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Population of a low-spin positive-parity band from high-spin intruder states in $^{177}$Au: The two-state mixing effect

M. Venhart $^{a,*}$, M. Balogh $^{a}$, A. Herzáň $^{a}$, J.L. Wood $^{c}$, F.A. Ali $^{b,d}$, D.T. Joss $^{b}$, A.N. Andreyev $^{e,f}$, K. Auranen $^{g}$, R.J. Carroll $^{b}$, M.C. Drummond $^{b}$, J.L. Easton $^{b,1}$, P.T. Greenlees $^{g}$, T. Grahn $^{z}$, A. Gredley $^{b}$, J. Henderson $^{e}$, U. Jakobsson $^{g,1}$, R. Julin $^{g}$, S. Juutinen $^{g}$, J. Konki $^{g,2}$, E.A. Lawrie $^{h,1}$, M. Leino $^{g}$, V. Matoušek $^{a}$, C.G. McPeake $^{b}$, D. O’Donnell $^{b,3}$, R.D. Page $^{b}$, J. Pakarinen $^{g}$, P. Papadakis $^{b,4}$, J. Partanen $^{g,5}$, P. Peura $^{g,6}$, P. Rahkila $^{g}$, P. Ruotsalainen $^{g}$, M. Sandzelius $^{g}$, J. Sarén $^{b}$, B. Saygi $^{j,k}$, M. Sedláčk $^{a}$, C. Scholey $^{g}$, J. Sorri $^{g,7}$, S. Stolze $^{g,8}$, A. Thornthwaite $^{b}$, R. Urban $^{a}$, J. Uusitalo $^{g}$, M. Veselský $^{l}$, F.P. Wearing $^{b}$

$^{a}$ Institute of Physics, Slovak Academy of Sciences, SK-84511 Bratislava, Slovakia
$^{b}$ Oliver Lodge Laboratory, University of Liverpool, Liverpool, L69 7ZL, United Kingdom
$^{c}$ Department of Physics, Georgia Institute of Technology, Atlanta, GA 30312, USA
$^{d}$ Department of Physics, College of Science Education, University of Sulaimani, P.O. Box 334, Sulaimani, Kurdistan Region, Iraq
$^{e}$ Department of Physics, University of York, Heslington, York YO10 5DD, United Kingdom
$^{f}$ Advanced Science Research Center, Japan Atomic Energy Agency (JAEA), Tokai-mura, Naka-gun, Ibaraki, 319-1195, Japan
$^{g}$ University of Jyväskylä, Department of Physics, FI-40014 University of Jyväskylä, Finland
$^{h}$ Thenka Laboratory for Accelerator Based Sciences, P.O. Box 722, 7129 Somerset West, South Africa
$^{i}$ Department of Physics and Astronomy, University of the Western Cape, P.O.B X17, Bellville 7535, South Africa
$^{j}$ Department of Physics, Faculty of Science, Ege University, Izmir, 35100, Turkey
$^{k}$ Department of Physics, Faculty of Science and Arts, Sakarya University, Sakarya, 54187, Turkey
$^{l}$ Institute of Experimental and Applied Physics, Czech Technical University, Prague, Czech Republic

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The extremely neutron-deficient isotopes $^{177,179}$Au were studied by means of in-beam $\gamma$-ray spectroscopy. Specific tagging techniques, $\alpha$-decay tagging in $^{177}$Au and isomer tagging in $^{179}$Au, were used for these studies. Feeding of positive-parity, nearly spherical states, which are associated with $2d_{5/2}$ and $3s_{1/2}$ proton-hole configurations, from the $1i_{13/2}$ proton-intruder configuration was observed in $^{177}$Au. Such a decay path has no precedent in odd-Au isotopes and it is explained by the effect of mixing of wave functions of the initial state.

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In this Letter we present results on in-beam $\gamma$-ray spectroscopic studies of $^{177,179}$Au, with various tagging techniques [1,2]. Previously, both isotopes were studied by means of in-beam $\gamma$-ray spectroscopy using the Gammasphere spectrometer coupled to the Fragment Mass Analyser (FMA) at Argonne National Laboratory. In $^{179}$Au [3], four rotational bands associated with $1h_9/2$, $2f_7/2$, and $1i_{13/2}$ proton-intruder configurations were observed. Transitions connecting these structures to the ground state were not observed. In $^{177}$Au, only the yrast $1i_{13/2}$ band together with its decay pattern was reported in the original publication [4]. Later, a data evaluation was published [5], which also contained a rotational band based on the $9/2^-$ state, probably associated with the
$^{1}h_{9/2}$ proton-intruder configuration. However, no relevant spectra documenting this band were ever reported in any refereed journal.

