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Monopole-Driven Shell Evolution below the Doubly Magic Nucleus ¹³²Sn Explored with the Long-Lived Isomer in ¹²⁶Pd

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A new isomer with a half-life of 23.0(8) ms has been identified at 2406 keV in ¹²⁶Pd and is proposed to have a spin and parity of 10⁺ with a maximally aligned configuration comprising two neutron holes in the $1h_{11/2}$ orbit. In addition to an internal-decay branch through a hindered electric octupole transition, β decay from the long-lived isomer was observed to populate excited states at high spins in ¹²⁶Ag. The smaller energy difference between the 10⁺ and 7⁻ isomers in ¹²⁶Pd than in the heavier N = 80 isotones can be interpreted as being ascribed to the monopole shift of the $1h_{11/2}$ neutron orbit. The effects of the monopole interaction on the evolution of single-neutron energies below ¹³²Sn are discussed in terms of the central and tensor forces.

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The nucleus, i.e., the highly dense central core of an atom, is an aggregate of two types of subatomic particles, protons and neutrons (nucleons). The stability of atomic nuclei, which is inherently linked to the presence of materials in nature, is much influenced by a shell structure and its resulting magic numbers; nuclei with specific numbers of nucleons (2, 8, 20, 28, 50, 82 for protons and, also, 126 for neutrons) near the β -stability line necessitate relatively high energies to remove one or two nucleons compared to the neighboring isotopes. Such a situation is analogous to the noble gases that have large ionization energies with closed-shell configurations of electrons.

The traditional nuclear magic numbers, which have been the microscopic basis for understanding nuclear structure for half a century since Mayer and Jensen [1,2], are nowadays recognized as being one aspect of a more generalized concept of the shell structure that can vary as functions of the proton number (Z) or neutron number (N) [3]. One of the underlying causes for shell evolution is based on the strong monopole part of effective nucleonnucleon interactions including the central and tensor forces, the latter of which plays an important role in the appearance or disappearance of the spherical magic numbers in lightmass nuclei with large N/Z ratios [4]. For heavier nuclei, similar mechanisms of the monopole interaction provide a reasonable explanation for the observed drift of singleparticle levels along the N = 51 isotonic and the Sb (Z = 51) isotopic chains [5,6].

Concerning the N = 82 magicity, the recent spectroscopic study of ¹²⁸Pd (Z = 46, N = 82) revealed that this shell closure is so robust as to give rise to the seniority isomerism in the semimagic nucleus [7]. Meanwhile, a reduction of the N = 82 shell gap toward Z = 40 was suggested by mean-field and shell-model (SM) calculations [8,9]. To tackle this issue from a microscopic viewpoint, it is crucial to elucidate the properties of single-neutron states below the doubly magic nucleus ¹³²Sn (Z = 50, N = 82). The ordering of single-particle orbitals and their energies in the vicinity of ¹³²Sn are reviewed in Ref. [10]. Of particular interest is the behavior of the $1h_{11/2}$ neutron orbit that locates just below the N = 82 shell closure, hereby being significant for the reduction of the N = 82 shell gap as protons are removed from the $1g_{9/2}$ orbit in the Z = 40-50subshell. In the present Letter, we focus on the characteristic isomers involving neutron-hole configurations with the $1h_{11/2}$ orbit in ¹²⁶Pd (N = 80). The systematic investigation of the analogous isomers in the N = 80 isotones uniquely embosses the evolution of this key orbit. This Letter presents, for the first time, an experimental implication for the monopole effects from the central and tensor forces on the shift of single-neutron energies below ¹³²Sn.

The knowledge of single-neutron states in this region also has a significant impact on the understanding of nucleosynthesis in the rapid neutron-capture (r) process [11], as the reaction rate of direct neutron capture after $(n, \gamma) \rightleftharpoons (\gamma, n)$ equilibrium, during the so-called freeze-out era, is expected to depend on the nature of single-neutron orbits around N = 82 [12]. Furthermore, the presence of isomeric states as long lived as the respective ground states in this neutron-rich region may affect the Solar System abundance distribution around the prominent $A \approx 130$ peak of the *r*-process isotopes.

