Letter

Three-quasiparticle isomers in odd-even ^{159,161}Pm: Calling for modified spin-orbit interaction for the neutron-rich region

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Neutron-rich Pm ($Z = 61$) isotopes were studied by delayed γ-ray spectroscopy at RIBF, RIKEN Nishina Center using the in-flight fission of a 345 MeV/nucleon ^{238}U beam. A cluster-type Ge detector array, EURICA, was used to measure the delayed γ rays from stopped ions. Isomeric γ decays were observed in ^{159}Pm and ^{161}Pm with half-lives of 4.97(12) μs and 0.79(4) μs, respectively. Level schemes for ^{159}Pm and ^{161}Pm were constructed in this study. The isomeric states of ^{159}Pm and ^{161}Pm could be interpreted as two quasiparticle excitations of neutrons with the configurations of $\nu(7/2[633] \otimes 5/2[523])$ and $\nu(7/2[633] \otimes 1/2[521])$, respectively. They are analogous to the isomers that have been observed systematically in other even-mass $N = 98$ and $N = 100$ isotones in this region. A projected shell model calculation was performed and it reproduced the order of three-quasiparticle states only if new Nilsson parameters with an N-dependent spin-orbit interaction were used.

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This work demonstrates that the strength of spin-orbit interactions in standard Nilsson parameters needs to be modified to study the properties of neutron-rich rare-earth nuclei around $A = 165$, and provides new evidence supporting the existence of the deformed $N = 98$ subshell gap in odd-mass nuclei for the first time.

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Nuclear properties of neutron-rich rare-earth nuclei around $Z = 60$ are the possible key to one of the longstanding astrophysical questions: the formation of the $A \approx 160$ peak observed in the elemental abundance distribution. The prominent features of the r-process abundance in the solar system are the pronounced peaks at $A \approx 130$ and ≈ 195, which are understood in terms of the enhanced stability of nuclei at the neutron magic numbers, $N = 82$ and 126. However, the production mechanism of the rare-earth peak at $A \approx 160$ is still under intensive investigations [1–5]. The question is how the rare-earth peak was produced. Simulations [6] found that the rare-earth peak is extremely sensitive to the nuclear-physics input such as nuclear deformation and β-decay properties in both hot and cold evolutions.

From the nuclear-structure point of view, the formation of the rare-earth peak requires extra stability of local nuclei. This is quite analogous to the stability associated with large gaps in the spherical picture, although in the case of the deformed rare-earth nuclei a large shell gap between Nilsson single-particle (SP) orbitals stabilizes the nuclear shape at large deformation. Based on the relativistic mean-field theory, calculations predicted a deformed shell gap at $N = 100$ around $Z \approx 62$ [7,8], and Ghorui *et al.* [9] argued that this gap will make the $N = 100$ isotones serve as a waiting point in the nucleosynthesis of the r process. Mumpower *et al.* identified a region of nuclei around $N \approx 100$ which is important to the rare-earth peak formation around $N \approx 100$ [4]. Thus experimental studies of the deformed gaps around $A \approx 160$ are important.

The location and size of deformed shell gaps are closely related to the behavior of deformed SP states. There is compelling evidence indicating that the orders of deformed SP states in neutron-rich regions are different from those in the stable region. It was revealed in Ref. [10] that, in some of the light rare-earth isotopes ($_{60}$Nd, $_{62}$Sm, and $_{64}$Gd) in the neutron-rich ($N = 98$–102) region, none of the preexisting potentials—the Woods-Saxon potential, the Nilsson modified oscillator potential with "universal" parameters, and the folded Yukawa potential—describe the correct ordering of the neutron SP states. This is a serious problem because these single-particle models have been regarded as basic and reliable tools for understanding the deformed structure. This suggests that, in an environment with extreme neutron excess, the nucleon spin-orbit force may need to be adjusted to reproduce the abnormal ordering of neutron SP orbits observed experimentally.

In principle, the variation of single-particle distribution with increasing neutron number is determined by the intricate interplay of the central, tensor, and spin-orbit forces in the shell model. However, there are not many discussions of the heavy, deformed, neutron-rich nuclei so far. The Nilsson model, on the other hand, treats all the force effects by absorbing them into parameters which are fitted to data. This method is a popular way of describing the single-particle distribution for the neutron-rich region.

