, the properties of these nuclei identify them as being among the most collective and deformed in their regions, and they display none of the properties associated with closed shells. Although often reported at the northern shores of islands of inversion⁵⁻⁸, shape coexistence, which is the occurrence of 0^+ states associated with different shapes within a small excitation-energy window near the ground state, has remained elusive. At the heart of the islands, clear evidence for shape-coexisting 0^+ states has been reported only in 12 Be (ref. 9), 30,32 Mg (refs. 10,11) and 44 S (ref. 12) following experimental tours de force, but not in the *N* = 40 island of inversion.

From an experiment motivated by the shell-model prediction of a stunning case of shape coexistence in ⁶²Cr, we report here direct evidence for a low-lying O⁺ excited state in this nucleus, just two neutrons away from the heart of the *N* = 40 island of inversion. This result was found in an in-beam γ-ray spectroscopy measurement performed at the newly operational Facility for Rare Isotope Beams (FRIB) at Michigan State University. The sensitivity of the two-proton knockout ⁹Be (⁶⁴Fe, ^{62}Cr + γ)X reaction to the final-state total angular momentum *J* of the residue¹³⁻¹⁶, combined with spectroscopy using the Gamma-Ray Energy Tracking In-beam Nuclear Array (GRETINA), a γ-ray tracking array^{17,18}, resulted in a detailed level scheme for 62 Cr at low excitation energy. The data have been verified with large-scale shell-model (LSSM) calculations and interpreted within the framework of the discrete non-orthogonal shell model (DNO-SM), which enables an analysis in terms of nuclear shapes 19 . The results highlight the importance of axial shape asymmetry (triaxiality) for the observed excited collective structures. This rare nuclear shape is not typically connected to islands of inversion.

The results reported here have broader implications. Only recently discovered²⁰, the *N* = 40 isotone ⁶⁰Ca plays a pivotal role on the nuclear chart, and competing predictions as to its deformed or doubly magic (spherical) nature can be found in the literature $21-25$. Based on our data, we provide the most informed prediction about its structure to date.

Experimental procedures

The 64 Fe projectiles were produced in-flight through fragmentation 26 of a primary beam of ⁷⁰Zn, accelerated in the FRIB linear accelerator to

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173 MeV/*u* and impinging on a stationary 4.2-mm-thick⁹Be production target within FRIB's advanced rare-isotope separator 27 . A 1.8-mm-thick Al wedge was used in the pre-separator to purify ⁶⁴Fe to 52%, with ⁶⁵Co (35%) being the only other intense component in the incoming beam. Using momentum slits and compression optics, the rare-isotope beam, which had a momentum width of Δ*p*/*p* = 0.4%, was delivered to the 100 mg cm⁻² thick ⁹Be reaction target in front of the S800 spectro $graph^{28}$. Reaction products were identified on an event-by-event basis in the spectrograph's focal plane. The particle-identification spectrum for 62Cr produced from the incoming 64Fe beam at a 100.4 MeV/*u* mid-target energy is provided in Fig. 1b. The inclusive cross section for two-proton removal from ⁶⁴Fe to ⁶²Cr was determined to be σ_{inc} = 0.59(4) mb.

The high-resolution γ-ray detection system GRETINA¹⁷, an array of 48 36-fold segmented high-purity germanium crystals assembled into modules of four crystals each, was used to measure the prompt γ-rays emitted by 62Cr residues. An online pulse-shape analysis was performed for its detector signals to determine the spatial coordinates of the γ-ray interactions within GRETINA $17,18$. These interaction points, together with the trajectory vectors of the reaction products as reconstructed with the spectrograph, enabled the event-by-event Doppler correction of the γ-rays emitted by 62Cr at *v*/*c* = 0.423. Figure 1a provides these spectra for 62 Cr with addback¹⁸ included, a method that corrects for the scattering of γ-rays between neighbouring crystals.

