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Spectroscopy of ^{193,195,197}Po

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Excited states built on the $13/2^+$ isomers of the odd-mass ^{193,195,197}Po isotopes have been observed via in-beam γ-ray spectroscopy. The α radioactivity of these isotopes has been used to tag γ-ray transitions following the ^{A}Er+164 MeV ^{32}S reactions, where $A=164$, 166, 167, 168, and 170. Prompt γ radiation was measured by ten Compton-suppressed Ge detectors at the target position and the Fragment Mass Analyzer was used to select evaporation residues. The results are compared with the first excited states of the heavier odd-mass polonium isotopes and of the even-mass cores. [S0556-2813(97)02708-8]

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I. INTRODUCTION

The onset and evolution of collective motion has a plethora of manifestations in the region of isotopes around the $Z=82$ proton shell closure. Different nuclear shapes coexist [1], driven by various collective modes, such as harmonic and anharmonic vibration, deformed rotation, and even superdeformed rotation at higher energies [2]. The observed deformation of this region is usually interpreted as due to excitations across the $Z=82$ proton gap [1]. A sudden phase transition from vibrational to rotational collective motion is expected to occur when the energy of the 2_1^+ state drops to a critical range (130–145 keV), as has been identified for a wide range of nuclei [3].

The polonium isotopes near the doubly magic ^{208}Pb exhibit nuclear properties dominated by the shell structure. As the neutron number decreases and approaches the middle of the shell, collective motion is expected to occur. Indeed, a trend towards collective vibrational motion sets in already at ^{205}Po and becomes better established in ^{199}Po, the lightest odd-A isotope with previously studied excitations [4]. The lowest state in all of these isotopes populated by prompt γ-ray spectroscopy is the $13/2^+$ isomer arising from the $i_{13/2}$ neutron orbital. The structure of the first excited states is interpreted as a weak coupling of the $i_{13/2}$ neutron to the even-even core. The excitation energies of the $17/2^+$ states relative to the $13/2^+$ levels are observed to remain almost constant in all odd-A Po isotopes heavier than ^{199}Po, while the $21/2^+$ and $25/2^+$ state energies in the lighter isotopes increase as the vibrational motion extends to higher spin. In the cases of ^{199,201}Po, where the collective vibrational motion is well established, the sequence $13/2^+$, $17/2^+$, $21/2^+$, and $25/2^+$ is obtained by the coupling of the $i_{13/2}$ neutron to the positive-parity yrast levels of the core, i.e., $\nu i_{13/2}^{-1}\times(0^+,2^+,4^+,6^+)$ states, respectively [5].

Recent studies of light even-mass polonium isotopes have identified a gradual downward trend in the excitation energies of the first excited states. This behavior sets in already in ^{196}Po [6] and continues in the lighter ^{194}Po [7] and ^{192}Po [8,9] isotopes. Theoretical calculations in these isotopes suggest that the drop of excitation energies is due to an increase in the contribution from the $\nu i_{13/2}$ orbital to the wave functions as the middle of the neutron shell is approached [9,10]. Possible four-particle, two-hole (4p-2h) proton excitations could also exist in these nuclei, but they do not appear to contribute to the configurations of the first excited yrast states [10]. Furthermore, the drop is not sufficient to reach the critical range for a phase transition from vibrational to rotational collective motion, even in ^{192}Po, the lightest polonium isotope with known excitations [8,9].

The downward trend in the excitation energies and the associated increase in the collectivity of the light, even-mass Po isotopes is now well established. In contrast, little is known about excitations in the odd-A isotopes and the effect that the odd neutron has on the collective motion. It is possible that the increased occupation of the deformation-driving $\nu i_{13/2}$ orbital could enhance the collectivity sufficiently for the energy difference $E_{17/2}=E(17/2^+)-E(13/2^+)$ to reach the critical range. No spectroscopic information for any odd-A Po isotope lighter than ^{199}Po was available previous to this work. It was our aim to identify low-lying excitations in the light ^{193,195,197}Po isotopes. The observation of these states will help to understand better the evolution of collective motion and the roles played by $i_{13/2}$ neutron and (4p-2h) proton excitations in such a trend.

