

Proton Single-Particle States In The Heaviest Actinide Nuclei

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Abstract. The level structure of ^{249}Bk has been investigated by measuring the γ -ray spectra following the α decay of a chemically and isotopically pure ^{253}Es sample. Alpha-gamma coincidence measurement was performed using a Si detector for α particles and a 25% Ge detector for γ rays. A gamma-gamma coincidence measurement was performed with the Gammasphere spectrometer. The Es sample was obtained by extracting the ^{253}Es which grew in a ^{253}Cf source material produced in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. Additional information on the ^{249}Bk levels was obtained from the study of γ rays produced in the β^- decay of ^{249}Cm . The ^{249}Cm sample was produced by neutron irradiation of ^{248}Cm . Using the results of the present study and the results of previous $^{248}\text{Cm}(\alpha,t)$ and $^{248}\text{Cm}(^3\text{He,d})$ reaction spectra, the following single-particle states have been identified in ^{249}Bk : $7/2^+$ [633], 0.0 keV; $3/2^-$ [521], 8.78 keV; $1/2^+$ [400], 377.55 keV; $5/2^+$ [642], 389.17 keV; $1/2^-$ [530], 569.19 keV; $1/2^-$ [521], 643.0 keV; $5/2^-$ [523], 672.8 keV; $9/2^+$ [624], 1075.1 keV. Four vibrational bands were identified at 767.9, 932.2, 1150.7 and 1223.0 keV with tentative assignments of $\{7/2^+[633]\otimes 1^-\}9/2^-$, $\{7/2^+[633]\otimes 0^-\}7/2^-$, $\{7/2^+[633]\otimes 1^-\}5/2^-$ and $\{7/2^+[633]\otimes 0^+\}7/2^+$, respectively.

INTRODUCTION

In order to test theoretical models which predict a gap in the proton single-particle spectra of superheavy nuclei, experimental energies of single-proton states in the heaviest nuclei are needed. The best way to determine these energies is to measure radiations associated with the decay of transfermium nuclei. However, at present, such measurements are not possible because these nuclides are produced in tiny amounts. The states which are of interest in transfermium nuclei occur as excited states in lower Z nuclei.

The intensity of an alpha group to an excited state in the daughter nucleus depends on the available decay energy (Q_α) and the wavefunctions of the parent and daughter nuclei. The exponential decrease in alpha intensity due to Q_α value for a favored transition is displayed in Fig. 1. From alpha decay systematics it is known that transitions between different single-particle states have hindrance factors between 10 and 1000. Using a hindrance factor of 100, we can estimate intensities of unfavored alpha groups which is shown in Fig. 1 as a dashed line. As shown in the figure, the alpha intensity to an excited state at ~ 1 MeV is of the order of $10^{-5}\%$ per decay. Thus, to study excited states up to 1 MeV excitation, sources with milliCurie strength are needed. The heaviest odd-Z nuclide available in such large strength is ^{253}Es ($t_{1/2}=20.47$ d). About a milligram of this isotope is produced in the High Flux Isotope Reactor (HFIR) in each irradiation campaign by multiple neutron capture on Cm. However, this Es sample contains $\sim 0.4\%$ ^{254}Es ($t_{1/2}=275.7$ d) whose daughter ^{250}Bk ($t_{1/2}=3.217$ h) decays by intense γ rays of 989.13 and 1031.85 keV. These γ rays produce a large Compton background thus reducing the sensitivity of the measurement. We therefore used second-chance Es, which was obtained by milking ^{253}Es which grew in a ^{253}Cf ($t_{1/2}=17.81$ d) sample and had much less ^{254}Es than the first-chance Es.

The level structure of ^{249}Bk has previously been studied by high-resolution alpha spectroscopy of ^{253}Es [1, 2], γ -ray spectroscopy of ^{253}Es and ^{249}Cm ($t_{1/2}=65.3$ min), and one-proton transfer reactions [3]. The level scheme

of ^{249}Bk deduced from these studies has been compiled in Nuclear Data Sheets [4]. Because of the availability of large germanium detectors and the large Compton-suppressed Ge detector array Gammasphere, we have investigated the level structure of ^{249}Bk . Three samples of second-chance ^{253}Es , ~ 20 mCi each, were obtained from Oak Ridge National laboratory in 1998, 2000, and 2003. The sources obtained in 2000 and 2003 were either sandwiched between quartz disks for measurement of low-energy γ rays or were sandwiched between two Pt disks for the high-energy spectrum. The source sandwiched between Pt disks did not have any light element or air. This reduced γ rays produced by the reaction of α particles on light elements. The sample produced in 2003 was extremely pure (chemically and isotopically) and provided the most sensitive measurements.