The 62Cr level scheme

Based on our previous low-statistical studies of ${}^{62}Cr$ (ref. 29), the decay of the first excited 0⁺ state to the 2^+_1 level was expected to be observed around 900–1,000 keV, with the 1,016 keV transition of 62 Cr then proposed as a candidate for this $0^+_2 \rightarrow 2^+_1$ decay. With the high statistics afforded by FRIB, a detailed γγ coincidence and γ-ray multiplicity analysis becomes possible. This revealed a previously unobserved transition at 933 keV, which is the best candidate for the decay from the predicted excited 0^{+}_{2} state and, furthermore, places the 1,016 keV transition as feeding the 4^+_1 rather than the 2^+_1 level. The most intense transitions in the spectrum are at 439, 728 and 1,016 keV, corresponding to the known decays of the 2^+_1 and 4^+_1 states and, as we will demonstrate here, from the $6₁⁺$ level, respectively. Figure 2 shows that the 933 keV γ-ray feeds the 2⁺ level but not the 4⁺ one and that it has a γ-ray multiplicity of not more than 2, consistent with the proposed $0^+_2 \rightarrow 2^+_1 \rightarrow 0^+_1$ cascade in the absence of appreciable discrete feeding. The 1,016 keV transition, on the other hand, is clearly demonstrated to

Fig. 2 | Spectra illustrating the γγ coincidence relationships for 62Cr. The bins of the histograms cover 8 and 5 keV in energy, respectively. The peaks are labelled by their energy. Spectra coincident with the transitions of the 2^+_1 , 4^+_1 and 6^+_1 levels, the strongest γ-rays in the spectrum of ⁶²Cr, are indicated in orange (filled), and by red and black lines, respectively. Inset: γ-ray multiplicity spectra, demonstrating that the 933 keV transition is part of a cascade of only 2 transitions (multiplicity 1 + 2, filled orange) whereas the neighbouring 1,016 keV transition has a multiplicity in excess of 3 (blue and red lines).

feed the 4⁺ level with a γ-ray multiplicity of 3 and higher, consistent with a $6^+_1 \rightarrow 4^+_1 \rightarrow 2^+_1 \rightarrow 0^+_1$ sequence markedly fed from higher-lying states. A similar coincidence analysis was performed for all transitions reported here with the results summarized in Extended Data Table 1. The efficiency-corrected γ-ray intensities and the level scheme resulting from the coincidence relationships were used to determine the partial cross sections included in the table.

Characterization of the 62Cr excited states

Two-proton (neutron) knockout from neutron (proton)-rich nuclei was shown to proceed as a direct reaction³⁰⁻³³ where the shape of the longitudinal momentum distributions of the knockout residues, *p*∥, encodes the total angular momentum^{13,14} as well as the total orbital angular momentum $15,16$ of the removed nucleons. When two-nucleon knockout occurs from a 0^+ ground state, an analysis of the final-state exclusive *p*∥ distribution of the residue directly revealed the total angular momentum of this state, *J*. Final-state exclusive momentum distributions were obtained through software cuts of the γ-ray spectra applied to the inclusive longitudinal momentum distribution, including background and feeder subtraction, when possible. The results are displayed in Fig. 3 for the three states for which such an analysis was possible in terms of statistics, peak-to-background ratio in the γ-ray spectrum and predominance of feeding. For the 2^+_1 level, the first excited state, for example, a large number of suspected unobserved feeders prevented such an analysis. Figure 3 also presents the calculated momentum distributions, based on the reaction dynamics and formalism of ref. 15, using two-nucleon amplitudes from LSSM calculations using the Lenzi-Nowacki-Poves-Sieja (LNPS) interaction²¹. The calculated distributions were boosted into the laboratory frame and folded with the experimental resolutions. They are superimposed on the measured ones in Fig. 3 and support the *J* assignments (0⁺) for the 1,379 keV level depopulated by a 933 keV γ-ray (Fig. 3b), 4^{+}_{1} for the state at 1,174 keV decaying through a 728 keV line (Fig. 3c) and 6⁺ for the 2,191 keV state de-excited through the 1,016 keV transition (Fig. 3d). As seen in Fig. 3a, the calculated 0^+_2 and 2^+_2 distributions are very similar in shape and, after folding in the experimental resolutions, overlap for all practical purposes. The assignment of the level, de-excited through

the 933 keV transition as the 0^+_2 rather than the 2^+_2 state, is discussed below based on comparisons with the LSSM calculations.