The formation and subsequent study of the neutron-deficient ^{193,195,197}Po isotopes is difficult, especially in the case of ^{193}Po which is the most neutron deficient of these. The small cross sections for fusion-evaporation channels compete with fission cross sections which can be 200 times larger. Therefore, in order to select preferentially these isotopes, additional triggering criteria are required in any

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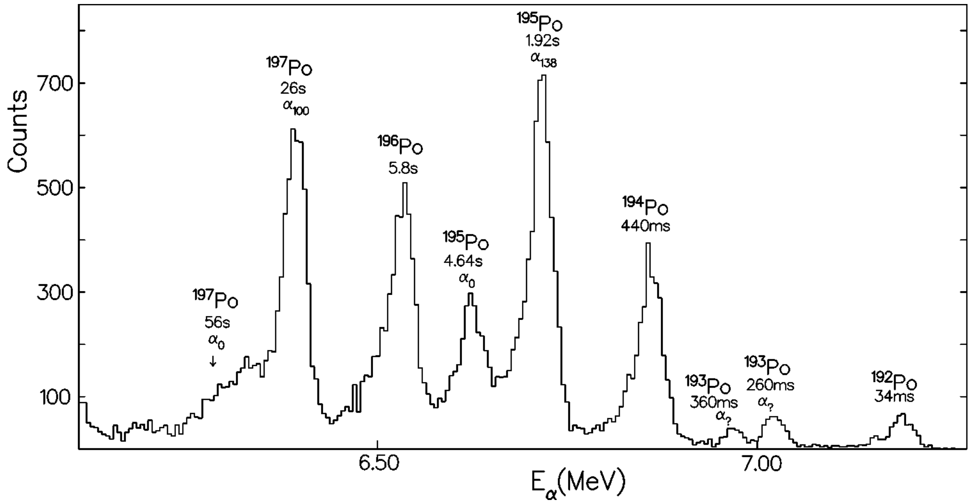

FIG. 1. Projection of the E_α vs γ matrix onto the E_α axis. Alpha-peak assignments together with the half-lives of the emitted α particles are given for all of the polonium isotopes produced in the reaction. Two α peaks are observed for each odd-mass Po isotope: the strong one from decay of the $13/2^+$ isomer and the weaker one from the decay of the $3/2^-$ isomer. For these peaks the excitation energies of the final levels in the daughter nuclei are also given. The final state excitation energies are not known for the ^{193}Po parent, hence, the question mark in the figure.

γ-ray spectroscopic study. The recoil-decay tagging (RDT) technique [11] has been used in this work to enhance the reaction channels of interest.

II. EXPERIMENTAL PROCEDURE

The ^{A}Er+^{32}S reactions at a beam energy $E(^{32}\mathrm{S})=164$ MeV were used to populate excited states of ^{193,195,197}Po at the Argonne Tandem Linac Accelerator System (ATLAS). The target, chosen for our study of ^{192}Po, was 724 μg/cm^2 thick and consisted of 73.6% ^{164}Er, 15.0% ^{166}Er, 5.5% ^{167}Er, 4.4% ^{168}Er, and 1.5% ^{170}Er. Prompt γ rays were recorded using an array of 10 Compton-suppressed Ge detectors at the target position. The Fragment Mass Analyzer (FMA) [12] was used to disperse the evaporation residues according to their mass/charge ratio. The position of the recoils at the FMA focal plane was determined using a position-sensitive parallel-plate grid counter (PGAC). These recoils were implanted in a double-sided Si strip detector (DSSD) located behind the PGAC, which was also used to detect the subsequent α decay of the implanted recoils. The 40×40 segmentation of the DSSD provided an effective space correlation between an implant and the subsequent decays.

We recorded recoil-γ and recoil-$\gamma\gamma$ coincidences, as well as α-decay events. The information included γ-ray energies (E_γ), position (x_{PGAC}) and energy loss (ΔE_r) of the recoils focused in the PGAC, and position (x,y) and energy (E_r) of recoils in the DSSD. The energy of decay radiations (E_α) in the DSSD and the time-of-flight (TOF) of the recoils between the PGAC and DSSD were recorded as well. In addition, timing from an absolute clock was recorded throughout the experiment to provide information on the time elapsed (T_{decay}) between the implantation and the decay of each recoil. Further details of the experiment are reported in [9].

