Band crossing observed in neutron-rich Pd isotopes via spontaneous fission of ^{252}Cf

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High spin excited states in neutron-rich nuclei ^{112,114,116}Pd have been investigated by measuring prompt γ rays emitted from a ^{252}Cf spontaneous fission source. Two different measurements were performed. First, γ-γ coincidence data, necessary to determine the decay schemes, were obtained by using an array of 20 Compton-suppressed Ge detectors. Second, Z identification and the enhancement of a desired neutron channel were carried out using a x-n-γ multiplicity spectrometer. Ground state bands have been extended to $J^\pi=10^+$ for ^{112}Pd, and $J^\pi=12^+$ for ^{114}Pd and ^{116}Pd. Band crossings were observed in all three isotopes. It was found from cranked shell model calculations that these backbends are due to the alignment of two $g_{9/2}$ protons. The measured energy levels were also compared with predictions of the interacting boson model.

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I. INTRODUCTION

Spontaneous fission and induced fission are so far the only means used to populate the excited states in very neutron-rich nuclei in the region of mass 90–160. The low spin states in many of these fission fragments have been investigated previously in beta decay studies by numerous authors and in prompt γ-ray deexcitations using spontaneous fissile materials, mainly ^{252}Cf (Ref. [1–9]). Recently, because of the existence of large and high resolution Ge detectors in very large solid angle array configurations, there has been a renewed interest in the study of the nuclear structure of neutron-rich nuclei at higher spins.

These fission-product nuclei exhibit a wide range of behaviors, from transitional to well deformed structures, and are amenable to interpretation by the spherical shell model and various collective and deformed models, including the interacting boson approximation (IBA). Thus, they can serve as useful tests for comparisons of the different models. The behavior of these nuclei at higher spins is an important new testing ground for some nuclear models.

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Recently, an Argonne-Manchester Collaboration has studied many of these nuclei, including both even-even and odd mass, especially for the Sr, Y, Zr, Nb, Mo, Ba, and Ce isotopes [10–14] via the spontaneous fission of ^{252}Cf and ^{248}Cm sources. Excited states of Sr and Zr isotopes were observed up to spin $10\hbar$ with a very large ground state deformation of $\beta_2 \sim 0.4$ and moments of inertia very close to rigid body values. For the first time, octupole deformation bands were observed in ^{144}Ba, ^{146}Ba, and ^{146}Ce isotopes, and in the case of ^{148}Ce and high spin states were extended to 14^+.

We have studied the nuclear structure of neutron-rich nuclei from the prompt γ ray emitted from spontaneous fission of ^{252}Cf and ^{242}Pu. From the analysis of coincidence data, excited states in ^{136}Te were identified [15] and high spin states were observed in several even-even light and heavy fragments such as $^{108-112}$Ru, $^{112-114}$Pd, $^{138-142}$Xe, ^{134}Te, and $^{152-154}$Nd. In most cases, bands have been extended to much higher spins than previously known (up to 12^+). In the case of ^{148}Ce, we have reported on the identification of a new γ ray deexciting from the 16^+ level, the highest spin seen in this high neutron-rich region [16,17].

In this paper we report on the observation of new excited states and also band crossings in three ^{112}Pd, ^{114}Pd, and ^{116}Pd isotopes. Information about low-lying levels in ^{114}Pd and ^{116}Pd has been reported previously from spontaneous fission of ^{252}Cf (Ref. [1]). Also, levels in the ^{112}Pd isotope were identified [18] via the ^{110}Pd$(t,p)^{112}$Pd reaction. Recently, Äystö *et al.* [19] have used the ^{238}U$(p,$F$)$ fission reaction to investigate the energy levels of even-

TABLE I. Experimental yields of some ^{252}Cf fission fragments as compared with predictions of Wahl *et al.* [20]. Both data are normalized to the yield of ^{148}Ce.

Fragment	Experiment (%)	Theory (%)
^{94}Sr	31	25
^{96}Sr	25	33
^{98}Sr	25	10
^{102}Zr	55	61
^{104}Zr	28	8
^{102}Zr	134	143
^{104}Mo	118	147
^{106}Mo	23	41
^{112}Ru	89	82
^{114}Pd	37	37
^{116}Pd	40	14
^{118}Cd	19	9
^{120}Cd	26	32
^{136}Xe	126	111
^{138}Xe	103	119
^{142}Ba	40	45
^{146}Ba	58	48
^{148}Ce	≡100	≡100

even $^{110-116}$Pd nuclei via β decay of odd-odd $^{110-116}$Rh isotopes using an on-line isotope separator. In these previous experiments, the levels up to spin $J^{\pi}=6^+$ have been reported in these isotopes. We, on the other hand, were able to extend the excited state of ^{112}Pd, ^{114}Pd, and ^{116}Pd to 10^+, 12^+, and 12^+, respectively. In the present work, we will discuss the results in terms of both the interacting boson approximation (IBA) and the cranked shell model calculations (CSM). The interpretation of our data in terms of band crossings compares favorably with theoretical predictions of CSM calculations.

