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Structure of neutron rich palladium isotopes produced in heavy ion induced fission

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Abstract. New band structures have been observed in ^{112}Pd, ^{113}Pd, ^{115}Pd and ^{118}Pd produced via the fusion-fission reaction ^{12}C+^{238}U at a bombarding energy of 90 MeV. $\gamma-\gamma-\gamma$ coincidence measurements have been performed with the Euroball III array. The first crossings observed in even-mass palladium up to A=118 are interpreted as the alignment of a $\nu h_{11/2}$ pair. This interpretation is supported by the rotational behavior of odd-mass palladium. Hartree-Fock-Bogoliubov calculations using D1S-Gogny force suggest a similar interpretation consistent with a prolate deformation.

PACS. 25.70.Jj Fusion and fusion-fission reaction – 23.20.Lv Gamma transitions and level energies – 21.60.Ev Collective models – 21.60.Jz Hartree-Fock – 27.60.+j 90 ≤ A ≤ 149

1 Introduction

Fission studies are known to provide neutron-rich nuclei in various states of excitation energy, spin and deformation. Very neutron-rich nuclei have been studied recently from spontaneous fission of ^{248}Cm and ^{252}Cf [1,2] with the new generation of large germanium arrays [3]. For example, octupole deformation [4] and K-isomerism [5] have been studied in the heaviest nuclei produced (A∼140). The nuclei near the shell Z=50 and N=82 are more spherical in shape and exhibit numerous isomeric states due to the high angular momentum of these subshells. For lighter fragments around A=100, shape coexistences and shape transitions have also been observed [6,7]. For neutron-rich palladium isotopes, a prolate-to-oblate shape transition has been predicted to occur at ^{111}Pd by Moller *et al.* [8,9]. Experimental studies in the lighter neutron-rich palladium isotopes (up to ^{108}Pd), produced via fusion-evaporation reactions, propose a prolate deformation in a neutron $h_{11/2}$ configuration [10]. While, the ground bands in the heavier nuclei ^{112,114,116}Pd are interpreted to be the result of a quasi-proton $(g_{9/2})^2$ alignment and an oblate deformation is proposed [11]. In contrast, from more recent experimental results, odd and even palladium isotopes, up to A = 112, have been interpreted as a neutron $h_{11/2}$ configuration driving the nucleus towards a prolate shape [12]. The search of the expected and debated shape transition in more neutron-rich odd and even palladium nuclei will provide an important test for theoretical predictions.

In the present work, the odd mass systematics are extended by new rotational bands in ^{113}Pd and ^{115}Pd, while for the even mass isotopes a partial level scheme for ^{118}Pd and new transitions in ^{112}Pd are proposed.

2 Experiment

The spontaneous fission of commonly used sources (eg ^{252}Cf or ^{248}Cm) gives access to two separate mass regions, whereas induced fission reactions exhibit a strong symmetrical fission component. In this work, palladium isotopes were abundantly produced by the fusion-fission reaction ^{12}C + ^{238}U. To minimize pre-fission neutron emission, the excitation energy of the compound nucleus ^{250}Cf was kept as low as possible, while still maintaining a large cross-section. A ^{12}C beam with an energy of 90 MeV was used, leading to an excitation energy of 72 MeV in the compound nucleus. At this energy, a fission cross-section of 970(110) mb has been measured [13]. The ^{12}C beam was delivered by the tandem of the INFN (Legnaro, Italy) with an intensity of ∼0.4 pnA. A thick target (47 mg/cm^2) was used in order to stop the fission fragments thus avoiding Doppler effect. The γ-rays emitted by the fragments were detected in the Euroball III array [14] which consists of (i) 15 cluster germanium detectors placed in the backward hemisphere (ii) 26 clover germanium detectors located in 2 rings around 90^o and (iii) 30 tapered single-crystal germanium detectors located at forward angles. Each germanium detector is surrounded by its own BGO Compton suppression shield. A cluster detector consists of seven encapsulated Ge crystals and a clover detector consists of four Ge crystals. In the experiment, about 1.9×10^9 coincidence events with a γ multiplicity greater or equal to three were obtained after about 60 hours of data acquisition. The very complex γ-ray spectra produced by the multitude of fission fragments have been unravelled by triple coincidence studies, using a symmetrized Radware cube [15].

