Gamma-ray spectroscopy approaching the limits of existence: a study of the excited states of ^{168}Pt and ^{169}Pt

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Excited states in the $N = 90$ and $N = 91$ Pt nuclei have been investigated using the JUROGAM and GREAT spectrometers in conjunction with the RITU gas-filled separator. These nuclei were populated via the reactions ^{92}Mo(^{78}Kr, 2n) and ^{94}Mo(^{78}Kr, 3n) at 335 and 348 MeV, respectively. The recoil-decay tagging technique has been used to correlate prompt γ radiation with the characteristic α decays of ^{168}Pt and ^{169}Pt. A γ-γ analysis has allowed a level scheme for ^{168}Pt to be reported for the first time and the level scheme for ^{169}Pt to be extended. The excitation energies of the proposed positive-parity yrast states of ^{168}Pt are compared with calculations based on the interacting boson model and found to be in excellent agreement. These data show a continuation of the trend towards vibrational nuclei as the $N = 82$ shell gap is approached. In addition, new excited states constituting two decay paths have been discovered in ^{169}Pt.

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I. INTRODUCTION

Investigations into the structure of nuclei approaching the proton drip line have advanced rapidly with the application of recoil-decay tagging (RDT) with highly efficient arrays of γ-ray detectors [1–3]. This is particularly true in the neutron-deficient W-Os-Pt region where this technique has been used to delineate the yrast sequences of a large number of nuclei (see Refs. [4–8] and therein). Experiments have shown [6, 8] that the even-even Pt isotopes evolve from the deformed regime near the neutron mid-shell ($N = 104$) through coexisting shapes to structures associated with γ-soft rotors and to weakly deformed vibrational nuclei as the spherical $N = 82$ shell closure is approached. The yrast configurations of these very neutron-deficient nuclei are sensitive to the relative positions of low-lying nucleon orbits to the Fermi surface. Near the closed shells, the low-spin yrast states are expected to be based on configurations formed by coupling the spins of a few aligned valence nucleons. For instance, in this region states of spin up to and including 6 $\hbar$ may be generated by the coupling of two valence neutrons occupying $f_{7/2}$ orbitals. In addition, states of 8 $\hbar$ may have their origin in $\nu(f_{7/2}, h_{9/2})$ or $\nu(h_{9/2})^2$ configurations. The spectroscopy of odd-A nuclei is also particularly important since the active single-particle orbitals can be identified and their core-polarizing influence explored.

The RDT technique has been used previously to study excited states of ^{168}Pt. Three transitions were reported by King *et al.* [7], however insufficient statistics were collected in order to perform a γ-γ coincidence analysis. Excited states were tentatively ordered through a comparison of the efficiency-corrected intensities of the observed transitions. In addition, the study of ^{169}Pt by Joss *et al.* [8] reported two transitions of 184 and 545 keV to be associated with the decay of excited states of ^{169}Pt. The 545 keV transition was assumed to depopulate the first excited state of ^{169}Pt but the origin of the 184 keV γ-ray could not be ascertained.

II. EXPERIMENTAL DETAILS

In the present study, the nuclei ^{168}Pt and ^{169}Pt have been populated using the reactions ^{92}Mo(^{78}Kr,2n) and ^{94}Mo(^{78}Kr,3n) at bombarding energies of 335 (162 hours) and 348 MeV (48 hours), respectively. The ^{78}Kr^{15+} beam was provided by the K130 cyclotron at the University of Jyväskylä with an average beam current of 7 pnA. Surrounding the self-supporting 550 μg/cm^2 thick targets, the JUROGAM Ge-detector array, comprising 43 escape suppressed spectrometers [9], was used to detect prompt γ radiation. The gas-filled separator RITU [10] was used

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to suppress scattered beam and transport recoiling fusion products to the focal plane where they were implanted into the double-sided silicon strip detectors (DSSDs) of the GREAT spectrometer [11]. The GREAT spectrometer allows the detection of charged particles and photons and comprises two adjacent DSSDs, a planar Ge detector, a large-volume Ge clover detector and an array of 28 Si PIN diode detectors. A multiwire proportional counter provided energy loss and (in conjunction with the DSSDs) time-of-flight information, which allowed the recoils to be distinguished from the scattered beam and subsequent radioactive decays. The Total Data Readout system [12] was employed in order to record time-stamped events observed in each of the constituent detectors. Subsequently, these data were sorted offline using the Grain analysis software [13] and the γ-γ coincidence data were analysed using the Radware software package [14].

