Nuclear Instruments and Methods in Physics Research A ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Prospects for measuring the ^{229}Th isomer energy using a metallic magnetic microcalorimeter

G.A. Kazakov [a,b,*], V. Schauer [b], J. Schwestka [b], S.P. Stellmer [b], J.H. Sterba [b], A. Fleischmann [c], L. Gastaldo [c], A. Pabinger [c], C. Enss [c], T. Schumm [b]

[a] Wolfgang Pauli Institute, Univ. Wien – UZA 4 Nordbergstrasse 15, A, 1090 Vienna, Austria
[b] Vienna Center for Quantum Science and Technology, Atominstitut, TU Wien, Stadionallee 2, 1020 Vienna, Austria
[c] Kirchhoff-Institute for Physics, Heidelberg University, INF 227, 69120 Heidelberg, Germany

ARTICLE INFO

Article history:
Received 14 June 2013
Received in revised form
6 September 2013
Accepted 8 September 2013

Keywords:
Thorium-229
Isomer energy
Gamma spectroscopy
Design of experiment

ABSTRACT

The Thorium-229 isotope features a nuclear isomer state with an extremely low energy. The currently most accepted energy value, 7.8 ± 0.5 eV, was obtained from an indirect measurement using a NASA x-ray microcalorimeter with an instrumental resolution 26 eV. We study, how state-of-the-art magnetic metallic microcalorimeters with an energy resolution down to a few eV can be used to measure the isomer energy. In particular, resolving the 29.18 keV doublet in the γ-spectrum following the α-decay of Uranium-233, corresponding to the decay into the ground and isomer state, allows to measure the isomer transition energy without additional theoretical input parameters, and increase the energy accuracy. We study the possibility of resolving the 29.18 keV line as a doublet and the dependence of the attainable precision of the energy measurement on the signal and background count rates and the instrumental resolution.

1. Introduction

The nuclear level scheme of the Thorium-229 isotope is expected to feature a long-lived isomer state, ^{229m}Th, extremely close to the nuclear ground state. The most recent value for the isomer energy E_{is}, 7.8 ± 0.5 eV, obtained from indirect measurements with a NASA x-ray microcalorimeter[1] [1,2], is within the reach of modern optical laser spectroscopy and could serve as a "nuclear frequency standard" [3]. This standard could reach an uncertainty level of 10^{-19} [4], and provide a new powerful instrument for testing the stability of fundamental constants [5,6]. It has been shown that an ensemble of Thorium nuclei doped into a transparent crystal may demonstrate superradiance with a non-trivial emission dynamics [7], and may be used for building an ultraviolet (UV) laser [8]. Finally, the frequency shifts and broadenings produced by such a crystal lattice environment might be used in studies of material properties, as is commonly done in Mössbauer spectroscopy [3]. The necessary step towards all of these exciting applications is a direct observation and precise determination of the isomer state energy.

The existence of the low-energy state in the ^{229}Th nucleus was first conjectured by Kroger and Reich based on studies of the γ-ray spectrum following the α-decay of Uranium-233 [9]. They concluded that this nucleus has a $J^{\pi} = 3/2^{+}$ isomer level lying within 100 eV above the $J^{\pi} = 5/2^{+}$ ground state level. The development of high quality germanium detectors (resolution from 300 to 900 eV) allowed Helmer and Reich to measure more precise γ-energies in 1989–1993 and to predict the energy of the nuclear transition to be $E_{is} = 3.5 \pm 1.0$ eV, placing it into the range of optical frequencies [10]. The decay pattern and combinations of transitions used by Helmer and Reich are presented in Fig. 1(a).

This unnaturally low value of E_{is} triggered a multitude of investigations, both theoretical and experimental, trying to determine the transition energy precisely, and to specify other properties of the $J^{\pi} = 3/2^{+}$ excited state (such as lifetime and magnetic moment). However, searches for direct photon emission from the low-lying excited state performed in the late 1990s [11,12] have failed to observe a signal [13,14]. In 2005, Guimãraes-Filho and Helene re-analyzed the data of Helmer and Reich, taking into account new information about the nuclear decay pattern [15]. They derived $E_{is} = 5.5 \pm 1.0$ eV.

