Electron capture decay of 58-min $^{229}_{92}$U and levels in $^{229}_{91}$Pa

I. Ahmad, R. R. Chasman, J. P. Greene, F. G. Kondev,* and S. Zhu
Argonne National Laboratory, Argonne, Illinois 60439, USA

(Received 15 July 2015; published 17 August 2015)

Electron capture decay of ^{229}U is investigated by measuring the γ-ray and conversion electron spectra of mass-separated and unseparated ^{229}U sources with high-resolution germanium and silicon detectors, respectively. Gamma-gamma coincidence measurements are also performed using germanium detectors. These studies provide level energies and level ordering in ^{229}Pa. Single-particle assignments are given to these levels which are in agreement with the systematics in this region and also with theory. In a previous study, we report the observation of a $5/2^{\pm}$ parity doublet in the ^{229}Pa ground state, which is a signature of octupole deformation. The present analysis of the data still shows a splitting of 60 ± 50 eV, but with this large uncertainty the existence of the doublet is not certain.

DOI: 10.1103/PhysRevC.92.024313 PACS number(s): 21.10.Pc, 23.20.Lv, 27.90.+b

I. INTRODUCTION

Incipient octupole deformation was predicted in 1980 by Chasman [1] in the ground state of ^{229}Pa. Its signature is a parity doublet consisting of two almost-degenerate states with the same spin but opposite parity and a large octupole matrix element between the connecting states. The existence of the parity doublet in ^{229}Pa was also found in later calculations [2]. Experiments performed [3] on the level structure of ^{229}Pa indicated the presence of a ground-state parity doublet. In these studies the ^{231}Pa(p,t) reaction was used to determine the excitation energy of the $I,K^{\pi} = 3/2,1/2^{-}[530]$ single-particle state by measuring the energies of the outgoing tritons with a magnetic spectrometer. (Note that the use of Nilsson quantum numbers is not strictly appropriate when there is a strong octupole correlation or octupole deformation in the nucleus. These quantum numbers should be understood as signifying the main single-particle component of the wave function.) The ^{231}Pa(p,t) reaction gave the energy of the $3/2,\ 1/2^{-}$ level as 128 ± 15 keV. In the electron capture (EC) decay of ^{229}U, we observed a strong 122.5-keV $M1$ transition, suggesting a level at 122.5 keV. Since the two energies are close, the 122.5-keV level was identified as the same state as populated in the (p,t) reaction and was given the $3/2,\ 1/2^{-}[530]$ assignment. The ground state of ^{229}Pa was established [4] as $5/2^{+}[642]$ on the basis of its EC and α decay. The $M1$ character of the 122.5-keV transition indicates a negative-parity state near ground, which we assigned to the $5/2^{-}[523]$ Nilsson state, establishing a parity doublet. The energy difference between the $5/2^{-}$ and the $5/2^{+}$ states was deduced from closed cycles of γ rays as 150 ± 100 eV.

The experiment in Ref. [3] was performed using the Argonne FN tandem Van de Graaff accelerator and the Enge split-pole magnetic spectrometer, measuring absolute energies of the outgoing tritons from the ^{231}Pa(p,t) reaction. Later, several careful experiments were performed at other laboratories to deduce the level structure of ^{229}Pa. In one experiment [5], the energies of tritons from the ^{231}Pa(p,t) reaction were measured with a Q3D magnetic spectrometer, which gave the energies of the members of the $1/2^{-}[530]$ band directly. The Q value to the $3/2^{-}$ level in ^{229}Pa was measured as -4145 ± 3 keV and the Q value between the ground states was calculated from known atomic masses [6] as -4126 ± 9 keV. The difference between the two Q values gave an excitation energy of 19 ± 9 keV for the $3/2^{-}$ state. Using a recent mass table [7], we calculate the Q value between the ground states as -4133 keV, which gives an excitation energy of 12 ± 5 keV for the $3/2^{-}$ level in ^{229}Pa.

In other experiments [8,9], γ rays from the ^{231}Pa$(p,t\gamma)$ and ^{230}Th$(p,2n\gamma)$ reactions were measured. These measurements gave the same level scheme as in Ref. [3] but the levels were given different single-particle assignments. The excitation energy of the $3/2,\ 1/2^{-}[530]$ state was measured [10] directly by particle-γ coincidence using the ^{231}Pa$(p,t\gamma)$ reaction. An 11.6 ± 0.3 keV γ ray was observed in coincidence with the tritons which was absent in the spectrum gated by protons or deuterons. This γ ray was assigned to the $1/2,3/2^{-} \rightarrow 5/2,5/2^{+}$ decay with an $E1$ multipolarity. In our experiments we do not see any γ ray or any difference between two γ rays at this energy. However, we observe several pairs of γ rays with an energy difference of 12.20 ± 0.04 keV.

The measured value of 19 ± 9 keV for the $3/2,\ 1/2^{-}[530]$ level in Ref. [5] is clearly in disagreement with the value of 128 ± 15 keV reported in Ref. [3]. Although we could not find any error in our (p,t) reaction measurements, it is possible that the calibration of the spectrometer changed before the time of the (p,t) reaction measurement. During the investigation of the ^{229}Pa level scheme in 1982, a large set of data was collected on the decay properties of ^{229}U which did not fit the level scheme proposed in Ref. [3]. These data included measurements of higher-energy γ rays, electron spectra in coincidence with Pa K x rays, and γ-γ coincidence measurements. All these spectra have now been analyzed in detail and have been used to construct a new level scheme for ^{229}Pa. By removing the constraint of the assignment of the $3/2,\ 1/2^{-}[530]$ configuration to the 122.71-keV level in Ref. [3], we have been able to construct a new reasonable level scheme which includes the original level scheme but with different spin-parity assignments. We still find a positive

*kondev@anl.gov

number in the $211.06 - (122.52 + 88.43)$ closed cycle but detailed analysis shows that the 211.06-keV γ ray is a doublet. One component is the $(122.52 + 88.43) - 0.0 = 210.95$ keV γ ray and the other component is the $241.80 - 30.67 = 211.13$ keV γ ray. The measured energy of 211.06 keV represents the energy of the mixed peak. Hence, this closed cycle cannot be used to determine the energy of the $5/2^-$ level. However, the $241.84 - (122.52 + 119.26) = 60 \pm 50$ eV closed cycle still gives a positive difference.

The level scheme of ^{229}Pa is best studied by measuring the radiations associated with the EC decay of the 58-min ^{229}U as done in Ref. [3]. However, it is quite difficult to produce a sufficient quantity of mass-separated ^{229}U because of the safety issues associated with radioactive targets. For this reason, no measurement of ^{229}U EC decay has been published since our experiments in the 1980s. In this article we present new experimental results on ^{229}U EC decay which are quite extensive and well established.

II. SOURCE PREPARATION

The nuclide ^{229}U was produced by ^{229}Th(^{4}He,4n) and ^{230}Th(^{3}He,4n) reactions. Milligram quantities of Th targets were irradiated with microampere currents of ^{4}He and ^{3}He ions at the Argonne 152-cm cyclotron. The uranium produced in the reaction was chemically isolated and run through the Argonne electromagnetic isotope separator [11]. For some measurements, where a large quantity of ^{229}U was needed, the chemically purified uranium fraction without mass separation was used. These sources had ^{230}Th and Pa isotopes as impurities because these elements could not be completely removed from the source due to the fast chemical separation needed for the short-lived ^{229}U. Mass separation reduced the ^{229}U activity produced at the end of irradiation by a factor of 100: a factor of 10 due to the ~10% isotope separator efficiency and a factor of ~8 due to the decay.