The Au isotopes play a unique role in our understanding of shape coexistence in that strongly-deformed structures intrude to become the ground state at mid shell ($^{183}$Au) and to exhibit a classical “parabolic” trend in excitation energy. The isotopes $^{177,179}$Au play a key role in that they establish the “left-hand side” of the parabola of intruder states. As we show in the present study, a clear picture now emerges regarding which states are intruder structures. Further, there is the subtle feature of changing spins of intruder bands, namely that decoupling effects result in changing spin order in each intruder band. This is established as occurring in a systematic manner in the positive-parity ($^{1}h_{13/2}$) band for the first time.

A major step in understanding the structure of $^{179}$Au, and of odd-mass Au isotopes in general, was the discovery of a 326 ns isomer, with spin-parity 3/2$^-$ [6]. The discovery required a combined analysis of data acquired at the University of Jyväskylä and at the CERN-ISOLDE facility, with application of various techniques such as high-statistics $\gamma$-ray spectroscopy, $\alpha$-decay spectroscopy, $\alpha$-electron summing effects, including GEANT4 simulations of atomic relaxation processes, and mass measurements. The 326 ns isomer in $^{179}$Au de-excites by either a strong 62.4-271 keV cascade or via a weak 89.5 keV cross-over transition to the ground state. The decay pattern suggested a positive-parity, proton-hole configuration to be the ground state. The in-source laser spectroscopy experiment, performed recently at ISOLDE has assigned ground state spin-parities for both $^{177,179}$Au unambiguously as 1/2$^+$ [7], confirming the previous conclusion. Measured magnetic moments suggest mixed $3s_{1/2} \oplus 2d_{3/2}$ proton-hole configurations for these ground states, in agreement with findings reported in [6]. The band head of structures reported in [3] was proposed to de-excite to the 326 ns isomer. Connecting transitions were not observed, due to their low energy and thus strong internal conversion. However, the data provided indirect evidence for them, see a detailed discussion in [6].

The data presented here prove that intruder configurations in $^{179}$Au, identified in the study [3], decay exclusively via the 326 ns isomer. A new level scheme for $^{177}$Au is constructed, which significantly differs from that reported in [4]. A major difference between the decay of the 1$^{13/2}$ band in $^{177}$Au and $^{179}$Au is observed. The head of this band also feeds positive-parity structures, which is unprecedented in odd-mass Au isotopes. The $^{177}$Au isotope is a unique case, with mixing of coexisting strongly and weakly deformed configurations, caused by their proximity. This mixing opens the decay path, which is otherwise suppressed.

Two separate experiments were performed at the Accelerator Laboratory of the University of Jyväskylä. First, the $^{177}$Au nuclei were produced via the $^{92}$Mo($^{88}$Sr,p2n)$^{177}$Au fusion-evaporation reaction. The bombarding energy of the $^{88}$Sr$^{10+}$ beam was 399 MeV with an average intensity of approximately 2 particle nA. For production of $^{179}$Au, the $^{82}$Kr($^{108}$Ru,p2n)$^{179}$Au reaction was used. The energy of the $^{82}$Kr$^{15+}$ beam was 352 MeV with an average intensity of approximately 5 particle nA. In both experiments, self-supporting metallic targets of isotopically enriched materials were used. Heavy-ion beams were delivered to the target chamber by the K = 130 MeV cyclotron.

Prompt $\gamma$ radiation following reactions in the target was detected by the JUROGAM-II array, which consists of 24 clover- and 15 single-crystal EUROGAM-type Compton-suppressed germanium detectors. Reaction products were separated in-flight from the primary beam by the RTU gas-filled separator [8] according to their magnetic rigidities. At the focal plane of the separator, nuclei were implanted into double-sided silicon strip detectors (DSSD) of the detection system GREAT [9]. Prior to the implantation, nuclei passed through the multwire proportional counter (MWPC), which provides discrimination of recoiling evaporation residues from the scattered primary beam particles (using energy losses in the gas and time-of-flight measured between the MWPC and the DSSD) and radioactive decays. The data were analyzed using GRAIN [10] and RADWARE [11] software. Isomeric transitions were detected at the focal plane of the separator with a planar double-sided germanium strip detector and three Clover-germanium detectors.

