

Improved Value for the Energy Splitting of the Ground-State Doublet in the Nucleus ^{229}Th

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Abstract

We have made an improved estimate of the $^{229\text{m}}\text{Th}$ isomer energy. The new value 7.8(5) eV includes an estimate of spectral contamination due to the out-of-band E2 transition from the 42.43-keV 7/2+ member of the [633] ground state band to the 3/2+ [631] $^{229\text{m}}\text{Th}$ bandhead. We estimate a 2% branching ratio for this unobserved transition in the 42.43-keV 7/2+ [633] deexcitation. The excitation of the $^{229\text{m}}\text{Th}$ level is increased from the previously reported value of 7.6(5) eV to the new value of 7.8(5) eV when this branch is included in the analysis.

1 Introduction

Beck, et al., report the energy difference between the ground state of ^{229}Th and the first excited state ($^{229\text{m}}\text{Th}$) as $\Delta E(^{229}\text{Th}) = 7.6(5)$ eV [1]. They measured γ -rays following the α -decay of ^{233}U to ^{229}Th , and particularly the γ -ray cascade from the ^{229}Th 71.82-keV level. This level decays predominantly by 2 step γ -ray cascades, populating both members of the ground state doublet with (1) an inband [631] two-step γ -ray sequence to the isomeric level (71.82 \rightarrow 29.19 \rightarrow $^{229\text{m}}\text{Th}$ keV), and (2) an out-of-band transition to the 42.43-keV member of the ground state band (71.82 \rightarrow 42.43 keV) followed by an inband [633] transition (42.43 \rightarrow 0 keV) (Fig. 1). The relevant portion of the measured γ -ray spectrum consists of 2 doublets, each with an energy splitting of ~ 200 eV. One doublet is composed of the 42.43 and 42.63-keV γ rays (ΔE_{42}), and the other doublet is composed of the 29.18 and 29.39-keV γ rays (ΔE_{29}). The difference in the energy sum $\Delta E = \Delta E_{29} - \Delta E_{42}$ yields a first approximation to the energy splitting of the ^{229}Th ground-state doublet. Beck, et al., obtained 7.0(5) eV for the raw centroid difference. They noted that the peak in the γ -ray spectrum corresponding to the inband decay of the 29.19-keV state to the upper (isomeric) doublet member may be complex, including a small contribution from an M1 ground state branch, 29.19 \rightarrow 0 keV. Therefore, a correction to the first-order centroid analysis is required and gives $\Delta E(^{229}\text{Th}) = (\Delta E_{29} - \Delta E_{42})/(1 - b_{29})$, where b_{29} is the branch 29.19 \rightarrow 0 keV. Beck, et al., assumed the rotational model, used measured nuclear data, and estimated the 29.19 \rightarrow 0 keV branching at 1/13; the correction for this unobserved branch amounts to + 0.6 eV, resulting in a value of the doublet splitting $\Delta E(^{229}\text{Th}, \text{eV}) = 7.0(5) + 0.6 = 7.6(5)$.

Singh [2] asked about the possible effect of a spectral contaminant due to the small out-band E2 branch 42.43 \rightarrow $^{229\text{m}}\text{Th}$, a branch also unresolved in the γ -ray spectroscopy but one that could have a small effect on the centroid analysis of ΔE_{42} . The effect of a 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV branch on the value of $\Delta E(^{229}\text{Th})$ was not included in the analysis presented in [1]. The issue may be important in this unique circumstance, because of the eV energy splitting of the doublet and the extraordinary resolving power of the NASA micro-calorimeter/spectrometer XRS [3, 4].

