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Does a $\frac{5}{2}^+$-$\frac{5}{2}^-$ ground-state parity doublet exist in ^{229}Pa?

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The $\frac{1}{2}$[530] decoupled band in ^{229}Pa has been identified up to the $\frac{19}{2}^-$ level in (p,t) and $(p,2n\gamma)$ experiments. It is found that the $\frac{3}{2}^-$ band head has an excitation energy of 19(10) keV, and can thus not be identified with a 123 keV level observed in the ^{229}U electron capture decay. This removes the evidence presented earlier for a spin-parity assignment of $\frac{5}{2}^+$-$\frac{5}{2}^-$ to a proposed nearly degenerate ground-state doublet in ^{229}Pa.

Nine years ago Ahmad *et al.* [1] suggested the existence of an $I=\frac{5}{2}$ ground-state parity doublet in ^{229}Pa with the $\frac{5}{2}^-$ level lying 220 eV above the $\frac{5}{2}^+$ ground state. These nearly degenerate levels were assigned to be the $\frac{5}{2}$[642] and $\frac{5}{2}$[523] Nilsson orbits, and a level at 123 keV was interpreted as $\frac{3}{2}^-$ band head of the $\frac{1}{2}$[530] decoupled band. The same Nilsson states are known in ^{231}Pa, but with inverted energy spacings: The $\frac{3}{2}^-$ level is the ground state, and the $\frac{5}{2}^-$ and $\frac{5}{2}^+$ levels lie at 174 and 184 keV, respectively. This shift in the position of the $\frac{1}{2}$[530] band relative to the $\frac{5}{2}^+$-$\frac{5}{2}^-$ doublet had been predicted earlier as arising from octupole correlation effects [2], and therefore Ahmad *et al.* interpreted their observations as evidence for a possible ground-state octupole deformation in ^{229}Pa.

The work of Ahmad *et al.* was among the first experiments suggesting octupole deformations in light actinide nuclei, and it has therefore contributed significantly to the revival of interest in the concept of intrinsic reflection asymmetry in nuclei [3]. The nearly degenerate opposite-parity ground-state doublet also has been discussed in connection with time-reversal nonconservation in nuclei. The mixing of the ground-state parity doublets in some deformed odd-A nuclei could enhance T-nonconserving nuclear moments, as compared to the moments generated by the unpaired valence nucleon. Haxton and Henley [4] estimate the largest enhancement of the electric dipole and magnetic quadrupole moments for ^{229}Pa, due to the extremely small energy splitting of the parity doublet.

The only evidence for a $\frac{5}{2}^+$-$\frac{5}{2}^-$ parity doublet in ^{229}Pa comes from the work of Ahmad *et al.* [1]. These authors have investigated the electron capture decay of ^{229}U to ^{229}Pa, from which they inferred the ground-state doublet and an excited state at 123 keV, which decays by a $M1$ transition to the upper level of the doublet. In addition, they studied the ^{231}Pa$(p,t)^{229}$Pa reaction, where they observed the $\frac{3}{2}^-$ member of the $\frac{1}{2}$[530] band—which is the ground state in ^{231}Pa, and therefore is preferentially excited in the (p,t) reaction [5]—at an excitation energy of 128(15) keV. Identifying this state with the 123 keV level observed in the electron capture decay led these authors to propose a spin and parity assignment of $\frac{5}{2}^+$-$\frac{5}{2}^-$ for the ground-state doublet, but the paper does not give the experimental evidence in sufficient detail to judge its reliability.

A sizable octupole deformation for ^{229}Pa would seem surprising, since this nucleus lies at the border of the accepted region of reflection asymmetry around $A\approx 224$, and therefore independent measurements seemed in place. The results of experiments described in this paper are inconsistent with the level scheme proposed by Ahmad *et al.* They establish that the $\frac{3}{2}^-$ bandhead of the $\frac{1}{2}$[530] band cannot be identified with the 123 keV level, and therefore remove the evidence for a $\frac{5}{2}^+$-$\frac{5}{2}^-$ assignment to the ground-state doublet.