3 Experimental results

The neutron-rich palladium isotopes with odd mass exhibit a $\nu h_{11/2}$ isomeric state [16]. Hence, yrast states in odd-mass palladium are built on these high-spin isomeric states. Before our studies, no transitions were known above the isomeric states in ^{113}Pd and ^{115}Pd. The lifetimes of these isomeric states are too long (> 300 ms) to allow a coincidence measurement with delayed γ-rays from the decay of the isomer, since the coincidence time window in our experiment was restricted to 1 μs. A coincidence analysis could therefore not be performed between transitions feeding these isomeric states and known transitions depopulating them. The analysis technique to assign γ-rays to a certain palladium isotope takes instead into account two characteristics of the fission process:

(i) The probability of charged particle evaporation is weak in this reaction. It has been checked that the sum of the fission fragment atomic numbers is equal to the proton number of the fissioning nucleus: for example, palladium isotopes are produced together with tellurium isotopes.

(ii) For a given palladium isotope a mass distribution of tellurium isotopes is observed, due to different numbers of emitted neutrons, either before fission, either after fission [17]. A total of ten neutrons are evaporated per fission events; we have observed, for example, that ^{108}Ru is mainly produced with ^{132}Xe.

Figure 1 illustrates these two points. The spectrum 1.a has been obtained by gating on the first two transitions $2^+\rightarrow0^+$ (743 keV) and $4^+\rightarrow2^+$ (754 keV) of ^{128}Te. Besides γ-rays of ^{128}Te feeding these two states, the other transitions can be associated with the complementary palladium isotopes. Gating, in addition to the 743 keV line of ^{128}Te, on a transition assigned to a certain palladium isotope (for example the 383 keV transition), the spectrum exhibits only γ-rays from ^{128}Te and from the chosen palladium isotope (Fig. 1b). Finally, a spectrum obtained by double gating on a particular palladium isotope shows γ-rays emitted from that palladium isotope, and γ-rays from all tellurium isotopes produced (Fig. 1c). With

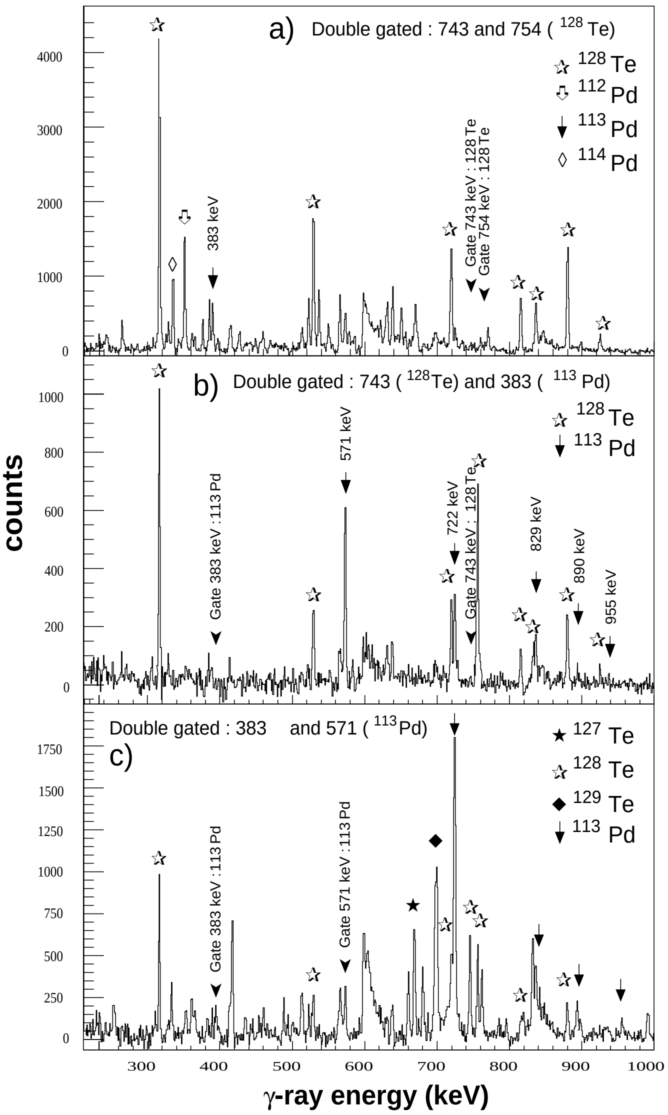

Fig. 1. Spectra obtained from double gates on the (a) the most intense transitions in ^{128}Te, (b) first transition in ^{128}Te and first transition in ^{113}Pd, (c) most intense transitions in ^{113}Pd