2 The 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV Branching Ratio

The branching of the 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV transition can be estimated with the rotational model parameters (Q_0 , Q_2 , $|g_K - g_R|$) which describe the electromagnetic transition strengths of inband and out-of-band γ -ray decays [5]. These parameters can be obtained from comparison with the measured properties of the 97.13-keV level [6]. The lifetime of the 97.13-keV $J^\pi = 9/2^+$ member of the ground state band is measured to be 0.147 (12) nsec [7]. The inband and cross-over γ -ray branching ratios are also known, as are level spins and parities [6]. The rotational model equations for the electromagnetic transition rate strengths are given in [5]. They are in standard notation:

$$B(E2; I_i \rightarrow I_f) = \left(\frac{5}{16\pi} \right) e^2 Q_0^2 \langle I_i K 2 0 | I_f K \rangle^2, \quad (1)$$

$$B(M1; I_i \rightarrow I_f) = \left(\frac{3}{4\pi} \right) (g_K - g_R)^2 K^2 \langle I_i K 2 0 | I_f K \rangle^2, \text{ and} \quad (2)$$

$$B(E2; I_i \rightarrow I_f) = \left(\frac{5}{16\pi} \right) e^2 Q_2^2 \langle I_i I_f 2 \Delta K | K_i K_f \rangle^2. \quad (3)$$

Eq. (1) is for inband E2 transitions, Eq. (2) is for inband M1 transitions, and Eq. (3) is for out-of-band E2 transitions. We use the equations above and deduce (1) the intrinsic quadrupole moment Q_0 from the average of the measured inband 97.13 \rightarrow 42.43 keV and 97.13 \rightarrow 0 keV E2 transition rates, (2) the g-factor $|g_K - g_R|$ from the inband 97.13–42.43 keV M1 transition, and (3) Q_2 from the cross-over transition 97.13 \rightarrow 29.19 keV. We use these parameters to predict the inband and cross-over transition rates for the 42.43-keV level. Numeric values are given in Table 1. The calculated 42.34 \rightarrow 0 keV transition rates are $\lambda[E2, s^{-1}] = (1.86 \times 10^{-6}) \times 10^{12}$ and $\lambda[M1, s^{-1}] (8.12 \times 10^{-6}) \times 10^{12}$, and for the 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV E2 transition rate $\lambda[E2, s^{-1}] = 0.222 \times 10^{-6} (x 10^{12})$. Thus, the magnitude of the 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV branch is $b_{42} = 0.22 / (0.22 + 8.12 + 1.86) = 0.02$. The sense of the energy correction (+) is the same as for contamination in spectral analysis due to the 29.19 \rightarrow 0 branching. The formula for the energy splitting when both branches are considered is $\Delta E(^{229}\text{Th}) = (\Delta E_{29} - \Delta E_{42}) / (1 - b_{29} - b_{42})$, and which gives the energy splitting of the ^{229}Th ground-state doublet $\Delta E(^{229}\text{Th}) = 7.8(5)$ eV.

3 Summary

The best value for the energy splitting of the ^{229}Th ground state doublet requires estimates of spectral contaminations due to the unobserved 42.43 \rightarrow $^{229\text{m}}\text{Th}$ keV and 29.19 \rightarrow 0 keV out-of-band transitions. We find $\Delta E(^{229}\text{Th}) = 7.8(5)$ eV after making these corrections. This extremely unusual and rare doublet may remain a scientific curiosity or else it may represent an important pathway in the field of atomic-nuclear coupling. Work on the nuclear and atomic properties of the excited doublet state continues to be reported. Inamura and Haba [8] report excitation of $^{229\text{m}}\text{Th}$ in a hollow cathode electrode discharge; they report the isomer half-life and excitation as $1 \leq T_{1/2}(\text{min}) \leq 3$ and $73 \leq E_x(\text{eV}) \leq 7$, respectively. Burke and colleagues [9] are mounting an experiment at LLNL to measure the nuclear properties of the isomer. Chapman *et al.*, have trapped and cooled $^{232}\text{Th}^{3+}$ ions [10]; currently they are mounting an experiment with the goal of measuring the hyperfine structure of $^{229}\text{Th}^{3+}$ ions.

Table 1: Rotational band parameters Q_0 , Q_2 , and $|g_K - g_R|$ for ^{229}Th ground state band [633] deduced from data summarized in [6], and from equations for $B(\text{ML})$ given in [5]. Entries for the 42.43-keV level transition rate are calculated with $B(\text{ML})$ values obtained from analysis of the 97.13-keV level.