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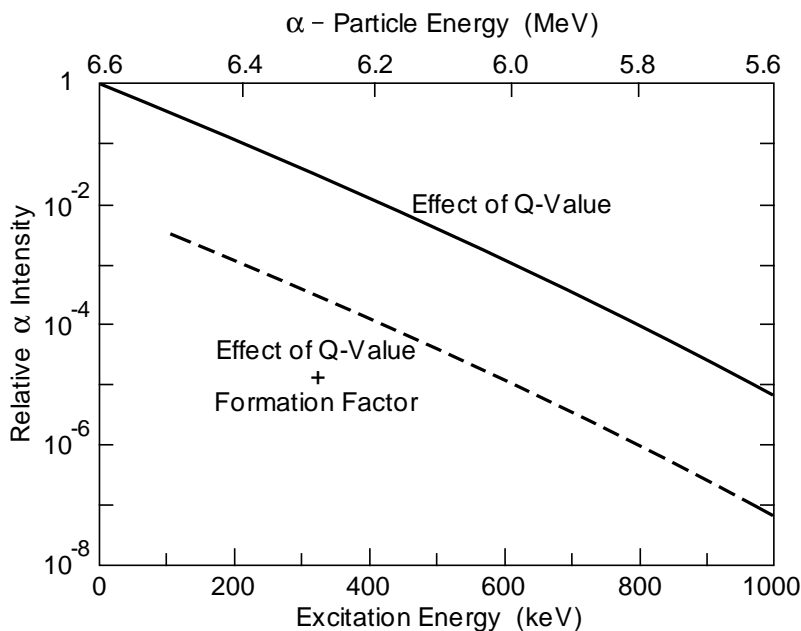


FIGURE 1. Dependence of alpha intensity on the decay energy of ^{253}Es . Also shown is the reduction due to formation factor of α particles.

EXPERIMENTAL DETAILS

Gamma-ray singles spectra of all three ^{253}Es samples were measured with two different germanium detectors. The low-energy γ -ray spectra measured with a $5\text{ cm}^2 \times 10\text{-mm}$ low energy photon spectrometer (LEPS) provided energies and intensities of γ rays up to 600 keV. The high-energy portion of the ^{253}Es spectrum was measured with a 25 % Ge detector and it is shown in Fig. 2. The energy was obtained by simultaneous measurement of the spectra of ^{253}Es and calibration sources. The detector efficiency was determined with a calibrated mixed source. Absolute γ -ray intensities were determined by measuring the α -particle rate of a thin ^{253}Es source with a Si detector of known solid angle and the γ -ray spectrum with a Ge spectrometer whose efficiency was determined with a calibrated source.

Alpha-gamma coincidence measurements were performed to confirm the assignment of γ rays to the ^{253}Es decay. α particles were detected with a 2.0 cm^2 Si detector and the γ rays with a 110% Ge detector. In the γ -ray spectrum gated by 4.45 - 6.20 MeV α particles, strong γ rays seen in the singles spectrum were identified. These include γ rays with energies 726.1, 767.9, 890.7, 900.0, 932.2, 946.3, 1040.2 and 1075.1 keV. The strong low-energy γ rays were observed in a spectrum gated by all ^{253}Es α particles.

In order to obtain further information about coincidence relationship among ^{253}Es γ rays, a 6-day long γ - γ coincidence measurement was performed at Argonne National Laboratory. The ~ 16 mCi source, obtained in 2003, was placed in the center of the Gammasphere spectrometer [5] consisting of 99 Compton-suppressed Ge detectors for this experiment. The data were sorted off-line into two-dimensional γ - γ coincidence matrices and γ -ray spectra were generated by placing 2-keV wide gates on various γ -ray peaks. The gated spectra contained three kinds of peaks. These

