

Discovery of a new isomeric state in ^{68}Ni : Evidence for a highly deformed proton intruder state

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We report on the observation of a new isomeric state in ^{68}Ni . We suggest that the newly observed state at 168(1) keV above the first 2^+ state is a $\pi(2p-2h) 0^+$ state across the major $Z = 28$ shell gap. Comparison with theoretical calculations indicates a pure proton intruder configuration and the deduced low-lying structure of this key nucleus suggests a possible shape coexistence scenario involving a highly deformed state.

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The atomic nucleus is a complex quantum system consisting of two kinds of strongly interacting fermions. A direct consequence of this fermionic nature, the Pauli principle, is the shell model of the nucleus, one property of which being the existence of magic gaps. Shell structures are present in a number of systems such as atoms, metal clusters, and quantum dots and wires, for instance, and are strongly linked to the symmetries of the mean field. How the shell gaps evolve in nuclei that are further and further away from stability is one of the key questions to which the radioactive beam facilities that are currently under construction hope to bring answers. Already today, the structure of moderately exotic nuclei such as ^{68}Ni allows one to pave the way toward a general answer to the problem of shell evolution. Unusual configurations which are expected to dominate in the ground-state structure of very exotic nuclei can be identified as excited structures in systems not very far away from stability. The strong contribution of the spin-orbit term in the nucleon-nucleon interaction affects in a major way the single-particle levels with the largest angular momentum, pushing it down in energy. This quenches significantly the $N = 40$ magic gap from the spherical harmonic oscillator. The intrusion of the $1g_{9/2}$ and the $2d_{5/2}$ neutron orbitals brings collectivity and enhances neutron-pair excitations across $N = 40$ from the fp shell into the $1g_{9/2}$. Conversely, however, this parity change hinders quadrupole excitation and mimics some properties usually associated to magicity. In ^{68}Ni , the observation of a first excited 0^+_2 state at low energy [1] and the high excitation energy of the 2^+_1 state [2] are examples of such properties. These competing consequences of shell quenching make ^{68}Ni a particularly suited case to study the evolution of shell gaps with isospin.

Reactions involving single-proton particle-hole excitations, $\pi(1p-1h)$, are an ideal tool to learn about the residual interaction. Unfortunately they lie at very high excitation energy. One possibility to circumvent this obstacle is to look for $\pi(2p-2h)$ states which are lowered in energy thanks to pairing correlations and proton-neutron residual interactions. Studying pair excitation across magic gaps means, therefore, studying these residual interactions. Pair excitations are revealed by the presence of excited 0^+ states. In ^{68}Ni , two such states are reported, mainly of neutron character, originating from the scattering of pairs into the $1\nu g_{9/2}$. A state corresponding to the excitation of two protons ($2p-2h$) has been predicted by Pauwels *et al.* [3,4] using the energy of the intruder $\pi(2p-1h)$ state in ^{69}Cu and, symmetrically, the $\pi(1p-2h)$ in ^{67}Co , which both lie at $N = 40$. The energy they derive leads to a low value of 2202 keV, which can be understood only with an important gain in binding energy from the $\pi - \nu$ residual interactions between the two proton-holes and the active valence neutrons across $N = 40$ [3,4]. The spin parity of the $\pi(1p-2h)$ state in ^{67}Co was proposed to be $(1/2^-)$ and corresponding to a prolate proton intruder configuration [3]. This shape isomer indicates therefore the presence of deformed low-lying proton intruder states below ^{68}Ni . Later on, evidences for such deformed ($\pi p_{3/2}$) intruder orbital were reported in the spectroscopy of odd mass Mn isotopes [5] in β -decay experiments. These results strongly suggest the presence of a deformed $\pi(2p-2h)$ intruder state in ^{68}Ni . However, despite the large number of experiments dedicated to ^{68}Ni , no evidence of proton-pair excitation has been reported so far, preventing us from a coherent understanding of the nuclear structure in this mass

region. In the present work, we report on the observation of a new isomeric state in ^{68}Ni which we propose to have a 0^+ character.