$E_i \rightarrow E_f$ (keV)	E_γ (keV)	$B(\text{E}2)$ ($\text{e}^2 \text{fm}^4$)	$B(\text{M}1)$ (μ_n^2)	Q_0 (e b)	Q_2 (e b)	$ g_K - g_R $ (μ_n^2)	$\lambda[\text{E}2]$ (s^{-1})	$\lambda[\text{M}1]$ (s^{-1})
97.13 \rightarrow 0	97.13	$3.4(8) \times 10^3$		5.86				
97.13 \rightarrow 42.43	54.70	$4.3(27) \times 10^3$		5.57				
97.13 \rightarrow 42.43	54.70		$9.2(19) \times 10^{-3}$			0.014		
97.13 \rightarrow 29.19	67.94	$1.6(4) \times 10^3$			2.20			
42.43 \rightarrow 0	42.43			5.71 ^a		0.014	1.8×10^6	8.2×10^6
42.43 \rightarrow $^{232\text{m}}\text{Th}$	42.43				2.20		0.22×10^6	

^a Unweighted average of Q_0^2 determined from $B(\text{E}2; 97.13 \rightarrow 0 \text{ keV})$ and $B(\text{E}2; 97.13 \rightarrow 42.43 \text{ keV})$.

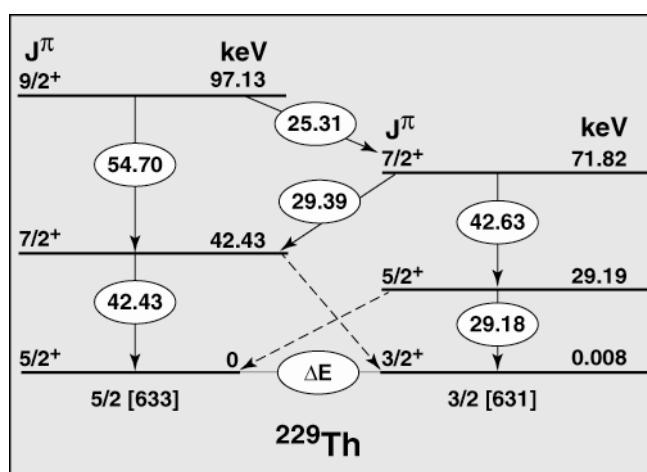


Fig. 1: Partial level scheme for ^{229}Th . The 71.82-keV level decays both to the ground state and to the excited member of the ground state doublet at 0.008 keV. Transitions suggested by the dashed lines are nearly degenerate with the known cascade decays of the 71.82-keV level.

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References

- [1] B. R. Beck, *et al.*, Phys. Rev. Lett. 98, 142501 (2007).
- [2] B. Singh, private communication (2007).
- [3] F.S. Porter, *et al.*, Rev. Sci. Instr. 75, 3772 (2004).
- [4] C.K. Stahle, *et al.*, Nucl. Instrum. and Meth. Phys. Res. **A520**, 466 (2004).
- [5] A. Bohr and B.R. Mottelson, in Nuclear Structure, Vol. II, W.A. Benjamin, Reading, Massachusetts, 1975. pp. 45, 56, 58.
- [6] Y. Akovali, Data Sheets 58, 555 (1989). See also R.E. Ruchowska, *et al.*, Phys. Rev. 73, 044326 (2006).
- [7] H. Ton, S. Roodbergen, J. Brasz, and J. Blok, Nucl. Phys. A155, 245 (1970).
- [8] T.T. Inamura and H. Haba, Phys. Rev. 79, 0343139 (2009). A large number of references are included in the manuscript.
- [9] J.T. Burke, Bull. Amer. Phys. Soc. DNP09, October 13-17, 2009, Hawaii.
- [10] C.J. Campbell, A.V. Steele, L.R. Churchill, M.V. DePalatis, D.E. Naylor, D.N. Matsukevich, A. Kuzmich, and M.S. Chapman, Phys. Rev. Lett. **102**, 233004 (2009).