Partial level schemes of both $^{177,179}$Au, constructed in the present work are presented in Fig. 1 and are discussed in detail and explained in the following text.

The relatively long half life of $^{179}$Au ($t_{1/2} = 7.1(3)\text{s}$ [12]) and high implantation rate into the DSSD prevented the use of the recoil-decay tagging technique for this isotope. Fortunately, the high production rate of the 326 ns isomer [6], which decays via low-energy electric-dipole (E1) transitions that can be detected with high efficiency, offers an excellent option for application of the recoil-isomer tagging technique. In the data analysis process, the recoil implantations that were followed by detection of the aforementioned isomeric $\gamma$ rays within the time window up to 1 $\mu$s, were selected. Prompt $\gamma$ rays, observed in the target position array, that preceded such implantations, were sorted into a $\gamma$-$\gamma$ matrix. Fig. 2a gives a projection of the matrix with the gate on the 353.8 keV transition, which is a known de-excitation of the 25/2$^+$ member of the yrast 1$^{13/2}$ band [3]. Except for other known in-band transitions of the yrast band, the unresolved 144.7-145.3 keV doublet together with the 220.3, 241.9, 349.9, and 370.5 keV transitions are evident. Energies of components of the doublet were determined using the “running” gate technique [13]. The 220.3 and 241.9 keV transitions are known de-excitations of the 11/2$^-$ and 13/2$^-$ members of the 2$f_{7/2}$ and 1$h_{9/2}$ proton-intruder configurations. The 144.7, and 145.3 keV transitions, as correctly assigned in the previous in-beam study [3], are de-excitations of the 13/2$^-$ state of the yrast band feeding nearly degenerate 11/2$^-$ and 13/2$^-$ states, see the level scheme in Fig. 1. In the study [3], the 349.9 and 370.5 keV transitions were assigned as de-excitations of 13/2$^-$ and 9/2$^-$ members of the 1$^{13/2}$ proton-intruder configuration feeding a floating state without spin assignment. Still the authors of the study [3] discuss the option that these $\gamma$ rays arise from a single initial state in the text.

Since the 349.9 and 370.5 keV transitions are observed in the spectrum tagged with the decay of the 326 ns isomer, they feed states associated with intruder structures above the isomer. In the previous study, no $\gamma$-ray detectors were employed at the focal plane of the FMA and thus this conclusion could not be made. The energy difference of the 349.9 and 370.5 keV transitions exactly matches the energy difference of known 9/2$^-$, and 7/2$^-$ band heads of 1$h_{9/2}$, and 2$f_{7/2}$ intruder configurations. This suggests that both transitions have a common initial state and feed these band heads, see the level scheme in Fig. 1.