Spectroscopic studies of ¹²⁶Pd have been performed at the RI Beam Factory (RIBF) facility [13], cooperated by RIKEN Nishina Center and CNS, University of Tokyo. Neutron-rich nuclei below ¹³²Sn were produced using inflight fission of a ²³⁸U⁸⁶⁺ beam at 345 MeV/nucleon with the intensity ranging from 7 to 12 pnA, impinging on a beryllium target with a thickness of 3 mm. The nuclei of interest were separated and identified through the BigRIPS separator and the following ZeroDegree spectrometer [14]. A total of 5.3×10^{4} ¹²⁶Pd fragments were implanted into a highly segmented active stopper, named WAS3ABi [15], which consisted of eight double-sided silicon-strip detectors (DSSSDs) stacked compactly. Each DSSSD had a thickness of 1 mm with an active area segmented into sixty and forty strips (1 mm pitch) on each side in the horizontal and vertical dimensions, respectively. The DSSSDs also served as detectors for electrons following β -decay and internal conversion (IC) processes. Gamma rays were detected by the EURICA array [16] that consisted of twelve Cluster-type detectors, each of which contained seven HPGe crystals packed closely. The γ -ray measurements were carried out with a time condition up to 100 μ s relative to the trigger signal generated either from a plastic scintillation counter placed at the end of the beam line or from WAS3ABi.

Isomeric states with (sub)microsecond lifetimes were investigated by means of particle- γ (delayed) coincidence on an event-by-event basis [7]. Meanwhile, to study the decay of long-lived states in the millisecond range, the implantation of an identified particle was associated with the subsequent electron events that were detected in the DSSSD pixels where the beam particle was implanted. The result presented in this Letter was obtained using the latter analysis method.

The decay schemes of the isomeric states in ¹²⁶Pd are exhibited in Fig. 1. For ¹²⁶Pd, the $J^{\pi} = (5^{-})$ and (7^{-}) isomers at 2023 and 2110 keV, respectively, were reported in Ref. [7]. In the present Letter, the γ rays below these isomers, except for the 86 keV line, have also been observed in coincidence with electrons that were associated with the prior implantation of ¹²⁶Pd, as demonstrated in Fig. 2(a). This observation implies the existence of a longlived, higher-spin isomer which decays via the cascades that include electromagnetic transitions with relatively large total conversion coefficients (α_T) . With gates on these γ rays, a prominent peak can be found in an electron spectrum [marked with "I" in the inset of Fig. 2(b)]; this corresponds to the conversion electrons for the 86 keV, E2 transition ($\alpha_T = 2.374$ [17]). In Fig. 2(b), a γ ray at 297 keV is clearly visible in addition to the γ rays below the (5^{-}) isomer by gating on the 86 keV IC peak. The appearance of the 297 keV peak is emphasized by taking a γ -ray time condition earlier than electron events, as is evident from the inset of Fig. 2(a), suggesting that this new γ ray precedes the highly converted 86 keV transition.

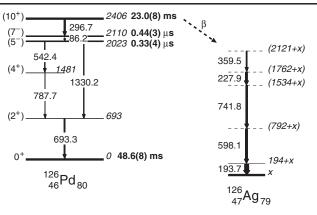


FIG. 1. Decay schemes of the $J^{\pi} = (10^+)$ isomer in ¹²⁶Pd. The widths of arrows represent relative intensities of γ rays summarized in Table I, except for the 86 keV transition in ¹²⁶Pd. In ¹²⁶Ag, the order of transitions between the levels at 2121 + x and 194 + x keV, which are populated via the β decay of the (10⁺) isomer, is ambiguous. (x denotes the unknown excitation energy of the lowest-lying state.)

Furthermore, the 297 keV γ ray is observed in coincidence with the other γ rays in ¹²⁶Pd [see Fig. 2(c) as an example]. Thus, the long-lived isomer can be identified at an excitation energy of 2406 keV. A peak marked with "II" in the inset of Fig. 2(b) is expected to arise from the conversion electrons for the 297 keV transition, being most likely of an *E*3 character ($\alpha_T = 0.1197$ [17]).