III. EXPERIMENTAL METHODS AND RESULTS

A. γ-ray spectroscopy

Gamma rays in the EC decay of ^{229}U were measured with a 5-cm^2 $\times$ 10-mm low-energy photon spectrometer (LEPS) and a 15% coaxial Ge(Li) detector, both placed in a very low-background shield. Both mass-separated and unseparated ^{229}U sources were used in these studies. The LEPS detector had a resolution [full width at half-maximum (FWHM)] of 600 eV at 122-keV energy. With this resolution, energies of low-energy strong γ rays were measured with an accuracy of ± 10 eV. In the measurement before the publication of our article [3], we did not make efforts to achieve a high precision in γ-ray energies. It was during data analysis that it was realized that a parity doublet energy of 0.2 keV could be derived from closed cycles of γ rays. We, therefore, analyzed the γ-ray spectra more carefully later and determined the energies with a higher precision. We checked the precision of our measurement by comparing the energies of Pa K x-ray lines with the precise energies measured with a curved crystal spectrometer [12]. We find that the difference between our measured K x-ray energies and those listed in the literature is, on average, less than 10 eV.

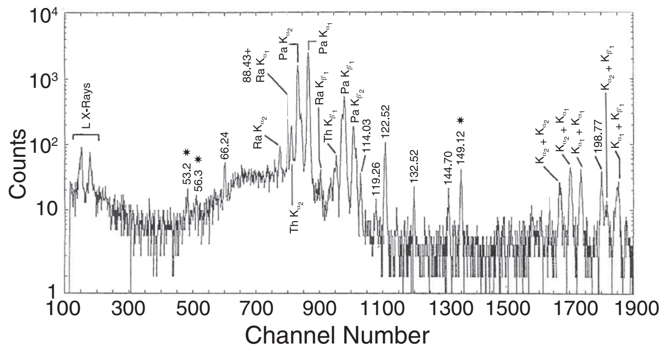

FIG. 1. The γ-ray spectrum of a mass-separated ^{229}U source measured with a 5-cm^2 $\times$ 10-mm LEPS detector. γ-ray energies (in keV) are indicated by the peaks. An asterisk denotes γ rays belonging to ^{225}Th or ^{221}Ra α decay.

For all the well-defined lines below 250 keV, the uncertainties are less than 40 eV.

Several γ-ray spectra of mass-separated and unseparated ^{229}U sources were used to determine energies and intensities. A γ-ray spectrum measured with the 5-cm^2 $\times$ 10-mm LEPS detector is shown in Fig. 1. The source for this spectrum was chemically purified after the mass separation. In this spectrum, in addition to the Pa K x-ray lines, Pa K x-ray–Pa K x-ray sum peaks are present. This shows that the ^{229}U decay generates high intensities of K conversion electrons, which cause the production of Pa K x rays. Additional evidence for their being sum peaks comes from the fact that the intensities of these peaks relative to that of the $K_{\alpha 1}$ peak are lower in the spectrum measured with the more efficient 15% Ge(Li) detector than those obtained from the spectrum measured with the LEPS detector. A spectrum of an unseparated ^{229}U source measured with the 15% Ge(Li) detector is displayed in Fig. 2. This spectrum has much higher counts in the peaks than the spectrum measured with the LEPS detector and was used to determine the energies of weaker transitions and higher-energy γ rays. There are several γ rays with energies around 240 keV. These γ rays are better resolved in the spectrum measured with the LEPS detector, shown in Fig. 3. The uncertainties in the energies of γ rays measured with the LEPS detector are between 20 and 40 eV and those measured with the Ge(Li) detector are between 50 and 300 eV. The γ-ray singles spectra also contained γ rays from the ^{229}U decay products. Gamma rays from the EC decay daughter ^{229}Pa (1.5 d) were studied in Ref. [4] and are too weak to be observed in the measured spectra. Gamma rays in the decay of the α-decay daughter ^{225}Th (8.72 min) and the granddaughter ^{221}Ra (28 s) have been reported in Ref. [13]. Gamma rays were assigned to the ^{229}U EC decay on the basis of their presence in the spectrum of a mass-separated ^{229}U source, their presence in the spectrum measured in coincidence with Pa K x rays, and their half-lives, and in Tables I and II, these are denoted by the letters m, c, and d, respectively. For some very weak γ rays, listed at the end of Table I, half-lives could not be determined. They may belong to the ^{229}U EC decay because they did decay with the short half-life of ~1 h.

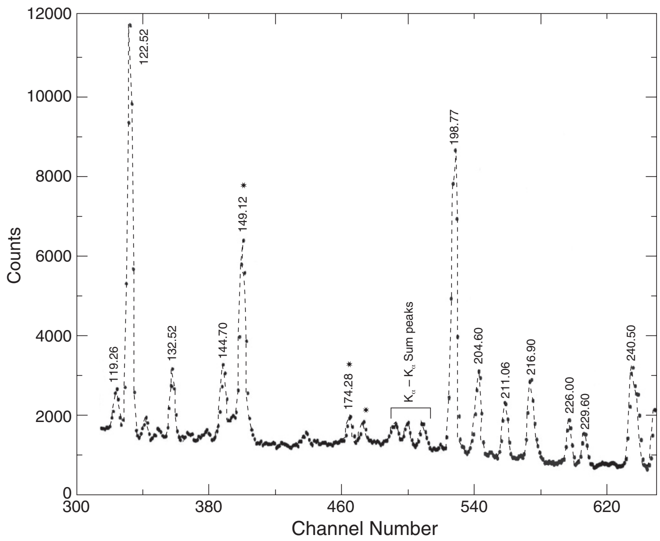

FIG. 2. The γ-ray spectrum of an unseparated ^{229}U source measured with a 15% Ge(Li) detector. A set of Cu and Al absorbers was used to reduce the counts in the low-energy γ-ray and x-ray peaks, thus reducing γ-γ summing. γ-ray energies (in keV) are indicated by the peaks. Counting was started ~2 h after the end of irradiation. An asterisk denotes γ rays belonging to ^{225}Th or ^{221}Ra α decay.

The energies of the ^{229}Pa γ rays (in keV) produced in the EC decay of ^{229}U and their intensities (in % per ^{229}U EC decay) are listed in Table I. Absolute intensities were obtained by normalizing the sum of all EC decays to 100%. These intensities depend on the assumed multipolarities of transitions and thus on the spin-parity assignments to the ^{229}Pa levels.

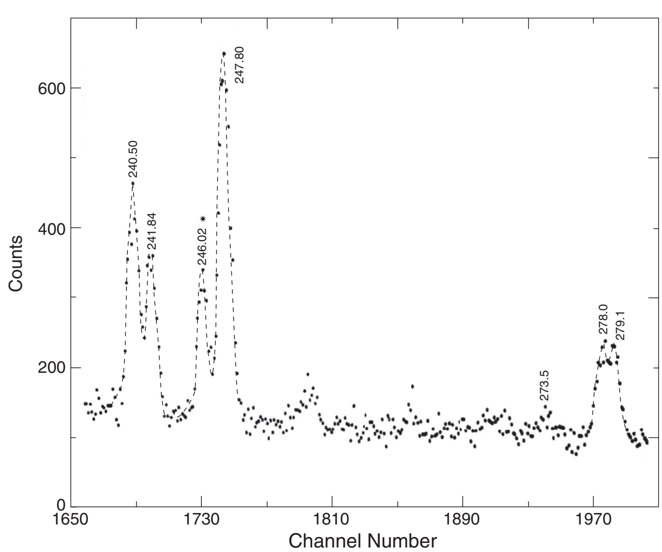

FIG. 3. The γ-ray spectrum of an unseparated ^{229}U source measured with a 5-cm^2 × 10-mm LEPS detector showing γ rays in the 240-keV region. A set of Cu and Al absorbers was used to reduce the counts in the low-energy γ-ray and x-ray peaks, thus reducing γ-γ summing. γ-ray energies (in keV) are indicated by the peaks. Counting was started ~2 h after the end of irradiation. An asterisk denotes γ rays belonging to ^{225}Th α decay.