Excited levels in ^{229}Pa were studied in the ^{230}Th$(p,2n)^{229}$Pa and ^{231}Pa$(p,t)^{229}$Pa reactions. In the $(p,2n)$ reaction, which was investigated at the Bonn cyclotron, conversion electrons and γ rays were measured with two iron-free orange spectrometers and four Compton-suppressed germanium detectors [6]. For the measurement of e^--n coincidences a 40 cm diam NE102A detector was used. The target was a ~300 μg/cm^2 thick deposit of thorium (91.5% ^{230}Th+8.5% ^{232}Th) on a carbon backing.

The (p,t) reaction was studied at the Munich Tandem accelerator using a target of 100 μg/cm^2 radioactive ^{231}Pa evaporated onto a 12 μg/cm^2 carbon backing. The tritons were detected in a Q3D spectrometer equipped with a 1.8 m position-sensitive focal-plane detector [7]. The calibration measurements were performed with targets of ~100 μg/cm^2 of ^{208}Pb and ^{235}U, and ~50 μg/cm^2 of ^{230}Th ($\leq$0.1% ^{232}Th), all on ~10 μg/cm^2 carbon backings, in addition to the 300 μg/cm^2 Th target.

a. The $\frac{1}{2}$[530] *band in the (p,2n) reaction.* The ^{230}Th$(p,2n)^{229}$Pa reaction was studied at $E_p=14$ MeV, where the cross section is expected to be large [8]. The singles electron spectrum, which is much less contaminated by background from fission fragments than the γ-ray spectrum [9], is dominated by the L and M conversion electrons of 110 and 160 keV transitions, for which the L-subshell ratios establish $E2$ multipolarity. Transitions with almost identical energies were found in ^{231}Pa as $\frac{11}{2}^-\rightarrow\frac{7}{2}^-$ and $\frac{15}{2}^-\rightarrow\frac{11}{2}^-$ transitions of the $\frac{1}{2}$[530]

ground-state band [10]. In addition, somewhat weaker lines appear in the electron spectrum, which we assign as L and M lines of the 123 keV $M1$ transition in ^{229}Pa mentioned above and the 121 keV $4^+ \rightarrow 2^+$ transition in ^{230}Th.

A γ-ray spectrum measured in coincidence with electrons corresponding to the L_1 conversion of the 123 keV transition in ^{229}Pa is shown in the upper part of Fig. 1. The dominating lines result from coincidences with the L_2 conversion electrons of the 121 keV transition in ^{230}Th. Two weak lines are seen, at 119.3 and 247.8 keV, which were also observed in the ^{229}U electron capture decay and were assigned to transitions in ^{229}Pa populating the 123 keV level [1]. The 110 and 160 keV lines assigned in the present work as $\frac{11}{2}^- \rightarrow \frac{7}{2}^-$ and $\frac{15}{2}^- \rightarrow \frac{11}{2}^-$ transitions of the $\frac{1}{2}[530]$ band in ^{229}Pa are not seen in this spectrum. They would, however, be expected if the $\frac{3}{2}^-$ ground state of this band was the 122.7 keV level depopulated by the 122.5 keV transition, as proposed by Ahmad *et al.* [1].

The γ-ray spectra measured in coincidence with L_2 conversion electrons of the 110 and 160 keV transitions are shown in the central and lower part of Fig. 1. As mentioned above, the 110 and 160 keV lines observed in the present work lie very close in energy to corresponding lines in ^{231}Pa, and they could thus be produced by the ^{232}Th$(p,2n)^{231}$Pa reaction on the 8.5% ^{232}Th content in the target. However, the corresponding γ-ray spectra measured with a ^{232}Th target look entirely different, except for the 110, 160, and 207 keV lines assigned [10] to transitions in the $\frac{1}{2}[530]$ band of ^{231}Pa.

