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Energy of the $3/2^{+}$ state of ^{229}Th reexamined

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^{229}Th has an isomeric state of unusually low energy, whose adopted value is by now 3.5(10) eV. This value was determined indirectly, based on several very precise γ-ray energies, between 25 and 217 keV, from the α decay of ^{233}U. Two recent works suggest that the decay pattern of the transitions that link the low-energy levels of ^{229}Th is different from that assumed in earlier works, but there also is a difference between them. In this article we investigate the effect on the value determined for the excitation energy of that isomeric state if those different assumptions regarding the γ-ray transitions in ^{229}Th are considered. We use published data and a statistical method that takes into account both variances and correlations between data. Adopting the statistically most acceptable assumption regarding the decay pattern of ^{229}Th, we deduced the value of 5.5(10) eV for the excitation energy of that isomeric state, with consequences for both theoretical and experimental studies related to that level.

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INTRODUCTION

The existence of a $\frac{3}{2}^{+}$ excited state quite close to the $\frac{5}{2}^{+}$ ground state of ^{229}Th was put into evidence about three decades ago [1]. In 1990, the energy difference between these levels was shown to lie below 7 eV (at the 2σ level) [2]. A few years later the excitation energy of the $\frac{3}{2}^{+}$ state, hereafter cited as Δ, was determined from a very detailed energy measurement of many γ rays emitted in the α decay of ^{233}U [2,3]. Thereafter, the accepted value, calculated by Helmer and Reich [3], is $\Delta = 3.5(10)$ eV.

This unusually low excitation energy is of great interest in many experimental and theoretical studies, for which the results depend critically on the knowledge of Δ [4–9]. For instance, the nuclear half-life of the $\frac{3}{2}^{+}$ level by an M1 electromagnetic transition depends strongly on Δ. Investigation of the consequence of different Δ values on the nuclear-spin mixing shows that the $\frac{3}{2}^{+}$ half-life, the energy of the emitted photon, and the mixing ratio of the $\frac{3}{2}^{+}$ level and the ground state in a hydrogenlike ^{229}Th^{89+} ion also strongly depend on Δ (see Ref. [4] and others cited therein). If the isomer excitation energy is greater than the ionization potential energy of Th, $\sim$5.9 eV, the $\frac{3}{2}^{+}$ state would rapidly decay to the ground state by internal conversion [7].

Because there is no an unambiguous measurement of the electromagnetic transition from the $\frac{3}{2}^{+}$ excited state to the ground state [8,9], the determination of the $\frac{3}{2}^{+}$ excitation energy depends not only on the precise measurement of the γ-ray energies, which feed each of those levels, but also on some assumptions about other relevant level energies and the γ-ray transition pattern of ^{229}Th. Recent experimental and theoretical studies of the nuclear structure of ^{229}Th [10,11] suggest a pattern of γ-ray transitions different from that assumed in Refs. [2] and [3], but there also is no complete agreement between their claims. Specially, the decays of the 29- and 71-keV levels must be considered. Formerly [2,3], these were assumed to feed only the $\frac{3}{2}^{+}$, whereas the nuclear model calculation of Ref. [10] indicates that both levels decay also to the ground state (see Fig. 1). To study the consequence of the different assumptions on the determination of the excitation energy of the $\frac{3}{2}^{+}$ level, we extended the work of Refs. [2] and [3], including more experimental data and new values of standard γ-ray energies [12]. We used the least-squares method in a matrix formalism that considers in the fit the totality of the available experimental information. So, the standard γ-ray energies, the experimental data from Refs. [3] and [13], and the energies of all the relevant levels and γ-ray transitions in the nucleus under study are taken into account on the same footing [14,15].

ANALYSIS

The energy of the $\frac{3}{2}^{+}$ level was fitted using the same general procedure adopted in Refs. [2] and [3], that is, considering multiple cascade/crossover relations between the γ rays of ^{229}Th. However, through the matrix formalism [14,15], both the calibration standards and the precise thorium γ-ray energies were considered in an equivalent manner. This method delivers best values, variances, and covariances that are consistent, in a statistical sense, for the whole set of experimental quantities subjected to the fit, as may be retrieved by consulting Refs. [14] and [15]. The use of the covariances is an important upgrading in the statistical analysis with respect to previous work. The level scheme of ^{229}Th is shown in Fig. 1: those transitions, which are by now well established, are identified by continuous arrows; dashed and dotted lines indicate the transitions that may be allocated to different final states, following the different experimental and theoretical results to be presented later. (The dotted lines correspond to transitions that were not used to the determination of Δ.)