In order to reduce the contamination from the scattered primary beam particles, two-dimensional gates were set on E_r vs TOF and ΔE_r vs x_{PGAC} matrices. The gated data were sorted into an x_{PGAC} vs E_γ matrix and an E_α vs E_γ matrix. Figure 1 shows the projection of the latter matrix onto the α-energy axis. In this figure one can clearly see that the heavy Po isotopes represent the larger percentage of the data, although the heavy Er isotopes represent only a small percentage of the target. This is because the formation of the heavier Po isotopes is characterized by larger fusion-evaporation cross sections with less competition from fission, compared to the lighter Po isotopes (especially ^{192}Po and ^{193}Po). The half-lives of the emitted α particles are also given in Fig. 1. For the odd-mass Po isotopes two α peaks are observed for each isotope: a strong peak from the deexcitation of the $13/2^+$ isomer, and a weak one from the $3/2^-$ isomer. These peaks are labeled by the excitation energies of the final levels in the daughter nuclei. The ^{192}Po events in these data have been analyzed in our report of the first excited states of this isotope [9].

Gamma-ray spectra in coincidence with the corresponding isotopes can be obtained by slicing the E_α vs γ matrix on the α-energy lines of the ^{193,195,197}Po isotopes. To obtain cleaner spectra, which facilitate unambiguous assignment of transitions in these isotopes, an upper limit was also set on the decay time of the events in coincidence with the α peaks of each isotope. In ^{193}Po we used $T_{\mathrm{decay}}<1$ sec, in ^{195}Po $T_{\mathrm{decay}}<10$ sec, and in ^{197}Po $T_{\mathrm{decay}}<80$ sec. The upper limits in all three cases were chosen to be 2–3 times the half-lives of the α decays of each isotope. This additional criterion resulted in spectra which are not contaminated by products with decay half-lives longer than the upper limits which characterize each isotope. The resulting γ-ray spectra in coincidence with the α decay of the $13/2^+$ isomers of the three polonium isotopes are shown in Fig. 2. Those in coincidence with the α decay of the $3/2^-$ isomers are shown in Fig. 3.

III. LEVEL SPECTRA OF ^{193,195,197}Po

The three most intense transitions above the $13/2^+$ isomers in the ^{193,195,197}Po isotopes can be identified from the

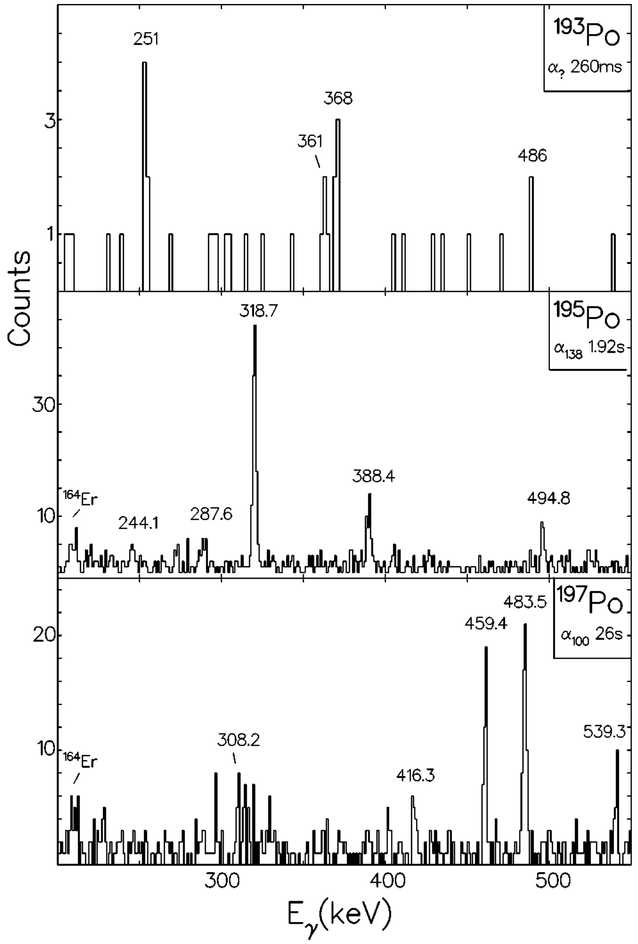

FIG. 2. Prompt γ-ray spectra in ^{193,195,197}Po. These spectra are gated on the α decay (energy and half-life) of the $13/2^+$ isomers in ^{193,195,197}Po (see Fig. 1). The corresponding assignments of the α peaks are given in the upper right insets. Errors on the γ-ray energies are 0.5 keV in ^{195,197}Po and 1 keV in ^{193}Po. The contaminant line associated with the Coulomb excitation of the ^{164}Er target is indicated.