In 2007, a cryogenic NASA x-ray microcalorimeter with instrumental resolution Δ_{inst} from 26 to 30 eV (FWHM) allowed Beck et al. [1] to perform a new indirect measurement of E_{is}, involving lower

* Corresponding author at: Vienna Center for Quantum Science and Technology, Atominstitut, TU Wien, Stadionallee 2, 1020 Vienna, Austria.
Tel.: +431 1 588 01 141859.
E-mail address: kazakov.george@gmail.com (G.A. Kazakov).

[1] We will refer to these devices as "x-ray" spectrometers, corresponding to their primary field of application. In the measurements described here, they detect both, x-rays and γ-rays.

0168-9002/$ - see front matter © 2013 The Authors. Published by Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nima.2013.09.012

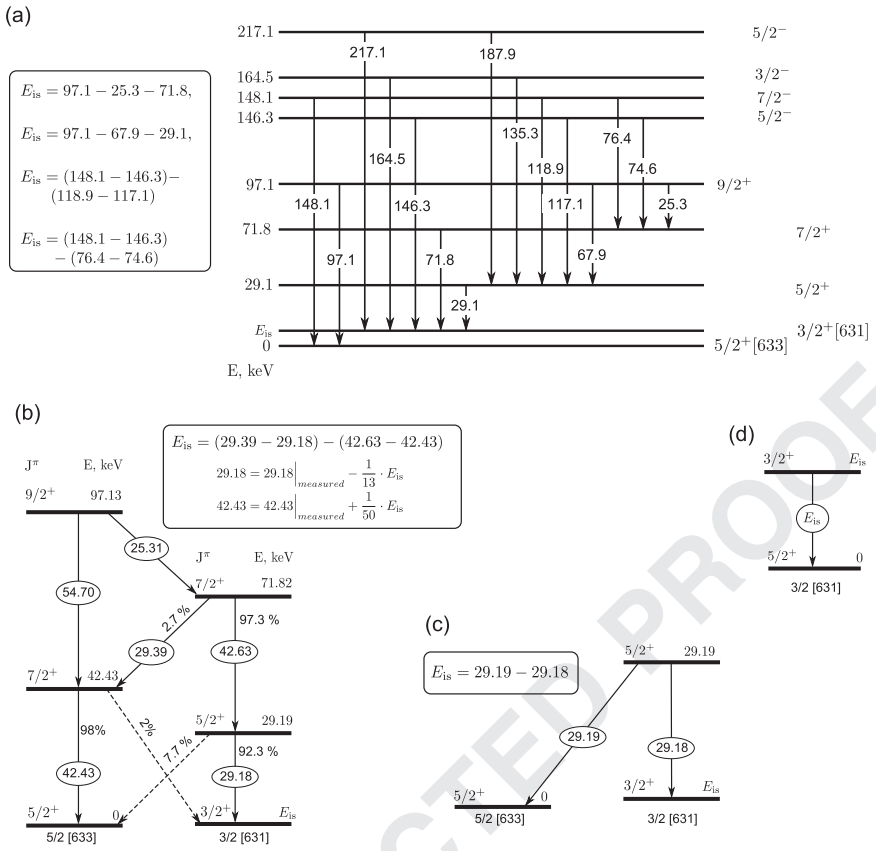

Fig. 1. Partial level schemes of the ^{229}Th nucleus with decay paths and energies (all in keV). Boxes in each panel denote the energy combinations used to derive E_{is} in the "indirect" methods discussed in the main text. (a) According to Helmer and Reich [10]; (b) according to Beck et al. [1,2], the interband transitions (dashed arrows) are taken into account; (c) approach discussed here using a high-resolution ($\Delta_{inst} \simeq 3$–9 eV) microcalorimeter to resolve the 29.1 keV doublet (first proposed in Ref. [31]); (d) direct detection of "nuclear light" (many unsuccessful attempts [11–14] and new proposal [17]). Schemes (a), (b), and (c) are *indirect* measurements, involving keV energy transitions whereas scheme (d) is *direct*, only measuring the isomer transition of a few eV energy.

