Electromagnetic properties of the excited states in ²³⁹Pu

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The electromagnetic properties of the 285.5- and 391.6-keV levels in ²³⁹Pu populated in the β^- decay of ²³⁹Np have been investigated. The half-lives of these levels have been measured using the delayed-coincidence technique to be $T_{1/2}(285.5) = 1.12 \pm 0.05$ nsec and $T_{1/2}(391.6) = 192 \pm 6$ nsec. The multipole mixing ratios of the 106.1-, 209.8-, 228.2-, and 277.6-keV transitions have been determined from γ - γ angular-correlation measurements to be $\delta (M2/E1)_{106.1} = -0.05 \pm 0.02$, $\delta (E2/M1)_{209.8} = -0.12 \pm 0.03$, $\delta (E2/M1)_{228.2} = -0.10 \pm 0.03$, and $\delta (E2/M1)_{277.6} = +0.17 \pm 0.02$. The mean precession angle $\omega\tau$ for the 285.5-keV level, in an external magnetic field of 20 kG, is measured to be $\omega\tau = 0.166 \pm 0.037$. From this the value of $g\beta$ is obtained to be 2.16 in the limit of L-S coupling. Using this the g factor of the 285.5-keV level has been deduced to be -0.49 ± 0.11 . The magnetic moment and the reduced transition probabilities have been compared with the theoretical estimates based on Coriolis-coupling calculations.

RADIOACTIVITY 239 Np; measured $\beta - \gamma$ and $\gamma - \gamma$ delay, $\gamma \gamma(\theta)$, $\gamma \gamma(\theta, H)$. 239 Pulevels, deduced $T_{1/2}$, δ , B(M1), B(E2), g.

I. INTRODUCTION

The energy levels of ²³⁹Pu, populated in the β^{-1} decay of ²³⁹Np and the electron-capture decay of ²³⁹Am, have been investigated by several work ers^{1-6} and its level scheme is well established. The lowest four excited states shown in Fig. 1 are interpreted as the members of the groundstate rotational band based on the $K = \frac{1}{2} [631]$ intrinsic state. The 285.5- and 391.6-keV levels are identified as the $\frac{5}{2}$ [622] and $\frac{7}{2}$ [743] Nilsson states, respectively. Earlier we had determined the mixing ratios of the transitions in ²³⁹Pu from γ - γ angular-correlation measurements using scintillation detectors.7 Krane, Olsen, and Steyert⁸ have recently determined the mixing ratios of the transitions in ²³⁹Pu from measurement of angular distributions of γ rays from oriented²³⁹Np. The mixing ratios of the transitions reported by them are in agreement with those of our earlier work.7 The ground state of ²⁴¹Pu is, like the 285.5keV level in ²³⁹Pu, the $\frac{5}{2}$ + [622] Nilsson state.⁹ The measured¹⁰ value of the magnetic moment of the ground state of ²⁴¹Pu is $\mu = -(0.718 \pm 0.017) \mu_N$, which can be compared with the Nilsson⁹ estimate $\mu = -0.5 \mu_N$ for this state. Bunker¹¹ has calculated the transition probabilities in ²³⁹Pu based on the Nilsson model with pairing correlations and Coriolis coupling taken into account.¹² It is therefore of interest to measure the magnetic moment of the 285.5-keV level in ²³⁹Pu and to compare it with different theoretical estimates.9,11

We report here the measurement of the magnetic

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moment of the 285.5-keV state using a perturbed angular-correlation technique. The half-life of this level has been measured with better accuracy. In addition the half-life of the 391.6-keV level has been measured. The γ - γ angular correlations have been remeasured using a Ge(Li)-NaI(T1) coincidence system in order to determine the mixing ratios of the transitions more accurately. Brief accounts of the present work have been reported earlier.¹³

II. EXPERIMENTAL PROCEDURES AND RESULTS

A. Source preparation

Sources of ²³⁹Np in the carrier-free form were obtained from the Radiochemistry Division of the Bhabha Atomic Research Centre, Trombay. Measurements were made with ²³⁹Np as neptunium nitrate in $8 M \text{ HNO}_3$. For lifetime measurements the carrier-free neptunium source was evaporated to dryness on a thin Mylar film.

