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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 like atoms, metal clusters, or 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 have to bring answers. Already today, the structure of moderately exotic nuclei such as $^{68}$Ni allows one to pave the way towards 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 bring 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 $0^+$ 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 of $^{68}$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 reef 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 $\nu 1g_{9/2}$. 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 \( ^{60}\text{Cu} \) and, symmetrically, the \( \pi(1p-2h) \) in \( ^{65}\text{Co} \), 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 protons and the active valence neutrons across \( N=40 \) [3, 4]. The spin-parity of the \( \pi(1p-2h) \) state in \( ^{67}\text{Co} \) 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 \( ^{68}\text{Ni} \). Later on, evidences for such deformed \( (\pi 3/2) \) intruder orbital were reported in the spectroscopy of odd mass Mn isotopes [5] in \( \beta \)-decay experiments. These results strongly suggest the presence of a deformed \( \pi(2p-2h) \) intruder state in \( ^{68}\text{Ni} \). But despite the large number of experiments dedicated to \( ^{68}\text{Ni} \), 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 \( ^{68}\text{Ni} \) which we propose to have a \( 0^+ \) character.

The experiment was performed at the Grand Accélérateur National d’Ions Lourds (GANIL) by using multi-nucleon transfer reactions in inverse kinematics. A \( ^{238}\text{U} \) beam at 6.33 A.MeV bombarded a 1.3 mg/cm\(^2\) thick \( ^{70}\text{Zn} \) target. The target-like 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 Multi Wire Proportional Chamber located 138 mm downstream the target. The flight path through the spectrometer is determined using Secondary Electron Detectors [9] and following the procedure described in [6]. We have obtained a mass resolution \( \Delta A/A = 1.2\% \) and an atomic number resolution \( \Delta Z/Z=1.1\% \). Prompt \( \gamma \)-rays emitted at the target position were measured by eleven clover detectors from the EXOGAM array [10] in coincidence with the recoils identified in VAMOS. Delayed \( \gamma \)-rays were also detected at the VAMOS focal plane by four HPGe detectors facing the silicon detectors where the recoils were implanted. With a recoil velocity from 22 to 45 \( \mu \text{m/ps} \), 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 \( \mu \text{s} \). Our delayed spectroscopy setup is therefore sensitive to lifetimes from \( \sim 100 \) ns up to \( \sim 10 \mu \text{s} \).

In this Rapid Communication, we focus on new results obtained on the \( ^{68}\text{Ni} \) key nucleus. Fig. 1 shows the delayed \( \gamma \)-ray spectrum observed in coincidence with its identification in VAMOS. Several isomers are known in \( ^{68}\text{Ni} \): 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 \( 0^+ \) state with \( t_{1/2} = 270(5) \) ns [13]. Given the mass and charge resolution we measured, the spectrum obtained after selection of \( ^{68}\text{Ni} \) is slightly contaminated by neighboring nuclei, \( ^{67}\text{Cu} \) and \( ^{69}\text{Ni} \), in which isomers are known. All the transitions from their decay have been identified (see Fig. 1). In particular, the transitions observed in \( ^{69}\text{Cu} \) correspond to the decay of a \( 13^2/2^+ \text{m} \) 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 keV 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 \( ^{68}\text{Ni} \) and in coincidence with the observed delayed \( \gamma \)-rays. The mass distribution given by the coincidence with the 168 keV line is \( A = 68.19(4) \) with \( \sigma = 0.32(4) \) i.e. 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 \( ^{68}\text{Ni} \). The 511 keV transition most probably arises from the internal pair creation in the decay of the \( 0^+ \) to the ground state. No isomeric state decaying by a 168(1) keV line was reported so far in \( ^{68}\text{Ni} \). The half-life of the new state decaying via the 168 keV transition was measured at \( t_{1/2} = 216(156) \) ns as shown in the inset of Fig. 1. No other transition is observed.