Fig. 2b depicts a projection of the $\gamma$-$\gamma$ matrix tagged with the decay of the 326 ns isomer, with the gate on the 370.5 keV transition. The spectrum shows only known in-band transitions of the 1$^{13/2}$ yrast band. Therefore, the initial state of the 370.5 keV transition is a member of this configuration. An alternative assignment with, e.g., 1$h_{9/2}$, or the 2$f_{7/2}$ intruders, would require an observation of corresponding rotational bands in coincidence with the 370.5 keV transition. This is not observed in the data and therefore the initial state is interpreted as the 9/2$^-$ member of the 1$^{13/2}$ proton-intruder configuration. The 9/2$^-$ state is produced by the anti-aligned coupling of the 1$^{13/2}$ proton with the first-excited 2$^+$ state in the $^{178}$Pt core. Several such anti-aligned states are known in odd-mass Au isotopes, see [13-15] and references therein, although all of them are associated with the $1h_{9/2}$, or the 2$f_{7/2}$ intruder configuration. The 15.1 keV transition with presumably electric-quadrupole ($E2$) character, connecting the 13/2$^-$ with the 9/2$^+$ state was not observed due to strong internal conversion.
States associated with the $3s_{1/2} \oplus 2d_{3/2}$ proton-hole configuration were studied in heavier odd-mass Au isotopes and well-developed systematics were established [14–16]. The first excited state above the $1/2^+$ ground state is the $3/2^+$ state, which was observed in $^{179}$Au at 27.1 keV [6]. According to known systematics, a spin-parity of the next excited state is $5/2^-$. This state, which is expected at approximately 270 keV in $^{179}$Au, could be fed by the $E2$ de-excitation of the $9/2^+$ state of the $1h_{11/2}$ configuration. Such a decay branch would not proceed through the 326 ns isomeric state and therefore cannot be observed in the spectrum presented in Fig. 2a. To identify this decay path, prompt $\gamma$ rays preceding all recoil implantations, i.e., without requiring the isomeric decay to be detected, were analyzed. To suppress contaminations from isotopes produced via different evaporation channels of the reaction, the data were sorted into a triple-$\gamma$ coincidence cube. This results in significantly reduced statistics, compared with double coincidences, but the influence of contaminations in a double gate is negligible and thus cleaner spectra can be projected. Fig. 2c gives a projection of the cube with gates on the 262.1, and 353.8 keV transitions. A signature of the direct decay path into the ground state would be observation of two parallel transitions that differ by 27.1 keV, and of the $E2$ de-excitation of the $9/2^+$ state. Parallel transitions would be decays of the $5/2^+$ to the $1/2^+$ ground state, and to the $3/2^+$ 27.1 keV first-excited state. No such transitions appear in the spectrum depicted in Fig. 2c.

The isotope $^{177}$Au has two $\alpha$-decaying states with well-separated energies of emitted $\alpha$ particles ($E_\alpha = 6.12$ MeV and $t_{1/2} = 1.18$ s for the ground state and $E_\alpha = 6.16$ MeV and $t_{1/2} = 1.46$ s for the isomeric state [4]). In contrast to the $^{179}$Au data, influence of randomly correlated events was found to be negligible and the recoil-decay tagging technique could be applied. The prompt $\gamma$-ray data were sorted into two separate $\gamma-\gamma$ matrices, tagged with the two different $\alpha$ decays.

The isomer that emits 6.12 MeV $\alpha$ particles is assigned as the $11/2^-$ state of the $1h_{11/2}$ proton-hole configuration. This assignment is supported by the recent observation of a pattern of transitions feeding this state [17] and comparison with well established systematics [13–15]. The excitation energy of 189(16) keV was determined by the $\alpha-\gamma$ decay spectroscopy of $^{181}$Tl [18]. Note that the $1h_{11/2}$ proton-hole configuration is not involved in the decay of intruder configurations in $^{179}$Au, because known systematics [14] suggests that the intruder band-head is located below the $1h_{11/2}$.
Fig. 2. Spectra of γ rays of 197Au a) tagged with decay of the 326 ns isomer and in prompt coincidence with the 353.8 keV transition, b) tagged with decay of the 326 ns isomer and in prompt coincidence with the 370.5 keV transition, and c) in prompt coincidence with the both 353.8 and 262.7 keV (without tagging). Transitions of the yrast 11/2+ band are denoted with an asterisk.

Fig. 3a gives the spectrum of γ rays tagged with the 11/2− isomeric state α decay and in coincidence with the 257.5 keV transition of the known yrast band, which is associated with the 11/2− proton-intruder configuration, see previous in-beam γ-ray study [4]. In addition to the yrast band transitions the 241.2, 289.9, and 319.7 keV transitions are observed. The 241.2 keV transition is the known magnetic-dipole (M1) de-excitation of the 9/2+ band head of the 1h9/2 intruder configuration to the 11/2− of the 11/2− proton-hole configuration [18].

Fig. 3b gives a spectrum of γ rays tagged with the 11/2− isomeric state α decay and in coincidence with the 319.7 keV transition, which shows only the 241.2 keV transition together with the yrast band members. Therefore, the initial state of the 319.7 keV transition is interpreted as the 9/2+ member of the 11/2− proton-intruder configuration, on the basis of the same arguments as were used for the 370.5 keV transition in 179Au, see the above discussion.