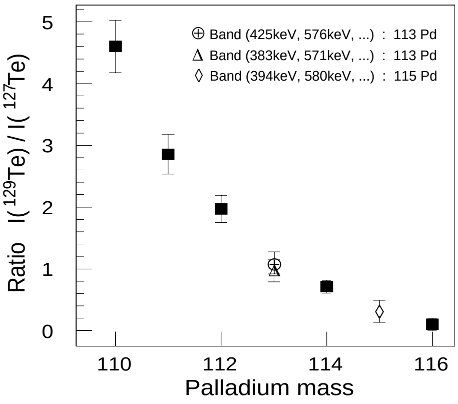

Fig. 2. Intensity ratios for transitions in ^{129}Te and ^{127}Te obtained from Pd double gated spectra. Ratios obtained for proposed new bands are displayed by open symbols

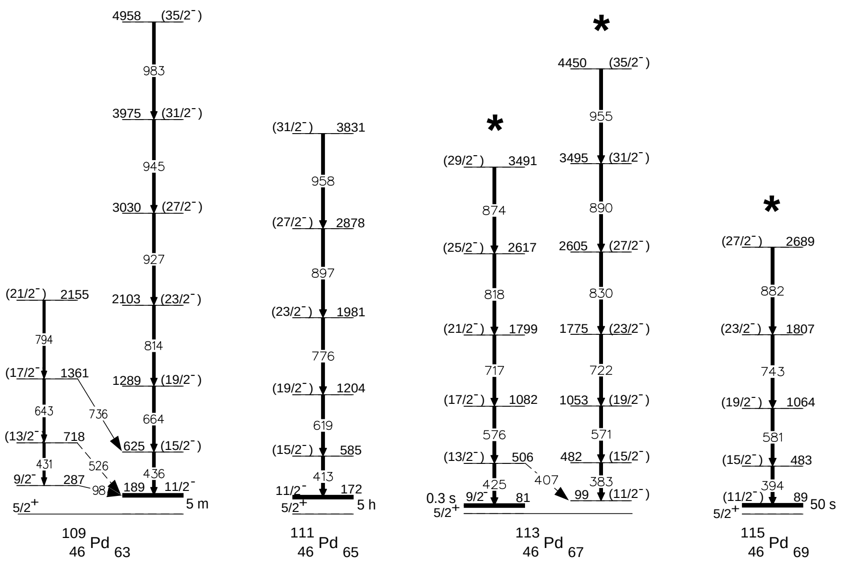

Fig. 3. Rotational bands observed in the present work for odd-mass palladiums ^{109}Pd to ^{115}Pd. Isomeric states are drawn by a thicker line. New proposed bands are built on isomeric states of ^{113}Pd and ^{115}Pd (labelled with stars)

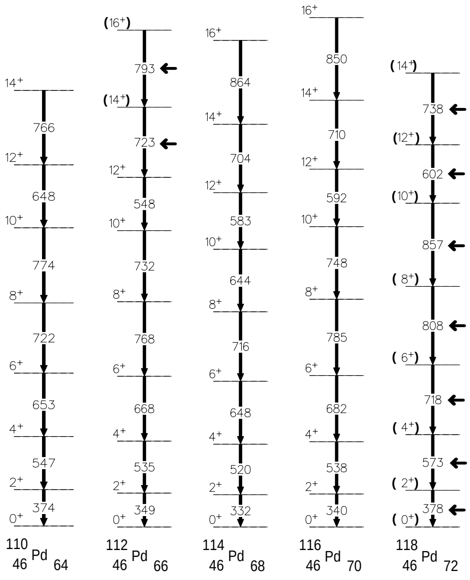

Fig. 4. Partial level schemes of even-mass palladium isotopes as observed in this experiment. New transitions are labelled by an arrow.

this method, four new γ-ray cascades have been identified in neutron-rich palladium isotopes. The mass assignment is based on the second fission characteristics presented above. The tellurium mass distribution produced with a given palladium isotope is a function of the palladium mass. The intensity ratios of γ-ray transitions in ^{127}Te and ^{129}Te obtained from different Pd-gated spectra are shown in Fig. 2. As expected, the intensity ratio of ^{129}Te to ^{127}Te becomes smaller for more neutron rich palladium isotope due to the reduced probability of neutron emission. The ratios obtained suggest that two bands are associated with ^{113}Pd and one with ^{115}Pd. The fourth new band has been assigned to ^{118}Pd because it is mainly in coincidence with ^{124}Te. Transitions up to the 6^+ state of ^{118}Pd have been recently confirmed by a β-decay experiment [18].

