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Isospin Properties of Nuclear Pair Correlations from the Level Structure of the Self-Conjugate Nucleus ^{88}Ru

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The low-lying energy spectrum of the extremely neutron-deficient self-conjugate ($N = Z$) nuclide $^{88}_{44}\text{Ru}_{44}$ has been measured using the combination of the Advanced Gamma Tracking Array (AGATA) spectrometer, the NEDA and Neutron Wall neutron detector arrays, and the DIAMANT charged particle detector array. Excited states in ^{88}Ru were populated via the $^{54}\text{Fe}(^{36}\text{Ar}, 2n\gamma)^{88}\text{Ru}^*$ fusion-evaporation reaction at the Grand Accélérateur National d'Ions Lourds (GANIL) accelerator complex. The observed γ -ray cascade is assigned to ^{88}Ru using clean prompt γ - γ -2-neutron coincidences in anticoincidence with the detection of charged particles, confirming and extending the previously assigned sequence of low-lying excited states. It is consistent with a moderately deformed rotating system exhibiting a band crossing at a rotational frequency that is significantly higher than standard theoretical predictions with isovector pairing, as well as observations in neighboring $N > Z$ nuclides. The direct observation of such a “delayed” rotational alignment in a deformed $N = Z$ nucleus is in agreement with theoretical predictions related to the presence of strong isoscalar neutron-proton pair correlations.

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Introduction.—Nucleonic pair correlations play an important role for the structure of atomic nuclei as well as for their masses. Some of the most well-known manifestations of the pairing effect in nuclei, which has strong similarities with superconductivity and superfluidity in condensed matter physics [Bardeen-Cooper-Schrieffer (BCS) theory [1,2]], are the odd-even staggering of nuclear masses [3], seniority symmetry [4–6] in the low-lying spectra of spherical even-even nuclei, and the reduced moments of inertia and backbending effect [7,8] in rotating deformed nuclei. Atomic nuclei, which are formed by the unique coexistence of two distinct fermionic systems (neutrons and protons), may also exhibit additional pairing phenomena not found elsewhere in nature. In nuclei with equal neutron and proton numbers ($N = Z$) enhanced correlations arise between neutrons and protons that occupy orbitals with the same quantum numbers. Such correlations have been predicted to favor a new type of nuclear superfluidity, termed isoscalar neutron-proton (np) pairing [9–12]. In addition to the normal isovector ($T = 1$) pairing mode based on like-particle neutron-neutron (nn) and proton-proton (pp) Cooper pairs that have their spin vectors antialigned and occupy time-reversed orbits, neutrons and protons may here also form np $T = 1$, $I = 0$ pairs. Of special interest is the long-standing question of the possible presence of a np pairing condensate [9–15] predicted to be built primarily from isoscalar $T = 0$, $I > 0$ np pair correlations that still eludes experimental verification. The occurrence of a significant component of $T = 0$ correlated np pairs in the nuclear wave function is also likely to have other interesting implications, e.g., the proposed “isoscalar spin-aligned np coupling scheme” in the heaviest, spherical, $N = Z$ nuclei [16].

Despite vigorous activity over the last decade or so, the fundamental questions concerning the basic building blocks and fingerprints of np pairing are still a matter of considerable debate. Even though until now there has been no substantial evidence for the need to include isoscalar, $T = 0$, np pairing to explain the known properties of