Gamma rays associated with the decays of ^{225}Th and ^{221}Ra are listed in Table II. These γ rays were not present in the spectrum measured in coincidence with Pa K x rays, indicating that they do not belong to the EC decay of ^{229}U. The energies and intensities of these γ rays are in excellent agreement with the published values [13].

B. γ-γ coincidence measurements

A γ-γ coincidence measurement was performed with an unseparated ^{229}U source and a 5-cm^2 × 10-mm and a 20% LEPS detector. Coincidence events were recorded in event-by-event mode and γ-ray spectra were later generated by gating on γ-ray peaks. A γ-ray spectrum produced by placing the gate on the Pa K x rays is displayed in Fig. 4. In this spectrum, K x-ray–K x-ray sum peaks are absent and hence peaks near 200 keV can be easily identified. Also, the Th K x rays from ^{229}Pa EC decay and the γ rays following the α decays of ^{229}U and its daughters are absent in this spectrum. Since most ^{229}U γ rays have low intensities, only strong γ rays were seen in the gated spectra.

C. Electron spectroscopy

Internal conversion electron lines in the EC decay of ^{229}U have low intensities; the strongest line (122.5 L_1 line) has an intensity of only 5.5% per ^{229}U EC decay. The source activity at the beginning of the measurement was only ~0.5% of the original activity at the end of the irradiation due to the mass separation and the time required to cool the Si(Li) detector. Hence, only strong transitions were observed in the electron spectrum. An advantage of using electron spectra is that, because the γ-ray energies are known, energies of their conversion electron lines are also known. Thus, one knows where to look for the peaks in the spectrum, and consequently, even the limit on the intensity can be used to determine the transition multipolarity. The conversion-electron spectrum of a mass-separated ^{229}U source was measured with a cooled Si(Li) detector [14] and the γ-ray spectrum of the same source was measured with a LEPS detector. The conversion coefficients of strong transitions in ^{229}U EC decay were determined using a ^{57}Co source as a standard, whose electron and γ-ray spectra were measured with the same detectors and at the same solid angles as the ^{229}U source.

The electron singles spectrum was also measured with an unseparated ^{229}U source. This source had an order of magnitude more ^{229}U activity than the mass-separated source but it contained impurities as mentioned earlier. Electron lines belonging to ^{229}U decay were identified using a spectrum obtained by subtracting a later spectrum from an early one. This difference spectrum, shown in Fig. 5, contains only lines which have decayed in the 3-h time interval between the measurements of the two spectra. Although the 198.8 K and 122.5 L lines stand out, the rest of the spectrum is quite complex. The region between 86 and 100 keV contains K x rays. The Si(Li) detector has an ~1 % efficiency for 100-keV photons and the electrons at this energy lose ~2 keV in the source and the detector window. For this reason, the K x-ray peak energies were ~2 keV higher

TABLE I. Gamma rays in ^{229}Pa from EC decay of ^{229}U measured in the present work. m: the γ ray was observed in a mass-separated source. c: the γ ray was observed in coincidence with Pa K x rays. d: the γ ray decayed with an $\sim$1-h half-life.

Energy (keV)	Intensity (% per ^{229}U EC decay)	Transition (keV; initial → final)	Remark(s)
66.24 ± 0.03	0.58 ± 0.07	210.95 → 144.70	m,c,d
88.43 ± 0.02	2.10 ± 0.20	210.95 → 122.52	m,c,d
92.28 ± 0.01	34 ± 2	Pa $K_{\alpha2}$	m,c,d
95.86 ± 0.01	53 ± 3	Pa $K_{\alpha1}$	m,c,d
107.59 ± 0.02	6.2 ± 0.4	Pa $K_{\beta3}$	m,c,d
108.42 ± 0.02	12.0 ± 0.7	Pa $K_{\beta1}$	m,c,d
111.50 ± 0.02	4.6 ± 0.3	Pa $K_{\beta2}$	m,c,d
112.37 ± 0.03	1.8 ± 0.2	Pa $KO_{2,3}$	m,c,d
114.03 ± 0.03	0.50 ± 0.04	144.70 → 30.67	m,c,d
119.26 ± 0.03	0.26 ± 0.02	241.80 → 122.52	m,c,d
122.52 ± 0.02	2.60 ± 0.13	122.52 → 0.0	m,c,d
132.52 ± 0.03	0.38 ± 0.03	144.70 → 12.20	m,c,d
144.70 ± 0.03	0.42 ± 0.03	144.70 → 0.0	m,c,d
184.2 ± 0.2	0.20 ± 0.05	210.95 → 26.7	c
198.77 ± 0.03	2.30 ± 0.13	210.95 → 12.20	m,c,d
204.60 ± 0.03	0.65 ± 0.05	304.7 → 100.1	m,c,d
211.06 ± 0.04	~0.13[a]	210.95 → 0.0	m,c,d
	~0.32[a]	241.80 → 30.67	m,c,d
216.90 ± 0.04	0.65 ± 0.05	458.75 → 241.80	m,c,d
226.00 ± 0.04	0.34 ± 0.03	252.7 → 26.7	m,c,d
229.60 ± 0.04	0.26 ± 0.02	241.80 → 12.20	m,c,d
240.50 ± 0.04	0.80 ± 0.06	252.7 → 12.20	m,c,d
241.84 ± 0.04	0.59 ± 0.05	241.80 → 0.0	m,c,d
247.80 ± 0.04	1.54 ± 0.10	458.75 → 210.95	m,c,d
273.5 ± 0.2	~0.10	285.7 → 12.2	
278.0 ± 0.1	0.42 ± 0.04	304.7 → 26.7	m,c,d
279.1 ± 0.1	0.42 ± 0.04	490.05 → 210.95	m,c,d
286.2 ± 0.1	0.12 ± 0.01	745.2 → 458.75	c,d
292.8 ± 0.1	0.15 ± 0.02	304.7 → 12.20	c,d
314.0 ± 0.1	0.11 ± 0.02	458.75 → 144.70	c,d
336.23 ± 0.06	0.22 ± 0.02	458.75 → 122.52	m,c,d
340.2 ± 0.3	0.027 ± 0.007	799.3 → 458.75	
345.2 ± 0.1	0.08 ± 0.01	490.05 → 144.70	d
350.80 ± 0.10	0.21 ± 0.02	561.72 → 210.95	m,c,d
425.7 ± 0.3	0.12 ± 0.012	884.2 → 458.75	d
446.60 ± 0.08	0.16 ± 0.02	458.75 → 12.20	m,c,d
458.82 ± 0.10	0.05 ± 0.01	458.75 → 0.0	
477.89 ± 0.10	0.05 ± 0.01	490.05 → 12.20	d
490.07 ± 0.10	0.032 ± 0.008	490.05 → 0.0	
497.2 ± 0.3	0.031 ± 0.008	1058.2 → 561.72	
534.3 ± 0.2	0.04 ± 0.01	745.2 → 210.95	d
549.50 ± 0.10	0.52 ± 0.05	561.72 → 12.20	m,c,d
557.1 ± 0.3	0.04 ± 0.01	799.3 → 241.80	
600.5 ± 0.2	0.09 ± 0.01	745.2 → 144.70	c,d
622.6 ± 0.2	0.22 ± 0.02	745.2 → 122.52	m,c,d
644.9 ± 0.3	0.027 ± 0.007	745.2 → 100.1	d
654.7 ± 0.3	0.030 ± 0.008	799.3 → 144.70	
733.0 ± 0.2	1.23 ± 0.10	745.2 → 12.20	m,c,d
761.2 ± 0.3	0.031 ± 0.008	884.2 → 122.52	d
787.2 ± 0.3	0.016 ± 0.003	799.3 → 12.20	
799.6 ± 0.3	0.05 ± 0.01	799.3 → 0.0	d
847.1 ± 0.3	0.09 ± 0.02	1058.2 → 210.95	d
884.2 ± 0.3	0.07 ± 0.01	884.2 → 0.0	d
1058.6 ± 0.3	0.06 ± 0.01	1058.2 → 0.0	d
414.5 ± 0.4	0.05 ± 0.01	Not placed in level scheme	d
433.6 ± 0.4	0.04 ± 0.01	Not placed in level scheme	d

