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Search for shape-coexisting 0^+ states in ⁶⁶Ni from lifetime measurements

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The lifetime of the 0_3^+ state in ⁶⁶Ni, two neutrons below the N=40 sub-shell gap, has been measured. The transition $B(E2;0_3^+ \rightarrow 2_1^+)$ is one of the most hindered E2 transitions in the Ni isotopic chain and it implies that, unlike ⁶⁸Ni, there is a spherical structure at low excitation energy. We have performed extensive shell-model calculations that correctly predict this result, obtaining a spherical 0⁺ state at the correct energy and with an extremely low $B(E2;0_3^+ \rightarrow 2_1^+)$ value.

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Shape coexistence in atomic nuclei occurs when lowlying states with similar energy show different intrinsic deformation. This leads to competing minima close in excitation energy. In even-even nuclei this phenomenon is revealed by low-lying 0^+ states, the classic example being ¹⁸⁶Pb, where a triplet of 0^+ states was found, associated with spherical, oblate and prolate deformation respectively [1]. The coexistence of nuclear shapes is closely linked to the shell gaps and how particle-hole excitations take place around them, eventually leading to deformation. Examples are abundant in regions of the nuclear chart in the proximity of shell closures and subshell closures [2].

The area around ⁶⁸Ni is specially interesting, since the N=40 subshell closure and the Z=28 spherical magic number cohabit. The N=40 subshell corresponds to a harmonic oscillator potential closure, and separates the pf negative parity shell from the spin-orbit positive parity g_{9/2} orbital. Therefore the presence of intruder states leading to isomers and shape coexistence may be expected. In the case of ⁶⁸Ni itself, 0⁺₂ is actually the first excited state, located at 1603.6 keV above the 0⁺ ground



FIG. 1. (Color online) Energy of the 0^+ states below 3 MeV in the even Ni isotopes. The 2^+_1 states have been included for reference.

state [3]. This 0_2^+ state has a known long half-life of 270(5) ns [4]. A third 0^+ state is located at 2511 keV, above the 2033.0-keV 2_1^+ state. See Fig. 1 for a systematics of all the 0^+ states below 3 MeV in the Ni isotopic chain.

Large-scale Shell Model calculations using the Lenzi-

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Nowacki-Poves-Sieja (LNPS) interaction [3, 5–7] accurately describe the ⁶⁸Ni experimental results, presenting a clear case of triple shape coexistence. The ground state is mainly comprised (60%) of a spherical configuration, corresponding to doubly magic closures at N=40 and Z=28. The calculations predicted an oblate-deformed 0_2^+ as the first excited state dominated by 2p2h neutron excitations to the intruder $g_{9/2}$ orbital, with the 2_1^+ state representing the first state of a band on top of it. The calculations also showed a prolate band built on the 0_3^+ state and characterized by 2p2h proton excitations across Z=28 and 4p4h/6p6h neutrons across the N=40 sub-shell. The highly deformed band continues with the 2_2^+ state and B(E2; $2_2^+ \rightarrow 0_3^+$)=46 W.u. or $\beta \sim 0.45$.

Tsunoda and collaborators [8] performed Monte Carlo shell model (MCSM) calculations for $^{68-78}$ Ni, producing predictions in good agreement with the LNPS results. From the potential energy surfaces and the basis vectors in the MCSM calculations they conclude that the 68 Ni ground state is spherical, the 0_2^+ state is oblate with a moderate deformation of the order of $\beta_2 \sim 0.2$, while the 0_3^+ state is of prolate character, with strong deformation of $\beta_2 \sim 0.4$ corresponding to an intrinsic quadrupole moment Q_0 of 200 fm². In the same manner the 2_1^+ level at 2033 keV is identified as member of the oblate band built on the 1604 keV 0_2^+ state, while the 2_2^+ state at 2743 keV, above the 0_3^+ level at 2511 keV, is consistent with a prolate nature. Nevertheless the most recent experimental results [9] seem in better agreement with the LNPS calculations.