B. Lifetime measurements

A time-to-amplitude converter (TAC) of the start-stop type was used for the measurement of the half-lives of the 285.5- and 391.6-keV levels. The start and stop pulses were obtained from two fast discriminators. The detectors were plastic scintillators mounted on RCA 8575 photomultipliers. Calibration of the TAC was done by introducing known delays using standard $50-\Omega$ cables.

The 106.1-277.6-keV γ - γ cascade was used for measuring the half-life of the 285.5-keV level. The 106.1-keV γ ray was detected in a 3.7-cm-diam \times 3.7-cm-thick 5% lead-loaded plastic scintillator and gated at the full energy peak. The 277.6-keV γ ray was detected in a 2.5-cm \times 2.5-cm NE 102A plastic scintillator and gated near the Compton edge of the 277.6-keV γ ray. The energy gates were selected using radioactive sources of ⁵⁷Co and ²⁰³Hg. The time spectrum obtained with the ²³⁹Np source is shown in Fig. 2. The prompt spectrum taken with a ⁶⁰Co source, with the same energy gates as for the ²³⁹Np source is also shown in Fig. 2. The half-life of the 285.5-keV level of ²³⁹Pu as obtained by a least-squares fit of the delayed part of the TAC spectrum is $T_{1/2} = 1.12$ ± 0.05 nsec; the error includes uncertainty in the time calibration. This is in agreement with the older value of 1.1 ± 0.1 nsec reported by Graham and Bell.14

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For the measurement of the lifetime of the 391.6keV level, the 328-keV- β -106.1-keV- γ delay was measured. The β rays were detected in a 2.5-cmdiam × 5-mm-thick NE 111 plastic scintillator. The energy gate was chosen at ~300 keV. The full energy peak of the 106.1-keV γ ray detected in the 5% lead-loaded plastic scintillator was chosen as the gate for the γ channel. The time spectrum is shown in Fig. 3. The least-squares fit of the delayed part of the spectrum gave the half-life of the 391.6-keV state as $T_{1/2} = 192 \pm 6$ nsec. This value agrees with $T_{1/2} = 193 \pm 4$ nsec reported by Engelkemeir and Magnusson.¹⁵

C. Angular-correlation measurements

A fast-slow coincidence setup with a 20-cm³ Ge(Li) detector and a 3.8-cm-diam $\times 3.8$ -cm-



FIG. 1. Partial decay scheme of ²³⁹Np.



FIG. 2. The decay curve of the 285.5-keV level as measured by the 106.1-keV- γ -277.6-keV- γ delayed coincidence. The "prompt" curve was measured using a ⁶⁰Co source.

NaI(Tl) detector along with a 512-channel analyzer was used for the angular-correlation measurements. The coincidence resolving time was 2τ = 60 nsec. The ²³⁹Np source was made in the carrier-free form as nitrate in 8 *M* HNO₃. The



FIG. 3. The decay of the 391.6-keV level as measured by the 328-keV- β -106.1-keV- γ delayed coincidence.

106.1-keV γ ray was detected in the movable NaI(T1) detector. The γ spectrum detected in the Ge(Li) detector in coincidence with the 106.1-keV γ ray was recorded in a multichannel analyzer. Data were collected at angles 90, 135, 180, 225, and 270°. The spectra at the equivalent angles of 90 and 270° and 135 and 225° were combined. The counts in the photopeaks of each of the 209.8-, 228.2-, and 277.6-keV γ rays in the coincidence spectra were integrated and corrected for chance coincidences. The 106.1-keV γ ray is known to be mainly E1 in nature from earlier studies^{2,6} and hence the coefficient of $P_4(\cos\theta)$ in the correlation function is expected to be negligible.⁷ The count rates of each γ -ray peak were fitted by a least-squares method to the correlation function

 $W(\theta) = 1 + A_2 P_2(\cos \theta)$.

The A_2 coefficients corrected for the finite solidangle attenuations are shown in Table I. These coefficients are analyzed graphically to get the multipole mixing ratios of the transitions (Fig. 4). The mixing ratio δ is the ratio of the quadrupole and dipole reduced matrix elements as defined by Biedenharn and Rose.¹⁶ The results of the internal-conversion measurements of Ewan *et al.*² were used to estimate the upper limits of the quadrupole admixture in the γ rays for the analysis. The mixing ratios of the transitions obTABLE I. The angular-correlation coefficients of the γ - γ cascades in ²³⁹Pu and the multipole mixing ratios of the γ rays deduced from them.