II. EXPERIMENTAL METHODS

A spontaneous fission source of ^{252}Cf (0.1 μg) having a strength of about 6×10^4 fissions/sec and with a 250 μm Be window was used for this study. This source was placed in the center of the 20 Compton-suppressed Ge-detector compact ball at the Holifield Heavy Ion Research Facility at Oak Ridge National Laboratory. Approximately 2×10^9 γ-γ coincidences were collected during a five day run. Both double- and triple-coincidence gates were sorted in the data analysis. Energy and efficiency calibrations were performed by using a National Bureau of Standards mixed ^{125}Sb, ^{154}Eu, and ^{155}Eu source. The total coincidence γ-ray spectrum observed was very complex because of several fission fragments having the same γ-ray transition energies. The $2^+\rightarrow0^+$ ground state transitions in even-even fission fragments are estimated to contain $>95\%$ of the independent yields of these isotopes. Therefore, their relative yields can be deduced from the γ-ray intensities of the $2^+\rightarrow0^+$ transitions. Table I shows the experimental yields of selected ^{252}Cf fission fragments compared with calculated values by Wahl *et al.* [20]. Both data are normalized to 100% for ^{148}Ce. With the exception of a few cases, very good agreement is obtained with the theoretical calculations.

As a compliment to the above experiment, another experiment was carried out at INEL with the x-γ-n multiplicity spectrometer to identify the Z of the fission fragments and to enhance a specific neutron channel. This setup consisted of a 2.5 cm^3 LEPS detector, a 25% n-type Ge detector and eight 5 cm$\times$5 cm neutron detectors. The source-to-detector distances were 2 cm for the LEPS and 10 cm for all other detectors. The neutron detectors were liquid scintillators (BC-501) and were utilized as a multiplicity filter to enhance a desired neutron channel. For n-γ discrimination, we employed charge-sensitive ADC's with combinations of short and long gates. This experiment ran continuously for three months during which time about 3×10^9 double-coincidence events were collected. A total x-ray spectrum in coincidence with γ rays and neutrons is shown in Fig. 1, together with the Z identification of each fragment. As seen in this figure, the $K\alpha$ x rays of the individual fragments are very well resolved. It should be mentioned here that the x-ray detector was used not only to identify the isotopes, but also to resolve close-lying doublets of low-energy γ rays from different isotopes.

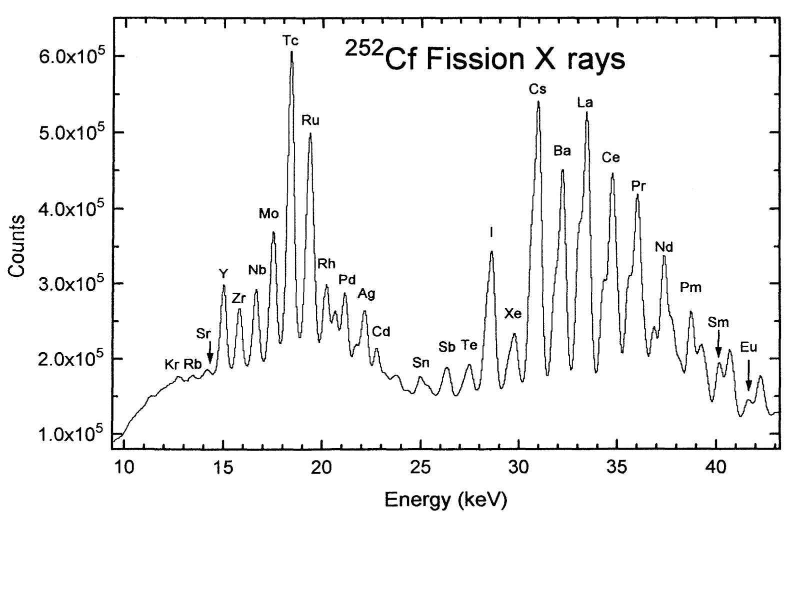

FIG. 1. Total coincidence x-ray spectrum of fragments from spontaneous fission of ^{252}Cf taken with a LEPS detector.