The experiment was performed at the Grand Accélérateur National d'Ions Lourds (GANIL) by using multinucleon transfer reactions in inverse kinematics. A ^{238}U beam at 6.33 A MeV bombarded a 1.3 mg/cm² thick ^{70}Zn target. The targetlike reaction products were detected and identified in the VAMOS spectrometer [6,7] used in a solenoid mode. The optical axis of the spectrometer was set at 45° with respect to the beam axis, such that the grazing angle was within the angular acceptance of the spectrometer. In this mode, the dipole of the spectrometer is not used and the transmission is increased by 50% with respect to the standard dispersive mode. The reaction products are refocused in a new detection setup [8] located at the focal plane of the spectrometer, which provides an unambiguous identification of the recoils on an event-by-event basis. The atomic number is measured by combining the energy loss in three successive ionization chambers and the residual energy deposited in four silicon detectors. The mass is determined from the total kinetic energy and the time of flight between silicon detectors and a multiwire proportional chamber located 138 mm downstream of the target. The flight path through the spectrometer is determined using secondary electron detectors [9] and following the procedure described in Ref. [6]. We have obtained a mass resolution $\Delta A/A = 1.2\%$ and an atomic number resolution $\Delta Z/Z = 1.1\%$. Prompt γ rays emitted at the target position were measured by 11 clover detectors from the EXOGAM array [10] in coincidence with the recoils identified in VAMOS. Delayed γ rays were also detected at the VAMOS focal plane by four high-purity germanium detectors facing the silicon detectors where the recoils were implanted. With a recoil velocity from 22 to 45 $\mu\text{m}/\text{ps}$, the flight time through the spectrometer was 200–400 ns. The lifetimes have been measured using a time-to-digital conversion module with a hardware gate of 3 μs . Our delayed spectroscopy setup is therefore sensitive to lifetimes from ~ 100 ns up to ~ 10 μs .

In this Rapid Communication, we focus on new results obtained on the ^{68}Ni key nucleus. Figure 1 shows the delayed γ -ray spectrum observed in coincidence with its identification

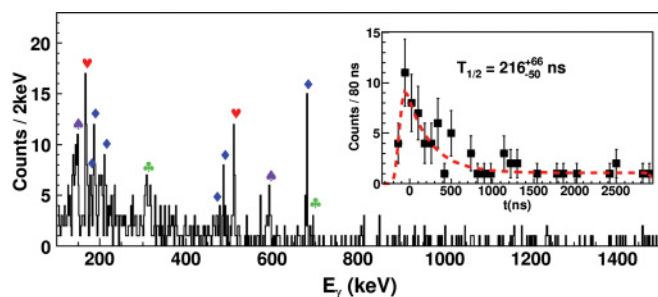


FIG. 1. (Color online) Delayed γ -ray spectrum in coincidence with ^{68}Ni . The purple spades, the blue diamonds, and the green clovers indicate, respectively, the ^{69}Ni , ^{69}Cu , and ^{67}Ni isomeric decay. The red hearts overscore the transition that belongs to ^{68}Ni . The inset shows the time spectrum from which the half-life of the 168(1)-keV transition is extracted.