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γ-γ cascade (energies in keV)	A 2	Mixing ratio ^a	
106.1-209.8 106.1-228.2 106.1-277.6	$-0.003 \pm 0.004 -0.027 \pm 0.004 -0.001 \pm 0.005$	$\delta_{106,1} = -0.05 \pm 0.02$ $\delta_{209,9} = -0.12 \pm 0.03$ $\delta_{228,2} = -0.10 \pm 0.03$ $\delta_{277,6} = +0.17 \pm 0.02$	

^a See text.

tained from the analysis of the angular correlations are also shown in Table I. These are in good agreement with the values reported by Krane, Olsen, and Steyert.⁸ The difference in sign batween the present value and that of Krane, Olsen, and Steyert⁸ for the mixing ratio of the 106.1-keV transition is due to the different sign conventions used for the first γ ray in the cascade.

The angular correlation of the 106.1-228.2-keV γ - γ cascade was measured earlier⁷ using a solid source in the form of NpO₂. The A_2 coefficient corrected for finite solid angle of the detectors was $A_2 = -0.021 \pm 0.006$. This coefficient is to be corrected for the interfering cascades accepted because of the poorer energy resolution of the NaI(T1) detectors. With these corrections⁷ the correla-



FIG. 4. Graphical analysis of the γ - γ angular-correlation coefficients. The upper limits of the quadrupole admixtures of the γ transitions were obtained from high-resolution internal-conversion measurements of Ref. 2. The mixing ratios of the transitions were obtained accurately from the analysis.

tion coefficient with a solid source becomes $A_2 = -0.025 \pm 0.008$. This value agrees with the one obtained using a liquid source (Table I).

D. g factor of the 285.5-keV state

As the 106.1-228.2-keV γ - γ cascade has the largest anisotropy (see Table I) it was used for the measurement of the g factor of the 285.5-keV state. The integral perturbed angular-correlation method was employed. The γ rays were detected in two 4.4-cm-diam×5.0-cm-thick NaI(Tl) scintillators. An external magnetic field of 20 kG was applied perpendicular to the plane of the detectors. The source was made from carrier-free ²³⁹Np in 8 M HNO₃. The fractional change R in the coincidence count rate with field up and field down was measured with the counters at $\pm 135^{\circ}$. The value of R obtained for $+135^{\circ}$ (0.0104 \pm 0.0021) and -135° (-0.0096 ± 0.0020) agreed with each other within statistical errors. The average value is |R| $= 0.0100 \pm 0.0014.$

The angular correlation of the 106.1-228.2-keV γ - γ cascade was also measured at five angles in the same geometry as was used for the measurement of R. A least-squares analysis of the data gave the correlation coefficient, uncorrected for geometry, to be $A_2 = -0.022 \pm 0.004$. This value was used to calculate $\omega \tau$ from the measured value of R using the relations:

$$R_{\pm 135} = \mp \frac{4C_2 G_2 \omega \tau}{1 + (2G_2 \omega \tau)^2} ,$$

where

$$C_2 = \frac{3A_2}{4+A_2}$$
,

and

$$\omega = -\frac{g\mu_N H_{\rm eff}}{\hbar}$$

Here τ is the mean life of the nuclear state, G_2 is the integral attenuation coefficient due to possible time-dependent perturbations and $H_{\rm eff}$ is the effective magnetic field at the nucleus. The value of $G_2\omega\tau$ obtained from the present measurements is $G_2\omega\tau=0.166\pm0.037$ rad.

Since the observed angular-correlation coefficient A_2 , is the same within statistical errors for solid and liquid sources (see Sec. II C), the attenuation coefficient G_2 is close to unity. Gunther and Parsignault¹⁷ have measured the differential α - γ correlation in the decay of ²⁴¹Am to ²³⁷Np. Assuming the extranuclear interactions to be similar in ²³⁷Np and ²³⁹Pu we get $G_2 = 0.97$ for the 285.5-keV level in ²³⁹Pu. This is consistent with the observed correlation using solid and liquid sources of ²³⁹Np. In the following we have taken the value of $G_2 = 1.0$. Since plutonium is paramagnetic, the magnetic field at the nucleus H_{eff} is different from the externally applied field: $H_{\text{eff}} = \beta H_{\text{ext}}$, where β is the paramagnetic correction factor.¹⁸ Using $H_{\text{ext}} = (20.0 \pm 0.4)$ kG and $\tau = 1.62 \pm 0.07$ nsec we get $g\beta = -1.06 \pm 0.25$.

