Lifetimes of the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ $E2$ transitions in some actinide nuclei*

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Lifetimes of the $E2$ transitions from the $\frac{1}{2}+[631]$ state to the $\frac{5}{2}+[622]$ state have been measured in several actinide nuclei using pulsed beam and delayed coincidence techniques. The $\frac{1}{2}+[631]$ level was excited in ^{239}U and ^{243}Pu by the (d,p) reaction and in ^{241}Pu by the (d,t) reaction, and the subsequent γ-ray decay to the $\frac{5}{2}+[622]$ level was observed between beam pulses. The corresponding $E2$ transitions were observed in ^{243}Cm and ^{245}Cm from the radioactive decay of ^{243}Bk and ^{245}Bk, respectively, and the lifetimes were determined from electron-photon delayed coincidence experiments. The half-lives from these measurements are: ^{239}U (0.78 ± 0.04 μs); ^{241}Pu (0.88 ± 0.05 μs); ^{243}Pu (0.33 ± 0.03 μs); ^{243}Cm (1.08 ± 0.03 μs); and ^{245}Cm (0.29 ± 0.02 μs). Comparisons are made between the experimental transition probabilities and theoretical estimates. The differences between the transition rates in these nuclei are understood when pairing interactions are taken into account.

[NUCLEAR REACTIONS ^{238}U$(d,p\gamma)$, $E_d = 12$ MeV; ^{242}Pu$(d,p\gamma)$ and ^{242}Pu$(d,t\gamma)$, $E_d = 16$ MeV; measured $t_{1/2}$ of 133.7-, 161.4-, and 384.0-keV levels in ^{239}U, ^{241}Pu, and ^{243}Pu, respectively. Deduced $B(E2)$ values. Enriched targets. RADIOACTIVITY ^{243}Bk [from ^{241}Am$(\alpha, 2n)$] and ^{245}Bk [from ^{243}Am$(\alpha, 2n)$]; measured E_γ, E_{ce}, $t_{1/2}$ of 87.4-keV level in ^{243}Cm by K x-ray–ce delayed coin; measured $t_{1/2}$ of 355.7-keV level in ^{245}Cm by γ-ce delayed coin. Deduced $B(E2)$ values. Mass-separated sources.]

I. INTRODUCTION

The β_2 deformation[1] of nuclei in the $Z = 92$ to $Z = 98$ region is fairly constant (0.27) and hence it is expected that the wave functions of low-lying single-particle states in the Nilsson model should not change from one nucleus to another. To check this hypothesis it is necessary to compare measured values of the same quantity in several nuclei. Previously, the ground state magnetic moments[2] have been used to characterize the single-particle stares. However, since the ground state configurations of nuclei change with the number of added nucleons, this approach is not entirely satisfactory. On the other hand, it is possible to measure γ-ray transition probabilities between the same two Nilsson[3] states in different nuclei. Since γ-ray transition probabilities depend on the details of the nuclear wave functions, experimental transition probabilities should afford a sensitive test for the constancy of these wave functions.

It is known from nuclear reaction studies[4,5] that the energy spacing between the $\frac{1}{2}+[631]$ and $\frac{5}{2}+[622]$ neutron states remains ~100 keV in ^{239}U, ^{241}Pu, ^{243}Pu, ^{243}Cm, and ^{245}Cm. Furthermore, the $\frac{1}{2}+[631]$ state is expected to decay to the $\frac{5}{2}+[622]$ single-particle state by a pure $E2$ transition. Thus by measuring the lifetimes of these transitions, $B(E2)$ values can be extracted. In the present paper we describe the measurement of these lifetimes and discuss the near constancy of the observed $B(E2)$ values.

II. EXPERIMENTAL PROCEDURES AND RESULTS

A. Lifetimes of states populated in the (d,p) and (d,t) reactions

The lifetimes of the $E2$ transitions between the $\frac{1}{2}+[631]$ and $\frac{5}{2}+[622]$ single-particle states in ^{239}U, ^{241}Pu, and ^{243}Pu were measured by the pulsed beam technique.[6] The $\frac{1}{2}+[631]$ state in ^{239}U, which has been identified[7,8] in (n,γ) and (d,p) studies at 134 keV, was populated by bombarding a 50 mg/cm^2 ^{238}U foil with 12-MeV deuterons from the Argonne National Laboratory tandem Van de Graaff accelerator. The $\frac{1}{2}+[631]$ states in ^{241}Pu and ^{243}Pu which were identified in charged-particle reaction studies[4] to be at 161 and 384 keV, respectively, were excited by the (d,t) and the (d,p) reaction with 16 MeV deuterons. In this case a target was fabricated by placing a piece of ^{242}Pu metal weighing approximately 0.5 g between two 1 mg/cm^2 Ni foils which provided support and minimized the chance of contamination of the target chamber. Special handling was also required to avoid exposing the Pu metal to atmospheric oxygen, because Pu metal is known to be pyrophoric.