The partial level schemes of odd-mass palladium isotopes, observed in this work, are presented in Fig. 3. Bands built on the isomers in ^{109}Pd and ^{111}Pd have been recently proposed [12]. Two bands have been observed in ^{109}Pd with a intensity ratio of five in favour of the band built on the $11/2^-$ state, located 98 keV below the $9/2^-$ bandhead. In a similar way, two bands are proposed for ^{113}Pd with the $9/2^-$ state located 18 keV below the $11/2^-$ state (because of the tentative 407 keV transition connecting the $13/2^-$ state to the $11/2^-$). For ^{113}Pd, the band built on $11/2^-$ is 3 times more intense than the band built on $9/2^-$ state. No unfavored bands in ^{111}Pd have been observed, probably due to pollution by tellurium transitions. Transitions above isomeric state $(11/2^-)$ in ^{117}Pd and unfavored band in ^{115}Pd have not been seen due to too weak intensities.

The even-mass palladium isotopes observed in this work are shown in Fig. 4. They have been previously studied up to A = 116 [11] [19] [20]. The most neutron-rich palladium isotope studied from our data set is ^{118}Pd. Seven γ transitions in mutual coincidence have been identified. They are proposed to form the band built on the ground state.
Several excited bands have been also observed in ^{110}Pd, ^{112}Pd, ^{114}Pd and ^{116}Pd. The level scheme of ^{112}Pd is presented in Fig. 5, as an example. The spin assignments are tentative and based on a comparison with other even mass palladium isotopes. New (negative and positive parity) bands are displayed as well as two yrast transitions beyond the 12^+ state (Fig. 5).

The identification method presented above requires a good knowledge of transitions in tellurium isotopes. In particular, the level scheme of ^{130}Te has been expanded with a complementary experiment using a fission fragment array to detect the fission products and a new

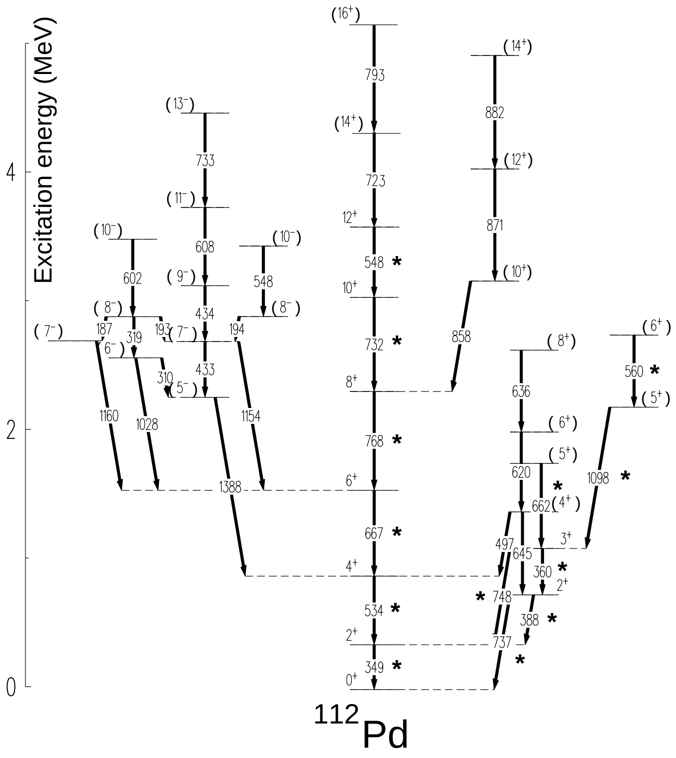

Fig. 5. Partial level scheme of ^{112}Pd. The already known transitions are labelled by a star

isomeric state is proposed at an energy of 4376 keV with a lifetime of 260(33) ns [21].