In the data evaluation for 177Au [5], the 290.3 keV transition was reported. It was interpreted as the first in-band transition of the 1h9/2 band. The observed coincidence between the 290.3 keV and transitions of the 11/2− band, which was reported already in the original publication [4], was explained by an unobserved E1 feeding from the 13/2+ state of the yrast band. Presently, the 289.9 keV transition is interpreted as a feeding of the 7/2− band head of the so-far unknown 2f5/2 band from the 9/2+ state. This interpretation is based on the analogy with the decay pattern of the 9/2+ state in 179Au, see the level scheme in Fig. 1. However, E1 decays of the 13/2+, analogous to 144.7, and 145.3 keV transitions in 179Au, can exist. There is a weak peak at 261.2 keV observed in coincidence with the 257.5 keV transition, see Fig. 3a. The rotational band associated with the 2f5/2 configuration is not known, but according to known systematics of 1h9/2 and 2f5/2 bands [3,19,20], 11/2− and 13/2− members are expected to be nearly degenerate. Therefore, the 261.2 keV γ ray is interpreted as the 11/2− to 7/2− transition of the 2f5/2 band. The 289.9 keV peak is probably an unresolved doublet. In this case it is not possible to apply the “running” gate technique, since there are no characteristic coincidences for both transitions. However, this does not affect the understanding of the nuclear structure of 177Au. The E1 transitions feeding the 11/2− and the 13/2− state were not observed because of their low energy and thus low detection efficiency of the JUROGAM-II array.

Transition probabilities for possible decays of 13/2+ states of the yrast bands were investigated for both isotopes. In the present work, fast collective E2 transitions feeding 9/2+ band heads are proposed as dominant de-excitation paths, see the discussion above. In the 179Au isotope, branching ratios of 31.6% for the 144.7 keV, 22.6% for the 145.3 keV, and 45.8% for un-observed the 15.1 keV transition, were determined. The branching ratio for the un-observed transition was deduced as the sum of intensities of the 370.5, and 349.9 keV γ rays in the 353.8 keV coincidence gate, see Fig. 2a. The reduced transition probability of 100−300 W.u. is assumed for the 13/2+ to 9/2+ transition. This assumption is based on the known reduced transition probability of 200(120) W.u. [21] of the 9/2− to 5/2− de-excitation in 185Au. Using the above branching ratios, this yields the reduced transition probabilities of (1−5)×10−5 W.u. for both E1 transitions de-exciting the 13/2+ state in 179Au. The information on the 1 transition strengths is scanty in odd-mass Au isotopes. In 185Au, the 7/2− state of the 1h11/2 proton-hole configuration feeds two 5/2− states of the mixed 3S1/2 + 2d3/2 proton-hole configuration via E1 transitions. These de-excitation have reduced transition probabilities of 3.0+15×10−5 W.u. and 2.1+5×10−5 W.u., respectively [22]. Strengths of E1 transitions connecting two states with proton-intruder character are not known in odd-mass Au isotopes. In 175,179Au, isomeric E1 transitions with the intruder-to-hole nature were observed, however their reduced transition probabilities were measured to be 10−6−10−8 W.u. [6,23]. Therefore, the above values, estimated for transitions in 179Au, corroborate the interpretation with the intruder character of both of its initial and final states. Adopting these values of reduced transition probabilities for the 177Au isotope suggests that 5−10% of de-excitation of 13/2+ state should proceed via unobserved E1 transitions, feeding the 11/2− and 13/2− states of intruder bands. This is in agreement with the observation of the weak 261.2 keV transition in coincidence with the 257.5 keV transition, see Fig. 3a.

Fig. 4a gives the spectrum of γ rays tagged with the ground-state α decay and in prompt coincidence with the 257.5 keV transition which, as already noted, is a member of the 11/2− band. In addition to the yrast band members, 264.5, 290.2, and 452.6 keV transitions are observed. Fig. 4bc give spectra of γ rays tagged with the ground-state α decay and in prompt coincidence with the
The unambiguous 1/2+ ground-state spin-parity assignment [7] provides another supporting argument for an unobserved transition in the de-excitation path of the 13/2− state of the yrast band. This path contains only two transitions in a cascade, since the 290.2 and 264.5 keV transitions are parallel, see the level scheme in Fig. 1. The energy difference of parallel 264.5 and 290.2 keV transitions identifies the first excited state of the 179Au isotope at 25.7 keV.