E. Paramagnetic correction

In order to deduce the g factor of the 285.5-keV state from the measured value of $g\beta$, the paramagnetic correction factor has to be estimated. In the case of the tripositive rare-earth ions the β factors have been calculated by Gunther and Lindgren¹⁸ and are found to be large. Such calculations are not available for the actinide ions. The actinide ions can exist in a number of ionic states in solution.

Plutonium in 8 M HNO₃ exists mostly in the Pu⁴⁺ state, although at lower acidities Pu³⁺ and PuO₂²⁺ (Pu⁶⁺) can also coexist.¹⁹ In the present case since the 285.5-keV state is populated through the isomeric state at 391.6 keV ($T_{1/2}$ = 192 ± 6 nsec) we assume that the stable state of plutonium is reached within this time. The electronic configuration of Pu⁴⁺ ion is 5 f^4 beyond the radon core. The paramagnetic correction factor may be estimated using the relation¹⁸

$$\beta = 1 + \frac{\langle J_{g} \rangle}{J} \frac{H_{5f}}{H_{\text{ext}}},$$

where $\langle J_{\bullet} \rangle / J$ describes the average orientation of the 5f shell in the external field and H_{5f} is the magnetic field due to the 5f shell at the nucleus for $J_{g} = J$. The β factors, calculated for rare earths in the limit of L-S coupling, compare well with the experimental results.¹⁸ Gunther and Parsignault have calculated the β factor for Np⁶⁺(5f¹) to be $\beta = 1.68 \pm 0.11$, which compares well with $\beta = 1.85 \pm 0.37$ derived indirectly from experimental results.¹⁷ In the case of actinides appreciable departures from Russel-Saunders coupling have been observed.²⁰ These departures can be taken into account in the intermediate coupling calculations. Moreover, appreciable crystal field effects can also be present due to complex formation by actinides in solution.²¹ In the present case the value of β has been estimated in the limit of L-S coupling neglecting crystal field effects. In this limit β is given by¹⁸

$$\beta = 1 + \frac{J(J+1)g_J}{3kT} \frac{\mu_0}{4\pi} 2\mu_B^2 \langle r^{-3} \rangle \langle J \| N \| J \rangle,$$

where g_J is the Lande g factor, $\langle r^{-3} \rangle$ is the expectation value of the inverse cube of the radius of the electron orbit, and $\langle J || N || J \rangle$ is the radial

matrix element. The values of the radial matrix elements are tabulated by Elliot and Stevens²² and of $\langle r^{-3} \rangle = 7.599a_0^{-3}$ has been taken from the relativistic calculations of Lewis *et al.*²³ Using these we get $\beta = 2.16$ for Pu⁴⁺ at T = 300 K.

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We have made an attempt to observe any change in β for plutonium in different chemical forms of the source by measuring $\omega \tau$ of the 285.5-keV level. The R measurements discussed in Sec. IID were repeated using (i) a solution of carrierfree neptunium in 1 M HNO₃ containing 0.1 M $K_2Cr_2O_7$ as an oxidizing agent (source B) and (ii) neptunium in 1 M HNO₃ containing 0.1 M FeSO₄ as a reducing agent (source C). The measured values of R are shown in Table II. In solutions containing macro amounts of neptunium, Np⁶⁺ is the main oxidation state present under conditions of source B, and Np⁴⁺ under conditions of source C. If the ionic state of plutonium is different in these two forms of the source and in 8 M HNO_3 , the quantity R is expected to be different due to differences in the β factors (Table III). However, because of the large statistical errors no definite conclusion could be reached regarding the ionic state of plutonium in the various conditions of measurement. The differential perturbed angular-correlation technique is ideally suited for seeing such changes. However, such measurements were not feasible in the present case due to the short half-life of the intermediate level and the small anisotropy of the angular correlation.

Assuming the ionic state of plutonium, after β^- decay of ²³⁹Np, to be Pu⁴⁺ in 8 *M* HNO₃ solution, the *g* factor of the 285.5-keV level has been obtained to be -0.49±0.11.

V. DISCUSSION

The reduced transition probabilities of the γ rays from the 285.5-keV state have been calculated from the present measurements of the lifetime and the mixing ratios (Table IV). The conversion coefficients of the γ rays have been taken from Hager and Seltzer.²⁴ The present experimental values of B(M1) and B(E2) are considerably

TABL I. The measured values of R for the 106.1-228.2-ke. cascade in an external field of 20 kG with different chemical forms of the ²³⁸Np source.