Two additional experiments further support the assignment of the 110 and 160 keV lines to the $\frac{1}{2}[530]$ band in ^{229}Pa: (i) In a measurement of e^--e^--coincidence spectra with the double-orange spectrometer we find that the 110 keV line is in coincidence with a 56.4(2) keV $E2$ line assigned to the $\frac{7}{2}^- \rightarrow \frac{3}{2}^-$ transition in ^{229}Pa. The corresponding transition in ^{231}Pa has an energy of 58.57(1) keV. (ii) e^--n coincidence measurements establish that the 110 and 160 keV conversion electrons, and also $\sim\frac{1}{3}$ of the intensity of the $123L_1(^{229}\mathrm{Pa})/121L_2(^{230}\mathrm{Th})$ peak, are in coincidence with neutrons, and thus cannot be produced in the ^{230}Th$(p,\gamma)^{231}$Pa reaction.

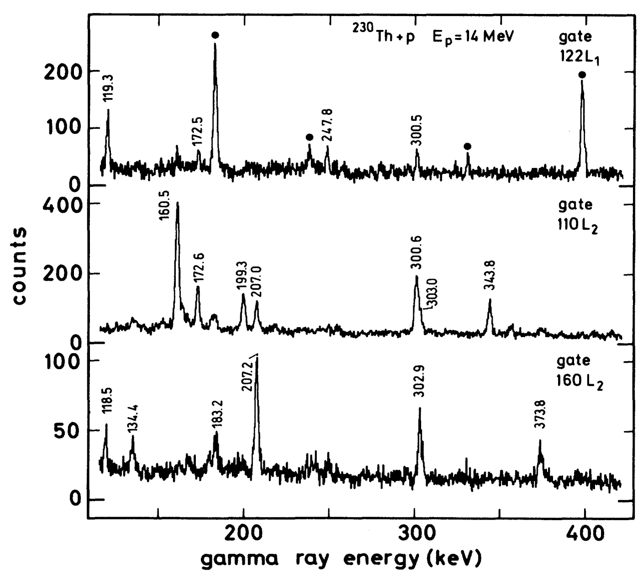

FIG. 1. e^--γ coincidence spectra with gates set on the $123L_1(^{229}\mathrm{Pa})/121L_2(^{230}\mathrm{Th})$ (top), $110L_2$ (center), and $160L_2$ (bottom) conversion electrons. Lines assigned to ^{229}Pa are marked by their energies. Lines marked by dots belong to ^{230}Th. In the spectrum with the $160L_2$ gate the 110 keV γ ray is weakly present, although largely masked by the strong K_β x rays.

The 123 keV γ ray is not present in the spectra coincident with the L_2 conversion electrons of the 110 and 160 keV transitions. From the center spectrum of Fig. 1 one obtains $I_\gamma(123)/I_\gamma(160) \leq 0.04$, which yields, for $M1$ and $E2$ multipolarities of the 123 and 160 keV lines, respectively, $I_{\mathrm{tot}}(123)/I_{\mathrm{tot}}(160) \leq 0.2$. Therefore we conclude that the 110 and 160 keV $E2$ transitions belong to the $\frac{1}{2}[530]$ band in ^{229}Pa, and are not in coincidence with the 122.5 keV $M1$ transition depopulating the 122.7 keV level proposed by Ahmad *et al.* [1].

A number of further strong lines are observed in the coincidence spectra. We assign the 207.1 keV line to the $\frac{19}{2}^- \rightarrow \frac{15}{2}^-$ transition of the $\frac{1}{2}[530]$ band, in accordance with a corresponding assignment in ^{231}Pa. Due to the lack of $\gamma\gamma$-coincidence data a reliable placement of the remaining lines in a decay scheme is not possible at present.

b. The $\frac{1}{2}[530]$ *band in the (p,t) reaction.* We have measured triton spectra in the ^{231}Pa$(p,t)^{229}$Pa reaction for 22-MeV-incident protons at five scattering angles between 5° and 45°. In order to obtain a reliable and precise energy calibration the (p,t) reactions on ^{230}Th, ^{232}Th, and ^{235}U targets were measured alternatingly with the ^{231}Pa target. The isotopes ^{230}Th and ^{235}U have Q values such that the $\frac{3}{2}^-$ level in ^{229}Pa is bracketed by excited levels with accurately known excitation energies, and which are strongly populated in the (p,t) reaction. The uncertainties in the reaction energies are then determined by the accuracy with which the ground-state Q values are known.