TABLE I. (*Continued.*)

Energy (keV)	Intensity (% per ^{229}U EC decay)	Transition (keV; initial → final)	Remark(s)
465.8 ± 0.4	0.03 ± 0.01	Not placed in level scheme	
543.3 ± 0.4	0.08 ± 0.01	Not placed in level scheme	d
552.6 ± 0.4	0.07 ± 0.01	Not placed in level scheme	d
663.0 ± 0.4	0.03 ± 0.01	Not placed in level scheme	
689.0 ± 0.4	0.04 ± 0.01	Not placed in level scheme	
712.4 ± 0.4	0.04 ± 0.01	Not placed in level scheme	
717.1 ± 0.4	0.06 ± 0.01	Not placed in level scheme	
749.4 ± 0.4	0.03 ± 0.01	Not placed in level scheme	

[a]The total intensity of the 211.06-keV γ ray is 0.45 ± 0.04% per ^{229}U EC decay.

than their real values relative to the electron energy. The 98.0-keV peak, which has the right energy for the 211.0 K electron line, also has the correct energy for the Pa $K_{\alpha 1}$ peak. Since we could not determine the contribution of the K x-ray component to the peak counts, the 98.0-keV peak was not used for the determination of the conversion coefficient.

In the spectrum shown in Fig. 5(b), the 246.0 K line of ^{221}Ra and the 149.1 L and M lines of ^{217}Rn are clearly visible. The multipolarities of these transitions are known [13] to be $M1$ and $E2$, respectively. As Fig. 3 shows, the 240.50-, 241.84-, and 247.80-keV γ rays have higher intensities than the 246.02-keV γ ray. Hence, if these transitions were $M1$, we would see their K electron lines as intense as the 246.0 K line. We do not see the K lines of these γ rays, which indicates that these are not $M1$ transitions. They could have $E1$ or $E2$ multipolarity. In heavy nuclei, $E2$ transitions between single-particle states are slower than $E1$ transitions. Hence, we assign $E1$ multipolarity to the 240.50-, 241.84-, and 247.80-keV transitions.

The electron spectrum of a mass-separated ^{229}U source was also measured in coincidence with Pa K_α x rays and is displayed in Fig. 6. Although this spectrum has low counts, it is very clean, and all strong ^{229}U electron lines seen in the singles spectrum are clearly visible. The intensities obtained from this spectrum have larger uncertainties but they agree with the values obtained from the singles spectrum. The 98.0-keV line, seen in the singles electron spectrum, is present in this spectrum also, but as mentioned earlier we cannot use it to determine the multipolarity of the 211.0-keV transition because it contains the Pa K x-ray line. There is an electron line at 89.8 keV in the spectrum which is assigned to the 110.9-keV transition between the 210.95- and the 100.1-keV levels. This γ ray is masked by the Pa $K_{\beta 2}$ line and hence its intensity was not measured. The electron intensities from all these spectra and the conversion coefficients deduced from them are listed in Table III along with the theoretical conversion coefficients [15]. From the upper limit on the intensities of electron lines we deduce the multipolarity of 66.24- and 88.43-keV transitions as $E1$. Also included in Table III are the electron intensities and conversion coefficients for transitions in the α decays of ^{225}Th and ^{221}Ra, and these are in agreement with literature values [13]. These electron intensities are normalized to the intensity of the 321.35-keV γ ray as 100.

TABLE II. ^{221}Ra γ rays from the α decay of ^{225}Th measured in the present work. m: the γ ray was observed in the spectrum of a mass-separated ^{229}U source. d: the γ ray decayed with a ~58-min half-life.

Energy (keV)	Intensity	Transition (keV; initial → final)	Remark(s)
53.2 ± 0.1	7.5 ± 0.6	53.14 → 0.0	m
56.3 ± 0.1	4.8 ± 0.6[a]	149.18 → 93.02	^{217}Rn, m
146.82 ± 0.08	3.4 ± 0.3	146.81 → 0.0	m,d
149.12 ± 0.03	35 ± 3	149.18 → 0.0	^{217}Rn, m,d
174.28 ± 0.08	5.8 ± 0.6	174.3 → 0.0	^{217}Rn, m,d
177.27 ± 0.08	4.9 ± 0.5	299.16 → 121.95	m,d
246.02 ± 0.05	18 ± 2	299.16 → 53.14	m,d
299.29 ± 0.10	5.8 ± 0.6	299.16 → 0.0	d
305.85 ± 0.08	18 ± 2	359.02 → 53.14	m,d
321.35 ± 0.08	100 (norm)	321.37 → 0.0	m,d
359.05 ± 0.08	20 ± 2	359.02 → 0.0	m,d
382.00 ± 0.10	2.5 ± 0.3	485.40 → 103.60	d
485.7 ± 0.1	0.9 ± 0.2	485.40 → 0.0	

[a]This intensity is higher than the value given in Ref. [13].

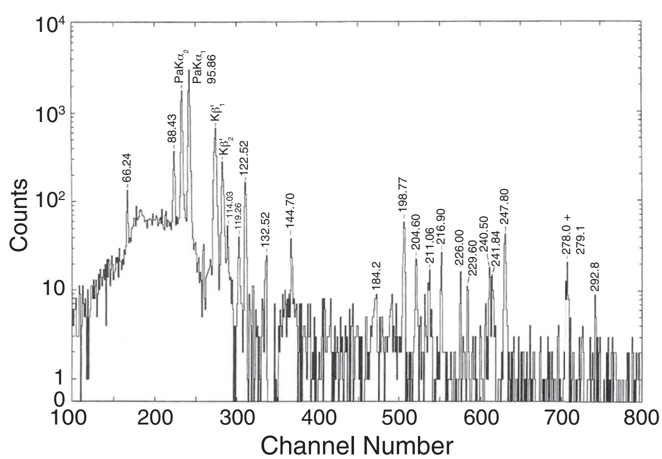

FIG. 4. The γ-ray spectrum of an unseparated ^{229}U source measured with a 5-cm^2 × 10-mm LEPS detector in coincidence with Pa K_α x rays. γ-ray energies (in keV) are indicated by the peaks. The K_α x rays were detected with a 20% LEPS spectrometer.

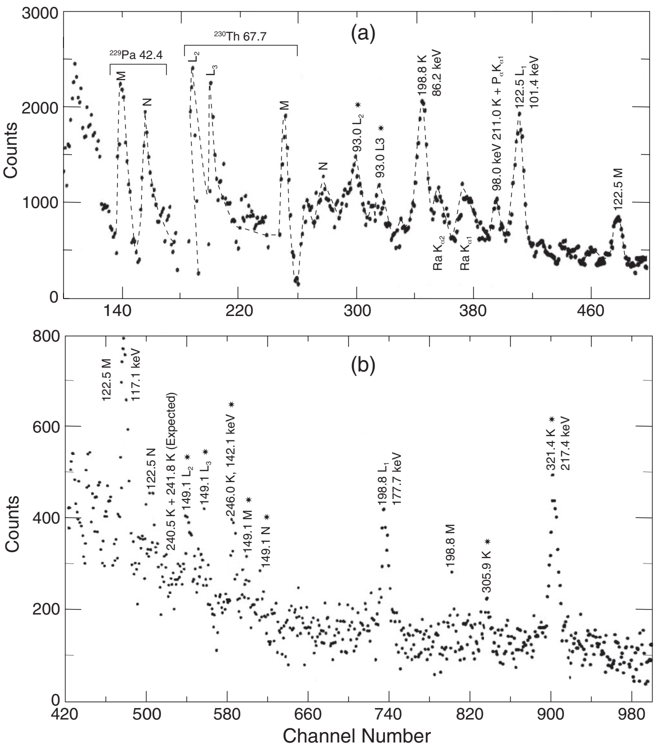

FIG. 5. Electron spectrum of an unseparated ^{229}U source measured with an 80-mm^2 $\times$ 3-mm cooled Si(Li) spectrometer with a resolution (FWHM) of 1.0 keV at 100-keV electron energy. The spectrum was generated by subtracting a later spectrum from an early spectrum. Strong electron lines from ^{230}Th and ^{229}Pa were present in both spectra. Electron and/or transition energies (in keV) are indicated by the peaks. An asterisk denotes electron lines belonging to ^{225}Th or ^{221}Ra α decay.