The low-lying ⁶²Cr level scheme extracted from the present measurement is compared with LNPS shell-model calculations in Fig. 4b. The latter indicates that for each firmly placed level below 2.3 MeV, its shell-model counterpart was calculated within less than 100 keV, showing notable agreement between experiment and theory. The figure also includes the 1,509 keV γ-ray that is proposed to correspond to the 2^+ \rightarrow 2⁺ transition. It forms an energy doublet with a peak at 1,492 keV feeding the 4^{+}_{1} state (inset of Fig. 4a). Obviously, no momentum distribution analysis was possible to confirm the 2^{+}_{2} assignment. Similarly, a candidate for the third $2⁺$ level is proposed, for which the decays to the ground and 2^{+}_{\perp} states are reported with a branching ratio close to the prediction. The notable good agreement with the shell-model calculations strongly supports the 0^+_2 assignment for the 1,379 keV state reported here as well as the 2^+_2 and 2^+_3 assignments. Note that, although both the 2^{+}_{2} and the 2^{+}_{3} levels are expected to decay to the 0^{+}_{2} state, the predicted branching ratios for these transitions are too small to be observed in the present experiment. For example, the γ-ray corresponding to the $2^+_3 \rightarrow 0^+_2$ transition would have a peak area of less than 16% that of the 933 keV one (see the level scheme in Fig. 4b for the branching ratios).

From the longitudinal-momentum-distribution analysis, together with the successful comparison of the proposed level scheme to shell-model calculations, these data establish the $2^+_1, 4^+_1, 6^+_1, 0^+_2, 2^+_2$ and 2^{+}_{3} states in ⁶²Cr, a nucleus close to ⁶⁴Cr, the centre of the *N* = 40 island of inversion. In the following, we will discuss the structure and shape

Fig. 3 | Measured and calculated longitudinal momentum distributions for 62Cr. a–**d**, For the two-proton knockout to individual final states in 62Cr, the shape of the longitudinal momentum distributions depends on the total angular momentum of the final state *J*, with the width increasing with *J*. **a**, The bare calculated momentum distributions transformed into the laboratory frame together with schematic configurations of the two protons that were knocked out in the reaction. **b**–**d**, Experimental data (black) compared with the calculated momentum distributions (red curves), folded with all experimental resolutions (Methods) and coincident to 933 keV (**b**), 728 keV (**c**) and 1,016 keV (**d**). Here, $\Delta p_{\parallel} = p_{\parallel} - p_0$, where $p_0 = 27.1$ GeV/*c* is the longitudinal momentum corresponding to a central path through the spectrometer and *p*∥ is the event-by-event reconstructed longitudinal momentum of ⁶²Cr. The error bars are the statistical standard deviations as propagated through the subtractions. The bins of the histograms cover 53.4 MeV/*c* in momentum.

Fig. 4 | Experimental data underlying the level scheme of 62Cr. a, γγ coincidence matrix and cuts of spectra in coincidence with 2^+_1 (blue) and 4^+_1 $\frac{1}{1}$ (orange, filled) states showing that the peak structure at 1,500 keV is a doublet. The 1,509 keV line is attributed to the $2^+_2 \rightarrow 2^+_1$ transition in coincidence with the 2+ ¹ level. The relevant peaks are labelled by their energy. **b**, Comparison of the

proposed (expt., experimental) low-lying level scheme (left) with the LNPS shell-model calculations discussed in the text (right). Shown are level energies and the associated *J*^π spins and parity (blue and grey for experiment and calculations, respectively) and γ-ray transition energies in black. Measured and calculated branching ratios are in red. *J*^m assignments in brackets are tentative.

of this key nucleus and present an informed prediction for the structure of the pivotal^{20,22-25} $N = 40$ isotone ⁶⁰Ca.

Calculations and evidence of shape coexistence

In this section, the present experimental data are discussed in the context of LSSM diagonalizations and interpreted within the recently developed DNO-SM¹⁹, which provides a description in terms of nuclear shapes. In both cases, the valence space is composed of the *pf* shell for protons and the $1p_{3/2}$, $0f_{5/2}$, $1p_{1/2}$, $0g_{9/2}$ and $1d_{5/2}$ orbitals for neutrons, on top of an inert⁴⁸Ca core.

The interaction is the well-known LNPS Hamiltonian²¹ with minor adjustments, which has enabled the successful description of many recent experimental findings in the neutron-rich region of the nuclear chart between 48 Ca and 78 Ni (refs. 29,34). As previously described, Fig. 4b compares the experimental and calculated level sequences; the agreement is excellent.

To characterize the spectroscopic shell-model results with respect to deformation and shapes, an analysis based on the DNO-SM model was undertaken. This approach enabled diagonalization of the same LNPS effective Hamiltonian and use of the same valence space within a relevant basis of deformed Hartree–Fock (HF) states from a potential energy surface represented in the (*β*, *γ*) plane. Here, *β* is the quadrupole deformation parameter and *γ* measures possible deviations from axial symmetry (*γ* = 0° and 60° for axial prolate and oblate shapes, respectively).