The half-life $(T_{1/2})$ derived from the time distribution of the 297 keV γ ray is in agreement with that of the 693 keV one within experimental errors, as illustrated in the insets of Fig. 2(c). Similar half-lives have been observed for six other γ rays that are listed in the second group of Table I; these transitions are expected to belong to high-spin states in ¹²⁶Ag (Z = 47, N = 79) which are populated through the β decay of the long-lived isomer in ¹²⁶Pd. Therefore, the isomeric half-life is determined to be 23.0(8) ms by taking a weighted average of the respective values listed in the first and second groups of Table I. Based on the observed mutual coincidence [see Fig. 2(d)] and γ -ray intensities, we propose the decay scheme from the long-lived isomer in ¹²⁶Pd to the high-spin states in ¹²⁶Ag as shown in Fig. 1.

The γ rays following the β decay of the ground state of ¹²⁶Pd have also been observed in Fig. 2(a). Prior to the present work, an isomeric state with $J^{\pi} = (1^{-})$ was known at 254 keV in ¹²⁶Ag [18]. This isomeric transition is visible in our analysis of delayed coincidence with β rays, as shown in the inset of Fig. 2(a). The γ rays which show half-lives longer than the 23 ms isomer are summarized in the third group of Table I and assigned for low-spin states in ¹²⁶Ag. A weighted average of the respective fits leads to $T_{1/2} = 48.6(8)$ ms for the ground state in ¹²⁶Pd, which is consistent with the value extracted from an independent analysis of β -decay half-lives [19]. It is worth remarking that the isomer half-life is nearly half of that of the ground state.

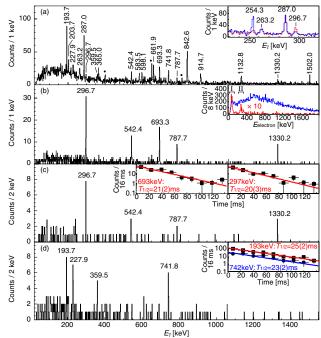


FIG. 2 (color online). (a) γ -ray spectrum measured within 50 ms after the ¹²⁶Pd implantation with a gate on an electron- γ time difference of $-0.5 \leq \Delta t_{e\gamma} \leq 0.5 \ \mu$ s. Contaminants from the granddaughter ¹²⁶Cd are marked with filled circles. The inset magnifies the energy region from 230 to 310 keV measured with $-4.0 \leq \Delta t_{e\gamma} \leq 0.5 \ \mu$ s (red) and $-0.5 \leq \Delta t_{e\gamma} \leq 50 \ \mu$ s (blue). (b) γ rays measured with $-4.0 \leq \Delta t_{e\gamma} \leq 0.5 \ \mu$ s, and additionally, with a gate on the electron peak marked with "T" in the inset, where the spectrum depicted with the red line (multiplied by a factor of 10) is obtained with a sum of gates on the γ rays below the (5⁻) isomer in ¹²⁶Pd, while the blue line represents an electron spectrum without γ -ray gates. (c) and (d) γ -ray coincidence spectra measured by gating on the 693 and 598 keV γ rays, respectively. The insets show the time distributions of γ rays as indicated.

A spin and parity of (10⁺) was assigned for the $T_{1/2} =$ 23.0(8) ms isomeric state on the basis of the arguments as follows: Assuming that all the intensities of the β decay from the long-lived isomer flow into the 194 keV transition in ¹²⁶Ag, the number of γ -ray counts can be approximated as $N_{\gamma}(194) \approx Y \times R_{\text{ext}} \times \varepsilon_{\beta} \times \varepsilon_{\gamma}(194)$. Here, Y denotes the isomeric yield that can be calculated by multiplying the total number of the ¹²⁶Pd ions by the isomeric ratio, which is estimated to be 7.7(2)% from the population of the $J^{\pi} = (10^+)$ isomer in ¹²⁸Cd [20] observed simultaneously in the present work. The γ -ray full-energy peak efficiency ε_{γ} is evaluated using ¹³³Ba and ¹⁵²Eu sources placed at the center of WAS3ABi, while $\varepsilon_{\beta} = 68(2)\%$ is derived from the ratio of the detected β events, which are associated predominately with the decay of the ¹²⁶Pd ground state, to the ¹²⁶Pd implants. The internal-decay branch, $R_{\text{int}} = 1 - R_{\text{ext}}$, can be estimated to be 28(8)% with the measured $N_{\gamma}(194)$. Meanwhile, R_{int} can be evaluated independently using the intensity of the 297 keV γ ray