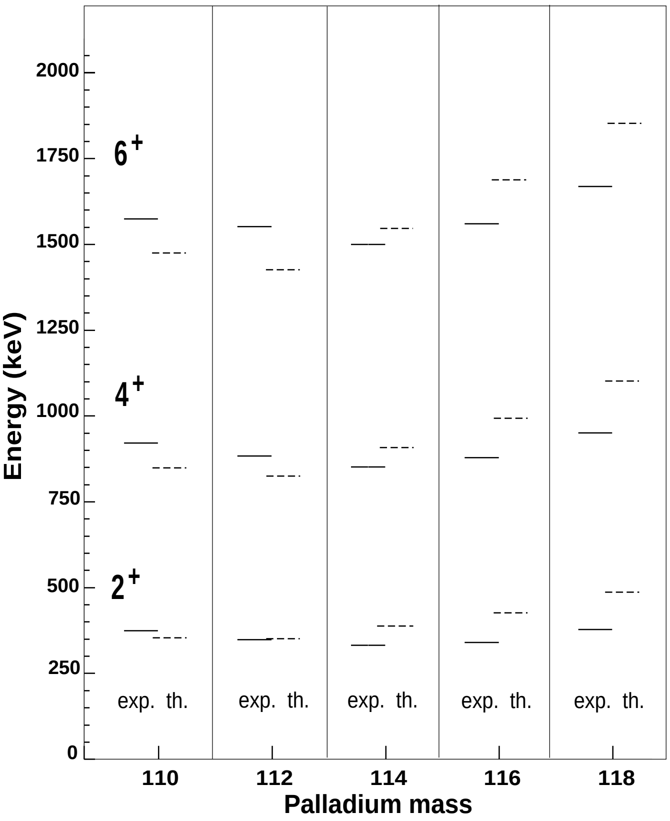

Fig. 6. The experimental energy levels for even mass palladium chain are compared with those obtained by solving collective Hamiltonian using HFB wave function

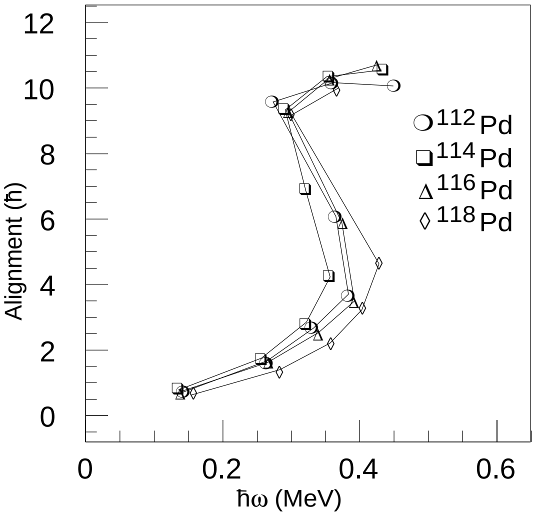

Fig. 7. Alignments calculated for even-mass palladium isotopes observed in this work using the Harris parameters $J_o=3\hbar^2/\mathrm{MeV}$ and $J_1=45\hbar^4/\mathrm{MeV}^3$

4 Discussion

4.1 Even mass systematic

In Fig. 6 the experimental energy levels are compared with those obtained from a GCM-GOA (Generator Coordinate Method with a Gaussian Overlap Approximation) calculation using HFB (Hartree-Fock-Bogoliubov) wave function and the D1S Gogny force as input [22] [23]. The agreement between theoretical and experimental level schemes is quite good.

In order to interpret the rotational band structures, the angular momentum alignment was studied as a function of the rotational frequency. The aligned angular momentum i_x is defined by [24]:

$$i_x = I_x - I_{ref},$$

$$with \qquad I_x = \sqrt{I(I+1) - K^2}$$

$$and \qquad I_{ref} = \omega(J_o + \omega^2 J_1).$$

I_x is the projection of the total angular momentum on the rotation axis and I_{ref}, the reference angular momentum, is parametrized using the two Harris parameters J_o and J_1 [25].Their values are chosen to give a mean constant value of i_x after the first band crossing for all the isotopes. This prescription leads to J_o=$3\hbar^2$/MeV and J_1=$45\hbar^4$/MeV^3. The alignments for even-mass palladium isotopes A=112 to 118 are summarized in Fig. 7. All curves display a back-