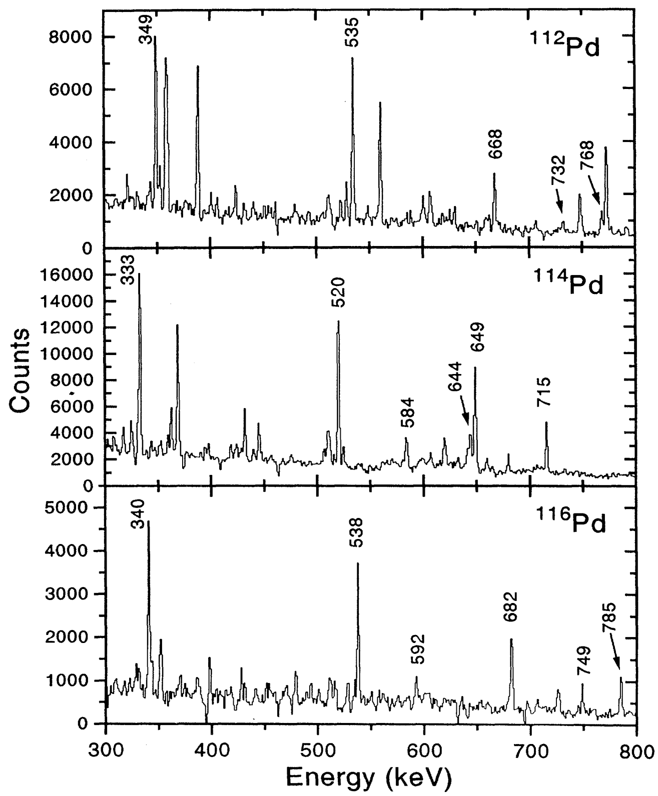

FIG. 2. Sum of the gates of yrast transitions in ^{112}Pd, ^{114}Pd, and ^{116}Pd nuclei.

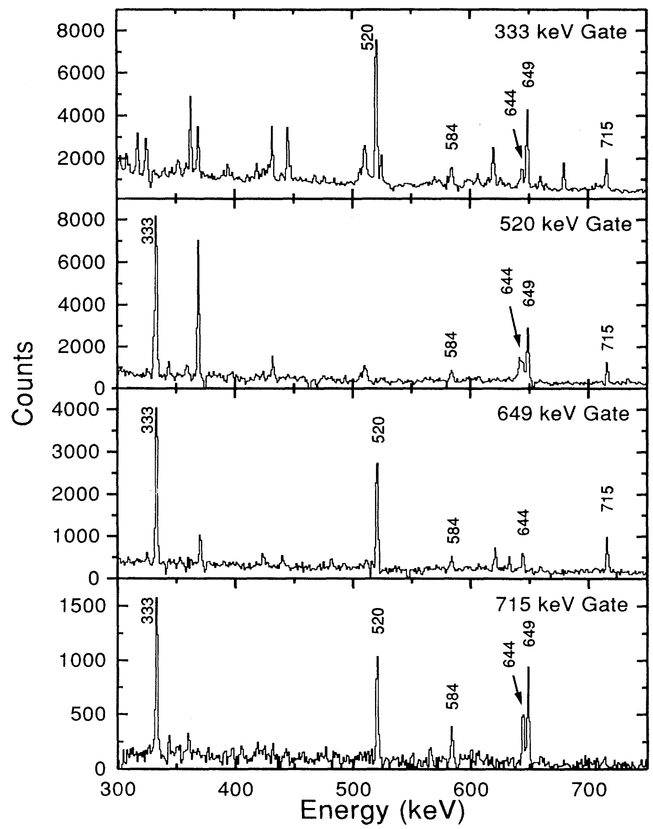

FIG. 3. Examples of γ-γ coincidence spectra gated on several transitions in ^{114}Pd.

III. EXPERIMENTAL RESULTS

Spectra of the sum of yrast γ-ray gates in ^{112}Pd, ^{114}Pd, and ^{116}Pd are shown in Fig. 2. Although only Pd γ rays are labeled in these spectra, all the other γ rays associated with the complementary Te isotopes were positively identified. These spectra were analyzed by fitting peaks with a Gaussian function for the peak and an exponential function for tails. A summary of transition energies and their relative intensities normalized to the $4^+\to2^+$ transition is given in Table II. The intensity measurements are based on an average of 20 detector efficiency curves.