Parent J^{π}	Daughter	E_{γ} (keV)	$T_{1/2}$ (ms)	I_{γ} (%)
¹²⁶ Pd, (10 ⁺)	¹²⁶ Pd	86.2		
		296.7	20(3)	7.5(8)
		542.4	20(3)	4.1(6)
		693.3	21(2)	6.6(9)
		787.7	18(4)	3.8(5)
		1330.2	23(5)	3.5(8)
¹²⁶ Pd, (10 ⁺)	¹²⁶ Ag, high spins	193.7	25(2)	26(3)
		203.7^{a}	27(2)	11(2)
		227.9	27(10)	12(2)
		359.5	17(4)	7(2)
		598.1	26(3)	13(2)
		741.8	23(2)	12(2)
¹²⁶ Pd, 0 ⁺	¹²⁶ Ag, low spins	254.3 ^a	54(3)	52(4)
		263.2 ^a	56(4)	21(3)
		287.0^{a}	48(2)	68(4)
		365.0 ^a	48(3)	30(3)
		583.5 ^a	46(5)	17(3)
		661.9 ^a	46(3)	35(3)
		842.6 ^a	48(2)	100(6)
		914.7 ^a	46(4)	30(3)
		1133.2 ^a	52(4)	30(4)
		1502.0 ^a	41(3)	27(4)

TABLE I. Summary of transitions in ¹²⁶Pd and ¹²⁶Ag.

^aNot incorporated in the level schemes of Fig. 1.

in ¹²⁶Pd as follows, $N_{\gamma}(297) \approx Y \times R_{\text{int}} \times [\alpha_T(86)/1 + \alpha_T(86)] \times \varepsilon_{\text{IC}} \times [1/1 + \alpha_T(297)] \times \varepsilon_{\gamma}(297)$. Given an *E*3 multipolarity for the 297 keV transition, $R_{\text{int}} = 0.23(3)/\varepsilon_{\text{IC}}$, being consistent with the value obtained above for $\varepsilon_{\text{IC}} \gtrsim 60\%$. A transition strength of B(E3) = 0.069(20) W.u. can be obtained for the 297 keV γ ray with $R_{\text{int}} = 28(8)\%$. Such a hindered *E*3 transition from the analogous 10⁺ isomer is known to be present also in ¹³²Te (Z = 52, N = 80) [21] and ¹³²Xe (Z = 54, N = 78) [22], as will be discussed later.

The main surprise in the present Letter is the small energy difference between the (10^+) and (7^-) isomers, $\Delta E_{7^-}^{10^+}$, in ¹²⁶Pd, compared to the analogous levels in the heavier N = 80 isotones, as demonstrated in Fig. 3(a). Since these two levels consist predominantly of maximally aligned two neutron-hole configurations, $(\nu 1h_{11/2}^{-2})_{10^+}$ and $(\nu 1h_{11/2}^{-1}2d_{3/2}^{-1})_{7}$, their level energies depend on the singleparticle energies (SPEs) of the $\nu 1h_{11/2}$ and $\nu 2d_{3/2}$ neutron orbits, as well as the strength of interactions between them. It can be seen in Fig. 3(a) that the energy of the $11/2^{-1}$ level relative to the $3/2^+$ ground state in the neighboring N = 81isotones decreases when approaching Z = 50. Note that these states are of one neutron-hole nature, and that the smooth reduction in energy can be essentially interpreted in terms of a short-range proton-neutron interaction [23] as follows: The $\pi 1 q_{7/2}$ proton orbit lies just above the Z = 50shell closure. The monopole interaction between $\pi 1g_{7/2}$ and $\nu 1h_{11/2}$, a spin-flip pair with $\Delta \ell = 1$ and $\Delta n = 0$, is