Thanks to the great effort put into studying this particular nucleus over the recent decades shape coexistence in ⁶⁸Ni is well established both from an experimental [3, 9– 12] and a theoretical point of view [3, 5, 8]. But the situation is not that clear for nuclei around it. Shape coexistence has been proposed for the more exotic ⁷⁰Ni (N=42) at even lower energy [9, 13, 14], and also for ⁶⁷Co and ⁷¹Cu (see Refs. [15, 16] and references therein), but surprisingly not for the nuclei closer to stability.

In this work we address shape coexistence in the next even neighbour to 68 Ni, the isotope 66 Ni with N=38. The excited structure of this nucleus is known from β -decay [10] and deep-inelastic [17] experiments. The 2^+_1 state is located at 1425 keV, with the second 0^+_2 state at 2443 keV and the 0^+_3 level immediately above it at 2671 keV (see Fig. 2 for the measured 66 Ni level scheme and calculations from this work). Recently Walters *et al.* [16] reported a transition connecting a new 2^+ state to the 0^+_3 bandhead, and proposed the levels at 2671 and 3312 keV as prolate intruder states.

The β decay of ⁶⁶Fe to ⁶⁶Co was studied by D. Pauwels et al. [18] and identified the ⁶⁶Co ground state as a proton intruder, with a newly assigned parity of (1⁺) based on the strong β feeding from the ⁶⁶Fe 0⁺ ground state. The decay of ⁶⁶Co only populates the ground and 0⁺₃ states and the first two 2⁺ states in ⁶⁶Ni. This result was later confirmed by Liddick et al. [19]. A β -decay scheme of ⁶⁶Co to ⁶⁶Ni is shown in Fig. 2.



FIG. 2. (Color online) (Left) ⁶⁶Ni level scheme populated in the β -decay of the (1⁺) ⁶⁶Co ground state. The dashed levels were not observed in this β -decay experiment, but have been included to help the discussion. The labels on the transitions are the energy in keV and the B(E2) in W.u., while the width is proportional to their absolute intensity. (*Right*) Results from the LNPS calculations from this work.

We have investigated experimentally the β^- decay of ⁶⁶Co to ⁶⁶Ni with the aim of measuring the half-life of the excited states populated in ⁶⁶Ni by means of the the Advanced Time-Delayed $\beta\gamma\gamma(t)$ method described in [20–22]. This experiment was part of a wider campaign to study the evolution of collectivity below ⁶⁸Ni [23–25].

The experiment was carried out at the ISOLDE facility at CERN [26]. ⁶⁶Ni isotopes were populated in the β decay chain of A=66 isobars, starting from ⁶⁶Mn. 1.4-GeV protons from the CERN Proton Synchrotron Booster incident on a UC_x target induced high energy fission. Produced radionuclides were thermally released from the target and manganese atoms were ionized by the ISOLDE Resonance Ionization Laser Ion Source. Mass A=66 ions were mass-separated and implanted on a thin aluminium foil in the center of the experimental setup. A fast plastic scintillator acted as β particle detector, and was placed just after the deposition point. Two truncated-cone shaped $LaBr_3(Ce)$ crystals coupled to Photonis XP20D0 photomultipliers were used for γ ray fast timing. The setup was completed by two HPGe detectors. Analog time-delayed $\beta \gamma(t)$ coincidences between the β and each one of the γ detectors were set up using time-to-amplitude conversion modules. The fast timing analysis is based on $\beta \gamma(t)$ distributions between the β and LaBr₃(Ce) detectors and $\beta \gamma \gamma(t)$ distributions including the former and an extra condition on HPGe energies. Further details on the experimental station and data acquisition strategy can be found in Ref. [23, 24].

There was no removal of the decay products and thus



FIG. 3. Singles HPGe energy spectrum. A time gate of 300-1200 ms after the proton impact on the target was selected to enhance the 66 Co decay activity. The long-lived 66 Ga contaminant was subtracted. This did not suppress the 66 Fe decay activity, so the transitions in 66 Co are present and labeled in the spectrum.