In order to make a minimal modification for the spin-orbit force to describe existing data for the neutron-rich $A \approx 160$ mass region, Liu *et al.* recently proposed an isotope-dependent spin-orbit term [11] for the "standard" Nilsson model suggested by Bengtsson and Ragnarsson in 1985 [12]. The new formula for the Nilsson parameters κ and μ of the neutron $n = 6$ shell,

$$(\kappa, \mu)_{\mathrm{New}} = (1 - 0.015|N - 102|)(\kappa, \mu)_{N=102}, \tag{1}$$

was introduced to apply to the neutron-rich nuclei with neutron number $N \geqslant 92$. The dependence is proportional to neutron number N, and the largest values with the 102 neutron occupation in Eq. (1) are $(\kappa)_{N=102} = 0.0713$ and $(\mu)_{N=102} = 0.391$. With the stronger spin-orbit interaction in the new Nilsson parameters, the neutron $i_{13/2}$ intruder orbital $\nu 7/2^+[633]$ is pushed down properly to lie between the normal orbitals $\nu 5/2^-[523]$ and $\nu 1/2^-[521]$, as indicated later in the insets of Fig. 5. The new Nilsson parametrization was tested thoroughly using available experimental data from ground and low-lying states in odd-mass nuclei in the neutron-rich region. It was emphasized [11] that, to understand the change of deformed SP orbitals around the neutron numbers $N = 98$ and 100, the study of isomeric states is of great importance because the excitation energies of two quasineutron isomers involving two different orbitals from the $N = 5$ shell of normal parity and the $N = 6$ intruder with unique parity are sensitive to the spin-orbit interaction and allow us to investigate its variation.

Experimental studies of $N \approx 100$ rare-earth region became available recently with the advent of new generation radioactive isotope (RI) beam facilities. A previous spectroscopic study for $Z \approx 62$ nuclei was carried out by using spontaneous fission of ^{252}Cf [13], and quasiparticle (qp) isomers were observed in Nd and Sm isotopes up to $N = 98$. Several years after, the observation was extended to $N = 102$ in Sm and Gd isotopic chains using in-flight fission, and a local maximum of the ground-band energies was revealed at $N = 100$ which seemingly indicated the predicted deformed shell gap at $N = 100$ [14]. Following studies discovered the 4^- $\nu(7/2[633] \otimes 1/2[521])$ isomers in the $N = 100$ isotones from $Z = 60$ to 64 [15,16] and found that their excitation energies and hindrance factors are not significantly different from those in the stable $_{68}$Er and $_{70}$Yb isotones. This means the energy space between the $\nu 7/2[633]$ and $\nu 1/2[521]$ orbitals at $N = 100$ is stable against the change of the proton number from 60 to 70 [16], which can be explained without the appearance of the $N = 100$ deformed shell gap around $Z \approx 62$. Recently, a 6^- $\nu(5/2[523] \otimes 7/2[633])$ state was discovered in ^{162}Gd$_{98}$ [10], indicating the large shell gap is actually present at $N = 98$.

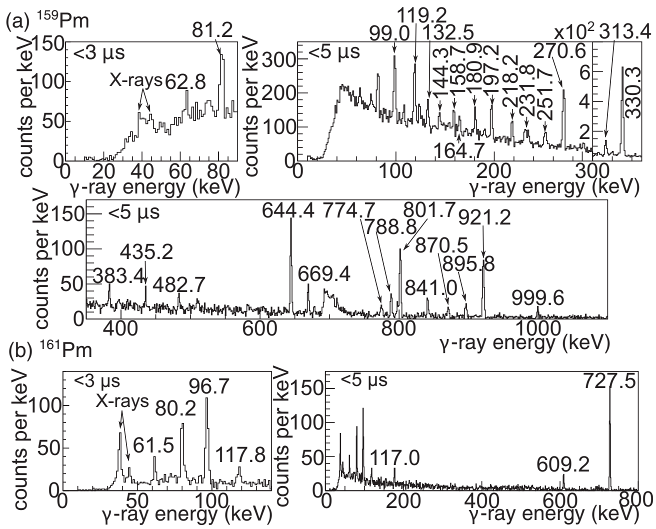

FIG. 1. Delayed γ-ray spectra of (a) ^{159}Pm and (b) ^{161}Pm. Time windows are up to 5 μs after the implantation. The low-energy parts of the spectra are drawn with 3 μs time window.

