NUCLEAR TRANSITIONS AND NEW STANDARDS OF LENGTH AND TIME*

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Modern standards of length and time are based on wavelength measurements of laser light sources, whose relative stability is limited by the temperature dependence of molecular or atomic transition frequencies and cannot be better than 10^{-15}. The cardinal way of improvement is to find nuclear transitions coincident with any laser line. This short review is devoted to the present status of investigations and available projects of using the 7.6 eV transition in ^{229}Th. Some new ideas are proposed and discussed.

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1. Introduction

The definition (1983) of the meter is based on the velocity of light $c = 299792458$ m/s and the product $c = \lambda\nu$. The wavelength of any light source, if its frequency is measured, becomes a standard of length with an adequate accuracy $\Delta\nu/\nu$. Presently, typically $\Delta\nu/\nu \approx 10^{-11}\div 10^{-13}$. The limit of $\Delta\nu/\nu \approx 10^{-15}$ is defined by laser stabilization methods based on molecular or atomic transitions, the frequencies of which particularly depend on the temperature. The increase of the accuracy up to $10^{-17} \div 10^{-21}$ is important for fundamental problems (tests of general relativity, revision of fundamental constants [1], astrophysics, measurements of gravitational waves *etc.*) and for applied fields (nanotechnologies, GPS, metrology *etc.* [2]). The cardinal way to improve the laser frequency stability and reproducibility is to find nuclear transitions coincident with any laser line or its harmonic. One of the main advantages of nuclear transitions relative to molecular ones is its independence of temperature and environment. Known nuclei with low-energy isomers are ^{99}Tc ($E^{\rm m} = 2150$ eV), ^{201}Hg (1561 eV), ^{183}W (544 eV),

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^{235}U (73 eV) and, with the lowest energy, ^{229}Th (7.6 ± 0.5 eV). These energies are comparable to those of electronic transitions in the shell and decay mainly via internal conversion (IC).

2. Known ^{229}Th isomer level characteristics

The commonly accepted candidate for a nuclear standard is the $^{229\mathrm{m}}$Th level. For metrology application of nuclear transitions, two characteristics are important — the lifetime of the isomeric level (τ) and the transition energy (ΔE). The lifetime τ defines the transition width, which can be of the order of 1 Hz and even 10^{-4} Hz. For a laser stabilization the nuclear level should be excited resonantly, so the precise knowledge of ΔE is needed, otherwise the laser frequency would scan a too wide region even for broadband lasers (for comparison the ± 0.5 eV correspond to $\Delta\nu \approx 10^{14}$ Hz).

Energy splitting of the ground state doublet in ^{229}Th. A partial level scheme of ^{229}Th is shown in Fig. 1. During 30 years considerable effort has been made to measure the energy splitting of the ground state doublet. Directly, *i.e.* by the observation of photon emission, it was not possible up to now and the available data have been obtained by the analysis of combinations of feeding transition energies. Up to 2007, the generally accepted value was around 3.5 eV (in [3] adopted as 3.5 ± 1.0 eV). Recently, the NASA X-ray spectrometer with an energy resolution of 6 eV [4] has been employed for a new measurement of the transition energies in ^{229}Th (shown in Fig. 1) and resulted in a new generally accepted value of $\Delta E = 7.6 \pm 0.5$ eV [5] (The most recent value is $\Delta E = 7.8 \pm 0.5$ eV [6]. Some discussion about the reliability of the error is presented in Ref. [7]). The new result places this transition energy into the deep UV range (*i.e.* VUV — Vacuum UV), whereas the old data corresponded to the visible range. It partly explains, why up to now nobody has unambiguously detected this transition, since VUV requires special optics, equipments and VUV transparent sources.

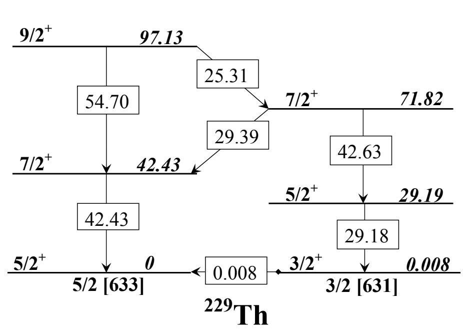

Fig. 1. Partial level scheme of ^{229}Th. The NASA X-ray spectrometer [4] has been employed for measurements of transition energies. Bold italic numbers mark level energies in keV, while transition energies are placed in boxes.