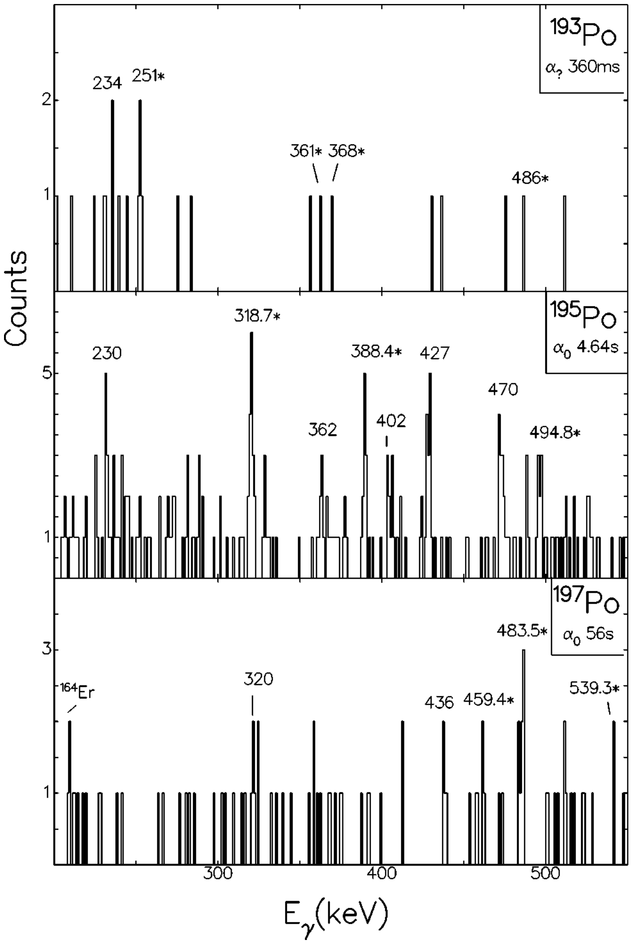

FIG. 3. Prompt γ-ray spectra in ^{193,195,197}Po. These spectra are gated on the α decay (energy and half-life) of the $3/2^-$ isomers in ^{193,195,197}Po (see Fig. 1). Transitions observed in coincidence with the α decay of the $13/2^+$ isomers are indicated by an asterisk. Gamma-ray energy uncertainties are 1 keV. See also caption for Fig. 2.

spectra shown in Fig. 2. Arranging these transitions according to their intensity, the three first excited states above the $13/2^+$ state in each isotope can be deduced, as summarized in the proposed level schemes for ^{193,195,197}Po in Fig. 4. No coincidence relationships between γ-ray transitions could be established because of the very low percentage of recoil-$\gamma\gamma$ events in our data ($<5\%$ of the total number of events). Hence, transitions in the spectra were placed in the level schemes based on intensity arguments. The only departure from this assumption is in the case of ^{193}Po, where we propose the 486-keV transition as the one deexciting the $(25/2^+)$ state, rather than the stronger 361-keV transition. This assignment was based mainly on comparison with the sequence of transitions in ^{195}Po, where we observe a 494.8-keV transition deexciting the $(25/2^+)$ state of ^{195}Po. Some weaker transitions are also present in the spectra, including the 361-keV line in ^{193}Po, the 244.1- and 287.6-keV lines in ^{195}Po, and the 308.2- and 416.3-keV lines in ^{197}Po. Placement of such transitions in the level schemes would require coincidence information, which would, in turn, require higher $\gamma\gamma$ statistics than were accumulated. Gamma-gamma coincidence data from a previous experiment [7] support the coincidences of the 483.5-, 459.4-, and 539.3-keV transitions in ^{197}Po and the 318.7- and 388.4-keV transitions in ^{195}Po; however, the poor statistics of these data do not allow definitive conclusions.