The fit was performed using all experimental data from Tables III and V of Ref. [3] and from Table 3 of Ref. [13], after reducing all energy values by 5.8 ppm to update them to the 1986 fundamental constant values [12]. (Data related to the

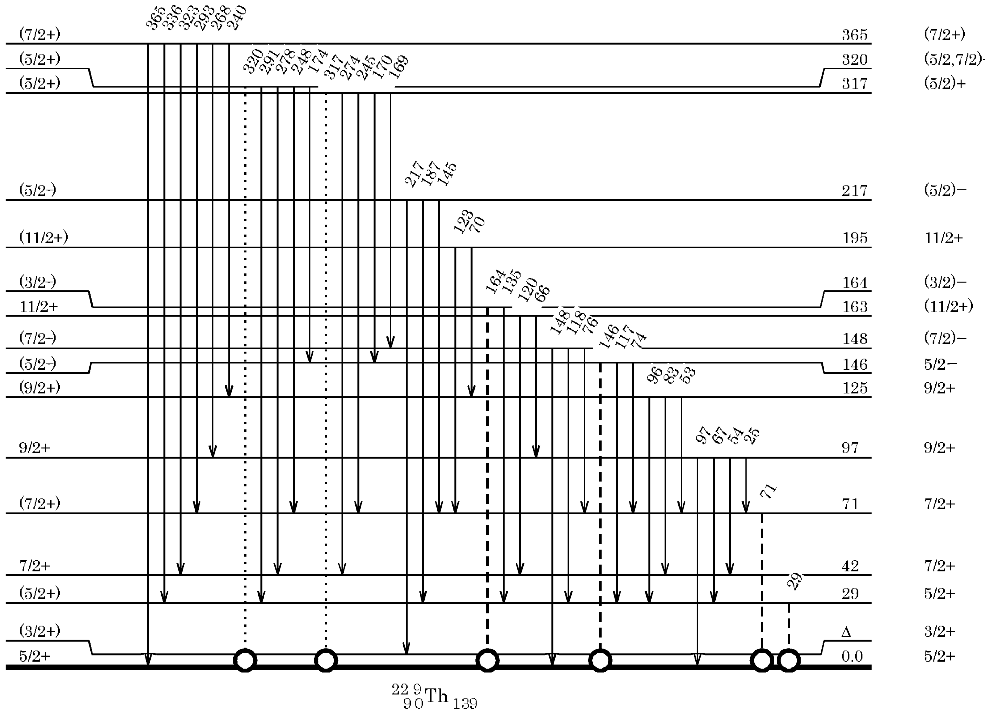

FIG. 1. Level scheme and γ-ray transitions of ^{229}Th. (Continuous arrows) Undisputed γ-ray transitions; (dashed and dotted lines) not well-established γ-ray transitions; (spins and parity) left side, Ref. [10]; right side, Ref. [11].

328-keV transition were not considered because of the very large inconsistency of the published results.) Some definitions necessary for the understanding of the method of analysis are given below.

The experimental data correspond to the differences between γ-ray energies, D_{ik}, and are related to the γ-ray energies by the following:

$$D_{ik} = E_i^0 - E_k^0 + \varepsilon_{D_{ik}}, \tag{1}$$

where E_i^0 and E_k^0 are the true (and unknown) values of the γ-ray energies, that is, the parameters to be fitted, and $\varepsilon_{D_{ik}}$ is the unknown error of D_{ik}. The error is related to the experimental uncertainty $\sigma_{D_{ik}}$ by the relation $\langle \varepsilon_{D_{ik}}^2 \rangle = \sigma_{D_{ik}}^2$, where the bracket stays for expectation value. When using Eq. (1) to analyze the data from Refs. [3] and [13], at least one of the γ rays comes from the ^{229}Th; the other γ ray is either a standard γ-ray transition or a ^{229}Th γ ray.