TABLE I. List of the γ-ray energies, relative intensities, and the half-lives of the ^{159,161}Pm isomer obtained in this study. I_{rel} is a number of detected γ rays normalized by the most intense one in the decay. The main source of the error on E_γ is the systematic error, which is 0.32 keV as described in the text.

E_γ (keV)	$T_{1/2}$ (μs)	I_{rel} (%)	E_γ (keV)	$T_{1/2}$ (μs)	I_{rel} (%)
			^{159}Pm		
62.8		13(6)	320.2		4.2(22)
81.2	2.7(14)	32(8)	330.3	4.51(16)	100(5)
99.0	4.5(23)	42(8)	383.4		4.5(14)
119.2	2.8(12)	33(6)	435.2		4.0(10)
132.5		13(3)	482.7		6.4(12)
144.3		7(2)	644.4	5.8(5)	30.1(19)
158.7		11.5(21)	669.4	2.9(9)	13.3(15)
164.7	4(4)	6.2(22)	774.7		5.4(15)
180.9	5(4)	9.5(19)	788.8	5.5(7)	
197.2	8.3(12)	14.3(23)	801.7	3.9(9)	36.2(24)
218.2	5.1(25)	9.3(17)	841.0		8.2(14)
231.8	5.4(23)	8.2(24)	870.5		6.5(12)
251.7	3.1(4)	8.8(20)	895.8		7.0(13)
270.6	4.3(9)	27.9(26)	921.2	5.8(5)	32.6(24)
313.4	3.1(4)	13.8(17)	999.6		5.9(12)
			^{161}Pm		
61.5	0.83(13)	12(4)	177.0	1.00(25)	9.9(19)
80.2	1.06(24)	36(6)	609.2	0.93(14)	16.0(27)
96.7	0.82(14)	44(6)	727.5	0.88(10)	100(6)
117.8	0.54(17)	5(2)			

In this Letter, we report new qp isomers in $^{159,161}_{\ \ 61}$Pm. Those 3-qp isomers involve one quasiproton plus two quasineutrons occupying the normal and intruder orbitals near the Fermi surface. The observation of the isomers cannot be explained by traditional Nilsson parameters. We demonstrate that the strength of the spin-orbit interaction needs to be changed in accordance with the neutron number in exotic nuclei to explain the observation. We also confirmed that the reported deformed shell gap at $N = 98$ [10] is present not only in even-even but also in odd-mass nuclei.

The spectroscopic study of neutron-rich Pm isotopes was carried out at the RIBF facility at RIKEN Nishina Center. The primary ^{238}U^{86+} beam at 345 MeV/nucleon bombarded a 4-mm-thick Be production target to induce in-flight fission. The typical intensity of the primary beam was ≈7 p nA. The total numbers of implantation were 4.8×10^5 and 5.5×10^4 ions for ^{159,161}Pm, respectively. Fission fragments were separated and identified in the BigRIPS in-flight separator [17]. Detailed explanations of the particle identification at the BigRIPS are shown in Refs. [18,19].

The measurements were conducted in two different stopper setups. One of them was optimized for isomer spectroscopy. Ions were implanted into a 1-mm-thick copper plate in order to accept a wide range of nuclides with high implantation rates up to ≈1 kHz. The other setup was optimized for β-γ spectroscopy by employing five layers of silicon-strip active stoppers, WAS3ABi [20]. Data sets from the latter setup were also used for the isomer spectroscopy but the total implantation rate was limited up to ≈100 Hz. The delayed γ rays from the implanted ions were detected by a cluster-type Ge detector array, EURICA [21], consisting of 12 EUROBALL clusters [22]. The energies and timings of delayed γ rays were measured in a time window of 16 μs following the ion implantation.