Spectroscopic results for both methods are displayed in Fig. 5. The calculated states can be grouped into three collective structures: the ground-state band and two sidebands, hereafter labelled the yrast, excited and *γ* bands. These three all exhibit collectivity with large intraband quadrupole transition strengths. In a traditional rotational picture, where the *K* quantum number is the projection of the total angular momentum onto the symmetry axis, the bands built on the 0^+_1 and 0^+_2 2 states would be assigned as *K* = 0 bands whereas that based on the 2^{+}_{3} level would correspond to $K=2$. Inspection of the properties of the 2^{+}_{2} and 2^{+}_{3} states indicates small spectroscopic quadrupole moments as well as a sizeable quadrupole transition between them. This points to *K* mixing and is associated with a degree of triaxial deformation. These findings were confirmed, as the first three states of each band expand into the (*β*, *γ*) plane, as shown in Fig. 6. The leading amplitudes are illustrated by orange circles. The following points are noteworthy: (1) There is a notable similarity in the expansion of the members in each band. (2) All three bands expand towards non-axial shapes. (3) The

Fig. 5 | Level schemes predicted by the DNO-SM and LSSM calculations. The calculations used the same valence space and the LNPS effective interaction. The levels are labelled by excitation energy and *J* π quantum numbers. Excited bands are predicted on top of the 2^+_3 and 0^+_2 states (labelled *γ* and excited band, respectively). Transitions within a band and out of the band structures are indicated by black and grey arrows, respectively, and are labelled by their transition strength. The Kumar invariants for the 0^+_1 and 0^+_2 states at fixed configurations (4p4h and 2p2h, respectively) are shown as well. The dots and shaded areas indicate the mean values of the *β* and *γ* deformations and their variances, respectively.

deformation is the largest for the yrast band. (4) The expansions on top of the 0^+_2 and 2^+_3 states closely mirror each other, indicating a clear correspondence between their respective characters.

The nature of the collectivity in ⁶²Cr can be understood further from the quadrupole properties derived from the Nilsson-SU3 model³⁵. An analysis following refs. 2,35, sketched in Extended Data Figs. 1 and 2 and Extended Data Table 2, revealed that the ⁶²Cr ground state can be characterized by four-particle, four-hole (4p4h) configurations associated with excitations across the *N* = 40 gap, which correspond to an axially symmetric solution. The excited bands, dominated by 2p2h configurations, correspond to a non-axially symmetric solution with *K* mixing. This is in agreement with the DNO expansion in Fig. 6, where the ground-state wavefunction is seen to develop around axial solutions (*γ* ≈ 0°) and where both excited bands expand towards non-axial

Fig. 6 | Potential energy surfaces in the *β***–***γ* **deformation plane.** For the states of⁶²Cr labelled by *J*^π, the potential energy surface (potential energy *E*, colourcoded on a scale of mega-electronvolts with solid and dashed lines representing the energy contours with 1 and 0.5 MeV distance, respectively) reveals the

importance of the triaxial degree of freedom (*γ* = 30°). The orange dots signal the dominant constrained HF configurations of the band structures. The area of an orange dot is directly proportional to the probability of the (*β*, *γ*) components in the total DNO wavefunction.

ones (*γ* ≈ 30°). Based on these considerations, the three bands observed in 62 Cr can be viewed as remnants of pure 4p4h excitations for the yrast band and pure 2p2h configurations for the excited bands, the first being axially symmetric and the latter two triaxial. This is also consistent with the Kumar plots at fixed particle–hole content shown in Fig. 5.

Given the successful description of 62 Cr, we now extrapolate towards the neutron-rich end of the key calcium isotopic chain. Indeed, within the present theoretical description, the intruder features dominating the structure of ⁶²Cr extend to the titanium and calcium chains. Recent discussions of $60,62$ Ti (refs. 23,36) indicated that there are quadrupole excitations with strong *n*p*n*h content in the low-lying states. In the proton-magic calcium chain, the proton–neutron quadrupole force is no longer at play, but the neutron *N* = 40 shell gap is reduced (Fig. 28 in ref. 2), hence favouring pairing excitations from the *pf* to the *sdg* shells. The 56,58,60 Ca isotopes are predicted to exhibit constant 2 $^\mathrm{*}$ energies, as perhaps hinted at by recent measurements²⁴. A large seniority *ν* = 0 component was calculated for their ground states (meaning that there will be almost no unpaired neutrons), a signature of a superfluid regime³⁷. A detailed decomposition of the ^{56,58,60}Ca ground-state wavefunctions is provided in Table 1.