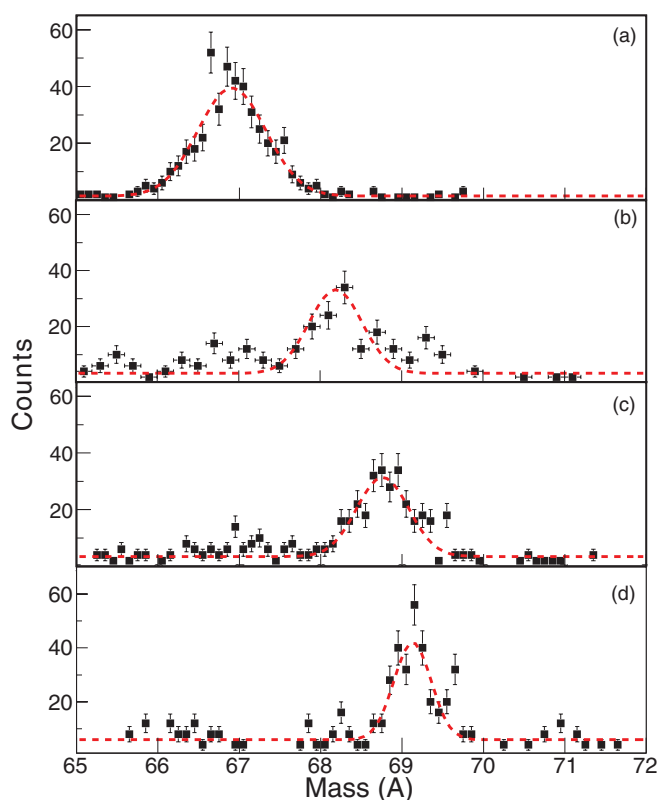


FIG. 2. (Color online) Mass spectra in coincidence with the observed delayed γ rays. The coincidence gates select (a) the 313 + 694-keV transitions in ^{67}Ni , (b) the 168-keV transition, (c) the ^{69}Cu lines, and (d) the 143 + 593-keV transitions in ^{69}Ni .

in VAMOS. Several isomers are known in ^{68}Ni : an 8^+ seniority isomer with a half-life $t_{1/2} = 23.3(11)$ ns [11], which decays in flight in the spectrometer; a 5^- state with $t_{1/2} = 0.86(5)$ ms [12], which is too long lived for its decay to be observed; and a 0_2^+ state with $t_{1/2} = 270(5)$ ns [13]. Given the mass and charge resolution we measured, the spectrum obtained after selection of ^{68}Ni is slightly contaminated by neighboring nuclei, $^{67,69}\text{Ni}$ and ^{69}Cu , in which isomers are known. All the transitions from their decay have been identified (see Fig. 1). In particular, the transitions observed in ^{69}Cu correspond to the decay of a $13/2^{+m}$ state. Its half-life is well known [$t_{1/2} = 360(30)$ ns] [11] and allowed us to fully control our setup and procedures. Our measurement yields the value of $t_{1/2} = 360(20)$ ns, in excellent agreement.

In addition, two peaks at 168 and 511 keV are clearly visible. The procedure to assign these lines to a given nucleus is described in Fig. 2. This figure shows the mass spectrum measured by VAMOS and in coincidence with the observed delayed γ rays. The mass distribution given by the coincidence with the 168-keV line is $A = 68.19(4)$, with $\sigma = 0.32(4)$, that is, 68. This procedure applied to the Z distribution confirms the assignment to nickel. The same is true for the 511-keV line. Therefore, they both have been assigned to ^{68}Ni . The 511-keV transition most probably arises from the internal pair creation in the decay of the 0_2^+ to the ground state. No isomeric state decaying by a 168(1)-keV line was reported so far in ^{68}Ni . The half-life of the new state decaying via the 168-keV transition

was measured at $t_{1/2} = 216({}_{-50}^{+66})$ ns, as shown in the inset of Fig. 1. No other transition is observed.