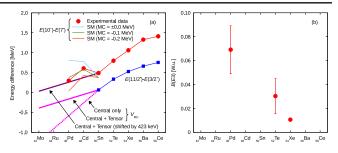


FIG. 3 (color online). (a) Energy differences between the 10⁺ and 7⁻ states, $\Delta E_{7^-}^{10^+}$, in N = 80 (filled circles) and between the 11/2⁻ and 3/2⁺ states in N = 81 (filled squares). The data are taken from Ref. [25] for $Z \ge 50$, Ref. [20] for ¹²⁸Cd, and the present work for ¹²⁶Pd. For $Z \le 50$, the differences in energy between the $1h_{11/2}$ and $2d_{3/2}$ neutron orbits calculated by $V_{\rm MU}$ interaction [6] and $\Delta E_{7^-}^{10^+}$ by SM calculations with monopole corrections (MC) are also depicted. (b) $B(E3; 10^+ \rightarrow 7^-)$ values in $\frac{132}{46}$ Pd, $\frac{132}{52}$ Te [21], and $\frac{132}{56}$ Xe [22].

stronger than the $\pi 1g_{7/2} \cdot \nu 2d_{3/2}$ pair, due to the larger overlap between the radial wave functions of the two orbits. Therefore, when emptying the $\pi 1g_{7/2}$ subshell, the $\nu 1h_{11/2}$ orbit is relatively less bound than the $\nu 2d_{3/2}$ one. The moderation in the slope at higher Z may be ascribed to the partial occupation of the $\pi 2d_{5/2}$ orbit, which exerts a weaker (stronger) monopole interaction on the $\nu 1h_{11/2}$ ($\nu 2d_{3/2}$) neutrons than $\pi 1g_{7/2}$. As is evident from the comparison in Fig. 3(a), a similar trend is observed for $\Delta E_{7^{-1}}^{10^+}$. This finding supports that these isomers can serve as sensitive probes for the evolution of the constituent neutron shell orbits.

Below Z = 50, protons in the $\pi 1 g_{9/2}$ subshell play a major role in changing the neutron SPEs, because this orbit is near the Fermi surface. The neutron SPEs are estimated by the monopole-based universal interaction, $V_{\rm MU}$, which consists of the Gaussian central force and the tensor force based on the $\pi + \rho$ meson exchanges, using the parameters fixed in Ref. [6]. The $V_{\rm MU}$ interaction has been applied to other regions of exotic nuclei and successfully reproduced various types of the shell evolution [6]. Figure 3(a) shows the evolutions of the relative $11/2^{-}$ energy starting from the experimental value in 131 Sn, predicted by the V_{MU} calculation with A = 130. In the calculation, the $\pi 1g_{9/2}$ subshell is considered to be well isolated between Z = 50and 40. In Fig. 3(a), only with the central force the $11/2^{-1}$ energy rapidly decreases to negative values as the proton number decreases, indicating that the $\nu 1h_{11/2}$ orbit moves away from the $\nu 2d_{3/2}$ one towards the N = 82 gap. If there is the tensor force effect, however, the slope becomes less steep. The observed reduction in $\Delta E_{7^-}^{10^+}$ from ¹³⁰Sn to ¹²⁶Pd is consistent with the expectation of $V_{\rm MU}$ including both the central and tensor forces, given the same trend as the $11/2^{-}$ relative energies. Based on these findings, it can be concluded that the tensor force slows down the upward drift of the $\nu 1h_{11/2}$ SPE when protons are removed from the $\pi 1g_{9/2}$ orbit below ¹³²Sn.

One can notice a kink in the systematics of $\Delta E_{7^-}^{10^+}$ at ¹²⁸Cd in Fig. 3(a). It should be noted that the above $V_{\rm MII}$ calculation is based on the monopole properties of the proton-neutron interactions. While the overall trend can be accounted for by $V_{\rm MU}$, other multipole components have to be introduced in a shell-model framework to describe the details of the level evolution. To confirm the effect of configuration mixing, a SM study based on the j-j coupling scheme was performed independently of the $V_{\rm MU}$ calculation. With the doubly magic nucleus ¹³²Sn as a closed core, the model space adopted includes the $2d_{3/2}$, $1h_{11/2}$, $3s_{1/2}$, $2d_{5/2}$, and $1g_{7/2}$ orbits for neutron holes, and the $1g_{9/2}$, $2p_{1/2}$, $2p_{3/2}$, and $1f_{5/2}$ orbits for proton holes. The SPEs were taken from the experimentally known levels in the single-hole nuclei, 131 Sn [24] and 131 In [9,26,27]. The surface-delta interaction (SDI) was employed for the proton-proton and neutron-neutron interactions, which were adjusted to reproduce the observed level energies of ¹³⁰Cd and ¹³⁰Sn, respectively. The proton-neutron twobody matrix elements (TBMEs) were derived from the empirical parametrization of interaction potentials [28]. For the $\pi 1g_{9/2}$ - $\nu 1h_{11/2}$ multiplet, the monopole correction was introduced by adding a constant to the diagonal TBME.