Standard calibration γ-ray energies, R_j, taken from Ref. [12], are related to their true values E_j^0 by the following:

$$R_j = E_j^0 + \varepsilon_{R_j}, \tag{2}$$

with $\langle \varepsilon_{R_j}^2 \rangle = \sigma_{R_j}^2$. In this procedure, E_j^0 is also considered a parameter to be fitted, whereas R_j is its experimental value.

The same standard γ-ray energy usually appears in more than one relation D_{ik}. For instance, the 71-, 74-, 76-, and 89-keV transitions in ^{229}Th were measured as energy differences with respect to the 84-keV transition from ^{182}Ta, which results in four relations of the type shown in Eq. (1) involving the 84-keV transition. A possibility widely employed until now is to use the standard value of the 84-keV energy from Ref. [12] to calculate the experimental values of the four γ-ray transitions of ^{229}Th. However, as already stated in Ref. [3], this procedure generates covariances between the results and a detailed calculation would be needed. In contrast, because we consider all D_{ik} as experimental results and E_i^0 and E_k^0 as parameters to be fitted, covariances are properly and automatically taken into account [14].

Equations (1) and (2) can be written in a matrix form as follows:

$$\begin{pmatrix} D_{12} \\ \vdots \\ D_{ik} \\ \vdots \\ R_i \\ \vdots \\ R_j \\ \vdots \end{pmatrix} = \begin{pmatrix} 1 & -1 & \cdots & 0 & \cdots & 0 & \cdots & 0 & \cdots \\ \vdots & \vdots & & \vdots & & \vdots & & \vdots & \\ 0 & 0 & \cdots & 1 & \cdots & -1 & \cdots & 0 & \cdots \\ \vdots & \vdots & & \vdots & & \vdots & & \vdots & \\ \vdots & 0 & \cdots & 1 & \cdots & 0 & \cdots & 0 & \cdots \\ \vdots & \vdots & & \vdots & & \vdots & & \vdots & \\ 0 & 0 & \cdots & 0 & \cdots & 0 & \cdots & 1 & \cdots \\ \vdots & \vdots & & \vdots & & \vdots & & \vdots & \end{pmatrix} \cdot \begin{pmatrix} E_1^0 \\ E_2^0 \\ \vdots \\ E_i^0 \\ \vdots \\ E_k^0 \\ \vdots \\ E_j^0 \\ \vdots \end{pmatrix} + \begin{pmatrix} \varepsilon_{D_{12}} \\ \vdots \\ \varepsilon_{D_{ik}} \\ \vdots \\ \varepsilon_{R_i} \\ \vdots \\ \varepsilon_{R_j} \\ \vdots \end{pmatrix}. \tag{3}$$

The covariance matrix of the experimental data was assumed to be diagonal, $V_{ll} = \sigma_l^2$.

The cascade/crossover relations were included in the fitting procedure as constraints and given by the following:

$$\frac{\left(E_i^0\right)^2}{2Mc^2} - N_a^0 + N_b^0 + E_i^0 = 0, \tag{4}$$

TABLE I. Different hypotheses about some γ-ray transitions in ^{229}Th.

γ-ray energy (keV)	Initial level (keV)	Final level (keV)	
		Ref. [10]	Ref. [11]
29	29	$\frac{3}{2}^+$ 75%; G.S. 25%	$\frac{3}{2}^+$
71	71	$\frac{3}{2}^+$ 60%; G.S. 40%	$\frac{3}{2}^+$
146	146	$\frac{3}{2}^+$	G.S.
164	164	$\frac{3}{2}^+$	$\frac{3}{2}^+$ 88%; G.S. 12%

G.S., ground state.

where the γ-ray energy E_i^0 linking levels a and b and the level energies N_a^0 and N_b^0 of ^{229}Th are the parameters to be fitted and M is the nuclear mass.

RESULTS AND DISCUSSION

We considered different assumptions regarding the decay scheme of γ rays linking excited levels of ^{229}Th in the fitting procedure. In Ref. [10] it was considered that the 146- and 164-keV transitions feed just the $\frac{3}{2}^+$ level and both the 29- and 71-keV transitions feed partially the $\frac{3}{2}^+$ level (with relative intensities 75 and 60%, respectively) and the ground state. Reference [11] indicates that the 29- and 71-keV transitions feed only the $\frac{3}{2}^+$ level, the 146-keV transition feeds the ground state, and the 164-keV transition feeds both the ground state (12% intensity) and the $\frac{3}{2}^+$ level. These different hypotheses are summarized in Table I.