As the energies of the first 2⁺ states in neutron-rich even-even Ca nuclei were some of the telltale observables in early experiments that aimed to understand the evolution of nuclear structure towards ⁶⁰Ca, it is interesting to explore the consequences of the present calculations for these $E(2^+_1)$ values. The absolute 2^+ energies in these isotopes have all been predicted to lie between 1.5 and 2.0 MeV, which is slightly above experimental values when available²⁴. A possible reason for this overestimation is the influence of the $s_{1/2}$ orbital computed to be at the Fermi surface in

Table 1 | Characteristics of 56,58,60Ca

Total occupation $n_{(g_9+d_5)}$ of the $g_{9/2}$ and $d_{5/2}$ neutron intruder orbitals and percentages for particle–hole excitations across the *N*=40 gap in the ground states of neutron-rich Ca isotopes. The last column is the seniority *ν* = 0 content in the ground-state wavefunction.

 $60Ca$ in ab initio coupled cluster calculations³⁸. Preliminary calculations extending the valence space used here to incorporate this orbital for the calcium isotopes depopulate the $g_{9/2}$ and $d_{5/2}$ orbitals somewhat and, indeed, reduce the 2⁺ excitation energies by about 300 keV, in agreement with recent experimental findings²⁴, pointing at another important factor in the description of the most neutron-rich Ca isotopes.

Outlook

Strong experimental and theoretical evidence has been provided for the observation of the excited 0^+_2 , 2^+_2 and 2^+_3 states in ⁶²Cr, a central tenant of the *N* = 40 island of inversion. Within the framework of LSSM calculations complemented by DNO-SM computations, an interpretation in terms of nuclear shapes emerges in which the calculated shape-coexisting structure built on the 0^+_2 level is associated with a non-axial shape and is dominated by 2p2h intruder configurations straddling the *N* = 40 shell gap. This structure should be contrasted with the axially symmetric, strong prolate one calculated for the collective band built on the ground state and predicted to be of dominant 4p4h character. The 4p4h components of the ground-state band then place ⁶²Cr as the portal to the $N=40$ island-of-inversion regime. Furthermore, the observed decay pattern together with the correspondence with the calculations leads to the assignment of the 2^+_3 state as the head of the predicted *γ* band. The calculations indicate sizeable mixing with the 0^+_2 band structure and a similar shape for both excitations. Armed with this agreement between experiment and theory, we extrapolate towards the nominally doubly magic 60Ca nucleus and predict a 2+ 1 excitation energy of the order of ~1 MeV as in the *N* = 36 and 38 isotones. Further tests of the detailed predictions for nuclear shapes and associated shell-model configurations in 62Cr will soon be possible, for example, by using low-energy Coulomb excitation and comprehensive transfer reactions, as FRIB's capabilities are ramped up.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at<https://doi.org/10.1038/s41567-024-02680-0>.

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Methods

In-beam γ-ray spectroscopy

Exotic light and medium-mass nuclei furthest away from stability are efficiently produced by fragmentation of stable beams impinging upon stable targets at high beam energies. The resulting secondary rare-isotope beam, here ⁶⁴Fe, is then available for experiments at velocities in excess of $v/c = 0.4$. In the experimental scheme employed here, the excited states of the reaction residue, 62 Cr, produced in the collision of the fast rare-isotope beam with the target, were tagged through γ-ray spectroscopy, a technique that is subject to large Doppler shifts due to the high velocity of the emitter. The GRETINA γ-ray tracking array is highly segmented and enables subsegment position resolution from online signal decomposition^{17,18}. The spatial coordinates *x*, *y* and *z* of the first interaction point of each γ-ray event within each germanium detector, as identified by the energy deposition being the largest, were then used to determine the γ-ray emission angle. The velocity of the emitting nucleus and the γ-ray emission angle were subsequently input into the event-by-event Doppler reconstruction, which transformed the γ-ray spectrum detected in the laboratory frame into the rest frame of the moving 62Cr ion. As detailed in ref. 29, in-beam γ-ray spectroscopy at these high velocities bears an inherent energy uncertainty related to the precise target location relative to the GRETINA centre, the excited-state lifetime and whether the associated γ-ray emission point is within or behind the target. Such systematic errors are included in the uncertainties quoted for γ-ray transition energies in addition to the typically small fitting uncertainty and are why we used for the 2^+_1 $\frac{1}{1}$ excited-state energy the precise value from β decay rather than the value determined from the transition energy measured here.