The unambiguous assignment of γ rays detected at the target position to ^{168,169}Pt was made through temporal and spatial recoil-decay correlations recorded at the focal plane. The decay properties of the characteristic α decay of ^{168}Pt was measured in the present work to be $E_\alpha = 6823(3)$ keV with a half-life of 1.98(16) ms which is consistent with previously measured values [15–17]. Similarly, new measurements for the α decay properties of ^{169}Pt, $E_\alpha = 6695(5)$ keV, $T_{1/2} = 6.99(10)$ ms, are in excellent agreement with earlier measurements [15–18]. The range of the α particles is greater than the implantation depth of the recoils. As a result, the full energy of the α decay of the nucleus of interest, depending on the direction of the α particle, may not deposited in the DSSDs and they escape into the surroundings. These escaped α particles contribute to the continuum, which can be seen at energies 1000-5000 keV in Fig. 1. It is possible to recover some of these γ-recoil-α_P(escape) correlations by demanding further conditions on the daughter α decay. By including γ-recoil-α_P(escape)-α_D correlations, an increase in statistics of 43% has been gained compared with using only γ-recoil-α_P correlations, where α_P and α_D represent parent and daughter decays. This technique has been used in the analysis of both ^{168}Pt and ^{169}Pt. Fig. 1 shows all alpha decays detected within 7 ms of a recoil implantation in the same DSSD pixel from the ^{78}Kr + ^{92}Mo data. The inset to Fig. 1 shows alpha decays, within 7 ms of recoil implantation, which preceded the detection of a second generation ^{164}Os decay (E_α = 6321(7) keV, $T_{1/2}$ = 21(1) ms [18]). The fact that the only peak observed corresponds to the α-decay of ^{168}Pt indicates the reliability of this method. In the analysis of ^{169}Pt the daughter nucleus was ^{165}Os ($T_{1/2} = 71(3)$ ms [18]).

III. RESULTS

Approximately 25,000 correlated recoil-α(^{168}Pt) events were detected at the focal plane. The cross section for the production of ^{168}Pt via the ^{92}Mo(^{78}Kr,2n)

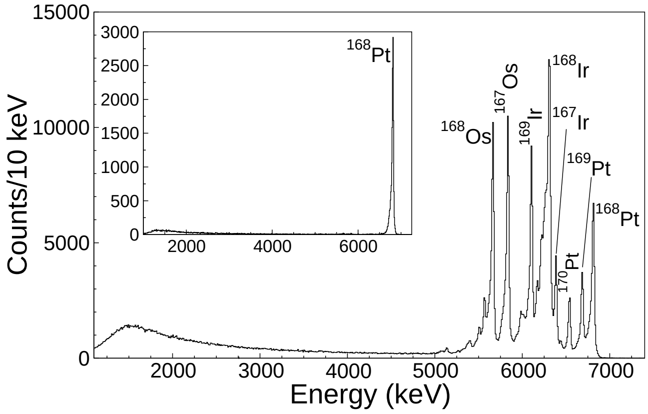

FIG. 1: Energy spectrum of α particles, detected in the DSSDs of the GREAT spectrometer, within 7 ms of a recoil implantation in the same pixel. These data were collected following the ^{78}Kr + ^{92}Mo reaction. The inset shows the energy spectrum of first generation α particles correlated with second generation ^{164}Os α decays.