low- or high-spin states in even-even $N = Z$ nuclei the available data for the heavier $N = Z$ nuclei are very limited due to experimental difficulties: No accurate information on masses for $N = Z$ nuclei above $A \approx 80$ is currently known, shape coexistence effects have muddled the analysis of rotational patterns of deformed $N = Z$ nuclei in the mass $A \sim 70$ region, and np transfer reaction studies on the lighter $N = Z$ nuclei are suffering from the complexity in the interpretation of the experimental results. Furthermore, correlations of this type are enhanced in heavier nuclei where more particles in high- j shells can participate. Many theoretical calculations suggest that the best place to look for evidence of an isoscalar pairing condensate is in nuclei with $A > 80$; for a recent review, see Ref. [17]. Calculations using isospin-generalized BCS equations and the Hartree-Fock-Bogoliubov (HFB) equation including pp , nn , np ($T = 1$), and np ($T = 0$) Cooper pairs indicated that there may exist a second-order quantum phase transition in the ground states of $N = Z$ nuclei from $T = 1$ pairing below mass 80 to a predominantly $T = 0$ pairing phase above mass 90, with the intermediate mass 80–90 region showing a coexistence of $T = 0$ and $T = 1$ pairing modes [18]. There are even predictions for a dominantly $T = 0$ ground-state pairing condensate in $N \sim Z$ nuclei around mass 130 [19] (although such exotic nuclei are currently not experimentally accessible).

The interplay between rotation and the like-particle pairing interaction has been studied in great detail in deformed nuclei where, normally, the neutron and proton Fermi levels are situated in different (sub-) shells; and hence the neutrons and protons can be considered to form separate Fermi liquids dominated by $T = 1$ pair correlations. However, the isoscalar, $T = 0$, np coupling has the interesting property of being less affected by the Coriolis interaction in a rotating system, which tends to break the time-reversed pairs with $T = 1$. Therefore, the presence of a np pairing condensate may reveal itself in the rotational states of deformed $N = Z$ nuclei where one might expect that the $T = 0$ pairing correlations are active while

the normal isovector pairing mode is suppressed by the Coriolis antipairing effect [20]. Calculations within the isospin-generalized HFB framework indeed also suggested such a mixed $T = 1/T = 0$ pairing phase with a transition from $T = 1$ to $T = 0$ dominance as a function of increasing angular momentum [21]. Hence, medium- to high-spin states of rotating $N = Z$ nuclei appear to be among the best places to search for the presence of $T = 0$ np pairing, and it is important to reach the heaviest possible $N = Z$ nuclei where, however, the experimental conditions are most challenging. One of the key signatures proposed for isoscalar pairing is a significant “delay” in band crossing frequency in deformed $N = Z$ isotopes compared with their $N > Z$ neighbors, which necessitates the study of such nuclei up to angular momentum around $I = 10\hbar$ or higher [17]. Such delays have previously been observed in the deformed $N = Z$ nuclei $^{72}_{36}\text{Kr}_{36}$, $^{76}_{38}\text{Sr}_{38}$, and $^{80}_{40}\text{Zr}_{40}$ but were not considered as conclusive evidence for isoscalar np -pairing effects due to the possible influence of shape coexistence on the alignment frequencies [22–24]. The nuclei $^{84}_{42}\text{Mo}_{42}$ and $^{88}_{44}\text{Ru}_{44}$ also have indications of delays in the rotational alignments; however in these cases the experimental data did not reach the required rotational frequency in order to draw firm conclusions [25,26]. The nucleus ^{88}Ru is here of particular interest, as it is predicted to be the last deformed self-conjugate nuclear system before the $N = Z = 50$ closed shells [27]. The structure of its intermediate-to-high-spin states constitutes one of the most promising cases for discovering effects of a BCS-type of isoscalar pairing condensate. However, due to the large experimental difficulties in producing and selecting such exotic nuclei in sufficient quantities excited states in ^{88}Ru were previously known only up to the $I^\pi = 8^+$ state [25], just where normal (isovector) paired band crossings are expected to appear in the absence of strong isoscalar pairing. In the present work the level scheme of ^{88}Ru has been extended to higher angular momentum states in the ground-state band, leading to a conclusive measurement of the rotational alignment frequency. The experimental difficulties have been overcome through the use of a highly efficient, state-of-the-art detector system and a prolonged experimental running period.