To allow for a discrimination of the consistency of those hypotheses with the experimental evidences, the fit was done in two steps. In a first step the level and γ-ray energies of ^{229}Th were fitted adopting the nondiscrepant part of the results of Refs. [10] and [11]. In this step a total of 178 input data (150 energy differences and 28 standard γ-ray energies) were used to fit 96 parameters (53 γ-ray transitions from ^{229}Th, 28 γ-ray standard energies, and the energies of 15 levels of ^{229}Th) with 34 constraints relations between γ-ray and level energies of ^{299}Th. The number of degrees of freedom is, thus, $178 - 96 + 34 = 116$.

In many cases, the same ^{229}Th γ-ray transition was determined in the original works [3,13] from comparisons with several standard γ-ray energies, being the final values presented in those studies calculated as a mean of the results obtained. To consider unidentified uncertainties, when averaging over the obtained results, Helmer and Reich [3] increased the uncertainties every time the reduced χ^2 value was deemed excessively high. Furthermore, to estimate the final recommended uncertainty of Δ, the result was multiplied by a factor 2 [3]. In this work the option was to multiply all data uncertainties from Refs. [3] and [13] by the same factor to obtain a reduced χ^2 value equal to one in the first step of the analysis. The resulting multiplicative factor was 2.8. The procedures used both in Ref. [3] and in this work are intended to correct for underestimated uncertainties. However, the procedure adopted in this work enable the use of the χ^2 test in the second step of the fit. If the fit were done in a single step, the final results would be the same, but a χ^2 test of the hypotheses would be impossible.

TABLE II. Level and γ-ray energies of ^{229}Th obtained in the first step (see text for details).

Transition (KeV)	Level energy (eV)	γ-ray energy (eV)	Energy difference (corrected for recoil) (eV)
29	29 188.1 (9)	29 185.7 (8)	2.4 (12)
71	71 817.1 (9)	71 813.9 (6)	3.2 (12)
146	146 354.0 (9)	146 346.9 (11)	7.1 (13)
164	164 527.7 (13)	164 522.9 (12)	4.7 (18)
217	217 154.2 (12)	217 151.1 (17)	3.0 (21)
320	320 539.9 (21)	320 543.1 (18)	−3.6 (23)

The excitation energy values for the 29-, 71-, 146-, 164-, and 217-keV levels and the corresponding γ-ray energies obtained in the first step are shown in Table II. Table II also presents the differences between level and γ-ray energy.

In this step we fitted also the energy of the 320-keV and the corresponding γ-ray transition, without constraints to study whether the transition feeds the ground state or the isomeric state. Because the results obtained give a difference between the level and the γ-ray energies, corrected for recoil, of $-3.6(23)$ eV (see Table II), it is suggested that the 320-keV level decays predominantly to the ground state.

In a second step we fitted the excitation energy of the $\frac{3}{2}^+$ level using the γ-ray and level energy results obtained in the first step. In this step we considered separately the different assumptions from Refs. [10] and [11]. In both cases the number of degrees of freedom of the fit is 4.

Considering the assumptions from Ref. [10] we obtained $\Delta = 5.5(10)$ eV with $\chi^2 = 6.3$, corresponding to a 18% confidence level, whereas taking the assumptions of Ref. [11] we obtained $\Delta = 1.7(10)$ eV with $\chi^2 = 35$, corresponding to a negligible confidence level $[P(\chi^2) \ll 1\%]$. This high χ^2 value is mainly due to the fact that Ref. [11] assumes that the 146-keV level decays exclusively to the ground state. It is to be noted that this hypothesis is highly inconsistent with the experimental results obtained in the first step, because the energy difference between the 146-keV level and the 146-keV γ ray (corrected for recoil) is 7.1(13) eV (see Table II).