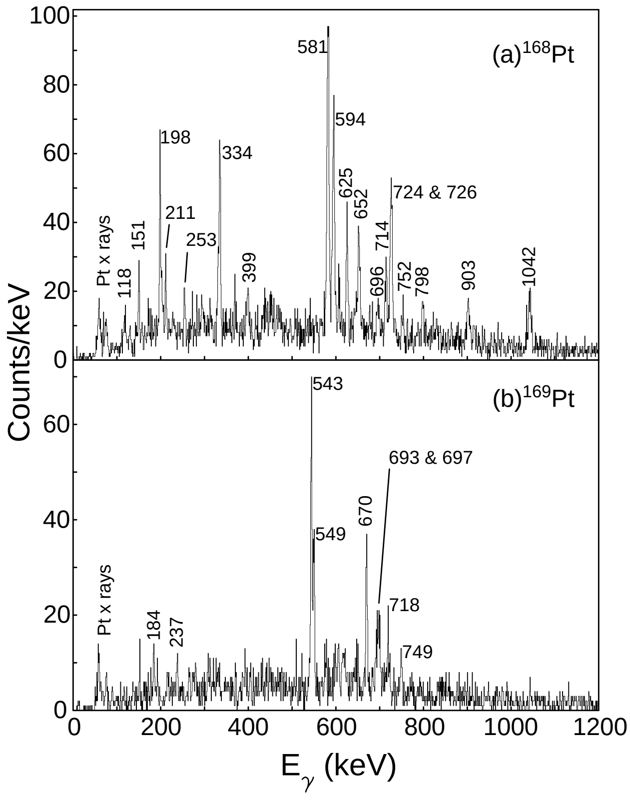

FIG. 2: Gamma-ray spectra detected at the target position with JUROGAM showing (a) γ rays correlated with the characteristic α decays of ^{168}Pt. The recoil-decay correlation time was limited to 7 ms; (b) γ rays correlated with the characteristic α decays of ^{169}Pt. The recoil-decay correlation time was limited to 25 ms

reaction at 335 MeV, assuming a transport efficiency of 35% for RITU, a recoil image coverage by the DSSDs of 70% and a 55% full-energy α detection efficiency for the DSSDs, is estimated to be $\sim$2 μb. Nineteen γ-ray transitions have been associated with the decay of ^{168}Pt as a result of the present study and are shown in Fig. 2(a). These include the 581, 594 and 726 keV transitions first identified by King *et al.* [7]. Table I lists the energies and relative intensities, as measured in singles analysis, of γ-ray transitions associated with the decay of excited states of ^{168}Pt. Gamma-ray coincidence spectra obtained from the summed γ-γ-recoil-α(^{168}Pt) and γ-γ-recoil-α(escape)-α(^{164}Os) correlations are shown in Fig. 3.

The most intense transition in ^{168}Pt at 581 keV is assumed to be the decay from the first excited state to the ground state and γ rays in coincidence with this transition are shown in Fig. 3(a). Several γ rays are clearly observed to be in coincidence with the 581 keV transition. Figure 3(b) shows γ rays in coincidence with the 726 keV transition. On the basis of this spectrum the 581, 724 and 726 keV are assumed to constitute the yrast sequence of ^{168}Pt. In an attempt to observe higher spin states in the yrast sequence, a summed spectrum of γ rays in coincidence with either the 581, 724 and 726 keV transitions is shown in Fig. 3(c). However, no additional transitions at higher spin could be established firmly. In addition to the yrast band, the 334 keV and 198 keV γ rays were established to be in coincidence with the 581 keV γ ray, see Fig. 3(d), forming a parallel decay sequence. The previously reported 594 keV γ-ray [7] feeds the first excited state, but is not in coincidence with the other transitions in the yrast sequence. The level scheme deduced from this analysis is shown in Fig. 5. The 581, 726 and 724 keV γ rays that form the yrast sequence are assumed to be of stretched E2 character, hence the band is tentatively observed to $I^\pi = 6^+$. Note that the γ-ray intensities deduced from the tagged singles spectrum for ^{168}Pt and presented in Table I suggest that the total intensity feeding the 2^+ state exceeds that of the 581 keV transition. This may indicate that one or more of the transitions feeding this state is a doublet, or that they may not all feed the 2^+ state directly and that part of their intensity then proceeds to the ground state by an alternative path. However, it was not possible on the basis of the present data to resolve this issue.