The deuteron beam was pulsed after acceleration by vertical electrostatic deflection. An 11-cm^3

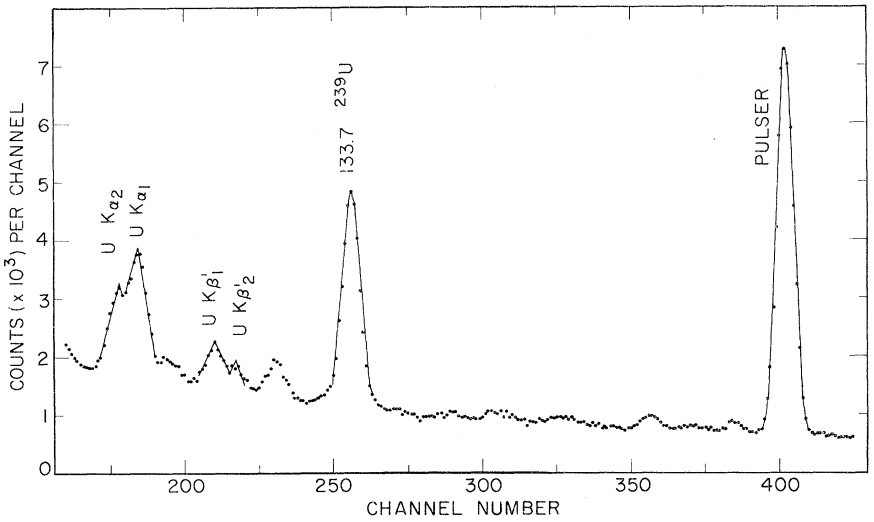

FIG. 1. The γ-ray spectrum from the $^{238}U(d,p\gamma)^{239}U$ reaction measured with an 11-cm^3 intrinsic Ge detector at 12 MeV bombarding energy. The spectrum was accumulated in the time interval 0.3 to 1.0 μs following the beam pulse for about 6 h of bombardment.

intrinsic Ge spectrometer having an energy resolution full width at half-maximum (FWHM) of 1.1 keV for the 122-keV γ rays of ^{57}Co was used in these measurements. The γ-ray pulse-height and time data were stored in a 512× 128 channel array in a magnetic core memory. The data were then examined by setting appropriate energy or time windows.

Figures 1 and 2 illustrate the γ-ray spectra which were accumulated in several hours while Fig. 3 shows the time spectra for decay of the $\frac{1}{2}$+[631] level to the $\frac{5}{2}$+[622] level in these nuclei.

The resolution of the ^{239}U spectrum (Fig. 1) is somewhat worse than that shown for the Pu isomers in Fig. 2 because of the experimental arrangement used to handle higher counting rates. In Fig. 2 two γ rays are observed in the decay of the $\frac{1}{2}$+[631] state in ^{243}Pu, since the $\frac{5}{2}$+[622] configuration is not the ground state in this nucleus as it is in ^{239}U and ^{241}Pu. Braid *et al.*[4] placed the $\frac{1}{2}$+[631] level, which appears in a complicated region of their (d,p) spectrum, at 397 keV, while the $\frac{5}{2}$+[622] state has been identified at 287.5 keV in the α-decay study[9] of ^{247}Cm. In the present in-