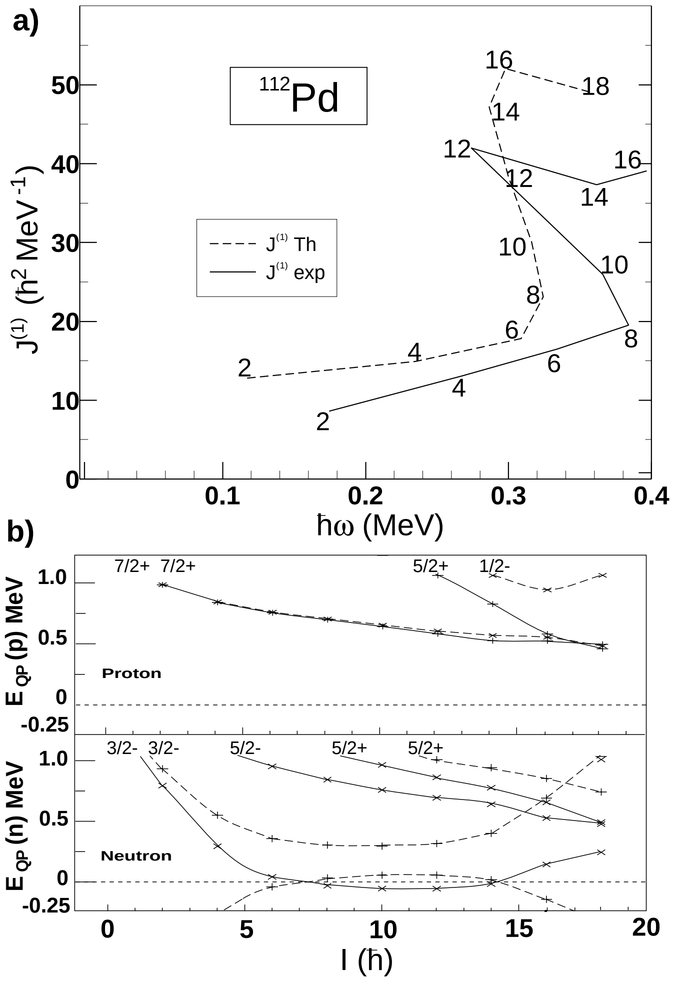

Fig. 8. Cranked Hartree-Fock-Bogoliubov calculations for ^{112}Pd. a) A comparison between theoretical and experimental moments of inertia as a function of rotational frequency; b) quasi-proton (top) and quasi-neutron (bottom) energies are displayed against angular momentum of nucleus excited states. The parity and the spin projection on the symmetry axis of the nucleus are given for the quasi-particle ground states

bending at a rotational frequency of $\hbar\omega \approx 0.38$ MeV with a gain in alignment of about $10\hbar$.

To determine whether the proton orbital ($\pi g_{9/2}$) or neutron orbital ($\nu h_{11/2}$) is responsible for the band crossings, cranked-HFB calculations have been performed [26]. In Fig. 8a the experimental and theoretical $J^{(1)}$ moment of inertia for the ground state band of ^{112}Pd are displayed as a function of the rotational frequency. The theory and the experiment results are in good agreement and both show a backbend occurring at I $= 8\hbar$ and $\hbar\omega = 0.33$-0.38 MeV. The calculated quasi-particle energies are shown as a function of the angular momentum for both quasi-protons and quasi-neutrons in Fig. 8b. The calculations predict that a neutron state from $h_{11/2}$ subshell becomes yrast at $I = 8\hbar$. Below $I = 18\hbar$, no band crossing caused by the alignment of two $g_{9/2}$ protons is expected.

The smooth evolution of low energy states with mass number (Fig. 6), the gain in angular momentum which is too large to be caused by the alignment of a pair of $g_{9/2}$ protons (Fig. 7) and the cranked-HFB calculations performed for ^{112}Pd (Fig. 8) suggest all that the observed band crossings in even-mass palladium isotopes from A=110 to A=118 are caused by a breaking of $h_{11/2}$ neutron pair.