From the ORNL data a two-dimensional ($4K\times4K$) γ-γ matrix was constructed to analyze double coincidences. Representative coincidence spectra in the ^{114}Pd isotope are shown in Fig. 3. As expected, in these gates there are peaks not only in coincidence with yrast transitions, but also in coincidence with several γ rays in complementary Te fragments, as well as with transitions in other isotopes with the same gate overlap. To eliminate most of the γ rays not associated with the isotopes of interest, we employed the triple coincidence technique [15]. Here, any γ rays in coincidence with two transitions known to be, or thought to be in coincidence, are histogrammed. This resulted in a substantial decrease in the statistics. However the decrease in backgrounds more than compensates for this decrease in statistics. This method, therefore, allows us to observe only the transitions in the two fission partners. Figure 4 shows a typical triple coincidence spectrum gated on both the 644- and

TABLE II. Energies and relative intensities of transitions in Pd isotopes. All intensities are measured with the $2^+\to0^+$ gate open.

	^{112}Pd		^{114}Pd		^{116}Pd	
$J_i^\pi\to J_f^\pi$	E_γ (keV)	Int (%)	E_γ (keV)	Int (%)	E_γ (keV)	Int (%)
$2^+\to0^+$	348.8	gate	332.9	gate	340.4	gate
$4^+\to2^+$	534.6	100	520.1	100	537.6	100
$6^+\to4^+$	667.7	37	648.5	62	682.1	62
$8^+\to6^+$	768.5	10	715.6	24	784.9	28
$10^+\to8^+$	731.9	4	644.1	15	748.5	17
$12^+\to10^+$			583.5	6	591.7	8

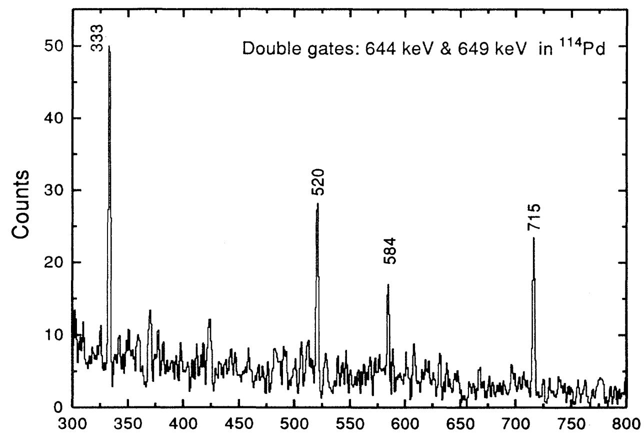

FIG. 4. A typical triple coincidence spectrum. This spectrum is gated on 644 keV and 649 keV transitions in ^{114}Pd.

649-keV transitions in ^{114}Pd. The other yrast transitions in ^{114}Pd show up prominently. Gamma-ray intensities, double γ coincidences, and triple γ coincidences were the primary factors used in constructing the level schemes. Secondary factors were x-γ coincidence data, predicted yields, and inputs from previous studies. The level schemes of ^{112}Pd, ^{114}Pd, and ^{116}Pd deduced from our data are shown in Fig. 5 where excited states were extended to 10^+, 12^+, and 12^+, respectively. Previous studies observed levels up to 6^+.

IV. DISCUSSION

High spin states in ^{102}Pd, ^{104}Pd, and ^{106}Pd have been studied by Grau *et al.* [21] using the reactions $^{92-96}$Zr(^{13}C,$3n\gamma$)$^{102-106}$Pd. They observed the yrast band spins up to 14^+, 18^+, and 16^+ in these isotopes, respectively. Energy levels in ^{108}Pd and ^{110}Pd have previously been investigated by Coulomb excitation using a ^{40}Ar beam [22]. Later, ^{16}O, ^{58}Ni, and ^{208}Pb projectiles were used to study the higher spin states in ^{110}Pd (Ref. [23]).