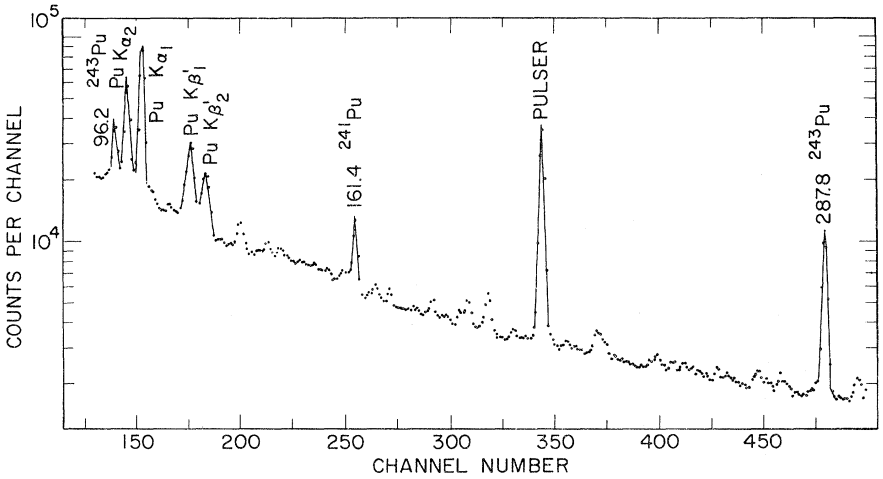

FIG. 2. The γ-ray spectrum from the $^{242}Pu(d,p\gamma)^{243}Pu$ and $^{242}Pu(d,t\gamma)^{241}Pu$ reactions measured with an 11-cm^3 intrinsic Ge detector at 16 MeV bombarding energy. Unidentified γ rays in the spectrum are attributed to fission products. The spectrum was accumulated in the time interval 0.3 to 1.0 μs following the beam pulse for about 10 h of bombardment.

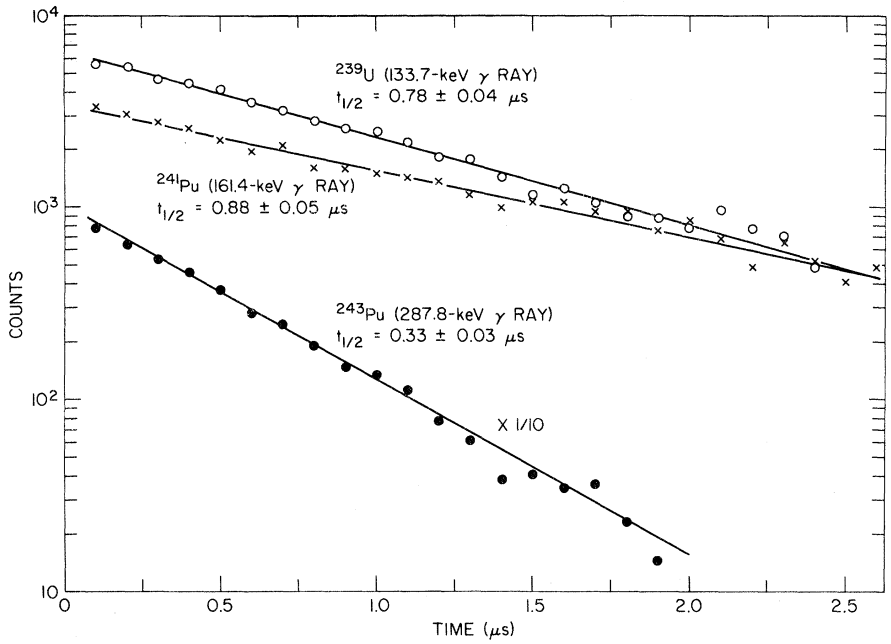

FIG. 3. Decay curves showing the decay of the $\frac{1}{2}+[631]$ state in ^{239}U, ^{241}Pu, and ^{243}Pu. Note that the curve for the 287.8-keV γ ray contained 10 times more counts than are shown on the ordinate.

vestigation the 96.2 ± 0.2 keV γ ray is identified as the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ transition and the $\frac{1}{2}+[631]$ level is placed at 384.0 keV. The measured half-lives of these γ rays are in good agreement with each other. The 287.8-keV γ ray was used to obtain the half-life of the $\frac{1}{2}+[631]$ isomeric state because of the difficulty in determining the background under the 96.2-keV γ ray. In addition to the γ rays attributed to the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ isomeric decays, additional peaks from the decay of fission products produced in these bombardments can be seen in the spectra of Figs. 1 and 2.

The experimentally determined half-lives are given in Table I. The time spectra were analyzed with the least-squares fitting computer code CLSQ.[10]

TABLE I. Summary of experimental half-life and transition probability data.