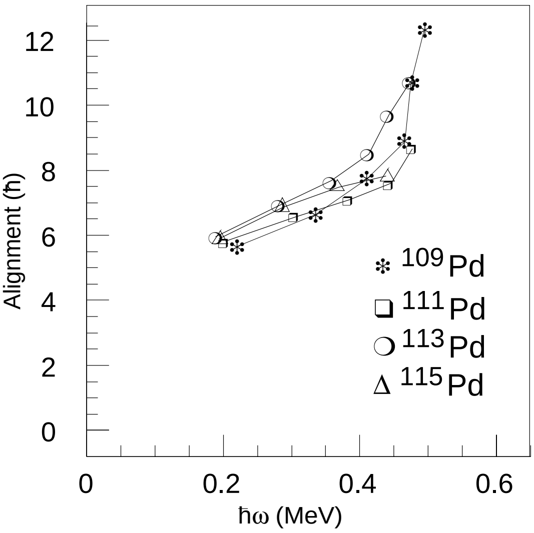

Fig. 9. Calculated alignment as a function of rotational frequency for bands built on the $11/2^-$ states in odd-mass palladiums. Harris parameters are the same as those used for even mass systematic

4.2 Odd mass systematic

The alignment plots for ^{109,111,113,115}Pd are displayed in Fig. 9. All bands exhibit a similar behaviour. The maximum angular momentum reached in this experiment does not allow to deduce the total alignment of the first non blocked pair of quasi-particles, but shows that the gain in angular momentum is greater than 6 $\hbar$. The crossing frequency occurs at 0.45 MeV which is higher than the crossing frequency for even nuclei (Fig. 7). This delayed alignment is due to the blocking of the lowest $h_{11/2}$ quasi-neutron. This blocking argument supports the interpretation of the backbendings in the neighboring even-mass palladium as due to a $\nu h_{11/2}$ pair.

A HFB calculation using the blocking method has been performed in order to study the deformation changes in neutron-rich palladium isotopes [5]. The results of this calculation are displayed in Fig. 10. The oblate-deformation states are plotted relatively to states with prolate deformation from the subshell $h_{11/2}$. The calculation predicts that the ground states of all palladium isotopes from A=109 to A=123 have a prolate shape. The deformation parameter (β) decreases with increasing mass, leading to a quasi-spherical shape for ^{121}Pd and ^{123}Pd. No prolate-to-oblate shape transition is predicted by our calculation but, instead a smooth trend towards spherical shape. An

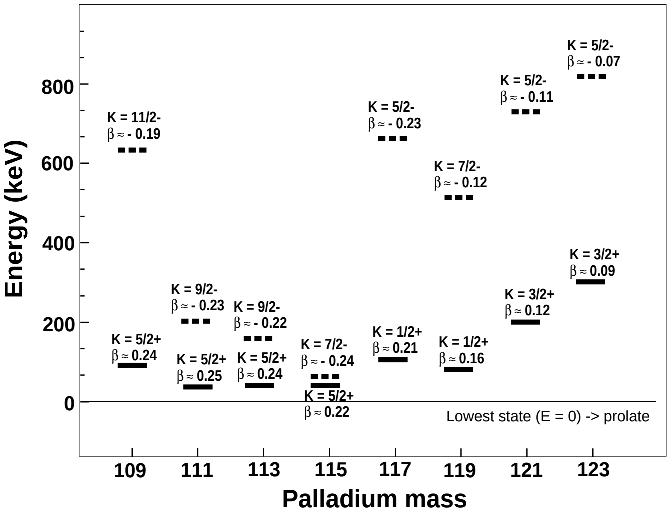

Fig. 10. Relative position of first oblate state (dashed lines) with respect to the first prolate state from the subshell $h_{11/2}$ (solid lines), using a HFB calculation in the blocking formalism. For each nucleus, the lowest state has a prolate deformation

oblate deformed state is however predicted in the isotopes ^{111,113,115}Pd at low excitation energies of 150 keV, 90 keV and 10 keV respectively.

5 Conclusion

The structure of the neutron-rich palladium isotopes with masses between 109 and 118 has been investigated via the fusion-fission reaction ^{12}C + ^{238}U. The study of prompt γ-rays emitted was enabled by the high efficiency and resolving power of the γ-array EUROBALL III. New rotational bands built on the $\nu h_{11/2}$ orbital have been identified in ^{113}Pd and ^{115}Pd. New γ-rays have been assigned to ^{112}Pd and ^{118}Pd. The systematics presented in this work do not display any experimental evidence for a prolate-to-oblate shape transition as suggested by Möller et al. [9]. The study of rotational bands in even-mass palladium as well as in odd-mass palladium support a quasi-neutron $h_{11/2}$ character of the first crossings in the even-mass palladium, up to A = 118. Moreover, new HFB calculations are consistent with an interpretation in terms of $\nu h_{11/2}$ configurations leading to a prolate deformation and no shape transition.

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