Helmer and Reich [3], when obtaining the value $\Delta = 3.5(10)$ eV, assumed that the 29- and 71-keV levels decay only to the $\frac{3}{2}^+$ level of ^{229}Th, although stating that a partial feeding to the ground state could be possible. To study the validity of the Helmer and Reich hypothesis, we repeated the second step, employing the results of the first step allowing, in addition to Δ, the relative intensities of the decay of those levels to the $\frac{3}{2}^+$ with respect to the ground state, $\phi_{E,\Delta}$, to be parameters of the fit. The results obtained are $\phi_{29,\Delta} = 49(15)\%$ and $\phi_{71,\Delta} = 59(16)\%$, respectively, for the 29- and 71-keV levels to decay to the isomeric level, with a corresponding

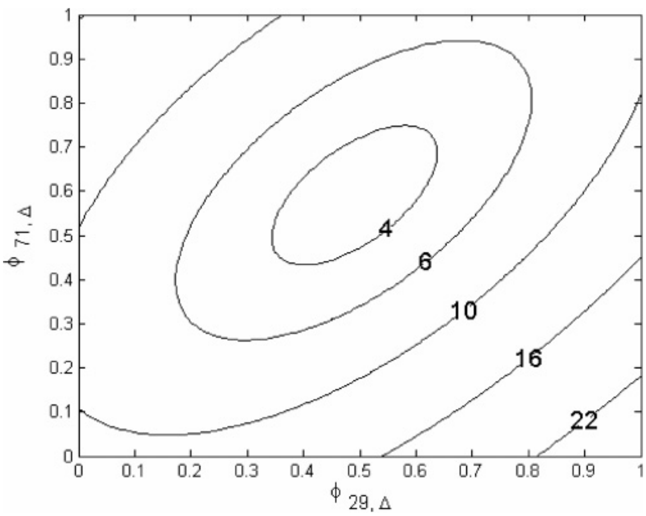

FIG. 2. Level curves of constant χ^2 value as function of the decay intensities of the 29- and 71-keV levels to the $\frac{3}{2}^+$ state (see text for details).

value of $\Delta = 5.8(10)$ eV. The contour lines for constant χ^2 values as function of $\phi_{29,\Delta}$ and $\phi_{71,\Delta}$ are shown in Fig. 2. The minimum value obtained for χ^2 was 3.4. Because three parameters (Δ and two relative γ-ray intensities) were fitted to the experimental data obtained in the first step, the number of degrees of freedom is 2 and the obtained χ^2 value corresponds to a 18% confidence level. Note that the fitted intensities agree well with the results of Ref. [10], as does the Δ value.

However, the assumption that the 29- and the 71-keV transitions feed just the $\frac{3}{2}^+$ level, adopted by Ref. [3], cannot be totally discarded, because when adopted for the second step of the fitting, a χ^2 value of 9.5 results, corresponding to $P(\chi^2) \approx 5\%$. In this case one obtains $\Delta = 4.7(9)$ eV. Note that, although statistically compatible with the Δ value adopted by Helmer and Reich, 3.5(10) eV, the value obtained in the present fit results somewhat higher, as a consequence of the consistent fitting procedure employed for all the data throughout steps one and two.

To stress the importance of a consistent analysis, it is to be observed that if the covariances obtained in the first step were neglected in the second step, the resulting values of Δ would be 5.1(7) and 3.2(7) eV when employing the assumptions of Refs. [10] and [11], respectively. Comparing these values with the results of the present work, 5.5(10) and 1.7(10) eV, it is seen that the covariances are responsible for a difference in the fitted values of, respectively, 0.4 and 1.5 eV, not negligible when compared to the experimental uncertainties. Also, omitting covariances, the uncertainties would be underestimated by about 30%.

As a final remark, it must be observed that any new theoretical interpretation affecting the decay pattern of ^{229}Th could change the adopted value for Δ. Conversely, if a still better precision became available for γ-ray energy measurements, the determination of the branching ratio to the $\frac{3}{2}^+$ isomeric state with respect to the ground state in the decay of each of the levels of interest would be improved and also a lower uncertainty for Δ would be obtained.

So, more theoretical and experimental studies are needed if narrower confidence intervals for Δ are to be obtained. For now, any studies of the interaction of atomic and nuclear degrees of freedom, optical experiments, and the $\frac{3}{2}^+$ level half-life must take into account the relatively broad range of possible values for Δ.

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