Nucleus	Transition energy (keV)	Half-life (μs)	Total $E2$ conversion coefficient α_T	Transition probability (s^{-1})
^{239}U	133.7 ± 0.2	0.78 ± 0.04	3.81	1.84×10^5
^{241}Pu	161.4 ± 0.2	0.88 ± 0.05	2.02	2.62×10^5
^{243}Pu	96.2 ± 0.2	0.33 ± 0.03	19.3	1.03×10^5
^{243}Cm	87.4 ± 0.1	1.08 ± 0.03	35.8	1.74×10^4
^{245}Cm	103.0 ± 0.2	0.29 ± 0.02	16.6	1.38×10^5

B. Lifetimes of states populated in radioactive decays

The delayed coincidence method was used to measure the $E2$ lifetimes of the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ transition in ^{243}Cm and ^{245}Cm. The $\frac{1}{2}+[631]$ state in these nuclei is populated by the electron capture (EC) decay of ^{243}Bk (4.3 h) and ^{245}Bk (5.0 d). The ^{243}Bk activity was produced by the irradiation of ~1 mg of ^{241}Am with 32 MeV α particles in the Argonne 60-in cyclotron. The irradiated Am was dissolved in 10 M HNO_3 – 0.1 M $KBrO_3$ and the Bk was extracted[11] three successive times with equal volumes of 0.14 F di-(2-ethylhexyl) orthophosphoric acid (HDEHP) in heptane. The fission products were removed by an extraction chromatographic column using tricapryl monomethyl ammonium thiocyanite as the stationary phase and NH_4CNS as the elutriant.[12] Additional decontamination from fission products was achieved by loading the activity on a column containing HDEHP on Celite and eluting[12] it with HCl. The chemically purified Bk fraction was mass-separated using the Argonne electromagnetic isotope separator[13] to prepare a thin sample for electron spectroscopy.

The ground state of ^{243}Cm is known to be $\frac{5}{2}+[622]$ from its α-decay studies,[14] and the Nilsson state $\frac{1}{2}+[631]$ was observed at 87 keV in the ^{244}Cm(d, t)-^{243}Cm reaction.[5] We measured the γ-ray spectrum of the mass-separated ^{243}Bk sample with a 25 cm^3 coaxial Ge(Li) detector and identified an 87.4 ± 0.1

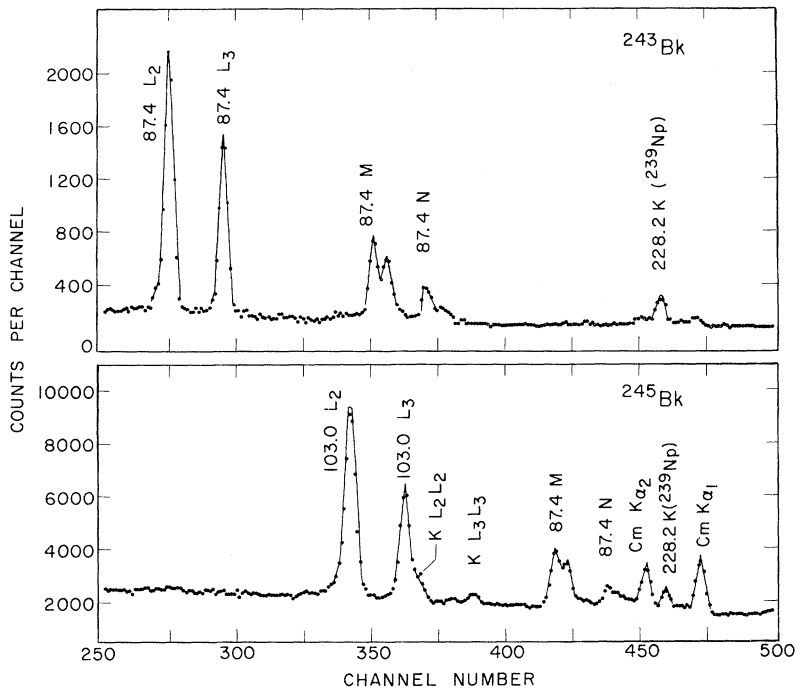

FIG. 4. Conversion-electron spectra of mass-separated ^{243}Bk and ^{245}Bk sources measured with a cooled Si(Li) detector. In addition to the electron lines due to the 87.4- and 103.0-keV transitions, the spectra also contain *KLL* Auger lines and Cm *K* x rays. The electron line at 106.4 keV is attributed to ^{239}Np contamination.