Experimental details.—Excited states in ^{88}Ru were populated in fusion-evaporation reactions induced by a ^{36}Ar beam produced by the CIME cyclotron at the Grand Accélérateur National d’Ions Lourds (GANIL), Caen, France. The ^{36}Ar ions were accelerated to an energy of 115 MeV and used to bombard target foils consisting of 99.9% isotopically enriched ^{54}Fe with areal density of 6 mg/cm², which was sufficient to stop the fusion products of interest. The beam intensity varied between 5 and 10 pA with an average of 7 pA during 13 days of irradiation time. Prompt γ rays emitted in the reactions were detected by the Advanced Gamma Tracking Array (AGATA) spectrometer [28] in its early phase 1 implementation [29], consisting of 11 triple

clusters of segmented HPGe detectors. Emission of light charged particles and neutrons was detected in prompt coincidence with the γ rays by the nearly 4π solid angle charged particle detector array DIAMANT [30,31], consisting of 64 CsI(Tl) scintillators, and the neutron wall [32] and NEDA [33,34] neutron detector arrays consisting of 42 and 54 organic liquid-scintillator detectors, respectively. The trigger condition for recording events for subsequent off-line analysis was that at least two of the high-purity germanium crystal core signals from the AGATA triple-cluster detectors were registered in fast coincidence with at least one neutronlike event recorded in the liquid scintillator detectors. The condition for the neutronlike events was determined by pulse-shape discrimination (PSD) via a firmware threshold set for the so-called charge comparison (CC) ratio between the charge integrated over the tail part of each liquid scintillator pulse and its total integrated charge. Similar PSD criteria made it possible to discriminate between different types of charged particles detected in the CsI(Tl) scintillators. The final discrimination between neutrons and γ rays was performed off line by setting two-dimensional gates on the neutron time of flight vs the CC ratio. The rare two-neutron evaporation events were separated from events where a neutron scattered between detectors by applying simultaneous cuts on the deposited energy and time of flight as a function of the distance between detectors that fired. For the off-line charged particle selection, individual two-dimensional gates on the particle identification and energy parameters of the DIAMANT detectors enabled the identification of γ rays as belonging to specific charged particle evaporation channels. A 50 ns wide time gate was applied to the time-aligned Ge detector timing signals in order to select prompt γ -ray emission. The γ -ray energy measurements with AGATA rely on tracking algorithms [35–39] that reconstruct trajectories of incident γ -ray photons in order to determine their energy and direction. This is achieved by disentangling the interaction points and corresponding interaction energies in the germanium crystals that are identified using pulse shape analysis of the detector signals and thereafter establishing the proper sequences of interaction points using the characteristic features of the interaction mechanisms (primarily the photoelectric effect, Compton scattering, and pair production). The energy calibration of the germanium detectors was performed using standard radioactive sources (^{60}Co and ^{152}Eu). Figure 1 shows projected spectra from the $2n$ -selected $E_\gamma - E_\gamma$ coincidence matrix obtained requiring anticoincidence with detection of any charged particle in the DIAMANT CsI(Tl) detector array. The spectrum in Fig. 1(a) was produced for events where γ rays coincident with the 616, 800, 964, and 1100 keV transitions assigned to ^{88}Ru were selected. The background spectrum was produced by using identical energy cuts on a selection of the data requiring coincidence with two neutrons and a charged particle summed with the background spectrum obtained by shifting the energy cuts a