Particle identification and reconstruction of momentum distributions in the S800 spectrograph

The projectile-like reaction residues emerging from the target were characterized event-by-event through their energy loss, measured in the S800 spectrograph's ionization chamber²⁸, their (x, y) positions and angles determined from two cathode read-out drift chambers, and time-of-flight measurements made between plastic scintillators in the DB3 position of the FRIB's fragment separator²⁷, the S800 analysis line object location and the back of the S800 focal plane. The energy loss was used to separate the observed isotopic chains by *Z*. The position and angle information measured in the focal plane, together with an ion optical transfer map that accounts for aberrations to fifth order, were used to reconstruct the longitudinal momentum distributions and were input into a trajectory correction of the time of flight, which was used to separate the masses *A* for a given isotopic chain with proton number *Z*. The S800 analysis beamline was operated in the so-called focus mode, where the beam was focused on the reaction target.

Reaction theory and comparisons of the measured and calculated parallel momentum distributions

A direct reaction model was used to predict the theoretical two-proton removal cross sections and the longitudinal momentum distributions of the residual ⁶²Cr nuclei^{13–15}. The high incident velocities $v/c \approx 0.4$ enabled use of the eikonal approximation for the reaction dynamics $31-33$, where the reacting constituents travel in straight-line paths for the short duration of their interactions. The protons were assumed to be removed in a single collision, as a result of elastic or inelastic interactions with a single ⁹Be target nucleus. The interactions of the removed protons and residual nuclei with the target were described by complex optical potentials. The wavefunctions of the projectile (the incident ⁶⁴Fe projectile in its ground state) and of the populated final states of the residual nuclei (${}^{62}Cr(E^*, J^{\pi})$) were computed using LSSM. The details of the orbital occupancies of the correlated, removed protons (their two-nucleon amplitudes) were obtained from the overlaps, $\langle ^{62}Cr$ (*J*ⁿ) | ⁶⁴Fe), of these many-body, shell-model wavefunctions. Combined, these physical inputs identified whether the two-proton

removal reaction events arose from surface-grazing collisions of the projectile and target. The spectator core assumption implies that the proton-removing collisions did not alter the state of the nucleons making up the heavy residual nuclei. The differential removal cross sections, with respect to the longitudinal momenta of the 62 Cr residues, were calculated consistently within the same model¹³⁻¹⁵.

The theoretical longitudinal-momentum distributions of the ^{62}Cr nuclei were calculated in the projectile rest frame and transformed into the laboratory frame, as seen in Fig. 3. They were then folded with a rectangular function to take into account the range of possible momentum losses through the thick ⁹Be target by reactions occurring at different depths. Finally, the theoretical distributions were convolved with the experimental longitudinal-momentum distribution from unreacted 64Fe projectiles (exploiting the charge state present in Fig. 1) in coincidence with γ-rays with energies above 500 keV. This approach empirically modelled the dissipative interactions taking place in the target and introduced the low-momentum tails ubiquitously observed in direct knockout $16,39$. The theoretical and experimental longitudinal-momentum distributions for different levels in 62Cr are compared in Fig. 3. The theoretical values have been scaled and slightly shifted to best fit the central bins. The fitting results for each *J*^{π} hypothesis $\{0^+_2; 2^+_2; 4^+_1; 6^+_1\}$ are as follows: 933 keV with $\chi^2_{\text{red}} = \{0.6; 1.0; 3.0; 4.3\}$, 728 keV with $\chi^2_{\text{red}} = \{28.1; 16.4; 1.5; 3.1\}$ and 1,016 keV with $\chi^2_{\text{red}} = \{9.8; 7.4; 1.9; 0.6\}$. The left tail, which was modelled only empirically, was omitted from the fit.