keV γ ray. The electron spectrum of the ^{243}Bk sample measured with a cooled Si(Li) detector[15] is shown in Fig. 4. The L_2 and L_3 electron lines of the 87.4 keV transition are the most prominent lines in this spectrum. The efficiency-geometry product of the electron detector was measured with a calibrated ^{203}Hg source. The absolute conversion coefficients of the 87.4-keV transition were experimentally determined for the L_2, L_3, M, and $N+O$ shells and were found to be 13.5, 8.8, 7.0, and 2.6, respectively. The corresponding theoretical values[16] for the 87.4-keV $E2$ transition

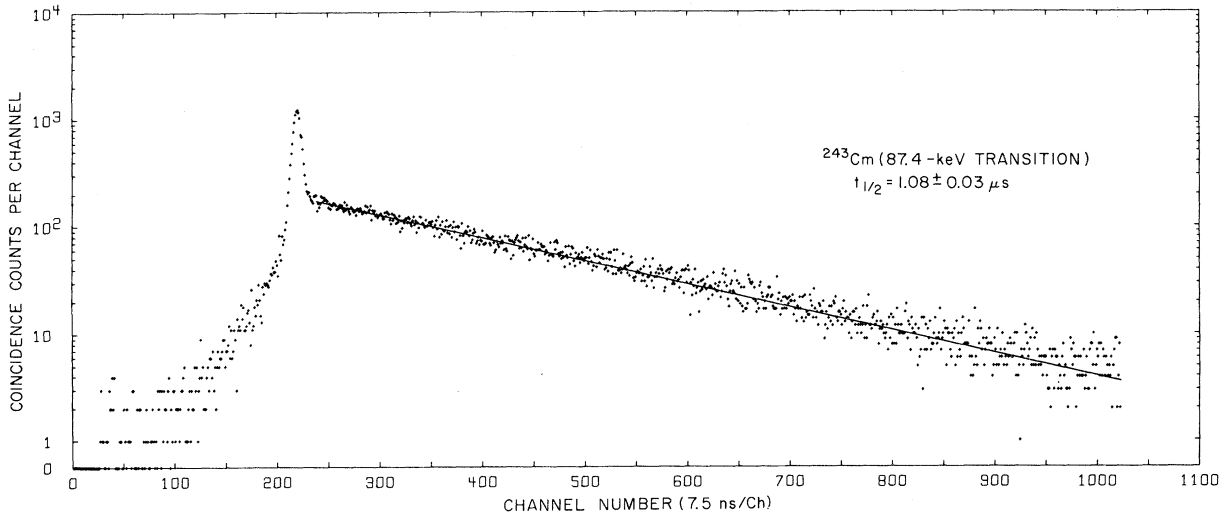

FIG. 5. Time-delay spectrum between Cm *K* x rays and 87.4 L_2+L_3 electrons from the decay of ^{243}Bk. The x rays were detected with a 7.6×7.6-cm NaI(Tl) crystal and the electrons were detected with a cooled Si(Li) detector. The geometry-efficiency product for the electron detector was 4%. The short lifetime to the left of the prompt peak is due to the decay of the 391.5-keV level in ^{239}Pu (^{239}Np contaminant).