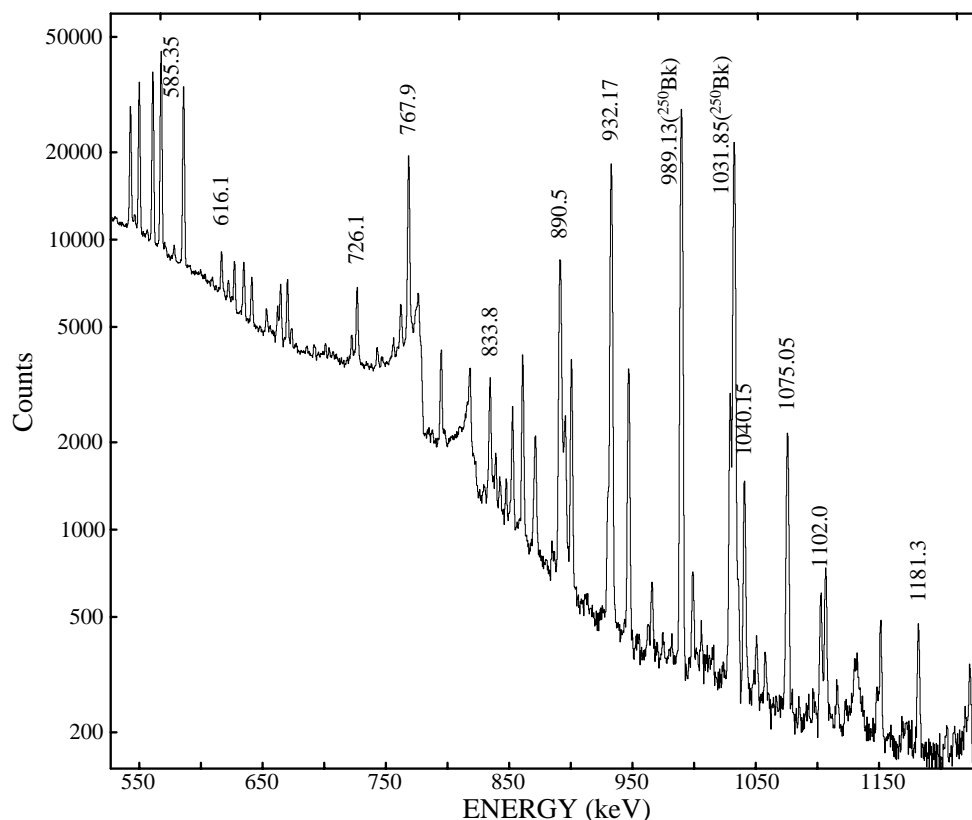


FIGURE 2. Gamma-ray spectrum of a ^{253}Es source measured with a 25% Ge detector. The intensities of γ rays can be estimated with respect to that of the 1181.3-keV γ ray which is measured to be $1.0 \times 10^{-6}\%$ per ^{253}Es α decay.

include the true coincidences between photopeaks, random coincidences, and Compton-Compton peaks. Each gate contained the full energy events from the γ ray of interest and Compton events from higher energy γ rays. The Compton scattered photon entered another Ge detector producing a signal in coincidence with Compton electron in the first detector. Since we know the singles spectrum, it was easy to identify random peaks. To distinguish real photopeak-photopeak events from Compton peaks, we used ^{250}Bk γ rays of 989.13 and 931.85 keV which have no γ rays in coincidence. These γ rays are the strongest peaks at high energies and they generate the strongest Compton-Compton peaks. In order to ensure that two γ rays were in coincidence with each other, spectra generated by placing a gate on each γ -ray peak were examined. We also compared γ -ray spectra gated by several γ rays deexciting the same level or feeding the same level to ascertain that the γ rays belonged to ^{253}Es decay. The sensitivity of Gammasphere was sufficient enough to identify γ rays with intensity as low as $1.0 \times 10^{-6}\%$ per ^{253}Es α decay in the gated spectra.

The analysis revealed γ rays deexciting high-lying states to the members of $5/2^+[642]$ and $1/2^+[400]$ bands. Examples of gated spectra are shown in Fig. 3. Energies of the weak γ rays were determined with respect to precisely known energies of stronger γ rays and have uncertainties of less than 0.2 keV. Only approximate intensities were obtained from the coincidence data.

Information on ^{249}Bk levels was also obtained from the β^- decay studies of ^{249}Cm ($t_{1/2}=65.3$ min). Samples containing ^{249}Cm were produced via neutron capture reactions in ^{248}Cm targets. Following irradiation, curium in the target was chemically purified to remove fission products. The procedure provided sufficient purification to permit measurement and identification of ^{249}Cm γ rays over three half-lives. The γ -ray singles spectrum of ^{249}Cm was measured with several Ge(Li) detectors. These detectors were calibrated for geometry and efficiency with standard sources. To obtain absolute intensities, the β^- particles were counted with a gas-flow proportional counter. The counter was calibrated for geometry and efficiency with a ^{198}Au standard whose absolute β^- disintegration rate was determined by gamma counting the 100% abundant 411.8-keV line.