The results of SM calculations with different values of the monopole correction for $Z \leq 50$ are displayed in Fig. 3(a). The kink manifests in the calculated $\Delta E_{7^-}^{10^+}$, as well as in the experimental result. Moreover, it can be seen that the variation in $\Delta E_{7^-}^{10^+}$ is very sensitive to the value of the monopole correction. According to the present SM calculation, the wave functions of the 10⁺ and 7⁻ states in each N = 80 isotone are scarcely affected by the monopole corrections. These arguments indicate that the lowering of $\Delta E_{7^-}^{10^+}$ in ¹²⁶Pd is ascribed to the monopole effect between the $\pi 1g_{9/2}$ and $\nu 1h_{11/2}$ orbitals rather than the configuration mixing of any kind.

Finally, it is worth commenting on the E3 transition depopulating the 10^+ isomeric state. If the 10^+ and $7^$ isomers had pure two neutron-hole configurations, $\nu 1h_{11/2}^{-2}$ and $\nu 1h_{11/2}^{-1}2d_{3/2}^{-1}$, respectively, an E3 decay would not occur between them. A possible explanation for the measured large hindrance, $B(E3; 10^+ \rightarrow 7^-) =$ 0.069(20) W.u., may be given in terms of a small admixture of different multiplets. In general, transition strengths are more susceptible to the configuration mixing than level energies. As can be inferred from the drift of the neutron SPEs observed for the N = 51 isotones with Z = 40 - 50[29], the $\nu 1g_{7/2}$ orbit is expected to be rapidly pushed up as removing the $\pi 1g_{9/2}$ protons, in consequence of the strong (attractive) monopole interaction between the spin-orbit partner. In Fig. 3(b), the observed rise in $B(E3; 10^+ \rightarrow 7^-)$ with decreasing proton number may suggest the admixture of different multiplets including the $\nu 1g_{7/2}$ hole, such as $\nu 1h_{11/2}^{-1}1g_{7/2}^{-1}$, in the 7⁻ state, to which the 10⁺ isomer decays. [The aforementioned SM calculation predicts that the wave function of the 7⁻ state contains such kind of configurations by ~2.6%.] This hypothesis, based on the monopole migration, will be verified experimentally by identifying the $7/2^+$ hole states in the N = 81 isotones, which can be populated in the β decay of odd-mass N = 82 nuclei through the $\nu 1g_{7/2} \rightarrow \pi 1g_{9/2}$ Gamow-Teller transition.

In conclusion, a new isomeric state with $J^{\pi} = (10^+)$, $T_{1/2} = 23.0(8)$ ms has been identified at 2406 keV in ¹²⁶Pd. The long lifetime, which is about half $T_{1/2}$ of the ground state, allows the β decay to compete with a hindered E3 transition. The energy difference between the 10^+ and 7⁻ isomeric levels is smaller in ¹²⁶Pd than in the heavier N = 80 isotones, being qualitatively explained by the proton-neutron monopole interaction that includes the central- and tensor-force effects. This result indicates that the tensor force suppresses the drift of the $\nu 1h_{11/2}$ neutron orbit towards the N = 82 gap when protons are removed from the $\pi 1 q_{9/2}$ subshell. Based on shell-model calculations with monopole corrections, we revealed that the energy shift of the 10^+ isomer is caused by the monopole part of the $\pi 1g_{9/2}$ - $\nu 1h_{11/2}$ interaction, but not by the configuration mixing.

Probing the nature of single-neutron states not only below but also above the N = 82 shell gap is essential to pin down whether the shell quenching takes place in this neutron-rich region. The knowledge about neutron orbitals obtained in the present Letter will prompt further study of the single-neutron states by means of β -decay and transferreaction experiments using radioactive-isotope beams.

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