It is unfortunately not possible to use \( \gamma-\gamma \) coincidences...
at the focal plane to build the level scheme. The non-
observation of the $2^+ \rightarrow 0^+$ transition is consistent with a
decay of the isomeric state to the $2^+$ state due 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 3 counts in the 2
MeV peak assuming a 100% decay to the $2^+$ state. This
is below our detection limit. All the other known tran-
sitions in $^{68}$Ni have an energy lower than 2 MeV and
should have been observed if lying in coincidence with
the isomer decay. Weiskopf estimates for the half-life of
an E1, E2, M1 and M2 168 keV transition 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) ≤ 23.5 e²fm⁴ ( ≤ 1.4 W.u.) (assuming a
pure E2 transition), indicates a single particle charac-
ter. Spin and parity of the new isomeric state can be
inferred from the direct transfer reactions $^{59}$Zn($^{14}$C,$^{16}$O)
performed in [1, 14]. Two states assigned to $^{68}$Ni have
been measured at 1770(30) keV and 2200(30) keV. The
angular distributions and Distorted Wave Born Approx-
imation calculations for the first excited state gave an
unambiguous $0^+_2$ assignment. The situation for the sec-
ond excited state was much more uncertain due to the
presence of a contaminant peak at E ~ 2.3 MeV arising
from the presence of oxygen in the target. For this rea-
son, the two data points closest to $0^+$ scattering angle
were removed from the analysis and the authors tenta-
vitely 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 letter. 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 [14], the angular distribution could also be
fitted as a $\Delta L=0$ transfer. Consequently, we tentatively
assign this $E_3=168(1)$ 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, i.e. $(1f_{7/2})^{-2}$, de-
duced 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 energy 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 par-
icle 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 using deep-inelastic reactions, fragmentation re-
actions and $\beta$-decay studies. The question of the non ob-
servation 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
depth-inelastic experiments [2, 11, 15] was suited to pos-
sibly detect it (no adequate isomer setup, high gamma-
ray-fold hardware trigger or high-lying transition gate in
the analysis). Fragmentation reactions dedicated to the
uest 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 compat-
ible 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 $0^+_{51}$ 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 $\beta$-decay of $^{68}$Co [18],
which has a ($7^-$) ground state. From the $\beta$-decay selec-
tion rules, it is clear that the $7^- \rightarrow 0^+$ transition is highly
forbidden. Another isomer in $^{68}$Co has also been ob-
served to decay in $^{68}$Ni which has been attributed a ($3^+$)
character in [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 $0^+$ charac-
ter. However, this decay would be a doubly-forbidden
Gamow-Teller transition. In addition, the $\pi f_{7/2} \rightarrow \nu f_{5/2}^{-1}$
transition from the $3^+$ state populates preferentially
neutron states in $^{68}$Ni as observed in [18]. More recently,
in [20], it has been proposed to reassign from $(3^+) \rightarrow (1^+)$
the spin and parity of this isomer with however some in-
consistency between apparent $\beta$-decay feeding of the first
excited state at 45 keV and its E1 decay to the isomer.
Another possibility that cannot be excluded however is a $\beta$-isomer hence the (3$^+$) configuration (see Fig. 3). Many examples of such $\pi(2p-2h)$ $0^+$ states are known in semi-magic nuclei [27]. They occur at lowest excitation energy when the number of active valence nucleons is at mid shell (e.g., $^{116}$Sn or $^{180}$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$^+_{3^-}$) state with active protons in the pf shell is very similar to the Fe case, i.e. with two holes in the $\nu(2p-2h)$ configuration. All states shown in this figure are therefore predominantly pf shell wave functions. However, a more coherent picture of the low-lying structure of $^{68}$Ni where the $0^+_{3^-}$ result from the scattering of pairs into the $1\nu g_{9/2}$ and the (0$^+_{3^-}$) 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 semi-magic nuclei [27]. They occur at lowest excitation energy when the number of active valence nucleons is at mid shell (e.g., around $^{116}$Sn or $^{180}$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$^+_{3^-}$) state with active protons in the pf shell is very similar to the Fe case, i.e. with two holes in the $\nu f_{7/2}$ orbital. Therefore one can reasonably con-