In the course of our measurements we found that the Q value for the ^{230}Th(p,t) reaction, as derived from the Atomic Mass Table [11], has to be revised. From the (p,t) calibration measurement with the 300 μg/cm^2 thorium target, where we observe lines from both the ^{230}Th(p,t) and ^{232}Th(p,t) reactions, we obtain $Q[^{232}\mathrm{Th}(p,t)] - Q[^{230}\mathrm{Th}(p,t)] = 493.5(10)$ keV. The Q value for the ^{232}Th(p,t) reaction is known from measured neutron binding and α-decay energies to be equal to [12] $-3076.0(13)$ keV, which yields $Q = -3569.5(17)$ keV for the ^{230}Th(p,t) reaction. For the ^{235}U(p,t) reaction we used $Q = -3659(3)$ keV, as derived from the Atomic Mass Table [11].

Two triton spectra for the ^{231}Pa(p,t) reaction, measured at 5° and 45°, are shown in Fig. 2 together with a spectrum recorded in the ^{235}U(p,t) reaction at 45°. The peaks which we assign to the members of the $\frac{1}{2}[530]$ decoupled band in ^{229}Pa are marked by spins and parities. For the $\frac{3}{2}^-$ bandhead we obtain a Q value of $-4145(3)$ keV and, with the ground-state Q value of $-4126(9)$ keV from the Atomic Mass Table, an excitation energy of 19(10) keV.

The level scheme, as derived from the present work, is

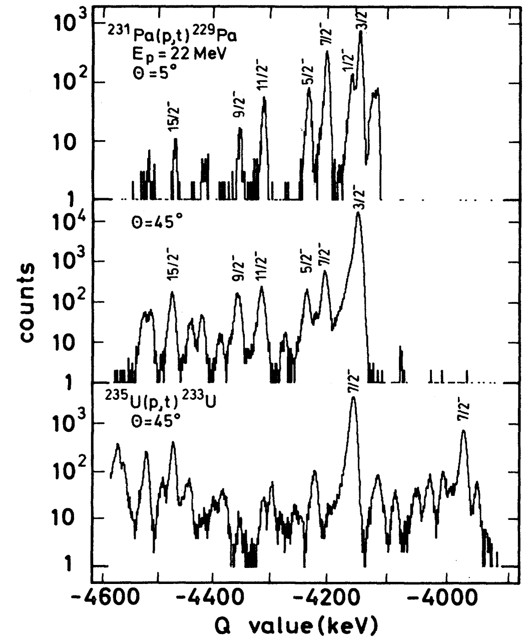

FIG. 2. Triton spectra observed in the (p,t) reaction on targets of ^{231}Pa and ^{235}U at $E_p=22$ MeV. The lines assigned to the $\frac{1}{2}$ [530] band of ^{229}Pa are marked by spins and parities. The two strong lines marked in the U spectrum are the $\frac{7}{2}^-$ members of the excited $\frac{5}{2}$ [752] and $\frac{7}{2}$ [743] bands in ^{233}U (Ref. [5]).