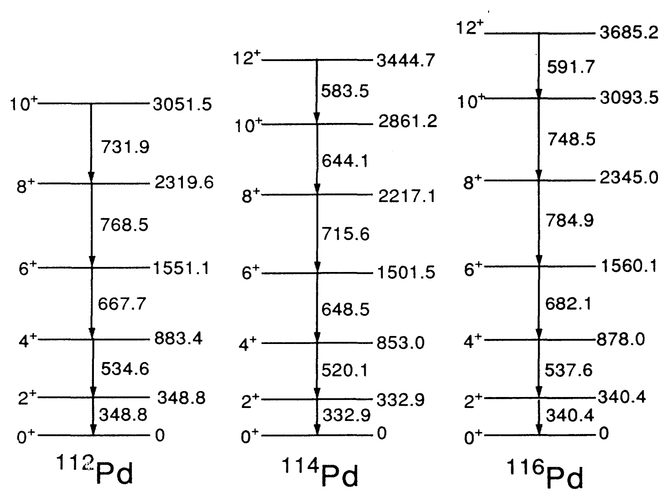

FIG. 5. Level schemes for neutron-rich even-even Pd nuclei.

Recently, new information about ^{108}Pd excited states was reported [24] and the level scheme was extended to $J^\pi = 10^+$

A. Systematics

The excitation energy systematics for known yrast states in ^{104}Pd to ^{116}Pd isotopes as a function of neutron number and mass are presented in Fig. 6. As seen in this figure, there is a decrease in the 2^+ through 12^+ energies as N increases until $N = 64$ (neutron subshell closure). Thereafter these energies are nearly constant until they reach a minimum energy at $N = 68$, then start to increase again. The onset of occupation of the $h_{11/2}$ neutron orbital may be the reason for the very slow change in energies from $N = 64$ to $N = 70$. The minimum in the ^{114}Pd nucleus can be understood because it has 68 neutrons which is close to the middle of the $N = 50-82$ shell.

The systematics of the level energies also show a definite transition from vibrator nuclei to more deformed ones. This is more apparent when plotting the experimental ratios of E_{4+}/E_{2+} and E_{6+}/E_{2+} as a function of neutron number as illustrated in Fig. 7. Sudden changes of these ratios at $N = 64$ in both curves indicate that there is a shape transition that may be interpreted as a change from SU(5) to O(6) symmetry that is from an anharmonic vibration to a γ-soft rotor. In fact, the E_{4+}/E_{2+} ratios for $^{112-116}$Pd are very close to a value of 2.5 for an O(6) nucleus of the IBA model, but considerably smaller than the value of 3.3 for a rigid rotor. The

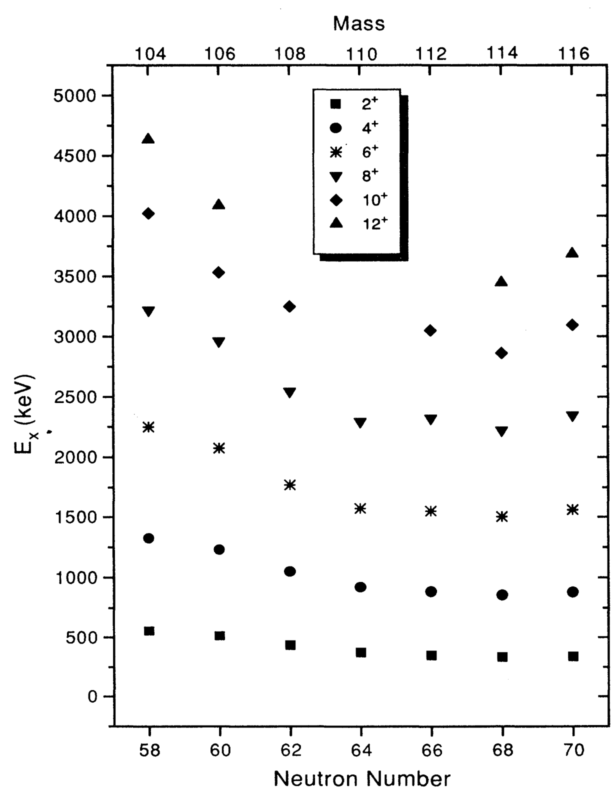

FIG. 6. Systematics of known ground states up to $J^\pi = 12^+$ in Pd isotopes.

same holds true for E_{6^+}/E_{2^+} ratios in these three isotopes which are closer to the IBA model prediction of 4.5 for O(6) symmetry than those of the rigid rotor. The shape transition in Pd isotopes has been confirmed by several different theoretical predictions including IBA calculations by Van Isacker *et al.* [25] and Stachel *et al.* [26], and microscopic Hartree-Fock-Bogolubov (HFB) calculations by Mattu and Khosa [27].