As expected from the low intensity of the α transitions deexciting the $3/2^-$ isomers of ^{193,195,197}Po, the statistics of the corresponding γ-ray spectra of Fig. 3 are insufficient to draw any definite conclusions on the level sequences above these isomers. A possible exception is in ^{195}Po, where the intensities of the 230-, 427-, and 470-keV peaks are sufficient to place unambiguously these lines above the $3/2^-$ isomer of ^{195}Po. However, due to lack of systematics for the levels above the $3/2^-$ isomers and the lack of adequate statistics for coincidence relationships, the relative placement of these transitions is uncertain. It should be noted that the energy gap between the $13/2^+$ and $3/2^-$ isomers in ^{195}Po is ~ 230 keV [4]. It is, however, unlikely that the 230-keV transition is a direct connection between these two levels because this would imply an $E5$ multipolarity. Such a transition is highly converted and the corresponding γ ray should not be visible in our experiment. The other transitions labeled in Fig. 3 (234-keV in ^{193}Po, 362- and 402-keV in ^{195}Po, and 320- and 436-keV in ^{197}Po) are tentative because

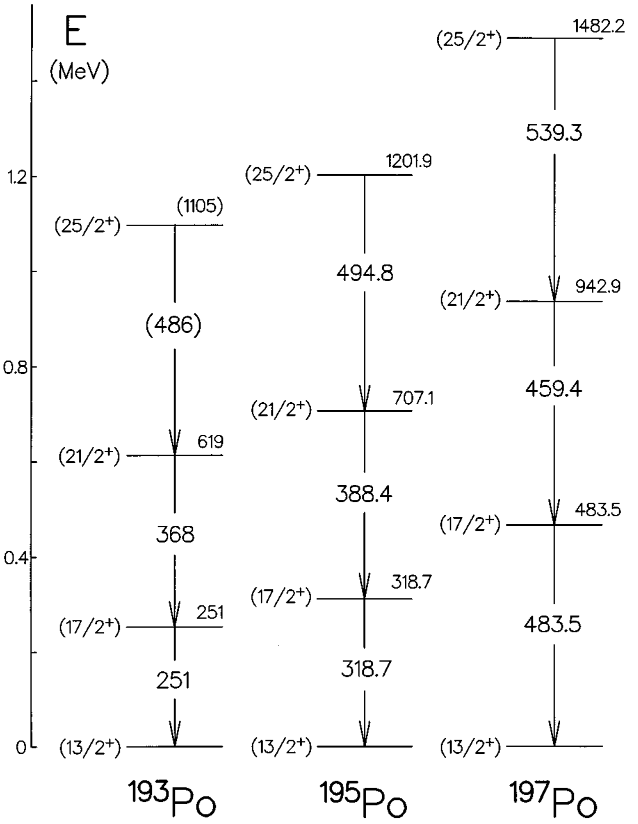

FIG. 4. Proposed level schemes above the $(13/2^+)$ isomers in ^{193,195,197}Po. Spin and parity assignments are based on systematics of the odd-mass polonium isotopes.

of poor statistics. However, if their assignment is correct, they would be involved in the deexcitation of levels above the $3/2^-$ isomer because they are not seen in coincidence with the α decay of the $13/2^+$ isomer.

The spin and parity assignments of the reported levels in Fig. 4 are based on comparisons to the corresponding levels in heavier odd-mass polonium nuclei, displayed in Fig. 5(a). As was the case for the in-beam spectroscopy of the heavier isotopes, we have assumed that the strongest lines are stretched $E2$ transitions built on the $13/2^+$ isomer.

IV. DISCUSSION

The interpretation of the structure of the excitations above the $13/2^+$ isomers in ^{193,195,197}Po is mostly based on the systematical behavior of the odd-mass isotopes and comparison to the excitations in the even-mass polonium cores summarized in Fig. 5(b). To follow the evolution of collective motion in the odd Po isotopes, we will define the following energy ratios:

$$R_1=[E(21/2^+)-E(17/2^+)]/[E(17/2^+)-E(13/2^+)], \tag{1}$$

$$R_2=[E(25/2^+)-E(21/2^+)]/[E(21/2^+)-E(17/2^+)]. \tag{2}$$

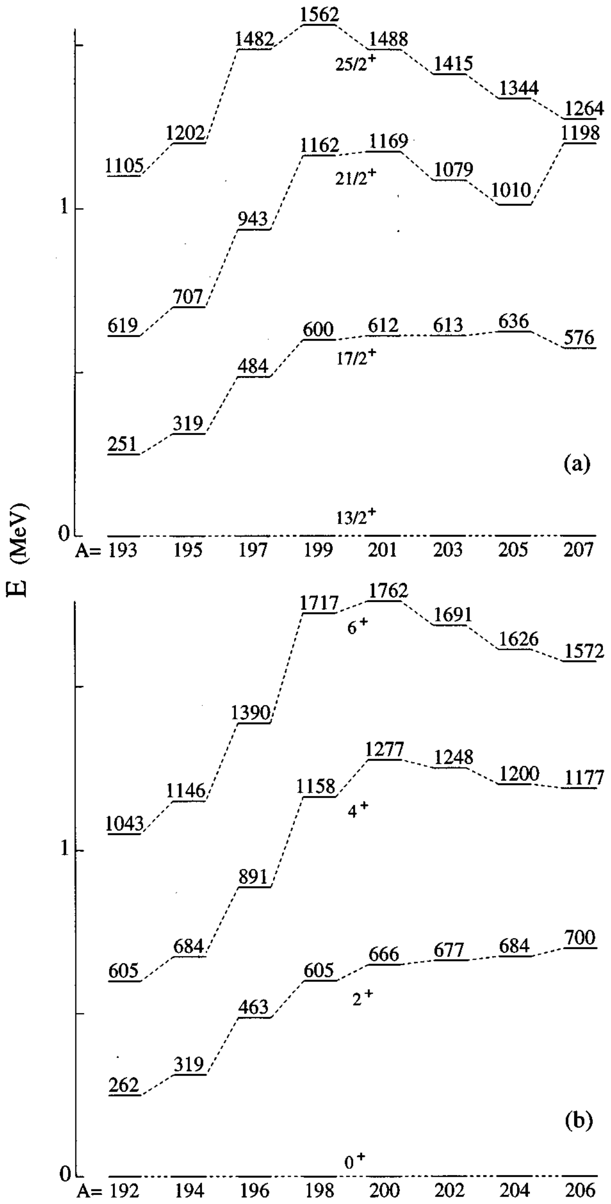

FIG. 5. (a) Systematics of the 13/2, 17/2, 21/2, and 25/2 positive-parity levels in odd-mass $^{193-207}$Po isotopes. The excitation energies relative to the $13/2^+$ isomers are given in keV. (b) Systematics of the 0^+, 2^+, 4^+, and 6^+ states in the even-mass $^{192-206}$Po isotopes. Data taken from [4,7–9] and the present work.

These ratios are analogous to the $R(4/2)=E(4^+)/E(2^+)$ and $R(6/4)=E(6^+)/E(4^+)$ ratios in the even-even cores. The systematics of the energy ratios in polonium nuclei are summarized in Fig. 6.

The systematics of the excitations built on the $13/2^+$ isomers in the odd-Po isotopes are summarized in Fig. 5(a). In odd $A>198$ Po isotopes, the excitation energies of the $17/2^+$ states are almost constant and the R_1 values are near vibrational at ~ 1.9. The energies of the $25/2^+$ states increase as the number of valence neutrons increases, but still the value of $R_2<1.5$, the value which characterizes a harmonic vibrator. The pattern suggests that (1) the $17/2^+$ states are collective excitations built on the $i_{13/2}$ neutron and (2) for

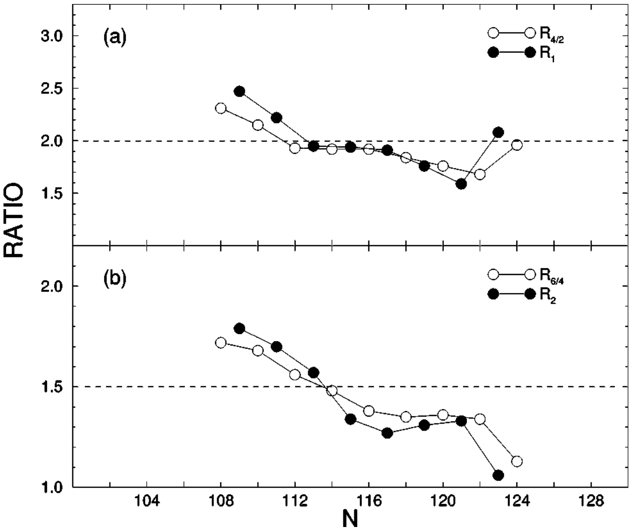

FIG. 6. Yrast energy ratios in the Po isotopes as a function of neutron number N. (a) R_1 and $R(4/2)$; (b) R_2 and $R(6/4)$. Filled circles are the values for odd-A isotopes, open circles for even A. The values for a harmonic vibrator are indicated by dashed lines.