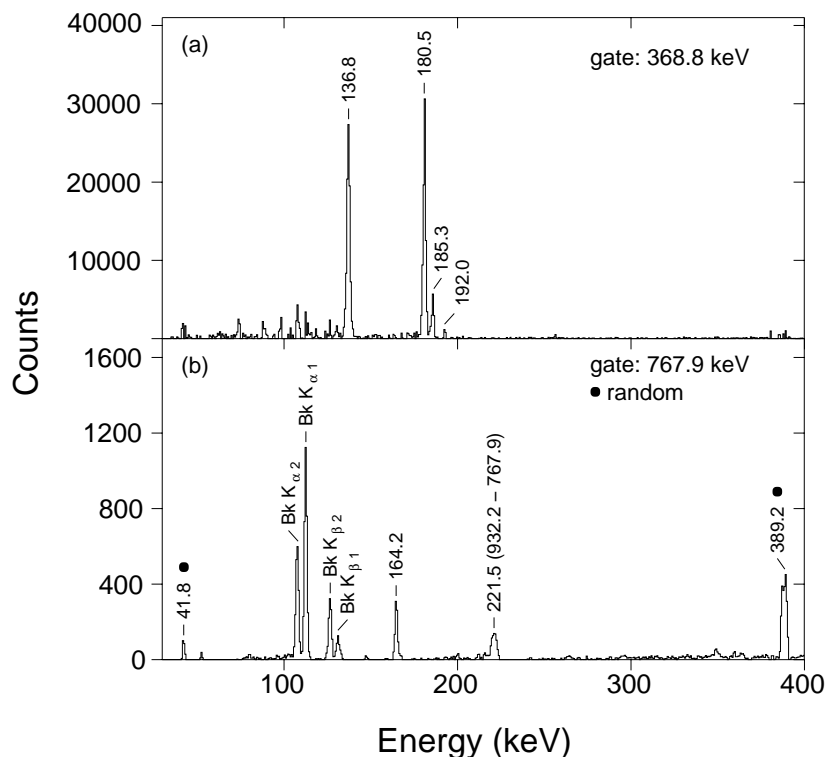


FIGURE 3. Gamma-ray spectra of a ^{253}Es sample measured with Gammashpere in coincidence with 368.8- and 767.9-keV γ rays.

DISCUSSION

The ground state spin of ^{249}Bk has been measured [6] to be $7/2$ and its measured magnetic moment agrees with the value calculated [7] for the $7/2^+[633]$ configuration. Its rotational members up to $I=17/2$ were identified [1] and in the present work we have observed the γ rays deexciting these levels. The coincidence relationships among intraband transitions observed in the present work clearly support the spin-parity assignments. The level at 8.78 keV was observed in the α singles spectrum [1, 2]. This level is also strongly populated in the β^- decay of ^{249}Cm suggesting spin $1/2$ or $3/2$. These observations plus the measured energies of its rotational members fix the spin-parity of this state as $3/2^-$. This state is assigned to the $3/2^- [521]$ Nilsson state [8]. Rotational members with spin up to $17/2$ have been identified. Both these bands were identified in $^{248}\text{Cm}(\alpha,t)$ and $^{248}\text{Cm}(^3\text{He},d)$ reactions [3, 9]. The state at 389.17 keV decays to the ground state by an M1 transition [4] and also to the $3/2^-$ band. This decay pattern fixes the spin-parity of the 389.17-keV level as $5/2^+$. Rotational members up to spin $15/2$ have been identified in the present work. The level scheme of ^{249}Bk deduced from the results of the present study is shown in Fig. 4.