IV. DISCUSSION

A. Level scheme

In electron capture or β^- decay, level energies in the daughter nucleus are deduced from sums and differences of energies of the observed γ-ray transitions. In the absence of coincidence and transition multipolarity information, different level sequences can be postulated as done by us [3,16]. However, with γ-γ coincidence information, one can generally determine the correct level orderings. We have used the criterion that an energy level is established only when two observed γ rays are associated with that level, either de-exciting it or populating it.

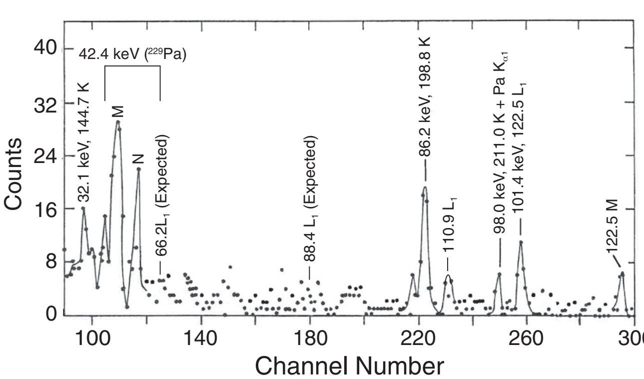

FIG. 6. Electron spectrum of a mass-separated ^{229}U source measured with a cooled Si(Li) spectrometer in coincidence with Pa K_α x rays. Electron and/or transition energies (in keV) are indicated by the peaks.

By balancing the total EC, γ-ray, and internal conversion electron intensities feeding each level, and the γ-ray and conversion electron intensities de-exciting it, we determine a $40 \pm 3\%$ EC population to the ^{229}Pa ground state and nearby states. Using the measured $M1$ multipolarity for the 122.52-keV transition, we deduce its transition intensity as $32 \pm 3\%$, which leaves only 28% intensity for the remaining γ rays. The γ-ray transitions in coincidence with the 122.52-keV γ ray have a total intensity of $<5\%$ and hence the 122.52-keV transition must decay to the ground state or to a nearby state. We interpret the 122.52-keV γ ray de-exciting a 122.52-keV level to the ground state because, as we show later, with this assignment all γ-ray transitions fit in a level scheme. In coincidence with the 122.52-keV γ ray, we observe 88.43- and 119.26-keV γ rays establishing levels at 210.95 and 241.80 keV, shown in Fig. 7. A gate placed on the 88.43-keV

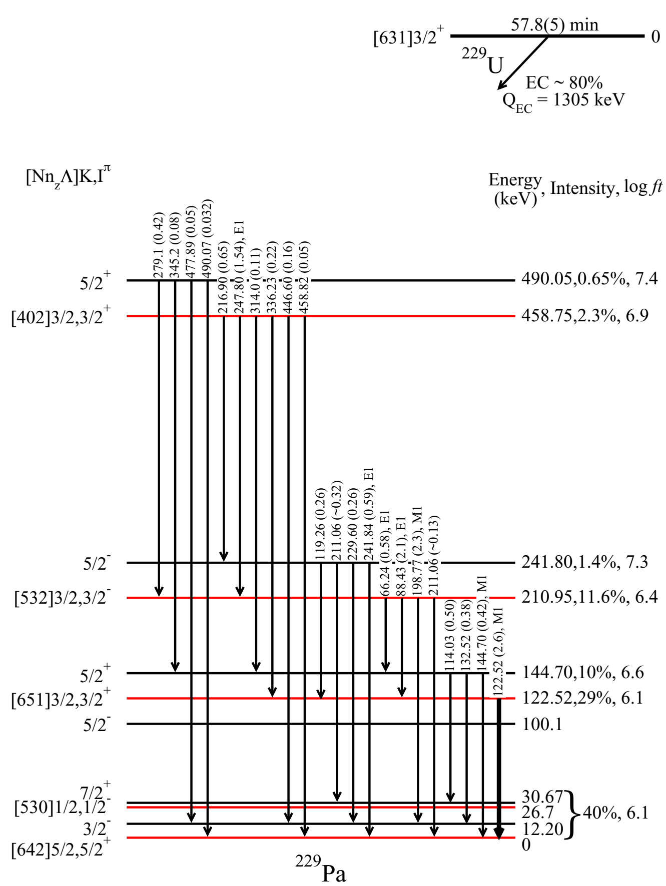

FIG. 7. (Color online) Low-energy portion of the ^{229}Pa level scheme. γ-ray energies are given in kilo–electron volts, and γ-ray intensities (in % per ^{229}U EC decay) are given in parentheses. The half-life in the figure was measured during the course of this work. Gray (red) horizontal lines represent the band heads.

TABLE III. Summary of ^{229}Pa electron lines in the EC decay of ^{229}U. The high intensity of the 122.5 M line could be due to the presence of the 229.60 K line.

Transition energy (keV)	Shell	Electron intensity (%)	Conversion coefficient	Theoretical conversion coefficient $E1$	$E2$	$M1$	Multipolarity
66.2	$L_1 + L_2$	<3.0	<5	0.19	36	10.4	$E1$
88.4	$L_1 + L_2$	<3.0	<2	0.09	9.4	4.5	$E1$
110.9[a]	$L_1 + L_2$	~1.0	—	0.054	3.4	2.3	$M1$
122.5	$L_1 + L_2$	5.0 ± 0.5	1.9 ± 0.2	0.042	2.2	1.75	$M1$
	$M_1 + M_2$	1.6 ± 0.2	0.6 ± 0.1	0.013	0.97	0.43	
	$N + O$	0.30 ± 0.04	0.12 ± 0.02	0.0043	0.32	0.14	
144.7	K	2.3 ± 0.6	5.5 ± 1.5	0.16	0.24	5.7	$M1$
198.8	K	5.5 ± 0.7	2.40 ± 0.33	0.077	0.158	2.3	$M1$
	$L_1 + L_2$	1.0 ± 0.2	0.43 ± 0.09	0.013	0.28	0.44	
149.1 (^{221}Ra)	$L_1 + L_2$	16 ± 4	0.46 ± 0.12	0.006	0.55	0.61	$E2$
149.1 (^{221}Ra)	L_3	12 ± 3	0.34 ± 0.09	0.005	0.35	0.004	$E2$
246.0 (^{225}Th)	K	15 ± 2	0.83 ± 0.14	0.044	0.11	0.99	$M1$
305.9 (^{225}Th)	K	~14	~0.8	0.027	0.068	0.64	$M1$
321.4 (^{225}Th)	K	44 ± 5	0.44 ± 0.05	0.025	0.062	0.47	$M1$
359.1 (^{225}Th)	K	~8	~0.4	0.019	0.049	0.35	$M1$

[a]The 110.9-keV transition is a 210.95 → 100.1 transition. No γ ray was seen.

peak shows the presence of the 122.52- and a 247.80-keV γ ray, indicating that the 210.95-keV level is fed by a 247.80-keV γ-ray transition. In coincidence with the 241.84-keV γ ray we observe a 216.90-keV γ ray, suggesting that the 241.80-keV level is populated by a 216.90-keV γ ray. By gating on the 247.80-keV γ-ray peak, we observe 66.24-, 88.43-, 198.77-, and 211.06-keV γ rays. By placing a gate on the 66.24-keV γ-ray peak we observe 114.03-, 132.52-, 144.70-, and 247.80-keV γ rays. Since the 66.24-keV γ ray originates at the 210.95-keV level, these coincidence γ rays define levels at 144.70, 12.20, and 30.67 keV. The presence of the 12.20-keV level is further supported by the observation of other pairs of γ rays with this energy difference. Thus, the coincidence measurements establish levels at 0.0, 12.20, 30.67, 122.52, 144.70, 210.95, 241.80, and 458.75 keV.