The spectra of ^{159,161}Pm are shown in Fig. 1 and the list of observed γ rays are summarized in Table I. Systematic errors on the γ-ray energies are estimated to be 0.32 keV, which is the standard deviation of the energy difference between current and previous measurements for the γ rays reported in Refs. [13,16].

In ^{161}Pm, the half-life of the isomer was obtained as 0.79(4) μs from the time spectrum of all the delayed γ rays associated with ^{161}Pm implants [see Fig. 2(b)]. Figure 3(b) shows the proposed level scheme of ^{161}Pm. The ground state of ^{161}Pm was assumed to be $(5/2^-)$ from the systematics [23] and from a theoretical prediction [24]. A rotational band structure up to the $(13/2^-)$ state at 357 keV was identified. The energy levels of the ground-state (g.s.) bands of ^{153}Pm and ^{155}Pm nuclei [25] are similar to those observed in ^{161}Pm. The isomer was assigned to a state at 966 keV which decays to

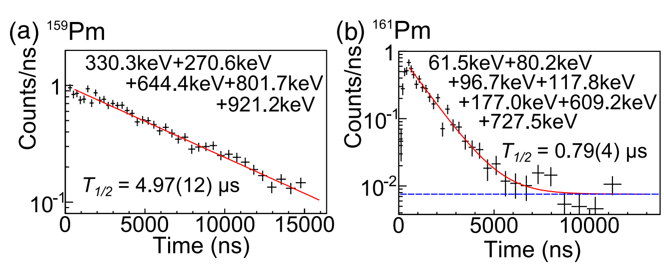

FIG. 2. Time spectra of delayed γ rays in (a) ^{159}Pm and (b) ^{161}Pm isomers. The decay curves were fitted by the function shown by the red curves. The dashed blue line in (b) shows the constant background component of the fit function.

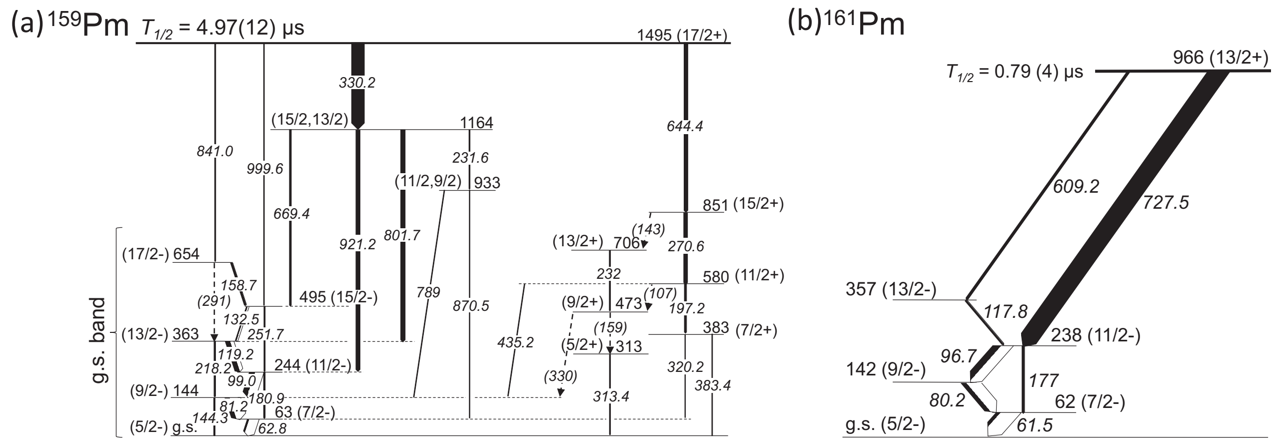

FIG. 3. Level schemes of (a) ^{59}Pm and (b) ^{161}Pm. The width of each arrow filled in black is proportional to the relative intensities of the observed γ rays. The widths of the white parts of the arrays show the intensities of internal conversions estimated by BrIcc [26].

both 357-keV ($13/2^-$) and 238-keV ($11/2^-$) states. This level scheme was deduced from the energy sums, relative intensities, and γ-γ coincidences. The sums of relative intensities of the decays into and out of each state agreed with this level scheme by making internal conversion corrections assuming $M1$ and $E2$ decays for the inter- and intraband transitions in the g.s. band. The mutual γ-γ coincidence relations between the 727.5-keV γ ray and 61.5-, 80.2-, 96.7-keV γ rays were confirmed, and are consistent with the level scheme.