$198<A<206$ the collectivity extends to the $21/2^+$ state, but not to the $25/2^+$. This is expected from a weak coupling picture of the odd-A structures, since the 2^+ and 4^+ states in the even-Acores are interpreted as vibrational states, but the 6^+ levels have large two-proton components for $A>198$ [10].

In ^{197}Po the observed levels extend up to ~1.5 MeV energy. As seen in Fig. 4, the excitation energies of the levels of this isotope drop, compared with the systematics of the heavier isotopes. The energy ratios $R_1=1.95$ and $R_2=1.57$ are quite close to the values of 2.0 and 1.5, respectively, expected for a harmonic vibrator. This again follows the weak coupling picture, because the yrast states in the ^{196}Po core have the equal spacing of a harmonic vibrator [6]. In other words, the drop of the energy of the $17/2^+$ state suggests that the collectivity increases, and the pattern suggests that this collective motion persists to the $25/2^+$ state, but this motion is still dominated by harmonic vibrations.

In ^{195}Po the yrast energies continue to drop in energy. Furthermore, the energies of the transitions increase with spin, resulting in a sequence which starts to evolve towards that of collective rotation. However, the $R_1=2.22$ and $R_2=1.70$ values in this isotope are far from the values of 3.33 and 2.10, respectively, which characterize well-deformed rotational motion. Moreover, the 318.7-keV energy of the $17/2^+$ state relative to the $13/2^+$ isomer is far from the critical range where the phase transition from vibrational to rotational collective motion is expected to take place [6]. Hence, vibrational motion, although anharmonic, persists in ^{195}Po. This anharmonic vibrational behavior has also been suggested for the ^{194}Po core [7].

The statistics displayed in Fig. 2 for ^{193}Po are near the limit of detection. This is expected because relative cross-section calculations for the formation of this isotope in all possible reactions [^{164}Er (^{32}S,3n) and ^{166}Er (^{32}S,5n)] yield values ~200 times lower than the fission cross section. However, we have identified three transitions to deduce the level scheme displayed in Fig. 4. Although the yrast energies in ^{193}Po continue the downward trend established in ^{195}Po and ^{197}Po, this trend appears to be quenched in ^{193}Po. A simple extrapolation of the drop of the energies between ^{195}Po and ^{197}Po would predict a $17/2^+$ energy of 155 keV, a value much less than observed in ^{193}Po. This quenching suggests that the anharmonic vibrations, observed in ^{195}Po and ^{194}Po, still play an important role in the behavior of this isotope, although the $R_1=2.46$ value has increased. This same quenching occurs in the ^{192}Po core [8,9], the lightest polonium isotope observed so far.

A summary of the energy ratios in polonium, $R(4/2)$ for even-A isotopes and R_1 for odd-A isotopes, is displayed in Fig. 6(a). These ratios at $N\sim122$ are <2, as is expected for a core structure dominated by two $h_{9/2}$ protons with a surface delta residual interaction. As the neutron shell opens there is a slow increase up to $R\sim2$ where the ratio remains constant over a region of six polonium isotopes (^{201}Po to ^{196}Po). This is the region where collective harmonic vibration in the one- and two-phonon states in the cores is manifested. For $A<196$ polonium isotopes the ratios increase. Extrapolating the increase in these ratios down to the middle of the shell ($N\sim104$), where the maximum collectivity is expected, still predicts values less than 3.0 for both odd- and even-mass polonium isotopes, i.e., the increase in collectivity is not strong enough to drive the lighter polonium isotopes toward collective rotation. The same result (ratios less than 3.0) holds if one extrapolates the trends in the excitation energies and then calculates the ratios, rather than directly extrapolating the ratios.

For $A>195$ the energy ratios in the odd-A isotopes are essentially identical to those in the even-A cores. However, for $A<196$ the energy ratios in the odd-A isotopes are larger than in the corresponding lighter even core, which suggests more collectivity in the excitations built on the deformation-driving $\nu i_{13/2}$ configuration.