The non observation of the 168 keV line in $\beta$-decay of $^{68}$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 $\beta$-decay. Another possibility that cannot be excluded however is a large fragmentation of the wave function of the (0$^+_{3^-}$) in $^{68}$Ni yielding to a very weak population, i.e. 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 $1\nu g_{9/2}$-$2\nu d_{5/2}$ orbitals and the valence protons in the pf shell. This interaction satisfactory 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$^+_2$ state [22] is found at 2.4 MeV (i.e. very close to the proposed (0$^+_{3^-}$)) for which the dominant configuration is a $\pi(2p-2h)$ i.e. $(1\pi f_{7/2})^{-2}$. 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$^+_2$ state. As a result, the calculated B(E2$_{1}$) 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 -7.6MeV of correlation energy, 899 $\varepsilon^2 fm^4$ quadrupole sum rule and 61% of closed shell configuration, whereas for the proton intruder 0$^+_2$ we obtain -30.9MeV of correlation energy, 4520 $\varepsilon^2 fm^4$ [23] of quadrupole sum rule and a wave function spread out over many spherical components. The calculated intrinsic quadrupole moment $Q_{int} \sim 195 \ vfm^2$ 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$^+_2$ state. They have very similar proton and neutron configurations and the $B(E2; 2^+_1 \rightarrow 0^+_2)$ is as large as 46 W.u., which confirms the highly collective character of the band built on top of the 0$^+_2$ state.

Complementary, 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 non-collective 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 $\beta$ deformation of $\beta \sim 0.45$ strongly suggesting a possible local minimum in the potential energy surface, hence a shape coexistence scenario involving a highly deformed state. The (0$^+_{3^-}$) state is obviously a natural candidate for such a minimum.

Finally, fully consistent QRPA calculations with DIS interaction [26] were performed for $^{68}$Ni. Three excited 0$^+$ states at 2.14 MeV, 2.24 MeV 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 due to the approximation used in the calculation of 2p-2h states in QRPA. This state is absent in the corresponding ph-RPA calculations and it has a pure proton character. Its wave function is dominated by the $\pi(2p-2h)$ 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$^+_2$ result from the scattering of pairs into the $1\nu g_{9/2}$ and the (0$^+_{3^-}$) 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 semi-magic nuclei [27]. They occur at lowest excitation energy when the number of active valence nucleons is at mid shell (e.g., around $^{116}$Sn or $^{180}$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$^+_{3^-}$) state with active protons in the pf shell is very similar to the Fe case, i.e. with two holes in the $\nu f_{7/2}$ orbital. Therefore one can reasonably con-

FIG. 3. Proposed level scheme deduced from the present work in $^{68}$Ni. The open (filled) arrows indicate respectively the observed delayed (prompt) $\gamma$-ray. The configurations which are discussed in the text and isomers half-life are indicated.
sider that the configuration corresponding to the \((0^+_m)\) state at 2202(1) keV in \(^{68}\text{Ni}\) migrates down to 491 keV excitation energy in \(^{67}\text{Co}\) to become the ground state in \(^{66}\text{Fe}\). The normal configuration, as opposed to the intruder one, might then appear as a low-lying \(0^+\) state in \(^{66}\text{Fe}\).

In summary, a new isomeric state has been observed at 168(1) keV above the first \(2^+\) state in \(^{68}\text{Ni}\). It is interpreted as the intruder \(\pi(2p\!-\!2h)\) \(0^+\) state across the major \(Z\!\!\!\!\!\!/28\) shell gap. This interpretation is supported by large scale shell model calculations which also indicate a highly deformed state with \(\beta_2 \sim 0.4\). Our observation fits extremely well with the prescription used in [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 \(Z\!\!\!\!\!\!\!\!/28\) strongly suggests that shape coexistence is occurring at low energy in \(^{68}\text{Ni}\).

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[23] Carrying the calculations for the present letter, it was found that the sum rule indicated in [22] is not correct and should be replaced by the present value of 4520 \(e^2fm^4\) for this \(0^+_m\) state.