The 177Au isotope is a unique case where the proton-intruder 11/2 yrast cascade splits at the bottom, feeding both the ground and the isomeric state (via the 1h9/2 intruder state). This is not the case with the 179Au isotope, see the discussion above and the level scheme in Fig. 1. The de-excitation probabilities of the bottom of the 11/2 intruder band were determined to be 69(13)% for the feeding the ground state, and 31(6)% for the feeding the α-decaying isomeric state, using the “inverse” tagging technique. The gate was set on the 160.2 keV transition detected at the target position, and the spectrum of corresponding α-particle energies was plotted. The intensities of 6.12 and 6.16 MeV peaks in the α spectrum (not shown here) gave corresponding de-excitation probabilities (after correction for known α-decay branching ratios, taken from [18]). Weak de-excitation paths through 13/2− and 11/2− states were neglected. Assuming the same reduced transition probabilities for E1 de-excitations as those estimated in 179Au, the B(E2) = 1− 20 W.u. is expected for the 9/2+ to 5/2+ transition. This is comparable with the reduced transition probability of 14.7 W.u. for the 5/2+ to 1/2+ E2 transition in 193Au [24], which is the nearest odd-mass Au isotope, where the strength of the E2 transition connecting positive-parity, proton-hole states is known.

The different de-excitation pattern observed in 177Au is explained by a configuration mixing. According to the systematics of the positive-parity bands in odd-mass Au isotopes, the 9/2+ state of the ground state band can be expected at a similar energy as in 187,189Au, i.e., between 700 and 760 keV [13–15]. Therefore, there are two 9/2+ states expected to be located close to each other in 177Au. One is the anti-aligned state of the 11/2− configuration, see the discussion above and one is the 9/2+ member of the ground-state band. Because these configurations are probably located close to each other, a strong mixing is expected. It is an intruder component in the wave function of both states that opens a decay path of the 13/2− member of the 11/2− configuration towards the ground state. Estimated unhindered nature of the E2 transition, see discussion above, corroborates such interpretation. Such a situation is not known not occur in any other odd-mass Au isotope, and therefore decays into proton-hole states were not observed. In 179Au, the effect is weaker, since both 9/2+ states are more separated in the energy. However, the non-observation of the 9/2+ to 5/2+ decay in 179Au can be explained by the parabolic pattern of the excitation energy of intruder configurations [6]. The energy of the 9/2+ to 5/2+ transition in 179Au can be expected to be approximately 215 keV. Since the E2 transition strength depends on the γ-ray energy in the fifth power, branching ratio of only approximately 2% for the feeding of the ground-state configuration can be expected. Such a weak transition cannot be observed in present experiment due to limited statistics.

The extensive systematics for intruder structures in odd-mass Au isotopes were established by means of in-beam γ-ray spectroscopy [3,19,20,26–29], and β-decay spectroscopy [13,14]. The states associated with the 11/2− band were observed to de-excite exclusively to states of the same configuration or into the 1h9/2 configuration via parity-changing E1 transitions. De-excitations into positive-parity states associated with the proton-hole configurations are strongly hindered due to the intruder-state-to-hole-state character of such transitions, or might be suppressed by the energy factor, as it is in 179Au. Another example of such hindrance is the observation of retarded M1 hole-state-to-intruder transitions with reduced transition probabilities of approximately 5 × 10−5 W.u. in 185,187,189Au [14], or the E3 isomerism in odd-Tl isotopes, see [18], and references therein.

The present work provides a key advance in understanding spin order associated with intruder bands. Thus, the 11/2− band exhibits the appearance of a spin{(j−2)} state below the spin-j state, similar to the 1h9/2 and 2f7/2 bands. Establishing this is critical for the organization of the decays of high-spin states. In this mass region, high-spin in-beam studies play a major role because it is very difficult to populate low-spin states: the standard approach via β decay is limited by competing α-decay channels. At the next level of study, the present work paves the road to adding the high level of detail expected in 181,183Au by comparison with 185,187Au. We note in particular that the strongly-coupled band observed in 177Au, reported earlier [17], indicates that there are structural changes occurring in the Au isotopes which point to multiple shape coexistence, even lying beyond such structures established in 187Au. Recent measurement [30] of lifetimes of excited states of the rotational band associated with the intruder 0+ configuration in 178Hg corroborates such interpretation.
Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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