The ground state of ^{249}Cm has been established to be the $1/2^+[620]$ Nilsson state from the neutron-capture studies [10] and the $^{248}\text{Cm}(d,p)$ reaction cross sections [11]. In ^{249}Bk , the only states expected to receive measurable β^- population are those with spin $1/2$ and $3/2$ because $\Delta I=0$ or 1 transitions are favored. These low spin levels were identified in ^{249}Cm decay. The level at 377.55 keV decays only to the $3/2^-$ level at 8.78 keV suggesting $1/2^+$ spin-parity. γ - γ coincidence data show that the 410.6- and 421.4-keV levels are its rotational members with spins $3/2$ and $5/2$, respectively. The $7/2$, $9/2$, and $13/2$ members have been identified at 498.6, 519.19, and 671.3 keV. It is observed that states with spin $5/2$ and higher decay predominantly to the ground state $7/2^+$ band supporting further the positive-parity assignment of this band. This band has been given $1/2^+[400]$ assignment on the basis of its deduced decoupling parameter.

Levels at 558.2 and 569.2 keV receive direct β^- population in ^{249}Cm decay and their deexcitation pattern suggests spin-parity of $3/2^-$ and $1/2^-$, respectively. The $5/2^-$, $7/2^-$, $9/2^-$ and $11/2^-$ members of this band are identified at 625.0, 606.7, 723.2 and 704.8 keV, respectively in the α decay of ^{253}Es . In the γ - γ coincidence experiment, a 136.8-keV E1 γ ray was observed to deexcite the 558.2-keV level to the $5/2^+$ member of the $1/2^+[400]$ band, which suggests

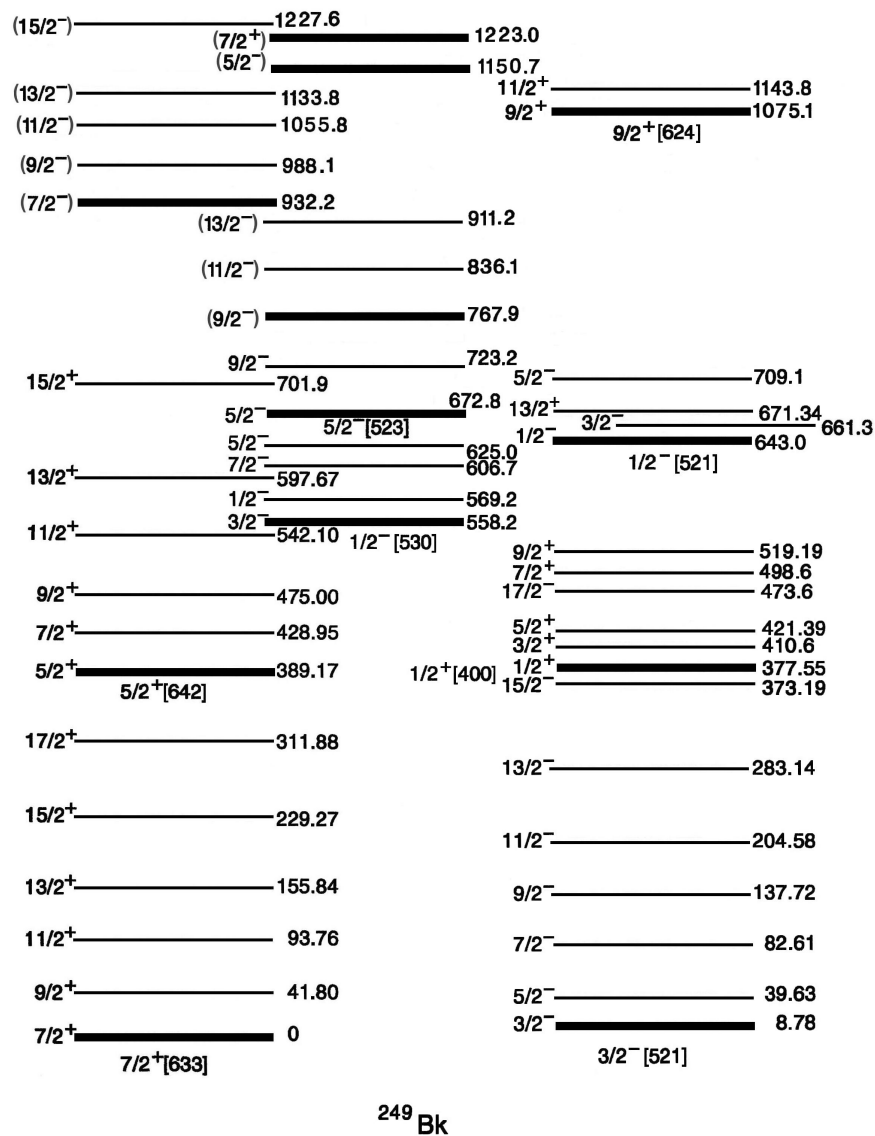


FIGURE 4. Level scheme of ^{249}Bk deduced from the study of ^{253}Es α decay and ^{249}Cm β^- decay. Bandheads are shown by thicker lines.

negative parity for the 558.2-keV level. This band is given the $1/2^- [530]$ assignment because the deduced decoupling parameter is in agreement with the value calculated for this configuration.