LSSM calculations

The LSSM calculations were performed within a valence space including the full *pf* shell for the protons and the $1p_{3/2}$, $0f_{5/2}$ and $1p_{1/2}$ orbitals together with the Δ*j* = 2, Δ*l* = 2, quasi-SU3 sequence 0*g*_{9/2}, $1d_{5/2}$ for the neutrons. The original LNPS effective interaction was used 21 with only minor modifications. Such calculations have provided full-space diagonalizations involving bases of more than one billion Slater determinants. In the calculation of the Kumar invariants, we followed the sum-rule method discussed recently in ref. 40 to obtain an exact extraction of quadrupole invariants and their variances. The seniority of the calcium isotopes was extracted with the *J*-coupled code NATHAN^{41,42}. This shell-model code expands the wavefunction into basis states of good seniority numbers using the quasi-spin formalism⁴³.

Discrete non-orthogonal shell model

In addition to the LSSM diagonalization, the recently developed DNO-SM19 was used with the same model space and effective Hamiltonian. This method enables the decomposition of the shell-model wavefunctions into different deformed HF configurations, which provides direct access to their respective contributions to the deformation and, hence, enables a quantitative analysis in terms of nuclear shapes in both the ground and excited states. It is important to keep in mind that, variationally, the absolute DNO-SM spectra represent an upper bound to the exact shell-model spectra and that DNO-SM wavefunctions should converge to exact shell-model solutions. In ref. 19, only the quadrupole degrees of freedom (*β*, *γ*) and, in some cases, cranking components were included. The results presented in this work were obtained within the recently extended DNO-SM[ph] framework, which also simultaneously takes into account deformed HF states and particle–hole excitations built on top of these. Each DNO-SM state is then a superposition of both the deformed HF states (visualized as single points in the potential energy surface of Fig. 6) and their particle–hole excitations (lying a priori outside the (*β*, *γ*) potential energy surface). The expansion of the DNO-SM[ph] states was optimized in a two-step procedure. First, relevant deformed HF states in the (*β*, *γ*) plane were selected with the Caurier minimization technique developed in ref. 19, which allowed us to include the fewest constrained HF configurations without having to perform a full discretization of the entire potential energy surface. Next, particle–hole excitations on top of

the constrained HF states found in the first step were incorporated in the same manner. The DNO-SM results in Fig. 5 indicate an almost perfect match with the shell-model spectrum for all three bands. For instance, in general, the ground-state band would be more compressed (if confined to the quadrupole degrees of freedom only), due mostly to the lack of ground-state pairing correlations. A study of this extended DNO-SM[ph] approach will be presented elsewhere.

Algebraic SU3 analysis

The interpretation of collectivity can be supplemented by algebraic models. SU3 symmetry 44 assumes a valence space composed of a main harmonic oscillator shell with degenerate orbits with a pure quadrupole interaction. Under these assumptions, the nuclear configuration maximizing the quadrupole moment becomes the ground state. In mid-mass nuclei, the spin–orbit interaction breaks the degeneracy of the orbits and destroys the SU3 symmetry. Nevertheless, its variants-pseudo-SU3⁴⁵ and quasi-SU3⁴⁶-can be applied in reduced shell-model spaces³⁵. The pseudo-SU3 dynamical symmetry is achieved in an harmonic oscillator shell when the lowest large *j* shell is closed. For quasi-SU3 dynamical symmetry, this is the case when Δ*j* = 2 orbits are present at the Fermi surface. In the present case, the LNPS valence space can be split into three groups of orbitals. For neutrons, the pseudo-SU3 symmetry applies in the space spanned by the $2p_{3/2}$, $2p_{1/2}$ and $1f_{5/2}$ orbitals, and the quasi-SU3 symmetry applies in the space formed by the 1g_{9/2}, $2d_{5/2}$ and $3s_{1/2}$ orbitals. For protons, the quasi-SU3 symmetry applies in the space formed by the $1f_{7/2}$ and $2p_{3/2}$ orbits. The single-particle quadrupole moments were obtained by diagonalizing the quadrupole force in the LNPS space. Normalized quadrupole moments, Q_0/b^2 , with *b* being the harmonic oscillator size parameter, are plotted in Extended Data Figs. 1 and 2 for different types of configuration.