It is unfortunately not possible to use γ - γ coincidences at the focal plane to build the level scheme. The nonobservation of the $2_1^+ \rightarrow 0_1^+$ transition is consistent with a decay of the isomeric state to the 2_1^+ state owing to the very low cross section to populate this new isomer (as given by the statistics in the 168-keV peak) and to the low efficiency of the four Ge detectors at the focal plane at 2 MeV: The efficiency ratio between 168 keV and 2 MeV is 16, which leads to less than three counts in the 2-MeV peak assuming a 100% decay to the 2_1^+ state. This is below our detection limit. All the other known transitions in ^{68}Ni have an energy lower than 2 MeV and should have been observed if lying in coincidence with the isomer decay. Weisskopf estimates for the half-lives of $E1$, $E2$, $M1$, and $M2$ 168-keV transitions are 0.1 ps, 300 ns, 6 ps, and 20 μs , respectively. The measured half-life indicates, therefore, an $E2$ character. The corresponding very low $B(E2 \downarrow) \leq 23.5 e^2 \text{fm}^4$ (≤ 1.4 W.u.) (assuming a pure $E2$ transition) indicates a single-particle character. Spin and parity of the new isomeric state can be inferred from the direct transfer reactions $^{70}\text{Zn}(^{14}\text{C}, ^{16}\text{O})$ performed in Refs. [1,14]. Two states assigned to ^{68}Ni have been measured at 1770(30) and 2200(30) keV. The angular distributions and distorted wave Born approximation calculations for the first excited state gave an unambiguous 0_2^+ assignment. The situation for the second excited state was much more uncertain owing to the presence of a contaminant peak at $E \sim 2.3$ MeV arising from the presence of oxygen in the target. For this reason, the two data points closest to 0° scattering angle were removed from the analysis and the authors tentatively identified this state as the 2_1^+ , not observed at that time, but later measured at 2033 keV [2]. The 2.2-MeV state might well be the one we report in this Rapid Communication. In fact, it is one of the properties of the angular distributions of the $0^+ \rightarrow 0^+$ to be strongly forward peaked. It is also clear that in Ref. [14], the angular distribution could also be fitted as a $\Delta L = 0$ transfer. Consequently, we tentatively assign this $E_\gamma = 168(1)$ -keV transition to the decay of a new (0_3^{+m}) state at 2202(1) keV to the 2_1^+ . The position of the (0_3^{+m}) level is in remarkable agreement with the 2202-keV $\pi(2p-2h)$ 0^+ intruder state, that is, $(1\pi f_{7/2})^{-2}$, deduced by Pauwels *et al.* [4]. The excitation energy at the nearly exact sum of the $\pi(2p-1h)$ and $\pi(1p-2h)$ intruder-state excitation energies in ^{69}Cu and ^{67}Co , respectively, is consistent with the fact that the wave function has a pure $\pi(2p-2h)$ character. This further supports the single-particle character deduced from our measured $E2$ transition rate. Any possible $E0$ decay would strengthen even more this conclusion.

The ^{68}Ni nucleus has been produced and studied by various means: transfer reactions, as already discussed, but also deep-inelastic reactions, fragmentation reactions, and β -decay studies. The question of the nonobservation of the 168-keV transition in these earlier works then arises. We carefully examined the previous studies known to us, and it first turns out that none of the deep-inelastic experiments [2,11,15] was suited to possibly detect it (no adequate isomer setup, high γ -ray-fold hardware trigger or high-lying transition gate in

the analysis). Fragmentation reactions dedicated to the quest of new isomers have been performed in particular at the LISE fragment separator. In these experiments, and when the flight path of the fragments was compatible with the new isomer half-life, neither the decay from the 8^+ seniority isomer nor the one from the new (0_3^{+m}) state is observed. This has been checked in data from the fragmentation of an ^{86}Kr beam [16]. It is clear from these data that the decay of ^{68}Ni proceeds essentially via the 5_1^- and the 0_2^+ isomers. This supports the conclusion of [17] showing that the feeding pattern of isomers from intermediate energy fragmentation is a complex interplay between reaction channels, beam energy and momentum distribution selected by the spectrometer.

Finally, ^{68}Ni was populated by β decay of ^{68}Co [18], which has a (7^-) ground state. From the β -decay selection rules, it is clear that the $7^- \rightarrow 0^+$ transition is highly forbidden. Another isomer in ^{68}Co has also been observed to decay in ^{68}Ni which has been attributed a (3^+) character in Refs. [18,19] with a $(\pi f_{7/2})^{-1}(\nu p_{1/2})^{-1}(\nu g_{9/2})^{+2}$ configuration. A small fraction of the flux is feeding a state which has been tentatively assigned a (0^+) character. However, this decay would be a doubly forbidden Gamow-Teller transition. In addition, the $\pi f_{7/2}^{-1} \rightarrow \nu f_{5/2}^{-1}$ transition from the (3^+) state populates preferentially neutron states in ^{68}Ni , as observed in Ref. [18]. More recently, in Ref. [20], it has been proposed to reassign from (3^+) to (1^+) the spin and parity of this isomer with, however, some inconsistency between apparent β -decay feeding of the first excited state at 45 keV and its $E1$ decay to the isomer. The nonobservation of the 168-keV line in β decay of $^{68}\text{Co}^m$ would rather confirm the neutron character of the isomer; hence, the (3^+) spin and parity. It is therefore very unlikely that the newly discovered state is fed by β decay. Another possibility that cannot be excluded, however, is a large fragmentation of the wave function of the (0_3^{+m}) in ^{68}Ni yielding to a very weak population, that is, below the sensitivity limit [20].