^{229}Th isomer lifetime studies. ^{229}Th isomer half-life calculations are unreliable, mainly due to internal electron conversion (IC) branches of the decay. From the point of view of systematics, a typical value of the reduced transition probability $B(M1)$ for the $3/2^+[631]$–$5/2^+[633]$ transition is about $B(M1) \approx 0.01$ W.u. and the range is $10^{-4} \div 1$ W.u. The corresponding lifetime is $\tau \approx 2$ h ranges from 1 min to 100 hours and the relative line width calculated from $B(M1)$ is $\Delta\nu/\nu \approx 10^{-20}$ and ranges from 10^{-22} to 10^{-18}. IC depends on the outer atomic shell structure and can drastically shift this evaluation to lower lifetimes and correspondingly larger $\Delta\nu/\nu$ values [8]. In these conditions, the measurement of τ is very important. Experimental studies give very different results: [9] 6 hr to 20 d; [10] 13.9 ± 3 hr; [11] ≈ 2.2 hr for 5.5 eV; [12] 1 min to 3 min. In Ref. [12] a hollow-cathode electric discharge was used to populate the isomer in ^{229}Th with the use of the Nuclear Excitation by Electron Transition (NEET) technique. Spectra of alpha-particles with different energies from the ground ($T_{1/2} = 7800$ y) and isomeric states have been studied. The lifetime of the isomeric level has been evaluated from the observed decrease of the decay intensity.

A direct determination of the energy and a reliable half-life measurement are needed before other experiments can be performed.

3. Projects on ^{229}Th studies and the nuclear frequency standard

Several projects on the investigation of the ^{229}Th isomer are supported by Nuclear Science Centers in USA, Germany, Finland, Canada *etc.*

LLNL project: ^{229}Th isomer lifetime studies. Experiments, which are attempting to measure the half-life by searching for IC electrons or photons emitted as a result of IC, have started at the Lawrence Livermore National Laboratory (LLNL) [13]. Using the population of ^{229m}Th via α-decay of ^{233}U, the recoiling ions are collected on a recoil catcher, which is subsequently placed between electron microchannel plate (MCP) and ultraviolet photo multitier (UV PMT) detectors (Fig. 2 (left)).

LMU-MLL project: ^{229}Th Δ E studies. A project of the Ludwig-Maximilians University (München) and Maier-Leibnitz Laboratory (Garching) (LMU-MLL) [14] aims at the direct observation of the VUV ^{229m}Th transition and the measurement of its energy with envisaged accuracy of 0.05 eV. Similar to the LLNL project, ^{229m}Th is populated via the ^{233}U α-decay. The ^{229m}Th recoils are stopped in a buffer gas stopping cell and after the extraction with a radio frequency (RF) plus direct current (DC) funnel (this system may be named isomer generator) is transferred into an RFQ ion guide before collecting the ions on a steel needle tip. The UV fluorescence light from the $^{229\mathrm{m}}$Th decay will be focused onto a microchannel plate (MCP) monitored by a charge coupled device (CCD) camera behind a screen (Fig. 2 (right)).

For laser experiments on resonance fluorescence a resolution ΔE of up to 10^{-4}–10^{-5} eV is required, which is about 3 orders of magnitude smaller than the resolutions of modern detectors. Some projects for resolving this problem and realization of metrology applications have been proposed.

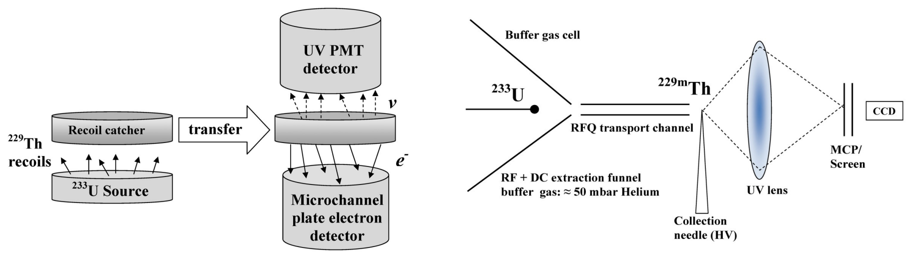

Fig. 2. Projects on the ^{229}Th isomer level study. Left: Schematic diagram of the experiment attempting to measure the half-life by searching for IC electrons and photons emitted as a result of IC [13]. After a population of $^{229\mathrm{m}}$Th via α-decay of ^{233}U, the recoiling ions are collected on a recoil catcher, which will be subsequently placed between MCP electron and UV PMT detectors. Right: Study of ΔE in $^{229\mathrm{m}}$Th nuclei [14]. The recoiling $^{229\mathrm{m}}$Th ions are extracted from the gas cell and collected on a needle tip. The UV fluorescence light from the $^{229\mathrm{m}}$Th decay will be focused onto an MCP monitored by a CCD camera behind a screen.