B. Spin-parity and configuration assignment

The ground-state spin of ^{229}Pa has been established as 5/2 on the basis of its EC and α decay [4] and the β^- decay of ^{225}Ra. The log ft values of ^{229}Pa EC transitions and the level spacings in the rotational bands of ^{225}Ac favor a $5/2^+[642]$ assignment. However, the $5/2^-[523]$ single-particle assignment is also possible.

The ground-state spin of ^{229}U is most likely 3/2, with the configuration $3/2^+[631]$ as determined from its α decay [13,17]. Since $\Delta K = 0, \pm 1$ and $\Delta I = 0, \pm 1$ EC transitions have the highest decay rates, only states with spin 1/2, 3/2, and 5/2 in ^{229}Pa are expected to receive a measurable EC population. The decay data establish a level at 12.20 keV. We assign it to the 3/2, $1/2^-[530]$ configuration because this energy is very close to the energy of 12 ± 5 keV measured by the ^{231}Pa(p,t) reaction [5] and theory [1] predicts its 1/2 member at 40 keV. The 3/2–1/2 and 5/2–3/2 energy differences for this band were measured as 15.1 ± 0.4 and 87.7 ± 0.3 keV in the (p,t) reaction [8]. From the decay of the $1/2^+$ and $3/2^+$ states at 252.7 and 304.7 keV (Fig. 8), we deduce these energy differences as 14.5 ± 0.1 and 87.9 ± 0.1 keV, respectively, in excellent agreement with the (p,t) reaction data.

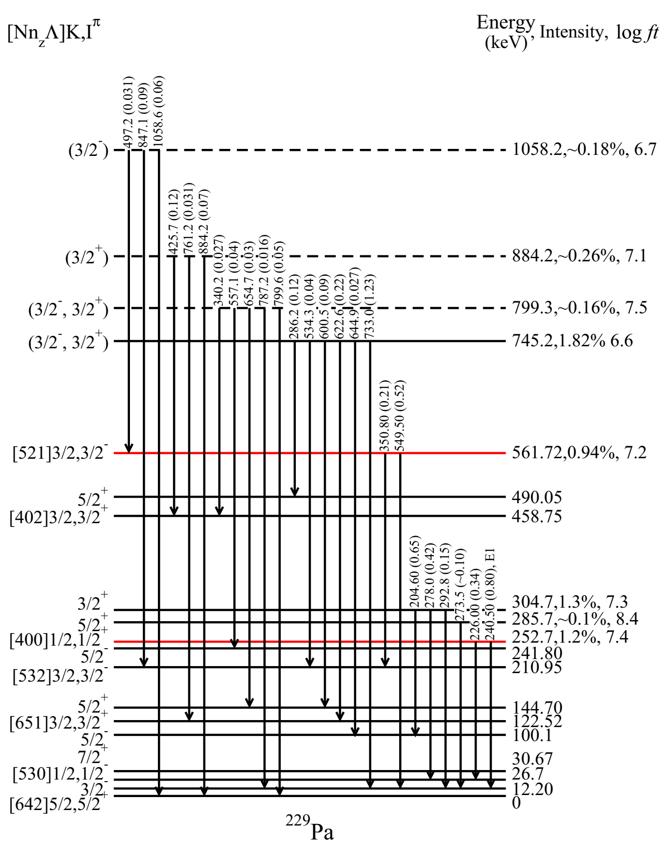

FIG. 8. (Color online) High-energy portion of the partial EC decay scheme of ^{229}U deduced in the present work. Gray (red) horizontal lines represent the band heads.

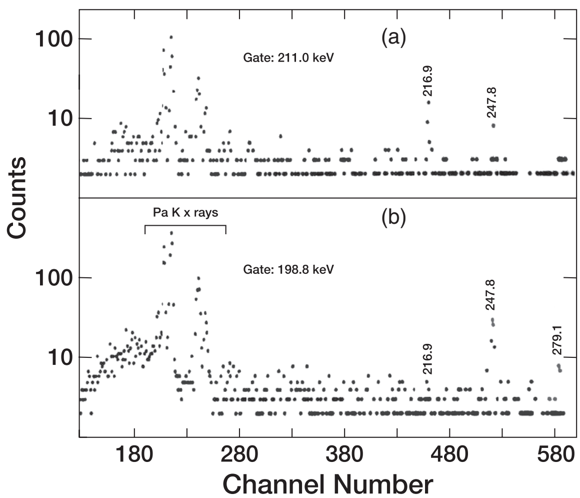

FIG. 9. The γ-ray spectra of a ^{229}U source measured in coincidence with 211.0-keV (top) and 198.8-keV (bottom) γ rays. γ-ray energies (in keV) are indicated by the peaks. The upper spectrum shows that most of the 211.0-keV γ-ray intensity originates at the 241.80-keV level (see Fig. 7).

The coincidence relationship establishes a level at 210.95 keV which decays to the 12.20-keV level by a 198.77-keV $M1$ transition. Thus the parity of the 210.95-keV level must be negative. We assign a $3/2^-$ spin parity to this state and a configuration of $3/2^-[532]$. The 241.80-keV level is interpreted as the $5/2^-$ member of this band because of the similarity of the level spacing in this band to the spacing in the $3/2^-$ band of ^{225}Ac [4] and ^{231}Pa [18]. The 210.95-keV level decays to the 122.52- and 144.70-keV levels by 88.43- and 66.24-keV $E1$ transitions, respectively, and hence these two states should have positive parity. We tentatively assign these levels to the 3/2 and 5/2 members of the $3/2^+[651]$ band. The level spacing in this band is comparable to the spacing in the $3/2^+$ band in ^{225}Ac. The $M1$ multipolarity of the 122.52- and 144.70-keV transitions establishes positive parity for the ground state and we assign it to the $5/2^+[642]$ single-particle configuration.

The 30.67-keV level, populated by the 114.03-keV γ ray, could be either the $5/2^-[523]$ single-particle state or the $7/2^+$ member of the ground-state band. The latter assignment is more likely because this level is expected around this energy and the intensity of the 114.03-keV γ ray is in agreement with the value calculated by the Alaga rules [19]. A measurement of the 114.03-keV transition multipolarity is needed to confirm this assignment. The transition energy from the 241.80- to the 30.67-keV level is 211.13 keV, whereas the energy from the 210.95-keV level to the ground state is 210.95 keV. These two γ rays are indistinguishable and the 211.06-keV peak in the γ singles spectrum contains both transitions. The contribution from each transition was determined from the coincidence data. Figure 9 shows two spectra gated by the 198.77- and 211.06-keV γ rays. The spectra have a large amount of Pa K x rays because the detector efficiency at the K x-ray energy is $\sim$4 times the efficiency at 200 keV and the background spectrum was not subtracted. If there were only one γ-ray transition of 210.95 keV, both spectra would be identical because both the 198.77- and the 210.95-keV γ rays originate at a common level at 210.95 keV. However, the two spectra are quite different. The data show that $\sim$70% of the 211.06-keV peak counts arise from the decay of the 241.80-keV level. For this reason the measured energy of the γ-ray peak in the spectrum is higher than 210.95 keV, giving the positive number in the $211.06 - (122.52 + 88.43)$ cycle.