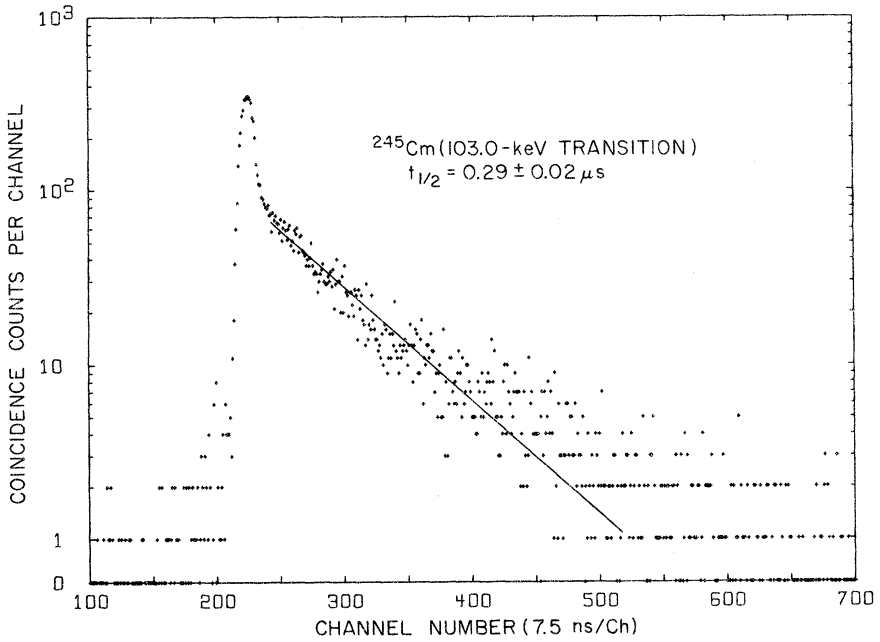

FIG. 6. Time-delay spectrum between the (320–450 keV) γ rays and 103.0 L_2+L_3 electrons from the decay of ^{245}Bk. The γ rays were detected with a 7.6×7.6-cm NaI(Tl) crystal and the electrons were detected with a cooled Si(Li) detector. The geometry-efficiency product for the electron detector was 4%.

are 15.0, 9.9, 7.2, and 2.6, respectively, which are in good agreement with the observed values.

The half-life of the 87.4-keV level in ^{243}Cm was determined by measuring the delayed coincidence spectrum between the Cm K x-rays and the L_2+L_3 electron lines of the 87.4-keV transition. The photons were detected with a 7.6×7.6-cm NaI(Tl) crystal and the conversion electrons were detected with a cooled Si(Li) detector. Timing single-channel analyzers (TSCA) were used to select the Cm K x-ray photopeak and the 87.4 L_2 and L_3 electron lines. The K x-ray pulses and electron pulses from the timing single-channel analyzers were used as start and stop signals for a time-to-amplitude converter (TAC). A time spectrum collected over a period of 7 h is shown in Fig. 5. The TAC was calibrated with a Tennelec model TC850 time calibrator. A least-squares fit to the observed data gave a half-life of 1.08±0.03 μs for the 87.4-keV level.

The ^{245}Bk activity was produced by the irradiation of ~20 mg ^{243}Am with 32-MeV α particles in the Argonne 60-in cyclotron. The Bk was extracted and purified by the same procedure as was used for ^{243}Bk. A thin sample of ^{245}Bk was prepared in the isotope separator and was used for the present

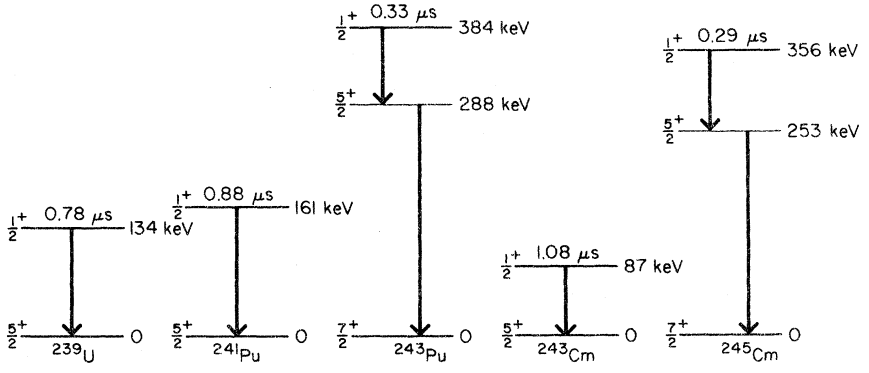

FIG. 7. Partial level diagrams showing the transition between the $\frac{1}{2}+[631]$ and $\frac{5}{2}+[622]$ states in ^{239}U, ^{241}Pu, ^{243}Pu, ^{243}Cm, and ^{245}Cm.

TABLE II. Transition probabilities of the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ γ ray.

Nucleus	$B(E2)_{\text{exp}}$ (e^2 fm^4)	$B(E2)_{\text{Nilsson}}$ (e^2 fm^4)	$(U_i U_f - V_i V_f)^2$	$B(E2)_{\text{theo}}$ (e^2 fm^4)	$\frac{B(E2)_{\text{exp}}}{B(E2)_{\text{theo}}}$	$\frac{T_{\text{exp}}}{T_{\text{Weisskopf}}}$
^{239}U	3.51	0.538	0.12	0.065	54	0.039
^{241}Pu	1.95	0.538	0.14	0.075	26	0.022
^{243}Pu	10.2	0.540	0.63	0.34	30	0.11
^{243}Cm	2.78	0.541	0.09	0.049	57	0.031
^{245}Cm	9.7	0.544	0.62	0.34	29	0.11

measurements.