Approximately 12,500 correlated recoil-α(^{169}Pt) events were detected at the focal plane. The cross section for the population of ^{169}Pt via the ^{94}Mo(^{78}Kr,3n) reaction at 348 MeV is estimated to be $\sim$ 3 μb. A γ-ray singles spectrum showing all prompt γ rays correlated with the characteristic ^{169}Pt α decay is shown in Fig. 2(b). Table I lists the energies and relative intensities, as measured in singles analysis, of the nine transitions associated with the decay of excited states of ^{169}Pt. In a previous study of ^{169}Pt, Joss *et al.* [8] observed two transitions of 184 and 545 keV, both of which have been observed in the present work. Figure 4 shows typical γ-ray coincidence spectra correlated with the decay of ^{169}Pt. Figures 4(a) and 4(b) indicate that the 543 keV transition is fed directly by two separate states depopulated by the 693 keV and 670 keV transitions. In addition, coincidences with the 549 keV γ-ray, shown in Fig. 4(d), suggest a sequence of transitions in parallel with the yrast structure. The relative excitation energy of these two structures could not be determined in this work. It is possible that both the 543 and 549 keV γ rays directly populate the ground state. However, it is also possible that a low-energy γ-ray transition connects the two lowest energy states of these bands and that the sensitivity of the present experimental set-up was insufficient to observe such a decay. The resulting level scheme for the decay of ^{169}Pt is shown in Fig. 5.

TABLE I: Properties of γ rays associated with the decay of ^{168}Pt and ^{169}Pt as observed in the recoil decay tagged γ-ray singles analysis. Relative intensities I_γ are normalized with respect to the most intense transition in each nucleus. Values indicated with superscript d are the result of a measurement of a known doublet.

^{168}**Pt**	E_γ (keV)	I_γ(%)	^{169}**Pt**	E_γ (keV)	I_γ(%)
	118.2 ± 0.7	11 ± 2		184.3 ± 0.2	17 ± 3
	150.6 ± 0.4	11 ± 2		237.0 ± 0.3	13 ± 2
	197.9 ± 0.4	26 ± 3		542.8 ± 0.4	100 ± 19
	210.5 ± 0.3	6 ± 1		548.7 ± 0.8	50 ± 15
	252.8 ± 0.7	4 ± 2		669.6 ± 0.1	69 ± 11
	333.6 ± 0.9	51 ± 9		695.5^d ± 1.8	73 ± 12
	399.0 ± 0.3	5 ± 2		718.0 ± 1.3	12 ± 16
	581.4 ± 0.1	100 ± 7		748.5 ± 0.5	34 ± 10
	593.9 ± 0.2	76 ± 6			
	625.3 ± 0.4	25 ± 6			
	652.1 ± 0.2	35 ± 3			
	695.9 ± 0.3	10 ± 1			
	714.0 ± 0.3	25 ± 3			
	725.9^d ± 1.2	67 ± 5			
	752.1 ± 0.4	11 ± 2			
	798.2 ± 0.3	17 ± 2			
	902.9 ± 0.5	30 ± 3			
	1041.7 ± 0.7	52 ± 17			

IV. DISCUSSION

Previous works by King *et al.* [7] and McCutchan *et al.* [19] have compared experimental data for Pt isotopes with $N = 90 - 106$ and $N = 94 - 116$, respectively, with predictions of the interacting boson model (IBM). The calculations of King *et al.* [7] were the result of a modification of the earlier work of Harder *et al.* [20] in which the vibrational term of the IBM Hamiltonian was linearly increased with decreasing neutron number. It was found that the IBM calculations were successful in reproducing experimental observations. However, deviations between experiment and IBM predictions were reported in the case of ^{168}Pt [7]. The excitation energies of ^{168}Pt which were compared in Ref. [7] were tentatively determined