a saturated source including the whole A=66 chain was created. Lines from the 66 Co $\rightarrow {}^{66}$ Ni decay were relatively enhanced by selecting the data between 300 ms and 1200 ms after proton impact, when most of the 66 Mn has already decayed away. A singles HPGe energy spectrum with this time condition is shown in Fig. 3. The identification of γ rays in ⁶⁶Ni is based on the existing information [10]. Despite having ~ 50 times more statistics than the previous β -decay experiment [10] only one new transition of 2231.9 keV has been observed. This transition is in clear coincidence with the 1425-keV γ -ray, but it is not observed in the singles spectrum, so its placement in the level scheme is uncertain. On a side note, we can confirm the unambiguous assignment of the 471-keV transition to the decay of 66 Fe to 66 Co, as was already proposed in [19], and in contrast to the suggestion made in [27]. We also found no trace of the 1020(1)-keV transition proposed in [27] nor the 1018 and 1478-keV lines suggested in [17]. These lines should be in coincidence with the 1425-keV transition to the ground state and, in spite of our sufficient amount of statistics, they were not found, see Fig. 4.

The ⁶⁶Co β^- decay mainly populates the ground state and the 0^+_3 level at 2671 keV with $I_{\beta} = 29(3)\%$ [18]. The 0^+_2 level at 2443 keV and the 0^+_4 at 2965(10) keV are not populated in the decay of the ⁶⁶Co (1⁺) ground state, pointing towards a substantially different nuclear structure. The 0^+_3 state de-excites to the 2^+_1 level at 1425 keV via a γ -ray of 1246 keV [10, 27], and the 2^+_1 feeds the ground state with a 1425-keV transition.

Figure 5 shows the β -LaBr₃(Ce) time difference with an energy gate in the 1246-keV $0_3^+ \rightarrow 2_1^+$ transition. This slope is identical to the one obtained with a condition



FIG. 4. Coincidence HPGe-HPGe energy spectrum with a gate on the 1425-keV transition. * A new transition of 2231.9 keV has been observed for the first time, see text for details.



FIG. 5. (Color online) Fit to the 2671-keV level half-life in 66 Ni using the time difference between a β and the 1246-keV decay transition detected in one of the LaBr₃(Ce) crystals. The fit was done to a prompt Gaussian including the Compton background under the peak and a decay exponential. The result of $T_{1/2} = 170(7)$ ps stems from the weighted average of four measurements. See text for details.

on the 1425-keV $2_1^+ \rightarrow 0_1^+$ peak and is not present if the gate is selected in the Compton background above each of the full-energy peaks. The half-life is therefore assigned to the 0_3^+ 2671-keV level unambiguously. The observed slope in the 1425-keV gate is consistent with feeding via the 1246-keV γ ray, since the direct β feeding to the 2_1^+ 1425-keV state is about 6 times lower than the β feeding to the 2671-keV state. In this manner the half-life is measured independently using two different γ transitions selected in the LaBr₃(Ce) detectors and using two different crystals. The four lifetime measurements are consistent and yield a weighted average value of $T_{1/2} = 170(7)$ ps. The calculated B(E2; $0_3^+ \rightarrow 2_1^+$) value is 1.11(5) e²fm⁴ or 0.070(3) W.u.



FIG. 6. Coincidence HPGe-HPGe energy spectrum with a gate on the 1246-keV transition, showing the 1425 keV line.

Since no transitions are observed feeding the 0_3^+ state from above (Fig. 6 shows only the $2_1^+ \rightarrow 0_1^+$ transition in coincidence), we can discard any significant contribution to the half-life from higher energy levels.

This result was further cross-checked in $\beta\gamma\gamma(t)$ coincidences. Either by selecting the 1246-keV transition in the LaBr₃(Ce) and the 1425-keV in the HPGe detector or vice versa, a slope compatible with a half-life of a few hundreds of picoseconds was observed in the delayed part. However, in this restrictive coincidence conditions the statistics were much lower and the resulting half-life, even if compatible with the $\beta\gamma$ result, had a much larger error bar.

The measured value can be compared to a similar state in ⁶⁸Ni, where a recent study by Crider *et al.* [9] reports a $T_{1/2} = 570(50)$ ps for the 2511-keV state via the 478-keV de-exciting transition. In terms of transition probabilities Crider et al. [9] obtained a B(E2; $0_3^+ \rightarrow 2_1^+)$ of 39.0(34) $e^2 f m^4$ using a 98.1% lower limit for the branching ratio. With this result they proposed the 0_3^+ state to be of prolate nature. From the lifetime in ⁶⁶Ni measured in this work, and neglecting any E0 decay branches to the ground and 0_2^+ states, the B(E2) obtained is 1.11(5) $e^2 fm^4$, around 35 times slower. Any E0 branch in ⁶⁶Ni would make the B(E2; $0_3^+ \rightarrow 2_1^+)$ value even lower.