The quadrupole moment of the nucleus was obtained by summing the single-particle moments. For 62 Cr, the maximum quadrupole moment, built from the four protons and ten neutrons above the ⁴⁸Ca core, was obtained by filling the Nilsson-SU3 'orbits'. Extended Data Fig. 1 illustrates the 4p4h ground state corresponding to four neutron particles in the quasi-*sdg* orbitals, four neutron holes in the pseudo-*sd* ones and four proton particles in quasi-*pf* orbits. The 2p2h configuration corresponding to the excited band is given in Extended Data Fig. 2. From the obtained configurations, notice that the 4p4h configuration corresponds to an axial solution with a unique filling of the Nilsson-SU3 levels, whereas the 2p2h configuration corresponds to a non-unique filling of the Nilsson-SU3 levels for the neutron holes and, hence, results in *K* mixing and a triaxial shape.

Data availability

This submission includes as additional supplemental files the relevant data supporting the findings of these studies. Other data are available from the corresponding author upon reasonable request.

Code availability

The techniques developed and exploited in the unpublished computer codes used to generate the results reported in this paper are presented in detail in published works. These works and reasonable requests for clarifications of the techniques or computational methods used are available from the corresponding author.

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Author contributions

A.G., R.V.F.J. and J.A.T. conceived and led the proposal for the experiment based on discussions with and calculations from F.N., S.M.L. and A.P. C.M.C., S.G., S.N., J.P. and D.W. led the operations of the experimental instruments used. A.G., B.L., R.V.F.J., A.D.A., M.J.B., C.M.C., M.P.C., J.C.-J., H.L.C., B.P.C., P.F., A.M.H., C.P. and E.R. stafed the shifts and monitored the online data quality. A.G. and B.L. led the data analysis. A.G., B.L., R.V.F.J., D.D.D., F.N. and J.A.T. led the interpretation of the data and wrote the first draft. J.A.T., D.D.D., F.N. and A.P. performed the reaction theory and nuclear structure theory calculations.

Competing interests

The authors declare no competing interests.

Additional information

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Extended Data Fig. 1 | Quasi- and Pseudo-SU3 plots for the 4p4h configuration in 62Cr. Left: neutron valence space and single-particle quadrupole moments characterized by the projection of the total angular momentum on the symmetry axis, *K*. Right: same illustration for protons. The 4p4h configuration corresponds to an axial solution with unique filling of the Nilsson-SU3 levels. The red dots indicate the occupation of the single-particle orbitals with neutrons and protons, respectively.

Extended Data Fig. 2 | Quasi- and Pseudo-SU3 plot for the 2p2h configuration in 62Cr. Left: neutron valence space and single-particle quadrupole moments characterized by the projection of the total angular momentum on the symmetry axis, *K*. Right: same plot for protons. The 2p2h configuration corresponds to a

non-unique Nilsson-SU3 levels filling for the neutron holes and, hence, *K* mixing and a triaxial shape. The red dots indicate the occupation of the single-particle orbitals with neutrons and protons, respectively.

Extended Data Table 1 | States populated in ⁶²Cr in the two-proton removal from ⁶⁴Fe projectiles

Listed are the excitation energy E_J and its spin-parity J, measured (exp) and calculated (th) cross sections σ , transition energy E_v, final-state energy E_v and spin-parity J_r. Spin-parity assignments in brackets are based on comparison with theory. There are unplaced *γ* rays at 2095, 2125, 2213, and 2965 keV that, in total, carry 0.029 mb of cross section. States in italic are tentative – a coincidence of the corresponding transition with the 4 $^+_1$ was firmly observed but, due to low statistics, the possibility that these transitions are actually feeding the 6 * state cannot be excluded. The partial cross section to the ground state was obtained from the inclusive one via subtraction. In addition to the uncertainty originating from unplaced, observed potential feeders, the possibility of unobserved feeding will make all partial cross sections upper limits except for σ(0 $_2^+$) for which the absence of any significant feeding is argued via Fig. 2. The inclusive cross section is 0.59(4) mb.

Extended Data Table 2 | Computed electric quadrupole transition strength, $B(E2;2^+_1\to0^+_1)$, using different models

Results for Quasi/Pseudo-SU3 estimations (QPSU3), Projection of Hartree-Fock solutions (PHF) having the same particle-hole *n*p*n*h configuration as the QPSU3 estimate, LSSM calculation using the fixed np-nh configurations (n fix), DNO shell-model calculations (DNO-SM), shell-model diagonalization (SM) for the case of ⁶²Cr, and the measured value (exp). Comparing the results from SM, DNO-SM, and n fix, one can see that the ground state is rather of 4p4h nature with a slight residual mixing in the wave function which reduces the SM B(E2) value with respect to the 'n fix' calculation.