In ^{245}Cm the $\frac{5}{2}+[622]$ state is known to be at 252.7 keV[17] and the $\frac{1}{2}+[631]$ state occurs at 358 keV.[5] In a recent study[18] of the ^{245}Bk decay scheme, the energy of the $\frac{1}{2}+[630] \rightarrow \frac{5}{2}+[622]$ transition has been found to be 103.0 ± 0.2 keV. Also, it has been observed that the 355.7-keV state is populated by several γ rays with energies between 350 and 410 keV.

The half-life of the 355.7-keV level was measured by the same experimental setup as was used for the half-life of the 87.4 keV level in ^{243}Cm. For start pulses the TSCA was set to select γ rays between 320 and 450 keV and for stop pulses the TSCA was set to select the L_2 and L_3 electron lines of the 103.0-keV transition. A time spectrum accumulated for about four days is shown in Fig. 6 and the measured half-life of the 103.0-keV transition was 0.29 ± 0.02 μs.

III. DISCUSSION

Partial energy level diagrams of ^{239}U, ^{241}Pu, ^{243}Pu, ^{243}Cm, and ^{245}Cm and the half-lives of the $\frac{1}{2}+[631] \rightarrow \frac{5}{2}+[622]$ transitions in these nuclei are shown in Fig. 7. The γ-ray lifetimes τ_γ were derived from the level lifetimes τ_l with the equation

$$\tau_\gamma = (1 + \alpha_T)\tau_l . \tag{1}$$

For conversion coefficients α_T the theoretical values of Hager and Seltzer[16] were used. Our measured conversion coefficients in ^{243}Cm agree well with theoretical values,[16] adducing that these are indeed normal $E2$ transitions. The reduced transition probability $B(E2)$ was extracted with the formula[3]

$$T(E2) = \frac{1}{\tau_\gamma} = \frac{4}{75}\frac{\pi}{\hbar}\left(\frac{E\gamma}{\hbar c}\right)^5 B(E2) . \tag{2}$$

In the above expression E_γ is the transition energy and $\hbar$ and c are the fundamental constants. The $T(E2)$ and $B(E2)$ values thus obtained are given in Tables I and II.

Theoretical $B(E2)$ values can be calculated from the nuclear wave functions and compared with the experimentally extracted quantities. According to Nilsson,[3] the $B(E2)$ value is given by the expression

$$B(E2)_N = \frac{5}{4\pi} e^2 \left(1 + \frac{Z}{A^2}\right)^2 \left(\frac{\hbar}{M\omega_0}\right)^2 \times |\langle I_i 2K_i (K_f - K_i) | I_f K_f \rangle|^2 G_{E2}{}^2 . \tag{3}$$

In the above equation G_{E2} is the $E2$ transition matrix element, the term in the angular brackets is the Clebsch-Gordan coefficient, and the subscripts i and f refer to the initial and final states. When the pairing correlations are taken into account, the above estimate for $E2$ transition rates between two different one-quasiparticle states becomes[19]

$$B(E2) = B(E2)_N P , \tag{4}$$

where the pairing factor is

$$P = (U_i U_f - V_i V_f)^2 . \tag{5}$$

In the above expression V is the occupation probability amplitude of the state.

The matrix element G_{E2} was calculated from Nilsson wave functions[3] at $\eta = 5.0$ and was found to be $+0.185$. A linear interpolation procedure was used to obtain the wave functions at this deformation, and the value of G_{E2} was assumed to be constant for the nuclei under investigation. The values of U^2 and V^2 for each nucleus were calculated with a BCS code using a standard method (e.g. Ref. 20) and Nilsson single-particle energies.[3]

The $B(E2)$ values calculated using Eq. (5) are given in Table II. Also included in this table are the ratio of the experimental transition probabilities to the Weisskopf estimate[21] of the transition rate and the ratio $B(E2)_{\text{exp}}/B(E2)$ which may be viewed as an enhancement factor for the transition.

From Table II it is apparent that the observed transition rates are more than an order of magnitude faster than the theoretical rates. The enhancement of the $E2$ transition rate could be caused by admixtures of collective components in the nuclear wave functions. These admixtures could arise either by second order Coriolis coupling between the $\frac{1}{2}+[631]$ and $\frac{5}{2}+[622]$ states or by γ-vibrational components in these states.