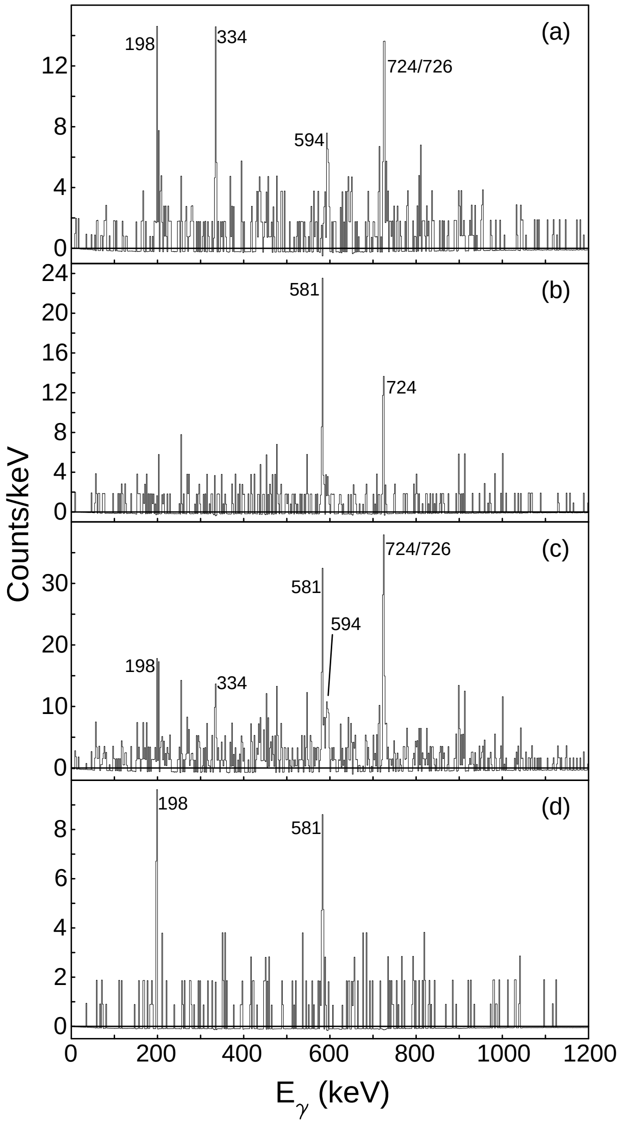

FIG. 3: Gamma-ray coincidence spectra correlated with the α-decay of ^{168}Pt. Individual panels show γ rays observed in coincidence with transitions of energy (a) 581 keV; (b) 726 keV; (c) sum of 581, 726 and 724 keV and (d) 334 keV.