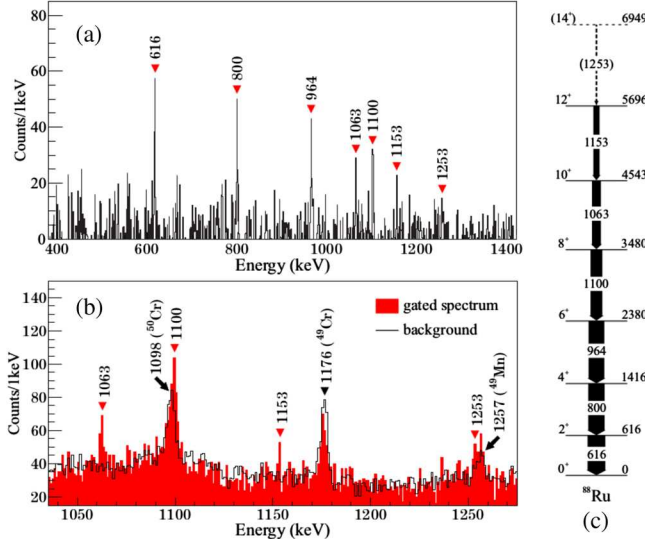


FIG. 1. (a) Gamma-ray energy spectrum detected in coincidence with the 616, 800, 964, and 1100 keV γ rays, with the additional requirement that two neutrons and no charged particles were detected in coincidence. (b) Expanded part of the unsubtracted gated spectrum around the new γ -ray transitions at 1063 keV ($10^+ \rightarrow 8^+$), 1153 keV ($12^+ \rightarrow 10^+$), and 1253 keV [$(14^+) \rightarrow 12^+$] is drawn in red together with the background spectrum (black) used to produce the spectrum shown in (a). Gamma-ray peaks due to contaminant reactions on oxygen leading to the population of excited states in $^{49,50}\text{Cr}$ and ^{49}Mn are indicated. (c) Level scheme of ^{88}Ru deduced from the present work. Relative intensities are proportional to the widths of the arrows.

constant offset of +20 keV in the two-neutron gated data requiring anticoincidence with the detection of charged particles. These transitions were previously identified as belonging to ^{88}Ru in a study involving a different reaction: $^{58}\text{Ni}(^{32}\text{S}, 2n\gamma)^{88}\text{Ru}^*$ [25]. All γ rays observed in prompt coincidence and assigned to the ground-state band of ^{88}Ru in this work are indicated with their energies in keV.

Discussion.—Figure 2 shows values of the kinematical moment of inertia ($J^{(1)}$) for the low-lying yrast level energy bands in the $N = 44$ isotones $^{88}\text{Ru}_{44}$ (this work), $^{86}\text{Mo}_{44}$ [40,41], and $^{84}\text{Zr}_{44}$ [42]. The ground-state bands in the even- Z , $N > Z$ isotones $^{86}\text{Mo}_{44}$ and $^{84}\text{Zr}_{44}$ exhibit a variation of $J^{(1)}$ (defined as the angular momentum, I , divided by the rotational frequency, $\omega = dE/dI$) as a function of rotational frequency that is characteristic of a normal paired band crossing in a rotating deformed nucleus of the isovector ($T = 1$) type. The band crossing frequency is $\hbar\omega_c \approx 0.47$ MeV in both cases (indicated by the black vertical dashed line in Fig. 2). For the $N = Z$ nucleus $^{88}\text{Ru}_{44}$ the increase in $J^{(1)}$ also resembles a paired band crossing, albeit at a significantly higher rotational frequency, $\hbar\omega_c \approx 0.54$ MeV, indicated by the red vertical dotted line in Fig. 2.