The Coriolis admixture of the $\frac{1}{2}+[631]$ band in the $\frac{5}{2}+[622]$ state was calculated using the computer code BANDMIX.[22] The calculations included the $\frac{3}{2}+[631]$ and $\frac{3}{2}+[622]$ states as intermediate states as well as the $\frac{1}{2}+[631]$ and $\frac{5}{2}+[622]$ states. For the particular case of ^{243}Pu the perturbed wave function for the $\frac{5}{2}+$ state was found to be

$$\Psi_{5/2} = -0.0036\Psi_{5/2,\,1/2[631]} + 0.095\Psi_{5/2,\,3/2[631]} + 0.0063\Psi_{5/2,\,3/2[622]} + 0.996\Psi_{5/2,\,5/2[622]} . \quad (6)$$

The admixture coefficient -0.0036 for the $\frac{1}{2}+[631]$ component would result in an enhancement of only 1.3×10^{-5} times the collective $B(E2)$ which for a $\frac{1}{2}+\rightarrow\frac{5}{2}+$ transition is typically around $9\times10^{4}\,e^{2}$ fm^4, as calculated from the average intrinsic quadrupole moment[23] obtained by Coulomb excitation and lifetime measurements in this region. The above estimate of the Coriolis admixture should also be taken as an upper limit, since the occupation probabilities were assumed to be unity and no Coriolis matrix element reduction factors were employed. In addition, the effect of Coriolis mixing on the transition rates in the isotonic series ^{239}U, ^{241}Pu, and ^{243}Cm was investigated by a calculation similar to that used for ^{243}Pu above. The admixture coefficients for the $\frac{1}{2}+[631]$ band in the $\frac{5}{2}+[622]$ state were 0.0023, 0.0020, and 0.0030 for ^{239}Pu, ^{241}Pu, and ^{243}Cm, respectively. The trend of ^{241}Pu having the smallest $B(E2)$ is thus reproduced; but, again, since the BCS pairing factor has considerable uncertainty for states near the ground state, the result of this analysis should not be considered complete. Also, in every case the calculated enhancements due to second-order Coriolis mixing contribute less than 30% as an upper limit to the observed enhancement of these transitions.

The particle-phonon interaction in the actinide nuclei has been considered by Komov, Malov, and Soloviev[24] where, for example, the $\frac{1}{2}+$ state in ^{239}U is described as

$$\Psi_{1/2+[631]} = 92\%\ \tfrac{1}{2}+[631] + 2\%\ \{\tfrac{3}{2}+[631]+Q(20)\} + 1\%\ \{\tfrac{5}{2}+[633]+Q(22)\}, \quad (7)$$

and the $\frac{5}{2}+$ ground state as

$$\Psi_{5/2+[622]} = 92\%\ \tfrac{5}{2}+[622] + 1\%\ \tfrac{5}{2}+[633] + 2\%\ \{\tfrac{5}{2}-[752]+Q(30)\} + 1\%\ \{\tfrac{1}{2}+[620\ +Q(22)\}. \quad (8)$$

The states are thus described as predominantly single-particle states with admixtures of particle-phonon coupled states where the appropriate phonon is given by $Q(\lambda\mu)$. A collective transition must thus occur from the $1\%\ \frac{5}{2}+[633]+Q(22)$ component in the $\frac{1}{2}+$ state to the $\frac{5}{2}+[633]$ single-particle component in the ^{239}U ground state. Clearly, this contribution to the transition rate is too small to account for the observed transition rates.

In conclusion, it should be remarked that this disparity of an order of magnitude between the theoretical and observed transition rates indicates that the theoretical $B(E2)$ matrix elements are only a factor of ~5 different from the experimentally deduced matrix elements. Furthermore, the $E2$ matrix element is small and its calculation involves cancellation of amplitudes for different l values, thus making it very sensitive to the accuracy of the wave functions. The enhancement in the $B(E2)$ value could, therefore, simply be due to systematic inadequacies in the Nilsson wave functions employed in the present calculations. The fact that the observed transition rates are nearly constant from one nucleus to the next after correction for pair correlation effects supports this conclusion. Another source of enhancement for the $B(E2)$ values could be from asymmetry in the nuclear shapes. However, little is known either experimentally or theoretically about the extent of nuclear asymmetry in odd-A deformed nuclei.

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