Figure 3(a) shows the proposed level scheme of ^{159}Pm. The half-life of the ^{159}Pm isomer was deduced to be 4.97(12) μs from intense γ rays [see Fig. 2(a)]. A g.s. band is constructed similar to that of ^{161}Pm but built up to higher spin with the excitation energy of 654 keV. The 330-keV γ ray has coincidences with 802-, 669-, 789-, 921-, and 871-keV γ rays as shown in Fig. 4(a). The 644-keV γ ray has coincidences with the γ rays belonging to the sideband shown on the right-hand part of the level scheme [Fig. 4(b)]. There was no coincidence between 330- and 644-keV γ rays. The γ rays shown by an arrow with a dashed line in Fig. 3 are observed only in γ-γ spectra. The 144-, 159-, and 330-keV γ rays are hidden by bigger peaks with close energies. The 313-keV γ ray has a coincidence with the 197-keV γ ray which shows that there is a 70-keV transition between the states at 383 and 313 keV. The 70-keV transition is not observed as a significant peak due to the large absorption and the high internal conversion rate. The 789-keV peak could contain the γ ray from the $LaBr_3$ detectors in the experimental setup but we concluded that it is mainly from the isomer because the peak has a decay curve with $T_{1/2} = 5.5(7)$ μs in its time distribution.

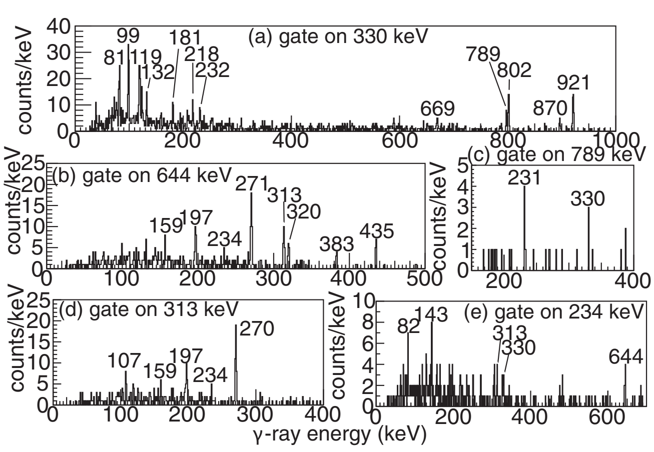

FIG. 4. Gamma-gated energy spectra for ^{159}Pm. The time windows are up to 5 μs after implantation and 300 ns between γ events.

The g.s. bands of ^{159,161}Pm are close in energy to each other and to those of ^{153,155}Pm [27]. The spins and parities of the ground states of ^{159,161}Pm are likely to be $5/2^-$ from the systematics of ^{153,155}Pm.

The sideband starting from 313-keV state in ^{159}Pm probably has a configuration with proton excitation to a positive parity orbital such as $\pi 5/2[413]$. In ^{153}Pm and ^{155}Pm, the $5/2^+$ states built from the same proton SP orbit are known at 32.2 and 180.6 keV, respectively. From the systematics, the 313-keV state in ^{159}Pm may have this configuration.

The hindrance factor (F) of the 609-keV decay in ^{161}Pm was 6.6×10^9 by assuming an $E1$ transition. This hindrance is similar to those in the decays from the $K^\pi = 4^-$ isomeric state to the 4^+ state in g.s. bands in $^{168}Er_{100}$ (2.7×10^9) [28] and $^{170}Yb_{100}$ (2.8×10^9) [29]. This suggests that the isomeric state of $^{161}Pm_{100}$ has 2-qp configuration of $\nu(7/2[633] \otimes 1/2[521])$ as in other $N = 100$ isomers, coupled to an odd proton on 5/2[523], giving total spin-parity $J^\pi = 13/2^+$. As for the isomeric state in $^{159}Pm_{98}$, the spin and parity were assigned to be $J^\pi = 17/2^+$ by coupling the odd proton to a neutron $\nu(7/2[633] \otimes 5/2[523])$ with $K^\pi = 6^-$ configuration. The $J^\pi = 6^-$ isomers with the same neutron configuration have been reported at 1453 keV in $^{162}Gd_{98}$ [10] and at 1648.1 keV in $^{158}Nd_{98}$ [15]. Assuming an $E2$ transition, the reduced hindrance factor ($f_\nu = F^{1/(\Delta K - \lambda)}$, where λ is multipolarity) of the 644-keV decay in ^{159}Pm is extracted as 22(2). This is within the same order of magnitude as the value in ^{158}Nd, 76(4), which is given by an $E1$ decay from the isomer to the g.s. band.