B. IBA calculations

In this section we present the results of calculations performed for the ^{112}Pd, ^{114}Pd, and ^{116}Pd isotopes within the framework of the IBA-1 model by using the program PHINT [28]. In the IBA-1 model, no distinction is made between neutron and proton degrees of freedom. Here, the even-even nuclei were assumed to consist of interacting *s* and *d* bosons [29–31]. The three $Z=46$ even-even Pd isotopes of interest have 66, 68, and 70 neutrons. Relative to the closed shell at $Z=50$ and $N=82$, we have $N_\pi=2$ proton pairs and $N_\nu=8$, 7, 6 neutron pairs, respectively. Thus the total numbers of bosons, $N=N_\nu+N_\pi$, are 10, 9, and 8 for the three nuclei. As discussed above, the energy spectra of these nuclei show definite rotational structures of an O(6) limit with the 4^+ at about 2.5 times the energy of the first 2^+ state. In the calculation, we adjusted the parameters for each isotope separately so as to give a good overall fit to the experimental excitation energies while insisting that parameters vary smoothly and systematically as a function of neutron number. Table III shows the complete list of parameters used in our calculations. The results of the calculated energy spectra are compared with the experimental values of our three Pd isotopes in Fig. 8. As seen in this figure, the experimental excitation energies for high spin states up to $J^\pi=8^+$ are reproduced very well in these calculations, although the predictions are somewhat lower than experimental values. Note that the 8^+ predictions are all below the experimental 8^+ levels while the 10^+ levels are all above the experimental 10^+ levels to support a change in structure. The results are less satisfactory for 10^+ states and become worse for the 12^+ state in the case of the ^{114,116}Pd isotopes. This change is related to the occurrence of backbendings at the $10^+\rightarrow 8^+$ transition which are not predicted by the IBA-1 calculations.

TABLE III. Parameters used in IBA-1 calculations.

Parameter	^{112}Pd	^{114}Pd	^{116}Pd
No. of bosons	10	9	8
EPS	0.87	0.82	0.78
ELL	0.004	0.004	0.004
QQ	−0.52	−0.52	−0.52
OCT	−0.01	−0.01	−0.01
HEX	−0.014	−0.014	−0.014
CHQ	0.0	0.0	0.0

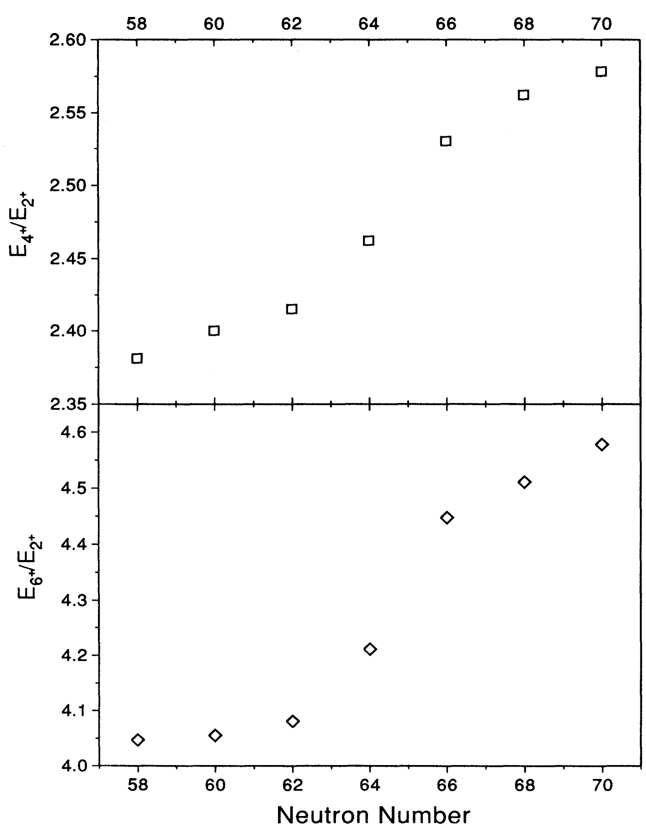

FIG. 7. Experimental excitation energy ratios of E_{4^+}/E_{2^+} and E_{6^+}/E_{2^+} as a function of neutron number.