Other levels are deduced from the γ-γ coincidence measurements and γ rays listed in Table I. In coincidence with the 88.43-keV peak we observe 247.80-, 279.1-, and 350.80-keV γ rays which require levels at 458.75, 490.05, and 561.72 keV. The multipolarity of the 247.80-keV γ ray has been measured as $E1$, and hence the 458.75-keV level should have positive parity. We assign the 458.75- and 490.05- keV levels to the 3/2 and 5/2 members of the $3/2^+[402]$ band, which is calculated [1] to be at 460 keV. The 561.72-keV level is given a tentative assignment of the $3/2^-[521]$ particle state because this level was observed in ^{231}Pa [20] at 604 keV. The $1/2^+[400]$ state, which is predicted [1] to be at 200 keV, was observed by Levon *et al.* [8] at 240.3 ± 0.5 keV relative to the energy of the 1/2, $3/2^-$ level and its 3/2 and 5/2 members were identified at 291.4 ± 0.3 and 272.5 ± 0.3 keV. We have observed a 240.50-keV γ ray which gives the energy of the $1/2^+$ level as 252.7 keV. A 226.00-keV γ ray is observed which is interpreted as the $252.7 \rightarrow 26.7$ transition, the 26.7-keV level being the 1/2 member of the $1/2^-[530]$ band. Three γ rays, of energy 204.60, 278.0, and 292.8 keV, define a level at 304.7 keV, which is interpreted as the $3/2^+$ member of the $1/2^+[400]$ band. Also, a weak 273.5-keV γ ray was observed, which has been assigned to the $285.7 \rightarrow 12.2$ decay, the 285.7-keV level being the $5/2^+$ member of the $1/2^+[400]$ band. The measured $E1$ multipolarity of the 240.50-keV transition further supports this assignment.

A level at 745.2 keV is proposed on the basis of γ rays connecting it to lower levels. This level decays predominantly to the 1/2, $3/2^-$ level at 12.20 keV and also to the $3/2^+$ and $5/2^+$ levels. This level could therefore be either the 1/2 or the 3/2 member of the $1/2^+[651]$ rotational band or the 0^+ state built on the 1/2, $3/2^-$ level. The β band is known in the neighboring ^{228}Th at 832 keV [21]. Three weakly populated levels, at 799.3, 884.2, and 1058.2 keV, are also shown as dashed lines in the level scheme. Their tentative spins and parities are shown in Fig. 8.

The level scheme deduced in this work is compared with the predictions of Chasman [1] in Fig. 10. The observed level ordering is, in general, in good agreement with theory, providing additional support for the proposed level scheme. The energy levels in ^{229}Pa are compared with the energy levels in ^{231}Pa in Fig. 11. Since both isotopes contain 91 protons, their level structures should be similar. However, there is a big change in the two nuclei; the $5/2^+$ level drops drastically in ^{229}Pa, indicating a difference in the structures of the two nuclei, in agreement with theory.

C. Gamma-ray transition probabilities

One of the observed properties associated with octupole correlations or octupole deformation is the enhancement of $E1$ transition rates. Since the center of charge and the center of

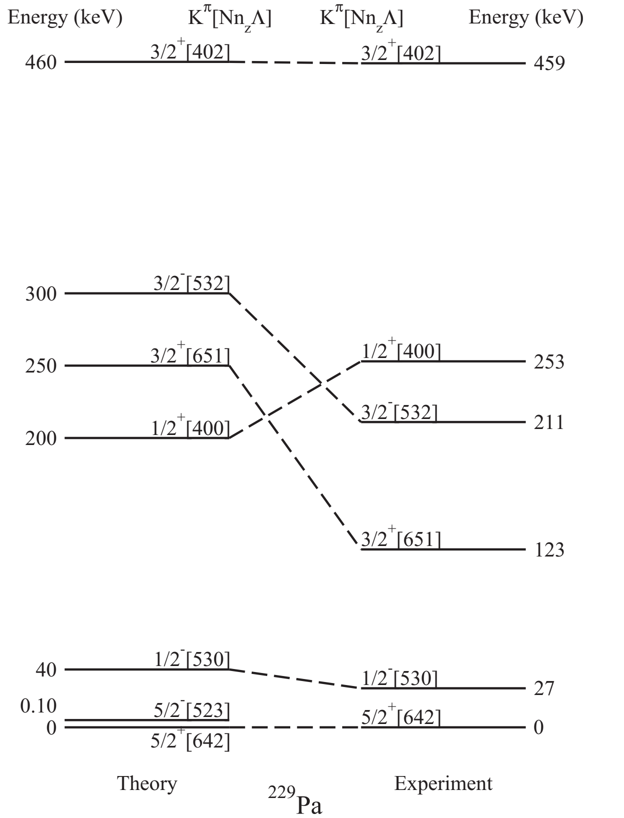

FIG. 10. A comparison of the ^{229}Pa energy levels deduced in the present work with the prediction of Chasman [1]. In Ref. [2], the energies of the $5/2^{+}[642]$, $5/2^{-}[523]$, and $1/2^{-}[530]$ single-particle states were calculated to be 0.0, 0.4, and 56 keV, respectively.

mass in an octupole shape do not coincide, they give rise to dipole and octuple moments which result in enhanced $B(E1)$ and $B(E3)$ values. It has been shown in nuclei in the $Z \sim 90$ region with octupole correlations that $E1$ rates between the members of the parity doublet are enhanced [22] over the respective values in Pu and heavier nuclei which have no octupole correlations. In nuclei with no octupole correlations, $M1$ transitions are found to be the fastest transitions. In ^{229}Pa, there are several enhanced $E1$ transitions (see Fig. 7). The ratio of the intensities of the 88.43- and 198.77-keV γ rays indicates that the energy-independent $E1$ rate is 10 times the energy-independent $M1$ rate. Similarly, the intensities of the 247.80- and 336.23-keV γ rays indicate that the $E1$ rate is 18 times the $M1$ rate. In the case of the $1/2^{+}$ band at 252.7 keV, we observe only $E1$ transitions. These enhanced $E1$ transition rates provide evidence for large octupole correlations in ^{229}Pa.

According to the Alaga rules [19], relative intensities of γ rays from any level to the members of a rotational band are proportional to the squares of the appropriate Clebsch-Gordan coefficients. Using the assignments in Figs. 7 and 8, we have calculated the relative intensities of γ rays de-exciting several levels. The excellent agreement between the measured and the calculated relative intensities further supports the single-particle assignments in Figs. 7 and 8.

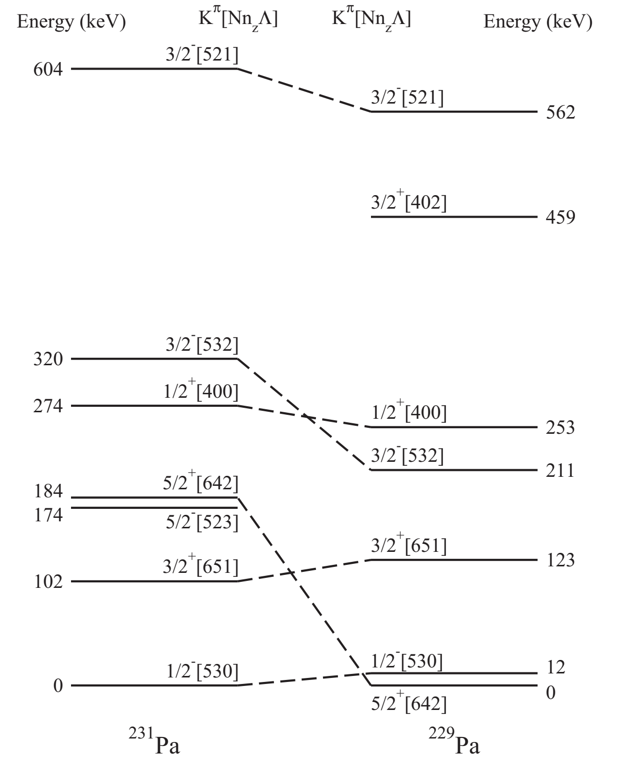

FIG. 11. A comparison of the experimental ^{229}Pa and ^{231}Pa energy levels. The figure shows that despite both nuclei having 91 protons, their structures are quite different.