Form of source	R		
A Nitrate in $8 M$ HNO ₃	0.0100 ± 0.0014		
B Nitrate in $1 M$ HNO ₃ containing			
$0.1 M \text{ K}_2 \text{Cr}_2 \text{O}_7$	0.0113 ± 0.0025		
C Nitrate in $1 M$ HNO ₃ containing	0 0110 / 0 0007		
$0.1 M$ re SO_4	0.0112 ± 0.0037		

TABLE III. Values of the paramagnetic correction factor β for various oxidation states of Pu in the *L*-S coupling limit (see text).

Oxidation state	Electronic con- figuration beyond the radon core	$\langle r^{-3} \rangle^{a}$ (a_0^{-3})	β	
Pu ⁷⁺ (Np ⁶⁺)	$5f^1$		1.68 ^b	
Pu^{6+} (Np ⁵⁺)	$5f^2$	8.854	2.64	
Pu^{5+} (Np ⁴⁺)	$5f^{3}$	8.223	2.82	
Pu^{4+} (Np ³⁺)	$5f^4$	7.591	2,16	
Pu^{3+} (Np ²⁺)	$5f^5$	6.942	1.24	

^a From Ref. 23.

^b From Ref. 17.

different from those given by Krane, Olsen, and Steyert⁸ even though the mixing ratios of the transitions are in agreement with their values. It appears that they have not taken the internal conversion of the γ rays into account in deducing the partial γ -ray transition probabilities. The B(M1)and B(E2) values obtained from the measurements have been compared with the calculations of Bunker.¹¹ The Coriolis matrix elements between the $\frac{3}{2}$ [631] and $\frac{5}{2}$ [622] bands are particularly large.¹² The $\frac{3}{2}$ [631] state is assumed⁸ to lie at ~700 keV in ²³⁹Pu. Bunker¹¹ has given two sets of admixed wave functions for ²³⁹Pu. It is found that the calculated energy spectrum for Set II compares better with the observed spectrum than the one with Set I. The transition probabilities calculated using both the Sets are given in Table IV. The present experimental values of the M1 transition probabilities agree well with the values given with Set II. The experimental values of B(E2) do not agree with these calculations. Krane, Olsen, and Stevert⁸ have compared their values with Set I.

The magnetic moment of the 285.5-keV $\frac{5}{2}$ [622]

TABLE IV. Reduced transition probabilities of γ transitions depopulating the 285.5-keV level in ²³⁹Pu.

γ-ray	$\frac{B(M1)}{(10^{-4}\mu_N^2)}$			B (e ²	(E2) fm ⁴)	
energy	Exp.	Calc. ^a		Exp.	p. Calc. ^a	
(keV)	_	Set I	Set II		Set I	Set II
209.8	1.36 ± 0.08	1.6	0.75	0.64 ± 0.32	5.3	2.50
228.2	3.53 ± 0.21	7.8	3.77	1.00 ± 0.65	1.6	0.67
277.6	2.29 ± 0.14	4.1	2.00	1.27 ± 0.27	0.01	0.03
285.5	•••	•••	•••	2.15 ± 0.13	3.8	2.52

^a Values calculated using the wave functions of Bunker (Ref. 11) taking pairing correlations and Coriolis coupling into account. The Nilsson parameters are taken to be $\delta = 0.24$, $\kappa = 0.0635$, $\mu = 0.325$, and the static moments $Q_0 = 11 \times 10^{-24}$ cm², $g_s = 0.8$ g_s^{free} , and $g_R = 0.1$.

state obtained in the present work is $\mu = -(1.23 \pm 0.25)\mu_N$. The Nilsson estimate for this state is $\mu_{\text{Nil}} = -0.49\mu_N$ using $g_R = Z/A$ and $g_s = g_s^{\text{free}}$. The value of the magnetic moment calculated using the Coriolis coupling wave functions¹¹ (Set II) is $\mu_{\text{Cor}} = -0.74\mu_N$ for $g_s = g_s^{\text{free}}$. The experimental value of μ differs considerably from these estimates.^{9,11}

Note added in proof. When internal conversion of the γ rays is properly taken into account in the work of Krane, Olsen, and Steyert,⁸ the reduced transition probabilities are in good agreement with those in the present work (K. S. Krane, private communication).

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