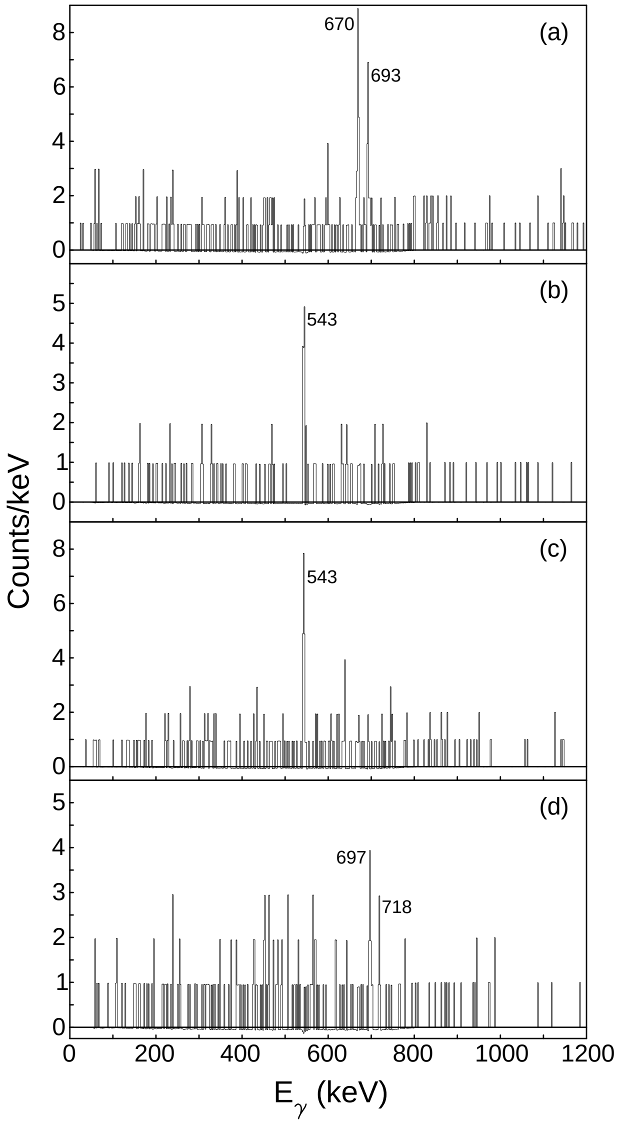

FIG. 4: Gamma-ray coincidence spectra correlated with the α-decay of ^{169}Pt. Individual panels show γ rays observed in coincidence with the following transitions (a) 543 keV; (b) 693 keV; (c) 670 keV and (d) 549 keV.

from the efficiency-corrected intensities from γ-ray singles spectra. In the present study, the γ-γ analysis has now established the level scheme of ^{168}Pt and permits a more meaningful comparison and the IBM level energies of Ref. [7] are reproduced in Fig. 5. Excellent agreement is observed with the IBM level energies calculated to be within 50 keV of experimental observations. Therefore, the modification of the IBM Hamiltonian by King *et al.* [7] is an appropriate choice to reproduce the structure of the neutron-deficient Pt isotopes down to $N = 90$.

The experimental ratio of the excitation energies of the first $I^\pi = 4^+$ and 2^+ states $E(4^+)/E(2^+)$ can reveal much about the underlying structure of a nucleus and highlights the evolution of this structure as a function of nucleon number. Such a plot is presented in Fig. 6 for the Pt isotopes with $A = 168 - 184$. The limiting values of $E(4^+)/E(2^+) = 2.5$ and 2.0 for the O(6) and U(5) symmetry groups of the IBM are also shown in Fig. 6. Macroscopically, the O(6) and U(5) symmetries are associated with triaxial γ-soft nuclei and spherical harmonic vibrators, respectively. The evolution of $E(4^+)/E(2^+)$, for the Pt nuclei, from O(6) to U(5) as neutron number is decreased is well described by the IBM calculations of McCutchan *et al.* [19]. The present work has established the excitation energies of the $I^\pi = 2^+$ and 4^+ states of ^{168}Pt for the first time. The resulting $E(4^+)/E(2^+)$ value of 2.25 continues the trend toward the U(5) limit, and indicates the increasing sphericity of the Pt nuclei, as the $N = 82$ spherical shell gap is approached.