Similar conclusions can be drawn by examining the pattern of the R_2 and corresponding $R(6/4)$ ratios, displayed in Fig. 6(b). Near the $N=126$ shell closure the 6^+ and 4^+ states (and their $25/2^+$ and $21/2^+$ counterparts in the odd-A isotopes) are nearly degenerate, as expected for a structure dominated by two $h_{9/2}$ protons with a surface delta residual interaction. These ratios remain small, <1.5, until $N<115$—for the lighter isotopes the collectivity appears to have extended to at least the 6^+ state of the core. The collective motion increases in the lighter isotopes, but the R_2 ratio never approaches the value of 2.1 expected for a rigid rotor. However, the R_2 ratios in ^{193,195}Po are greater than the $R(6/4)$ ratios in the lighter core, and much greater than the average values of the adjacent even-even isotopes. Again, this suggests that the occupation of the $i_{13/2}$ orbital in the yrast excitations in the odd-Po isotopes increases the deformation. The R_2 values in ^{193,195}Po of 1.79 and 1.70, respectively, are what would be expected for a collective nucleus with R_1 [or the analogous $R(4/2)$] values of 2.47 and 2.22, respectively [13].

The α decay of the light even-A Po isotopes, especially that of ^{192}Po, suggests that there is strong mixing between 4p-2h (deformed) and spherical configurations in these isotopes [14]. In our earlier work [9], we noted that the yrast energies in ^{192}Po followed the expectations for a moderately collective, anharmonic vibrator—the perturbations of the level spacings, that would suggest that the structure of the ground state and the excited states are different, are not observed. The striking similarities between the energies and energy ratios of excitations in the odd and even isotopes suggests that the important degrees of freedom in the even core are also important in the odd neighboring isotope. In particular, any role that 4p-2h configurations play in the even cores would be similar in the adjacent odd isotope.

Particle-core model (PCM) [15] and quasiparticle random phase approximation (QRPA) [16] calculations have been used to understand the structure of the even-mass $^{192-208}$Po isotopes [9,10]. In the PCM the yrast excitations and the non-yrast 2_2^+ and 4_2^+ states could be explained by the coupling of two $h_{9/2}$ particles to a phenomenological vibrational core. The change in the particle-core coupling strength for $A < 200$ gives rise to the increase in collective motion in the lighter isotopes and the persistence of this collectivity to at least the 6^+ yrast state. In particular, the energies of these yrast and non-yrast states could be understood without invoking 4p-2h excitations across the $Z = 82$ shell gap. The QRPA calculations suggest that it is the increased role played by $i_{13/2}$ neutrons, and their attractive interaction with $h_{9/2}$ protons, that gives rise to the increase in collectivity. That the $i_{13/2}$ neutron plays an even more important role in the yrast states in the odd isotopes may explain why the energy systematics suggest an increase in collectivity in these nuclei, compared to the lighter even core.

V. CONCLUSIONS

Transitions in ^{193,195,197}Po isotopes have been identified using the RDT technique. This technique is based on preferential selection of the isotopes of interest using their characteristic α radioactivity. The first three excited states of all three isotopes have been observed and compared with the corresponding excited states in the lighter even-mass core.

In contrast to the pattern in the heavier odd-A polonium isotopes, where the excitation energies of the first three excited states either remain constant or exhibit a gradual upslope, a systematic drop in the energies is observed in ^{193,195,197}Po. This indicates an increase in collectivity towards collective rotation, measured by the gradual increase of the R_1 and R_2 ratios in ^{193,195,197}Po. An increase in collectivity is expected as the middle of the neutron shell is approached. However, the collective motion has not yet reached the phase transition from vibrational to well-deformed rotational modes. Vibrations, harmonic in ^{197}Po and anharmonic in ^{193,195}Po, still dominate the structure of the first excitations in these nuclei.

The downward trend of the energies observed in ^{193,195,197}Po has previously been observed in ^{192,194,196}Po. The similarity in the behavior of the odd- and even-mass isotopes is striking. PCM and QRPA theoretical calculations performed in ^{192,194,196}Po suggest that the increase in collective motion in these lighter isotopes comes from the increased role of the neutron $i_{13/2}$ orbital and the attractive proton-neutron interaction between $i_{13/2}$ neutrons and the valence $h_{9/2}$ protons. The similar features established in ^{193,195,197}Po suggest that the same interpretation is also valid for these isotopes. The slight increase in the energy ratios R_1 and R_2 for ^{193,195}Po compared to the ^{192,194}Po cores suggests that the increased occupation of the $\nu i_{13/2}$ configuration for the excitations built on the $13/2^+$ isomers gives rise to an increase in the collective motion, which remains predominantly anharmonic vibrational in character.

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