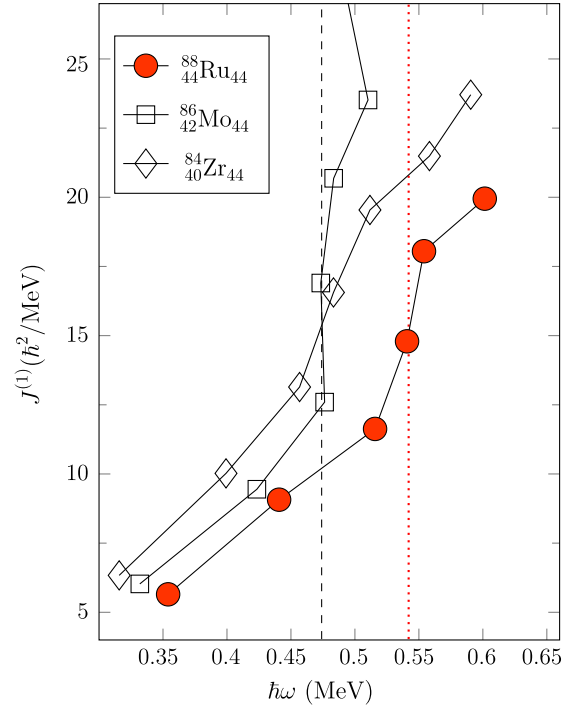


FIG. 2. Experimental values for the kinematical moment of inertia ($J^{(1)}$) for the low-lying yrast bands of the $N = 44$ isotones $^{88}\text{Ru}_{44}$ (this work), $^{86}\text{Mo}_{44}$ [40,41], and $^{84}\text{Zr}_{44}$ [42]. The black dashed vertical line indicates the approximate rotational frequency of the first isovector-paired band crossing due to $g_{9/2}$ protons as predicted by standard cranked shell model calculations [43,44]. The red dotted vertical line indicates the band crossing frequency for the ground-state band in $^{88}\text{Ru}_{44}$ observed in this work.

Theoretical predictions of the rotational response of excited states and the associated spin alignment can be provided by cranked shell model calculations [45], which predict the first proton two-quasiparticle $(\pi g_{9/2})^2$ alignment to occur at $\hbar\omega_c \approx 0.45$ MeV followed closely by a neutron $\nu(g_{9/2})^2$ alignment [43,44]. Mountford *et al.* have demonstrated that the first alignment in ^{84}Zr is due to $g_{9/2}$ protons by means of a transient-field g -factor measurement [46]. The slopes of the $J^{(1)}$ curves around the crossing point also exhibit an expected variation, reflecting the change in interaction strength between the ground-state band and the broken-pair S band as the proton Fermi level changes within the $g_{9/2}$ subshell. The large delay in band crossing frequency for $^{88}\text{Ru}_{44}$ compared with its closest $N = 44$ isotones can not readily be explained using standard mean field models.

Developments of computational methods in recent years enable shell model calculations to be performed with large model spaces, providing nuclear structure predictions for medium-mass nuclei away from closed shells. Large-scale shell-model (LSSM) calculations with an isospin-conserving Hamiltonian are also the method of choice

for theoretical investigations of the isospin dependence of nucleonic pair correlations [17]. In Ref. [26], projected shell model calculations following the approach of Ref. [47] predicted a delay in the band crossing frequency in the $N = Z$ nuclei $^{84}_{42}\text{Mo}_{42}$ and $^{88}_{44}\text{Ru}_{44}$ as an effect of enhanced neutron-proton interactions. Kaneko *et al.* [48] employed LSSM calculations using a “pairing-plus-multipole” Hamiltonian [49] in the $(1p_{1/2}, p_{3/2}, f_{5/2}, g_{9/2}, d_{5/2})$ (often denoted as *fpgd*) model space for studying $^{88}_{44}\text{Ru}_{44}$, $^{90}_{44}\text{Ru}_{46}$, and $^{92}_{44}\text{Ru}_{48}$ and concluded that $T = 0$ *np* pairing is responsible for the distinct difference in rotational behavior between the $N = Z$ and $N > Z$ nuclei. These calculations also predicted a significant delay in the band crossing frequency for $N = Z$ and their prediction for the $J^{(1)}$ moment of inertia of $^{88}_{44}\text{Ru}_{44}$ revealed a sharp irregularity at a rotational frequency $\hbar\omega_c \approx 0.65$ MeV [48]. We therefore conclude that the delayed alignment of $g_{9/2}$ protons observed in the ground-state band of ^{88}Ru in the present work is likely not to be in agreement with the response of a deformed rotating nucleus in the presence of a normal isovector pairing field and that isoscalar pairing components may be active in this self-conjugate nucleus.

Summary.—In summary, new γ -ray transitions in the self-conjugate nuclide $^{88}_{44}\text{Ru}_{44}$ have been identified, extending the previously reported level structure. The observed ground-state band exhibits a band crossing that is significantly delayed compared with the expected behavior of a rotating deformed nucleus in the presence of a normal isovector ($T = 1$) pairing field. The observation is in agreement with theoretical predictions for the presence of isoscalar neutron-proton pairing in the low-lying structure of ^{88}Ru .

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