D. Splitting energy of the $5/2^{\pm}$ doublet

So far the $5/2^{-}$ member of the parity doublet has not been identified. In this work we have established many excited states which would decay to the $5/2^{-}$ level expected below 80 keV from the systematics. The fact that we have placed all observed transitions in the level scheme presented here leaves no γ-ray transition which could be attributed to the decay to the $5/2^{-}$ level. From these observations we conclude that the $5/2^{-}$ level is almost degenerate with some level below 80 keV, possibly the ground state. The $5/2^{\pm}$ doublet in ^{229}Pa was identified in Ref. [3] from two closed cycles of γ rays. One of the closed cycles, $211.09 - (122.51 + 88.43)$, cannot be used because the 211.06-keV γ ray is a doublet. The other closed cycle, $241.84 - (122.52 + 119.26) = 60 \pm 50$ eV, still gives a positive number that makes $5/2^{-}$ the ^{229}Pa ground state. However, the large uncertainty in the energy difference makes the assignment of the $5/2^{-}$ level uncertain.

E. Summary

We have presented a large set of data on ^{229}U EC decay which establishes a detailed level scheme for ^{229}Pa. The decay scheme is constructed on the basis of γ-γ coincidence data and conversion electron measurements. The energy levels presented here are well supported by the data and single-particle assignments are in agreement with theoretical predictions. The fact that the $5/2^{+}[642]$ single-particle state is the ground state in ^{229}Pa, as predicted by theory [1], and the $E1$ transition rates are enhanced suggests large octupole correlations in ^{229}Pa. The presence of large octupole correlations in ^{229}Pa is also supported by the near-equality of the decoupling parameters for the $1/2^-$ band ($a = -1.63$) and the $1/2^+$ band ($a = +1.53$) [8], as predicted by theory [2]. However, the $5/2^-$[523] level, the other member of the parity doublet, has not been located. The data reported in this article make it possible to search for the $5/2^-$ member of the parity doublet. Since many γ rays have been shown to belong to the ^{229}U EC decay, mass separation is not necessary for detailed spectroscopy. A ^{229}U sample can be produced by the irradiation of a ^{229}Th target with α particles or by the irradiation of a ^{230}Th target with a ^{3}He beam. Chemical purifications can produce a quite pure ^{229}U sample, free of fission products and other actinide elements. The only other uranium isotope produced in the reactions which generates γ rays is the 4.2-d ^{231}U. However, transitions in ^{231}U decay are known [23] and can be easily distinguished from ^{229}U transitions because of their low intensities and long ^{231}U half-lives. The use of high-efficiency Ge detectors can improve the γ-ray sensitivity. Measurements that can be used to determine the energy of the $5/2^-$[523] level include the following:

(a) A precise measurement of γ-ray energies with a LEPS detector or a microcalorimeter so that the energy difference between the $5/2^-$ and the $5/2^+$ levels, if there is a ground-state doublet, could be measured with a higher precision.
(b) Measurement of the electron spectrum. The multipolarity of the 114.03-keV transition will determine whether the 30.67-keV level is $7/2^+$ or $5/2^-$. The energy of the 12.2 M conversion line for the $M1$ transition is $\sim$0.4 keV less than for $E1$ multipolarity. The $M1$ multipolarity will provide evidence for the $5/2^-$ state near ground. The low-energy conversion electrons could be detected with a high-resolution cooled 100-μm-thick silicon detector or with a magnetic spectrometer.
(c) A measurement of the lifetime of the 12.20-keV level. If a $5/2^-$ level is near the ground state, the lifetime of the 12.20-keV level will be short; otherwise it will be in the microsecond range.
(d) Study of the α decay of the 36.2-min ^{233}Np, which has the small α-decay branch of $\sim$0.001%. Even when the $5/2^+$ and $5/2^-$ levels in ^{229}Pa are degenerate, their $7/2$ members will have different energies and will receive a measurable α population.

ACKNOWLEDGMENTS

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics, under Contract No. DE-AC02-06CH11357.

[1] R. R. Chasman, Phys. Lett. B **96**, 7 (1980).
[2] G. A. Leander and Y. S. Chen, Phys. Rev. C **37**, 2744 (1988).
[3] I. Ahmad, J. E. Gindler, R. R. Betts, R. R. Chasman, and A. M. Freiedman, Phys. Rev. Lett. **49**, 1758 (1982).
[4] I. Ahmad, J. E. Gindler, A. M. Friedman, R. R. Chasman, and T. Ishii, Nucl. Phys. A **472**, 285 (1987).
[5] V. Grafen, B. Ackermann, H. Baltzer, T. Bihn, C. Günther, J. de Boer, N. Gollwitzer, G. Graw, R. Hertenberger, H. Kader, A. Levon, and A. Lösch, Phys. Rev. C **44**, R1728 (1991).
[6] A. H. Wapstra, G. Audi, and R. Hoekstra, Atom. Data Nucl. Data Tables **39**, 281 (1988).
[7] G. Audi, F. G. Kondev, M. Wang, B. Pfeiffer, X. Sun, J. Blachot, and M. MacCormick, Chin. Phys. C **36**, 1157 (2012).
[8] A. I. Levon, J. de Boer, G. Graw, R. Hertenberger, D. Hofer, J. Kvasil, A. Lösch, E. Müller-Zanotti, M. Würkner, H. Baltzer, V. Grafen, and C. Günther, Nucl. Phys. A **576**, 267 (1994).
[9] A. Lösch, J. de Boer, J. Kvasil, A. I. Levon, M. Würkner, H. Baltzer, V. Grafen, and C. Günther, Z. Phys. A **348**, 235 (1994).
[10] A. I. Levon, J. de Boer, M. Loewe, M. Würkner, T. Czosnyka, J. Iwanicki, and P. J. Napiorkowski, Eur. Phys. J. A **2**, 9 (1998).
[11] J. Lerner, Nucl. Instrum. Methods **102**, 373 (1972).
[12] G. Barreau, H. G. Börner, T. von Egidy, and R. W. Hoff, Z. Phys. A **308**, 209 (1982).
[13] B. Ackermann, T. Bihn, P. A. Butler, V. Grafen, C. Günther, J. R. Hughes, G. D. Jones, Ch. Lauterbach, H. J. Maier, M. Marten-Tölle, R. Tölle, R. Wadsworth, D. L. Watson, and C. A. White, Z. Phys. A **332**, 375 (1989).
[14] I. Ahmad and F. Wagner, Nucl. Instrum. Methods **116**, 465 (1974).
[15] I. M. Band, M. B. Trzhaskovskaya, C. W. Nestor, Jr., P. O. Tikkanen, and S. Raman, Atom. Data Nucl. Data Tables **81**, 1 (2002).
[16] I. Ahmad and P. A. Butler, Annu. Rev. Nucl. Part. Sci **43**, 71 (1993).
[17] C. P. Ruiz, University of California Report UCRL-9511, unpublished (1961).
[18] E. Browne and J. K. Tuli, Nucl. Data Sheets **114**, 751 (2013).
[19] G. Alaga, K. Adler, A. Bohr, and B. R. Mottelson, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. **29**, 9 (1955).
[20] J. R. Erskine, G. Kyle, R. R. Chasman, and A. M. Friedman, Phys. Rev. C **11**, 561 (1975).
[21] K. Abusaleem, Nucl. Data Sheets **116**, 163 (2014).
[22] I. Ahmad, R. R. Chasman, J. E. Gindler, and A. M. Friedman, Phys. Rev. Lett. **52**, 503 (1984).
[23] E. Browne, I. Ahmad, K. E. Gregorich, S. A. Kreek, D. M. Lee, and D. C. Hoffman, Nucl. Instrum. Methods A **339**, 209 (1994).