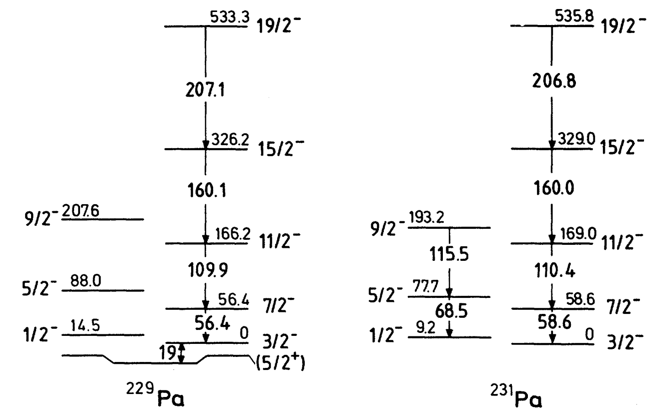

FIG. 3. Levels in ^{229}Pa as observed in the present work. For comparison the $\frac{1}{2}$ [530] ground-state band of ^{231}Pa is also shown. The level energies of the $K^\pi=\frac{1}{2}^-$ band are given relative to its $\frac{3}{2}^-$ ground state, which has an excitation energy of 19 ± 10 keV in ^{229}Pa (see text). The excitation energies obtained from the (p,t) measurement are 56.3(5), 165.9(5), and 324.7(10) keV for the σ positive branch and 14.5(15), 88.0(5), and 207.6(8) keV for the σ negative branch of the $K^\pi=\frac{1}{2}^-$ band. The $\Delta I=2$ transition energies have uncertainties of ± 0.2 keV.

shown in Fig. 3. To point out the similarity, a partial level scheme of ^{231}Pa is also displayed. The energies of the members of the $\frac{1}{2}$ [530] band above the $\frac{3}{2}^-$ state in ^{229}Pa obtained from the (p,t) measurements are listed in the caption of Fig. 3. The level energies of the positive-signature branch are in good agreement with those derived from the transition energies measured in the $(p,2n)$ experiment.

In summary, we have shown that the $\frac{3}{2}^-$ band head of the $\frac{1}{2}$ [530] decoupled band in ^{229}Pa cannot be identified with the 123 keV level observed in the ^{229}U electron capture (EC) decay. The excitation energy of the $\frac{3}{2}^-$ level is determined from its Q value in the (p,t) reaction as 19(10) keV, where the large error stems from the uncertainty in the ground-state mass of ^{229}Pa. The tabulated value of this mass has changed from the 1983 Atomic Mass Table to the 1986 Midstream Mass Table [11] by 11 keV. This indicates that the uncertainty in the excitation energy of the $\frac{3}{2}^-$ level might be even larger than the quoted error, so this level might lie very close to the ground state.

The assignment [1] of $\frac{5}{2}^+$ to the ground state of ^{229}Pa, based on the logft values for the ^{229}Pa EC decay to levels with established spins and parities in ^{229}Th, seems fairly conclusive. Ahmad *et al.* associate this level with the $\frac{5}{2}$ [642] orbital. An assignment as the $\frac{5}{2}^+$ member of the Coriolis-distorted $\frac{3}{2}$ [651] band [13] would be equally consistent with the β-decay data, although the α decay of ^{229}Pa to ^{225}Ac seems to favor the $K=\frac{5}{2}$ configuration [14]. The $\frac{5}{2}^-$ assignment to the proposed 220 eV level was based on the identification of the $\frac{3}{2}^-$ member of the $\frac{1}{2}$ [530] band with the 123 keV level [1]. The present work shows this to be wrong. Only one further experimental observation had been invoked to support the $\frac{5}{2}^-$ assignment: Ahmad *et al.* have measured a 420 ns half-life in the decay of ^{229}U, which they assign to the 220 eV level on the basis of the electron spectrum associated with this half-life. If negative parity is assumed for the 220 eV level the resulting $E1$ transition to the $\frac{5}{2}^+$ ground state would be strongly enhanced relative to the usual $E1$ rates between one-quasiparticle states [14,15]. Such fast $E1$ transitions are observed only between members of rotational bands build on parity doublets in nuclei with octupole deformation. However, there is a problem with this interpretation in the present case: The enhancement in $B(E1)$ is generally interpreted as resulting from the large intrinsic electric dipole moment Q_1 induced by the polarizing electric field of the nuclear octupole deformation. As noted by Leander and Chen [16] the observed Q_1 values for odd-A nuclei are always close to those of their even neighbors, with the exception of ^{229}Pa. With the value of $B(E1)\sim6\times10^{-2}$ W.u. extracted by Ahmad and co-workers from the 420 ns half-life [14,15] one obtains $Q_1\sim0.6$ fm, as compared to a value of 0.11 fm for ^{228}Th. Thus the suggested $\frac{5}{2}^-\rightarrow\frac{5}{2}^+$ transition would be almost 2 orders of magnitude faster than expected from octupole correlations. There is no such difficulty with the half-life if the 220 eV transition has $M1$ or $E2$ multipolarity, and we therefore conclude that the presently available experimental data favor positive parity for the 220 eV level.