energy nuclear states, as depicted in Fig. 1(b). In this measurement, the obtained transition energy (7 eV) was corrected by accounting for the theoretical branching ratios 29.19 keV→^{229g}Th estimated as 1/13, and 42.43 keV→^{229m}Th estimated as 2% in Ref. [2]. This correction yields the currently most accepted value $E_{is} = 7.8 \pm 0.5$ eV, now placing the transition into the vacuum UV range (≈ 160 nm).

In the experiments described above [1,9,10] the isomer transition energy E_{is} is not measured directly but is derived from the spectrum of higher-energy (keV) γ-radiation of a spontaneously decaying ^{233}U source. We will refer to these measurements as *indirect passive*. Possible alternatives are *direct passive* and *active* approaches.

In the *direct passive* schemes (Fig. 1(d)), the aim is to perform spectroscopy of the ultraviolet radiation emitted from the isomer appearing in the α-decay of ^{233}U (2% of the nuclei decay is expected to lead into the isomer state). This method has two main difficulties: a relatively high false count rate caused by the Uranium sample radioactivity, and a high probability of non-radiative decay (quenching) of the isomer state in neutral Thorium atoms (up to 10^9 times higher than the radiative decay rate [16]). To overcome these problems, it was proposed in Ref. [17] to extract α-recoil Thorium ions ejected from an Uranium sample, and collect them in a small spot on a MgF_2 coated surface to minimize the quenching rate. Vacuum ultraviolet spectroscopy of the emitted fluorescence radiation may then allow to measure the isomer transition energy.

On the contrary, in *active approaches*, Thorium nuclei (in the ground state) will be illuminated by tunable radiation to excite them to the isomer state. In the *solid-state approach* a macroscopic ($10^{12} - 10^{18}$) number of Thorium ions doped into UV transparent crystals can be excited, for example, by synchrotron radiation, and the emerging fluorescence signal can be studied [18–20]. Apparent advantage of this approach is the huge number of simultaneously excited nuclei. At the same time, crystal fluorescence can cause difficulties in identifying the Thorium isomer transition, and various crystal effects can hamper the precise determination of E_{is}. Another approach is the *spectroscopy of trapped Thorium ions*. At PTB, Germany, work is under way to excite nuclei of Th^+ ions into the isomer state using a two-photon scheme, exploiting the electronic bridge mechanism [21,22]. In Georgia University of Technology, USA, the laser manipulation of Th^{3+} ions is under investigation [4,23]. Detection of the excitation of the Thorium into the isomer state may be based on a change of the electronic hyperfine structure [3]. Studies of the hyperfine structure of Thorium are also performed at the IGISOL facility in Jyväskylä, Finland, in collaboration with a group of the University of Mainz, Germany [24].

We should also mention a number of studies aimed to measure the lifetime of the isomer state without a determination of E_{is}. In Ref. [25], the half life of the isomer state for a *bare nucleus* was derived theoretically based on the calculations of the matrix element of the nuclear magnetic moment and on the experimental data concerning transitions at higher energies. They predict a half-life of $T_{1/2} = (10.95\ \text{h})/(0.025E^3)$ for the isomer transition, where E is given in eV, which yields $T_{1/2} = 55$ min for $E = 7.8$ eV. Direct measurements of this lifetime were performed in several groups [26–28]. The obtained results vary from 2 min [26] to 6 h [28]. This discrepancy may be explained either by an incorrect interpretation of the observations [29] or by a difference in chemical composition of the Thorium resulting in different internal conversion rates.

All active approaches and eventually all nuclear spectroscopy applications require irradiation of the sample with some external narrow-band tunable radiation, and study of the emerging