The shell evolution of $N = 40$ nuclei presents many similarities with the $N = 20$ region around ^{32}Mg and was presented as a new *island of inversion* [21]. The large-scale shell model (LSSM) calculations performed with the most recent interaction (LNPS) tailored for the mass region and in an extended valence space [22] show that the strong deformation in the Cr chain is the result of the proton-neutron correlations between the neutrons populating the $1g_{9/2}-2d_{5/2}$ orbitals and the valence protons in the pf shell. This interaction satisfactorily reproduces experimental data in the mass region [22]. We performed LSSM calculations using the LNPS interaction for ^{68}Ni . In the calculations, a third 0_3^+ state [22] is found at 2.4 MeV [i.e., very close to the proposed (0_3^{+m})] for which the dominant configuration is a $\pi(2p-2h)$, that is, $(1\pi f_{7/2})^{-2}$. This *intruder* state has only *normal* parity neutron states as neighbors. The 2_1^+ wave function is calculated to be a $\nu(2p-2h)$ configuration and hence with a negligible overlap with the theoretical 0_3^+ state. As a result, the calculated $B(E2 \downarrow)$ is 0.9 W.u., in good agreement with the experimental value. The correlation energies, quadrupole sum rule, and fragmentation of the calculated wave functions reflect the difference in nature of low-lying 0^+ states. The spherical ground state is characterized by -7.6 MeV of correlation energy, $899 e^2 \text{fm}^4$

quadrupole sum rule and 61% of closed-shell configuration, whereas for the proton intruder 0_3^+ we obtain -30.9 MeV of correlation energy, $4520 e^2 \text{fm}^4$ [23] of quadrupole sum rule and a wave function spread out over many spherical components. The calculated intrinsic quadrupole moment $Q_{\text{int}} \sim 195 e \text{fm}^2$ corresponds to a very large quadrupole deformation of $\beta_2 \sim 0.4$, a value which is comparable to the one measured in superdeformed bands of the zinc region [24]. In addition, the second 2^+ level is calculated just 234 keV above the 0_3^+ state. They have very similar proton and neutron configurations and the $B(E2; 2_2^+ \rightarrow 0_3^+)$ is as large as 46 W.u., which confirms the highly collective character of the band built on top of the 0_3^+ state.

Complementarily, the collective structures in $N = 40$ isotones have been recently investigated within the Hartree-Fock-Bogoliubov approach using the Gogny D1S effective interaction [25]. Coupling between collective and noncollective degrees of freedom are not included in the approach and therefore any two-proton state is beyond reach of the model. However, one can extract hints on the deformation from the calculated pairing energies. In ^{68}Ni , proton and neutron pairing energies vanish simultaneously at large β deformation ($\beta \sim 0.45$), strongly suggesting a possible local minimum in the potential energy surface, hence a shape coexistence scenario involving a highly deformed state. The (0_3^{+m}) state is obviously a natural candidate for such a minimum.

Finally, fully consistent quasiparticle random phase approximation (QRPA) calculations with D1S interaction [26] were performed for ^{68}Ni . Three excited 0^+ states at 2.14, 2.24, and 3.47 MeV, respectively are predicted. The two first states most probably correspond to a single physical state at 2.14 MeV. This is attributable to the approximation used in the calculation of 2p-2h states in QRPA. This state is absent in the corresponding particle-hole random phase approximation (ph-RPA) calculations and it has a pure proton character. Its wave function is dominated by the $(1\pi f_{7/2})^{-2}$ configuration, in excellent agreement with the proposed interpretation. The 0^+ excited state at 3.47 MeV has a pure neutron configuration and might correspond to the first experimental excited state.