An interesting trend in the excitation energies of yrast

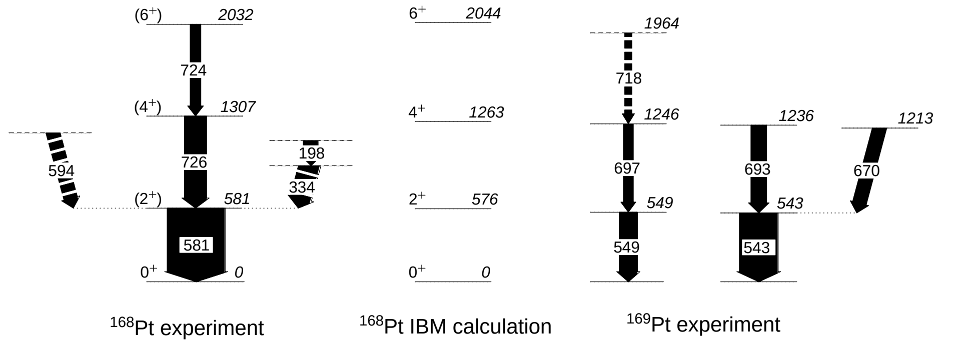

FIG. 5: Proposed level scheme for ^{168}Pt and ^{169}Pt as deduced from the present study. Arrow widths indicate the estimated relative intensity of transitions as observed from γ-γ coincidence spectra. In the case of ^{169}Pt, the excitation energies shown are relative to the lowest-energy state in each of the bands. Also shown, for comparison, is a ^{168}Pt level scheme calculated within the framework of the IBM and extracted from Ref. [7].

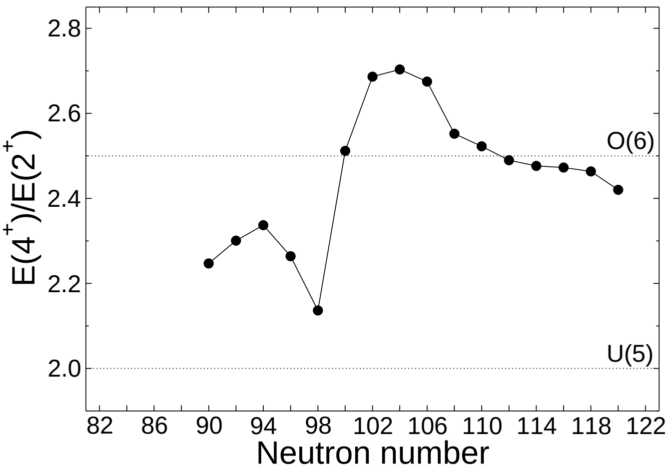

FIG. 6: The ratio E(4^+)/E(2^+) for the $^{168-198}$Pt isotopes. The horizontal dotted lines mark the limiting values of E(4^+)/E(2^+) for two symmetry groups of the IBM: O(6) (triaxial γ-soft) and U(5) (harmonic vibrator)

8^+ states of even-even $_{66}$Dy-$_{78}$Pt isotones has been uncovered and discussed in previous works such as Refs. [5, 8]. According to the trends identified, it is expected that the excitation energy of the 8^+ state in ^{168}Pt lies lower than that observed in ^{170}Pt (E(8^+) = 2437 keV). Since the energy of the $I^\pi = 6^+$ state has been established in the present work as being 2032 keV, it is reasonable to assume that the $8^+ \rightarrow 6^+$ transition energy in ^{168}Pt to be $\lesssim$ 350 keV. A number of candidate transitions that fulfil this criterion have been observed and can be seen in Fig. 2(a). However, due to the lack of statistics the decay of the 8^+ state could be not confirmed in the γ-γ analysis.

The $I^\pi = 8^+$ states of nuclei in this neutron-deficient region of the nuclear chart have been associated with the occupation of single-particle configurations involving the neutron $f_{7/2}$ and $h_{9/2}$ orbitals [5, 8, 21]. Assuming a similar origin for an 8^+ state in ^{168}Pt, a Weisskopf estimate for an E2 transition of $\sim$ 350 keV gives a half-life of the order of nanoseconds. Such a relatively long-lived γ-ray transition would make its observation in JUROGAM less probable due to the finite opening angle of the collimated germanium detectors causing reduced detection efficiency away from the target position. The opening angle limits the maximum half-life of an observable transition from a recoil traveling at $0.04c$ to be the order of 5 ns. This may also explain why it has not been possible to observe the yrast 8^+ state in the present study.