C. Cranked shell model calculations

The plot of the kinematic moment of inertia as a function of rotational frequency shows a backbending behavior in all three Pd isotopes studied in the present work. This is shown in Fig. 9. The crossings occur at a rotational frequency between 0.32 and 0.34 MeV/$\hbar$. Similar band crossings have been observed in ^{104}Pd, ^{106}Pd, and ^{108}Pd, but not in ^{102}Pd (Ref. [21]). There is also an indication of backbending in ^{110}Pd, as seen in

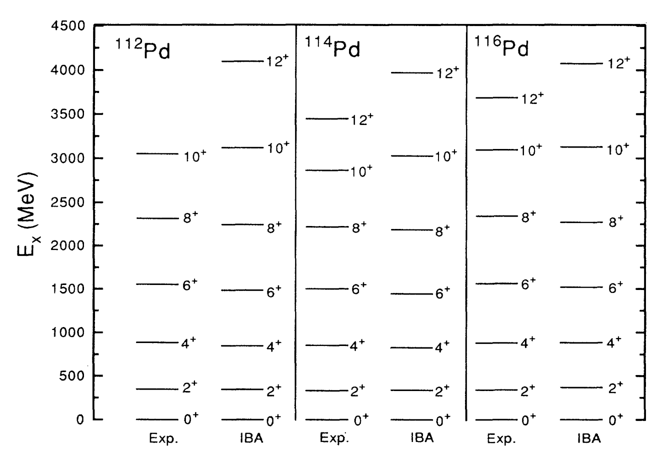

FIG. 8. Calculated excitation energies for ground states of Pd nuclei compared with IBA-1 calculations.

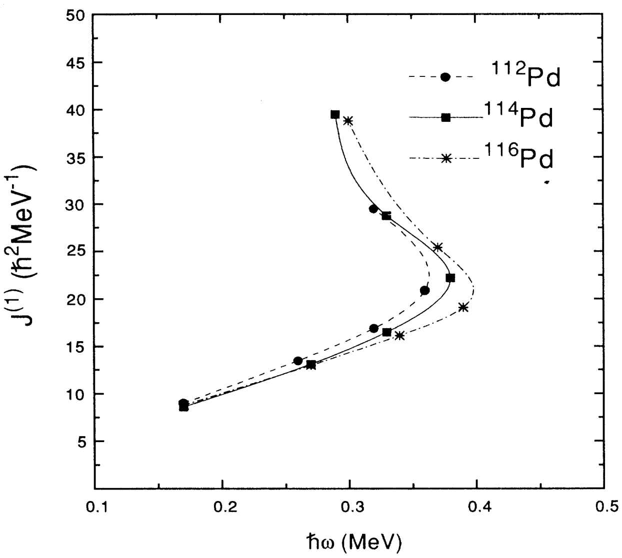

FIG. 9. Plot of moment of inertia as a function of rotational frequency.

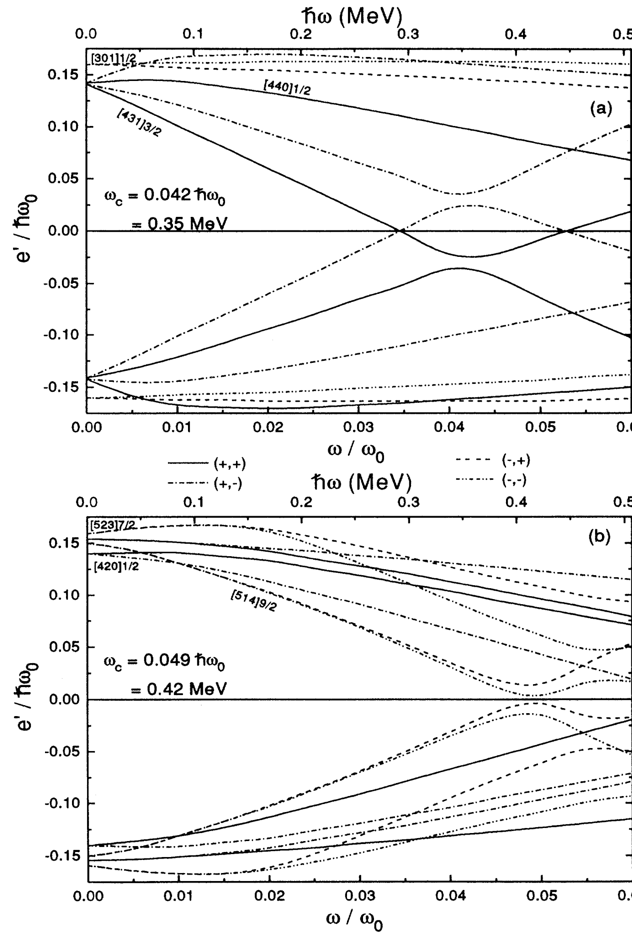

FIG. 10. Cranked shell model calculations for both quasiprotons (a) and quasineutrons (b) are plotted against rotational frequency. The parameters used were $\varepsilon_2 = -0.23$, $\varepsilon_4 = 0.0$, $\gamma = 0°$, $\Delta_p = \Delta_n = 0.14\hbar\omega_0$, $\lambda_p = 5.103\hbar\omega_0$, and $\lambda_n = 5.746\hbar\omega_0$.