Both experimental and theoretical investigations give, therefore, a more coherent picture of the low-lying structure of ^{68}Ni , where the $0_{2,4}^+$ result from the scattering of pairs into the $1\nu g_{9/2}$ and the (0_3^{+m}) is interpreted as a $(1\pi f_{7/2})^{-2}$ configuration (see Fig. 3). Many examples of such $\pi(2p-2h)$ 0^+ states are known in semimagic nuclei [27]. They occur at lowest excitation energy when the number of active valence nucleons is at midshell (e.g., around ^{116}Sn or ^{186}Pb) because neutron-proton correlations are maximal at that point. What makes ^{68}Ni unique is that, despite the doubly magic character of the dominant component of its ground state, it has a low-lying $\pi(2p-2h)$ 0^+ state as a result of an unusually large neutron-proton correlation energy. The configuration of the newly observed (0_3^{+m}) state with active protons in the pf

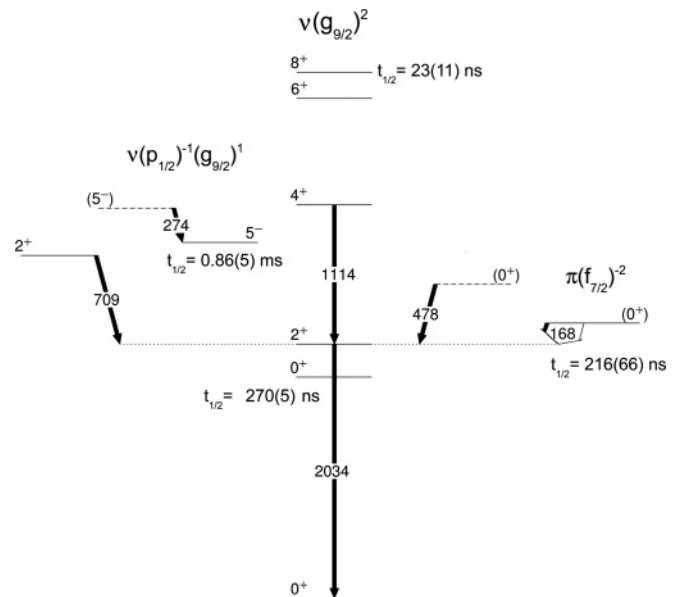


FIG. 3. Proposed level scheme deduced from the present work in ^{68}Ni . The open (solid) arrows indicate, respectively, the observed (prompt) γ ray. The configurations which are discussed in the text and isomer half-lives are indicated.

shell is very similar to the Fe case, that is, with two holes in the $1\pi f_{7/2}$ orbital. Therefore, one can reasonably consider that the configuration corresponding to the (0_3^{+m}) state at 2202(1) keV in ^{68}Ni migrates down to 491 keV excitation energy in ^{67}Co to become the ground state in ^{66}Fe . The *normal* configuration, as opposed to the *intruder* one, might then appear as a low-lying 0^+ state in ^{66}Fe .

In summary, a new isomeric state has been observed at 168(1) keV above the first 2^+ state in ^{68}Ni . It is interpreted as the intruder $\pi(2p-2h)$ 0^+ state across the major $Z = 28$ shell gap. This interpretation is supported by LSSM calculations, which also indicate a highly deformed state with $\beta_2 \sim 0.4$. Our observation fits extremely well with the prescription used in Ref. [4] to predict the energy of the $\pi(2p-2h)$ and also with the most recent theoretical calculations, indicating a pure proton excitation character for this state. The systematics of intruder states along the $N = 40$ isotonic chain with $Z \leq 28$ strongly suggests that shape coexistence is occurring at low energy in ^{68}Ni .

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