We should mention one alternative explanation for the low energy, isomeric electron transition. Ahmad *et al.* find the 420 ns half-life to be associated with 220 eV electrons in protactinium, as established from the observation of coincidences of these electrons with Pa K x rays. But their measurements do not seem to yield direct information on the energy of the actual transition, and thus on the location of the isomer. The 220 eV electrons could for example result from the L_3 conversion of a 17 keV $E1$ transition, which could be the $\frac{3}{2}^{-} \rightarrow \frac{5}{2}^{+}$ transition expected from the present work. This would give a $B(E1, \frac{3}{2}^{-} \rightarrow \frac{5}{2}^{+}) = 9\times10^{-6}$ W.u. in remarkable agreement with the values known for the corresponding transitions in ^{231}Pa. This hypothesis could perhaps be checked by a search for the isomeric 17 keV γ rays and L x rays in the ^{229}U EC decay.

The suggestion of a possible ground-state octupole deformation in ^{229}Pa was based on the observation of a dramatic change in the low-energy level ordering in going from ^{231}Pa to ^{229}Pa. The present results eliminate this evidence to a large extent, and thus indicate that octupole correlations might be less important in ^{229}Pa than suggested earlier. For example, the experimental data are not inconsistent with an interpretation of the ground-state doublet as $\frac{5}{2}^{+}$ and $\frac{3}{2}^{+}$ members of the $\frac{3}{2}$[651] band, coupled to the higher-lying $\frac{5}{2}$[642] band by the Coriolis interaction, as is the case in ^{231}Pa where the $\frac{3}{2}^{+}$ and $\frac{7}{2}^{+}$ levels are almost degenerate [13]. If this interpretation is adopted there would be a close similarity between ^{231}Pa and ^{229}Pa, and thus little need to invoke an octupole deformation in ^{229}Pa. Clearly more experimental information on the low-energy level structure of ^{229}Pa is required before safe conclusions can be drawn on possible octupole correlations in this nucleus.

Additional information on the shape of ^{229}Pa might come from the structure of the $\frac{1}{2}$[530] band. For the rotational energies of the $K=\frac{1}{2}$ band we used the form

$$E_{\rm rot} = A\{I(I+1) + (-1)^{I+1/2}(I+\tfrac{1}{2}) \times a[1-\varepsilon^2(I-\tfrac{1}{2})(I+\tfrac{3}{2})]\} .$$

The spin-dependence of the decoupling term, which is necessary to reproduce the energies, can be interpreted as first-order correction due to the coupling of the $\frac{1}{2}$[530] band to higher-lying $K=\frac{3}{2}$ bands. From the data in Fig. 3 one obtains

$$A=6.34(4)\ \text{keV},\ a=-1.79(2),\ \text{and}\ \varepsilon=0.061(4)\ \text{for}\ ^{229}\text{Pa},$$

$$A=6.26(4)\ \text{keV},\ a=-1.50(2),\ \text{and}\ \varepsilon=0.058(4)\ \text{for}\ ^{231}\text{Pa}.$$

For the decoupling parameter of ^{231}Pa a value of $a=-2.27$ has been calculated [17]. The decoupling parameter might be very sensitive to the octupole deformation, depending on the Nilsson orbits involved [18]. A calculation of this quantity may thus yield additional information on a possible octupole deformation of ^{229}Pa.

One open question remains: If the ground state of ^{229}Pa is the $\frac{5}{2}^{+}$ level, the rotational members of the ground-state band would be the yrast levels, and they should be strongly populated in the $(p,2n)$ reaction. We observe, however, no clear lines which might possibly be assigned to the lowest transitions in this band. The reason is probably that the rotational levels of the $\frac{3}{2}$[651] or $\frac{5}{2}$[642] band decay predominantly by intraband $\Delta I=1$ $M1$ transitions, as follows from the corresponding bands in ^{231}Pa and ^{233}Pa. These highly converted transitions are too low in energy to be seen in the present measurements. Their identification would require the detection of low-energy conversion electrons, made difficult by the large background from δ electrons.

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