Long-lived 8^+ states have been observed in other nuclei in the region. For example, in the case of ^{152}Er$_{84}$, a half-life of 1.8 ns [22] has been reported for the 8^+ state while in ^{154}Yb$_{84}$ $T_{1/2}(8^+) = 28$ ns [23]. In both cases, the excitation energy of the yrast $I^\pi = 8^+$ state is lowered, relative to its position in the heavier isotopic neighbor, while the energy of the yrast $I^\pi = 6^+$ state continues to increase with decreasing neutron number. It is also worth noting that the estimated cross sections for ^{168}Pt and ^{169}Pt are comparable in this work. However, the α-tagged γ-ray statistics obtained for each of them are similar despite the fact that the beam time used to populate them was different, 162 and 48 hours, respectively. The presence of a relatively long-lived $I^\pi = 8^+$ state in ^{168}Pt could explain these observations since reduced feeding of the lower energy yrast states, via the long-lived state, would be observed. The fact that the observed population of the yrast states in the present work may be due mainly to side-feeding could also explain why in both this study and the similar work of King *et al.* [7] the side feeding transition at 594 keV is observed with an efficiency-corrected intensity comparable to that of the 726 keV (4^+) $\rightarrow$ (2^+) transition.

In the case of the odd-A Pt isotopes with $N = 93-97$, the yrast sequence is built upon a state which has been tentatively assigned spin and parity of $13/2^+$ [6, 8, 24, 25] and is associated with the unpaired neutron occupying an intruder $i_{13/2}$ orbital. In each case, the decay of this state is thought to be isomeric. Indeed, the decay of a $I^\pi = 13/2^+$ state via an M2 transition has been observed in the case of ^{171}Pt [26].

The use of the GREAT spectrometer in the present work should permit any isomeric decay in ^{169}Pt with a lifetime greater than the time-of-flight through the separator ($\sim 0.5\,\mu$s) to be identified. However, no delayed γ-ray transitions associated with ^{169}Pt have been observed.

It is possible that if an isomer exists it is sufficiently short to decay in-flight. Indeed, recent measurements of the $I^\pi = 13/2^+$ isomer in ^{171}Pt reveal a half life of 901 ns [26] and systematic trends established for neighboring nuclei suggest the half-life decreases with decreasing neutron number.

An alternative interpretation may be that, in contrast with the heavier odd-A isotopes, the excited states of ^{169}Pt are not based on an $i_{13/2}$ configuration since it is much higher in excitation energy relative to the Fermi surface. Instead, the observed band structures, presented in Fig. 5, may be interpreted as arising from single quasiparticle configurations based on the $\nu f_{7/2}$ or $\nu h_{9/2}$ orbitals. Based on the systematics of the region, the most likely ground state of ^{169}Pt is $7/2^-$. Nonetheless, the near-degeneracy of the states in ^{168}Pt and ^{169}Pt suggests that the latter consists of a neutron which is weakly coupled to the ^{168}Pt core. In addition, the similarity of the ^{169}Pt level scheme reported here and ^{170}Pt level scheme reported by Joss *et al.* [8] indicates that ^{169}Pt can also be treated as a neutron-hole decoupled from the ^{170}Pt core.

In summary, excited states in the nuclei ^{168}Pt and ^{169}Pt have been populated via fusion-evaporation reactions and their decay studied through recoil-decay tagged γ-γ coincidence measurements. In the case of ^{168}Pt, excited states up to $I^\pi = (6^+)$ are reported. The observed structure of this nucleus suggests a tendency towards vibrational behavior and this conclusion is supported by excellent agreement with previously reported IBM calculations. In addition, the non-observation of the yrast $I^\pi = 8^+$ state is discussed in terms of systematics and comparisons with neighboring nuclei.

Two distinct structures in ^{169}Pt have been observed each with level excitation energies similar to those of the yrast states in the neighboring ^{168}Pt and ^{170}Pt isotopes suggesting that the odd-neutron may be decoupled from the core rotation. The non-observation of γ rays from isomeric states has been discussed in terms of a possible configuration change from the $\nu i_{13/2}$ yrast configurations established in the heavier ($N > 92$) odd-A isotopes to configurations based on the $\nu f_{7/2}$ and/or $\nu h_{9/2}$ orbitals at greater neutron deficiency.

Acknowledgments

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