The 643.0- and 661.3-keV levels receive direct β^- population and their decay patterns suggest $1/2$ and $3/2$ spin, respectively. The $5/2^-$ member of this band is identified at 709.1 keV in the α decay of ^{253}Es . This band is given a tentative assignment of $1/2^- [521]$ because this is the only single-particle state available in this energy region. However, the small decoupling parameter of -0.19 (theoretical value is +1.02) suggests that this band has appreciable mixing with the $1/2^- [530]$ band. This mixing is supported by the difference in the deduced value of the decoupling parameter of -0.141 and the calculated value of -2.2 for the $1/2^- [530]$ band.

In the γ singles spectrum we observe a strong γ ray with energy of 1075.1 keV which we assign to the decay of a 1075.1-keV level. On the basis of the decay pattern of this level, it has been given a spin of $9/2$ and single-particle configuration $9/2^+ [624]$. Its $11/2$ member has been identified at 1143.8 keV and the $13/2$ member was observed in the $^{248}\text{Cm}(\alpha, t)$ and $^{248}\text{Cm}({}^3\text{He}, d)$ reactions at 1225 keV [9].

A level at 672.8 keV was deduced from the observation of 672.8- and 664.0-keV ($672.8 \rightarrow 664.0$) γ rays in the singles spectrum. Coincidence relationships establish the spin of the 672.8-keV level as $5/2$. The relative intensities of γ rays deexciting the 672.8-keV level are very similar to the relative intensities of γ rays from the decay of the $5/2^- [523]$ state at 447.8 keV in ^{247}Bk [12]. On the basis of this similarity, we assign the $5/2^- [523]$ Nilsson state configuration to

the 672.8-keV level.

The level at 767.9 keV decays to the 7/2 and 9/2 members of the ground-state band suggesting spin value of 7/2 or 9/2. This level does not decay to the 5/2⁺ band at 389.2 keV, thus favoring a 9/2 spin. We have tentatively identified the 11/2 and 13/2 members of this band at 836.1 and 911.2 keV, respectively. The three levels give a rotational constant of 6.2 keV, which is reasonable in this mass region. This band is interpreted as the 1⁻ octupole vibrational band built on the 7/2⁺[633] ground state.

The level at 932.2 keV decays to the 7/2 and 9/2 members of the ground-state band. This level also decays to the 767.9-keV level by a 164.4-keV M1 transition. Thus this level should have the same parity as the 767.9-keV level. The negative parity is also supported by the lack of transition to the 5/2⁺[642] band. On the basis of its decay, the most likely spin value for this state is 7/2. The 9/2, 11/2, 13/2 and 15/2 members of this band are tentatively identified at 988.1, 1055.8, 1133.8 and 1227.6 keV, respectively. These levels fit as members of a K=7/2 rotational band and give a value of 6.15 keV for the rotational constant. This band is interpreted as the 0⁻ octupole vibrational bands built on the 7/2⁺[633] ground state.

The decay patterns of the 1150.7- and 1223.0-keV levels suggest 5/2 and 7/2 spins, respectively. Because of very low hindrance factor for the 1223.0-keV level, we interpret it as the β -vibrational band built on the 7/2⁺[633] state. The 1150.7 keV level could be the {7/2⁺[633]⊗1⁻}5/2⁻ state. These vibrational bands lie at approximately the same energies in ²⁴⁸Cm [13].

In the present work we did not identify the 7/2⁻[514] orbital which is expected to lie in the 0.5-1.0 MeV range. This state is not expected to receive measurable α population because its intrinsic spin is antiparallel to the intrinsic spin of the ²⁵³Es ground state. The 9/2 member of this band is expected to have large cross section in the ²⁴⁸Cm(α ,t) reaction and this level was observed at 750 keV by Erskine *et al.* [3].

The single-particle energies measured in the present work are in fair agreement with level energies calculated with a Woods-Saxon potential [14] and also with a single-particle potential plus quasiparticle-phonon interaction [15].

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