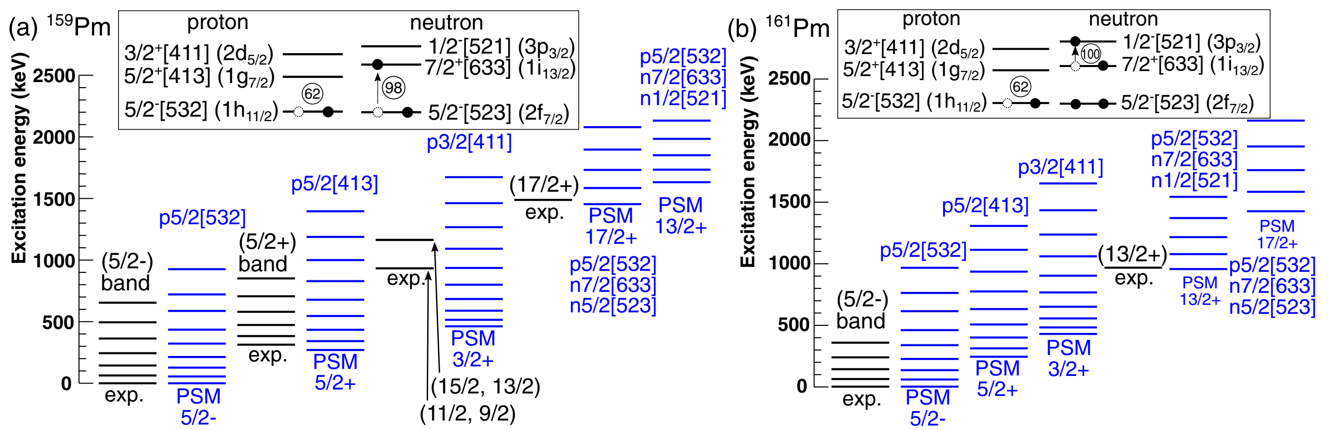

FIG. 5. Calculated excitation energies of (a) ^{159}Pm and (b) ^{161}Pm by PSM. Levels drawn in black with a label "exp." show experimental values.

In order to understand the observed 3-qp isomeric states in ^{159,161}Pm, a calculation employing the projected shell model (PSM) [30,31] is carried out. The PSM is a shell model with its multi-qp configurations constructed in a deformed SP basis. Since the deformation is related to the breaking of the rotational symmetry, angular-momentum projection calculation is performed to recover the broken symmetry in the configurations, which ensures that the final results to be compared with data are calculated in the laboratory frame. The PSM, which is suited for the description of well-deformed nuclei, has been successfully applied for different neutron-rich mass regions [32,33]. However, prior to the present work, the 3-qp configurations were not included in the original PSM configuration space [34] because this 3-qp state consists of quasiparticles originating from three different N shells. Only very recently, the configuration space was greatly extended for odd-mass nuclei [35] thanks to the introduction of the Pfaffian algorithm for fast computation [36]. In particular, this development allows us for the first time to calculate 3-qp states of this kind (see their configurations in Fig. 5) within the PSM framework, and to study the microscopic mechanism of the isomers observed in this work.

In the calculation, we used deformation parameters $\varepsilon_2 = 0.30$ and $\varepsilon_4 = 0.03$ to construct the deformed basis for ^{159}Pm, and $\varepsilon_2 = 0.30$ and $\varepsilon_4 = 0.01$ for ^{161}Pm, which are consistent with those for other nuclei in this well-deformed mass region [11]. All other parameters including the monopole-pairing strength and quadrupole-pairing strength appearing in the Hamiltonian are the same as those used in previous works [11,32].