This result clearly implies that the 0_3^+ states in ⁶⁶Ni and ⁶⁸Ni are not of the same nature, eliminating the possibility that the 0_3^+ is the prolate state in ⁶⁶Ni contrary to the proposal in [16]. One final argument that can be made, is the different population of the 0^+ states by the ⁶⁶Co β -decay. It strongly populates the 0_1^+ and 0_3^+ , while no population has been observed for the 0_2^+ and 0_4^+ . If the ground state of ⁶⁶Co is a deformed proton intruder state from the pf orbitals as argued in [18, 19], the selection rule requires that β decay populates states in ⁶⁶Ni by allowed transitions, therefore neutrons in the pf orbitals, as opposed to population of neutrons in the $g_{9/2}$ orbital, which would require a first forbidden transition. In this region states based on neutrons in the pf orbital are of spherical nature, contrary to the $g_{9/2}$ orbital, which carries deformation.

To gain insight in the nuclear structure of ⁶⁶Ni, we performed extensive shell-model calculations using the LNPS interaction [5]. The configuration space was extended up to 14*p*-14*h* excitations across the Z=28 and N=40 shell gaps to achieve energy convergence. The corresponding basis dimension amounts to 2×10^9 Slater Determinants basis. Effective charges of 1.31 for the protons and 0.46 for the neutrons were adopted from the microscopic calculation of Ref. [28]. Our results, summarized in Tab. I, clearly show that the 0_3^+ state is dominated by the spherical two neutron-hole configuration in the *pf* orbitals. This 0_3^+ state has the lowest deformation of the 0^+ levels in ⁶⁶Ni, and can be compared to the spherical ground state in ⁶⁸Ni.

The shell-model calculations showed in Tab. I also hint that ${}^{66}\text{Ni}$ is not a clear-cut case of shape coexistence. The 0_2^+ presents a weakly deformed oblate structure, based on a two-neutron excitation from the pf to the $g_{9/2}$ orbital, and can be considered to have a similar structure than the 0_2^+ level in ${}^{68}\text{Ni}$. The 0_4^+ state shows a high prolate deformation of $\beta = 0.39$. Only in this state the $d_{5/2}$ occupation is not negligible. This state can be matched to the 0_3^+ state in ${}^{68}\text{Ni}$. Finally, the ${}^{66}\text{Ni}$ ground state would be an admixture of the 0_2^+ and 0_3^+ states, with a slight oblate deformation.

These results are consistent with the ${}^{64}\text{Ni}(t,p){}^{66}\text{Ni}$ experiment described in [29], where an enhanced relative yield of the 0^+_1 and 0^+_3 states in the reaction are reported, while the 0^+_4 population is suppressed, pointing to a possible deformed configuration for the later level, compared to a spherical one of the other two.

As a stringent test of the shell-model calculations, the calculated B(E2) values were compared to the experimental results, see Fig. 2. It is worth noting the excellent agreement between the calculated energies and the experimental ones, as well as the B(E2; $2_1^+ \rightarrow 0_1^+$) and B(E2; $0_3^+ \rightarrow 2_1^+$) values.

Our result can be compared with neighboring isotopes. Table II shows all the experimental values of the $B(E2;0_i^+ \rightarrow 2_1^+)$ transitions measured in the Ni isotopic chain. The result presented in this paper is more than one order of magnitude smaller than that of similar transitions in the region. This slow transition clearly points out that ⁶⁶Ni is comprised of different nuclear structures than the rest of the isotopic chain. If we compare the $B(E2;0_2^+ \rightarrow 2_1^+)$ (the E2 transition from the prolate 0⁺ state to the first 2⁺) in ⁷⁰Ni (2 neutrons above ⁶⁸Ni), we can see that it is, at least, 50 times faster than the $B(E2;0_3^+ \rightarrow 2_1^+)$ in ⁶⁶Ni (2 neutrons below ⁶⁸Ni). We can safely discard this 0₃⁺ in ⁶⁶Ni as the same prolate state as in ⁷⁰Ni.