Coulomb excitation experiments [23]. However, for ^{104}Pd and ^{106}Pd the crossing frequencies are higher, at about 0.35 and 0.38 MeV/$\hbar$, respectively. This may be because of the shape transition in this region at $N=64$ as previously mentioned. The occurrence of shape change is also evident from the comparison of a $B(E2;\, 0^+ \to 2^+)$ value of 0.51 $e^2\mathrm{b}^2$ in ^{104}Pd to a $B(E2;\, 0^+ \to 2^+)$ value of 0.91 $e^2\mathrm{b}^2$ in ^{110}Pd. The same is true for Q_{2+} value which is -0.25 e b for ^{104}Pd as compared to 0.82 e b for ^{110}Pd. Recent macroscopic-microscopic calculations by P. Möller *et al.* [32] predict positive quadrupole deformations (prolate) for the $^{104-108}$Pd nuclei, whereas negative deformation (oblate) parameters were predicted for the $^{112-116}$Pd isotopes.

The observation of backbends in the Pd isotopes indicates that an appropriate interpretation should be possible in terms of the cranked shell model (CSM) described in detail by Bengtsson and Frauendorf [63–36]. We have performed such calculations to determine whether the proton orbital ($\pi g_{9/2}$) or neutron orbital ($\nu h_{11/2}$) is responsible for band crossings at the observed rotational frequencies. Our results for ^{114}Pd are presented in Fig. 10(a) for protons and Fig. 10(b) for neutrons, respectively. The standard parameters used in the calculations were as follows: quadrupole deformation, $\varepsilon_2 = 0.23$; hexadecapole deformation, $\varepsilon_4 = 0.0$; asymmetry parameter $\gamma = 0°$ (the Lund convention); pairing gap parameters for protons and neutrons, $\Delta p = \Delta n = 0.14\hbar\omega_0$ (where $\hbar\omega_0 = 41 A^{-1/3}$); and Fermi levels for protons and neutrons taken from Nilsson diagrams in Ref. [37] at $\omega = 0$, $\lambda_p = 5.103\hbar\omega_0$, $\lambda_n = 5.746\hbar\omega_0$. The x axes in these figures are shown in both harmonic oscillator units (bottom) and MeV units (top). The various quasiparticle orbitals are labeled by their asymptotic Nilsson quantum numbers $[N, n_z, \Lambda]\Omega$ at $\omega = 0$.

The calculations predict a band crossing due to the alignment of two $g_{9/2}$ protons at $\hbar\omega = 0.35$ MeV, which is very close to the experimental values of 0.32–0.34 MeV in the $^{112-116}$Pd isotopes. The crossing caused by the alignment of two $h_{11/12}$ neutrons is predicted to occur at $\hbar\omega = 0.42$ MeV, which is much higher than the observed values. Hence, we believe that the $g_{9/2}$ proton orbital is responsible for backbendings in this region. To support this argument further, one is obliged to look at the energy spectra of neighboring odd-Z even-N nuclei, such as ^{113}Rh and ^{115}Ag, where crossing corresponding to $g_{9/2}$ protons is blocked because of the odd number of protons. Unfortunately, high spin states in these nuclei are not yet established, so blocking arguments cannot be utilized at this time.

V. CONCLUSION

In this work we have studied the energy levels of even-even Pd isotopes up to moderately high spins. A study of the level systematics in this region reveals that there is a shape transition around $N=64$ which is theoretically predicted to go from prolate to oblate. It was found that the energy of all levels decreases as a function of N and reaches a minimum at $N=68$ (^{114}Pd). Our IBA-1 calculations reproduce the experimental energy levels quite well up to spin 8^+, but a change occurs for higher spins of 10^+ and 12^+. This change is related to the occurrence of band crossings which are not predicted in the framework of the IBA-1 model. The backbends observed at $J^\pi=10^+$ have been interpreted based on our cranked shell model calculations as the alignment of two $g_{9/2}$ protons along the rotational axis. These calculations were also consistent with the prediction of oblate deformations for these three isotopes.

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