Figure 5 compares the calculated and experimental levels in ^{159,161}Pm. For both isotopes, the calculation yields the correct g.s. configuration $\pi 5/2^-[532]$ with a rotational band based on it. The calculation also reproduced a sideband built on top of $\pi 5/2[413]$ in ^{159}Pm, as observed at 313 keV experimentally. In ^{159}Pm, a $17/2^+$ state with 3-qp configuration $(\pi 5/2^-[532] \otimes \nu 7/2^+[633] \otimes \nu 5/2^-[523])$ is predicted at 1.465 MeV, which is in good agreement with the isomer in the experiment. A $13/2^+$ state is calculated at slightly higher energy, which is consistent with the fact that it is not observed as an isomer. It is noteworthy that this good agreement can only be achieved by using the newly proposed Nilsson parameters for the $N = 6$ neutron shell [11]. If we use the SP states generated by the standard Nilsson parameters in Ref. [12], the $17/2^+$ state is calculated at much higher energy above 2 MeV, and higher than the $13/2^+$ state. In this case, one would expect a γ decay from $17/2^+$ to $13/2^+$, which contradicts our observation.

In ^{161}Pm, the 3-qp configuration $(\pi 5/2^-[532] \otimes \nu 7/2^+[633] \otimes \nu 1/2^-[521])$ gives a $13/2^+$ state at 0.955 MeV in the calculation, which also nicely agrees with the experimental result. In this isotope, the ordering between the 3-qp $13/2^+$ and $17/2^+$ states is reversed from that in ^{159}Pm, giving rise to the observation of the $13/2^+$ isomer instead of the $17/2^+$ one in the experiment.

The PSM calculation predicted $B(M1)/B(E2)$ ratios from 0.15 to 0.30 $\mu_N/e^2\mathrm{b}^2$ for the g.s. bands of ^{159,161}Pm in the spin range from 9/2 to 17/2. This is consistent with the experimental values within the error bars which are 20% to 40% when we fix the mixing ratio, δ, to 1.

This work provides strong experimental evidence showing the validity of new Nilsson parameters for understanding the delicate relative positions between the $17/2^+$ isomer in ^{159}Pm and the $13/2^+$ isomer in ^{161}Pm. The results indicate that the strength of the spin-orbit interaction is changed by increasing neutron numbers and the $\nu 7/2^+[633]$ orbital is pulled lower in energy than in the standard Nilsson model at $N = 98$ and 100.

The experimental energy difference $E(17/2^+)_{N=98} - E(13/2^+)_{N=100}$ is found to be 529 keV for $_{61}$Pm isotopes. This value is comparable with the values $E(6^-)_{N=98} - E(4^-)_{N=100} = 541$ keV for $_{60}$Nd and 459 keV for $_{62}$Sm in Ref. [10], suggesting that the argument for the large energy gap between the $\nu 7/2[633]$ and $\nu 5/2[523]$ orbitals at $N = 98$ is valid also for odd-mass nuclei.

In summary, we identified new isomers in ^{159,161}Pm via γ-ray spectroscopy. These isomers are attributed to an odd proton in $\pi 5/2[532]$ coupled to 2-qp neutron configurations that have been observed in $N = 98$ and 100 isotones in this region. Together with the data for the 6^+ isomers in the $N = 98$ isotones and the 4^+ isomers in the $N = 100$ isotones (see [10,11] and references cited therein), a deformed gap at $N = 98$ can be established in both even-even and even-odd nuclei in this region. A PSM calculation was performed to understand the observed 3-qp isomers and reproduced the experimental data well by adopting the newly proposed Nilsson parameters [11]. This is the first demonstration that the "standard" Nilsson parameters for the stable nuclei need to be modified in exotic nuclei, showing that the spin-orbit interaction may be changed significantly in accordance with neutron number. This work provided a stringent test of Nilsson models in exotic nuclei and raised the necessity of the modification to the model. This is of great importance in obtaining reliable predictions on the properties of the *r*-process nuclei outside the current reach of experiments, and thus in understanding the formation of the rare-earth peak in the abundance distribution.

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