We need to go to 58 Ni to find a transition with such retardation as the one observed here. The 0^+_2 presents one

Level	Exp. $E(MeV)$	Calc. $E(MeV)$	$\pi f_{7/2}$	$\nu g_{9/2}$	$\nu d_{5/2}$	β_s	γ_s	$Q_s \ (efm^2)$
0_{1}^{+}	0	0	6.6	1.0	0.1	0.20	39°	12.4
0^{+}_{2}	2.443	2.15	6.0	2.0	0.2	0.25	36°	11.2
0^{+}_{3}	2.671	2.87	6.7	0.5	0.1	0.17	36°	6.4
0_{4}^{+}	2.965	3.22	5.1	3.1	0.7	0.39	12°	-44.7

TABLE I. Summary of the key results of the LNPS calculations performed for ⁶⁶Ni. Columns 4 to 6 give the particle occupation of the orbitals. β_s and γ_s are the deformation and the deviation from axial symmetry as defined by Kumar [30]. Q_s is the intrinsic quadrupole moment of the sum-rule 2⁺ state built upon each 0⁺ state.

Isotope	Transition	B(E2) (W.u.)	Reference
⁵⁸ Ni	$0_2^+ \to 2_1^+$	$4.0(6) \times 10^{-4}$	[31]
	$0_3^+ \to 2_1^+$	5.6(18)	[32]
⁶⁰ Ni	$0_2^+ \to 2_1^+$	<71	[33]
	$0_3^+ \to 2_1^+$	9(3)	[33]
⁶² Ni	$0_2^+ \to 2_1^+$	42(22)	[34]
	$0^+_3 \rightarrow 2^+_1$	< 0.84	[35]
⁶⁴ Ni	$0_2^+ \to 2_1^+$	110(60)	[33]
⁶⁶ Ni	$0_3^+ \to 2_1^+$	0.070(3)	This work
⁶⁸ Ni	$0_3^+ \to 2_1^+$	2.4(2)	[9]
⁷⁰ Ni	$0_2^+ \to 2_1^+$	>3.4	[9]

TABLE II. Summary of the known $\mathcal{B}(\mathcal{E}2;0^+_i\to 2^+_1)$ in the Ni isotopic chain.

of the smallest $\rho^2(\text{E0})$ observed [36]. But this level also has the lowest B(E2) of the Ni isotopes, with B(E2;0_2^+ \rightarrow $2_1^+)=4.0(6) \times 10^{-4}$ W.u. (moreover, it is one of the most hindered transitions ever observed). There is some parallelism between the two isotopes, as ⁵⁸Ni can be explained as a core with two extra neutrons in the $p_{3/2}f_{5/2}p_{1/2}$ orbitals while ⁶⁶Ni would be also a core with two neutron holes in the same orbitals.

Möller *et al.* [37] calculated the existence of nuclear shape isomers for a vast range of nuclei and predicted a spherical ground state for ⁶⁶Ni with a prolate 0^+ at 2.67 MeV. Despite the excellent agreement with the experimental energy and the long lifetime of the 0_3^+ state, our results clearly indicate that this state is spherical. Thus the data and calculations presented in this work clearly discard the third 0^+ as the predicted shape isomer, leaving the second and fourth 0^+ states as candidates.

In conclusion, we present direct evidence through the measurement of the 0^+_3 state lifetime and by large scale

shell model calculations, of the presence of a spherical structure at low energies in ⁶⁶Ni. Moreover, these calculations also suggest that, unlike the neighboring ⁶⁸Ni, ⁶⁶Ni is not a clear case of shape coexistence, with a significant mixing of the different bands at low energy. The measured $B(E2;0_3^+ \rightarrow 2_1^+)$ is exceptionally low and is the second lowest B(E2) transition rate in the Ni isotopic chain.

Note added: During the review process, we haven been made aware of a very recent publication [38] reporting an independent measurement of the same quantity by a complementary method, and including lifetime measurements of the other 0^+ states. Their measured lifetime for the 0_3^+ is more than 2σ away from our value. While statistically speaking these two values are not in agreement, the underlying physics interpretation of the state and the nucleus is the same in both cases. The Monte Carlo Shell Model calculations in [38] are in remarkable accordance with the calculations presented in this work.

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