A nuclear frequency standard with trapped and placed into crystals ^{229}Th ions. A possible laser excitation scheme utilizing the electronic level scheme of $^{229}\mathrm{Th}^{3+}$, allowing to apply a double-resonance method for laser cooling and fluorescence light detection in a linear Paul trap is described in Ref. [15]. More recent publications [16] and [17] are also devoted to solid-state nuclear frequency standards with ^{229}Th ions placed into VUV transparent crystals with a high transition quadrupole field to avoid recoiling of $^{229\mathrm{m}}$Th nuclei. It is assumed that the resonance radiation can be measured, even if the resonant scattering rate is only of the order 10^{-4}/s per nucleus.

Berkeley Labs Advanced Light Source project. The projects presented above have the disadvantage of extremely low rates, which can be too small for a practical laser stabilization. New possibilities are connected with high intensity photon beams from the Berkeley Labs Advanced Light Source (ALS), based on a 1.9 GeV synchrotron (10^{19} ph/sec/mm^2 as compared to 10^{10} from the Sun!). A novel approach to directly measure the energy of the low-lying isomeric state in ^{229}Th has been proposed [18]. $^{229\mathrm{m}}$Th nuclei produced by ALS should be driven into a host crystal and ALS-induced fluorescence has to be studied. To locate the transition, the frequency will be scanned in steps of 0.05 eV. As a result, the wavelength λ can be determined with an accuracy of $\Delta\lambda \approx 0.1$ nm. The experimental effort has a focus on LiCaF crystals which are transparent down to 110 nm (7.6 ± 0.5 eV corresponds to 163 ± 11 nm). A further increase in the accuracy and the creation of a standard based on ^{229}Th is planned. $\Delta\nu/\nu \approx 2 \times 10^{-18}$ is expected.

4. Alternative proposals

Population of the ^{229}Th isomer via Coulomb Excitation (Coulex). The cross-section of a level population via Coulex is $\sigma(E2) \sim B(E2)\, f_{E2}(\Delta E/E)$. For ^{229}Th the expected $B(E2)$ value is ≈ 10 W.u. with $\mathrm{f}_{E2}(\Delta E/E)$ sharply increasing when $\Delta E/E \to 0$. Here ΔE is the level energy (≈ 7.6 eV) and E is the energy transferred to recoils (≈ 10 MeV). So, due to the extremely low ΔE, Coulex using as isomer generator, instead of using the above described ^{233}U α-decay, should allow to populate $^{229\mathrm{m}}$Th with much higher efficiency, even more than ALS, which due to a very narrow line width can produce not more than 10^5 excited $^{229\mathrm{m}}$Th nuclei per sec. Our proposal is illustrated in Fig. 3 (left). High energy ^{229}Th recoils should lose energy in a stopper layer and be caught by a host crystal. Photon irradiation (or photon plus IC) can be investigated off-beam (similar to above described LLNL project) or on-beam in anti-coincidence with beam bunches.

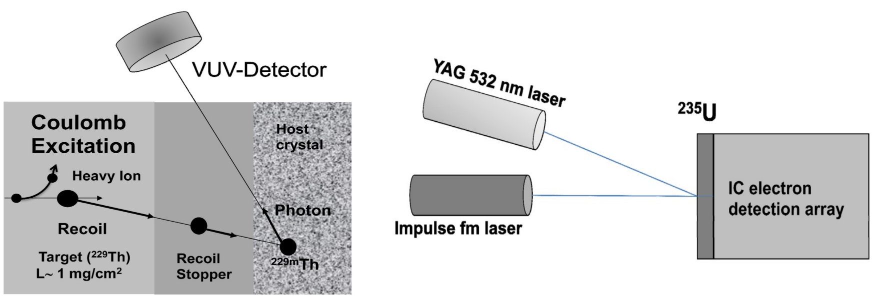

Fig. 3. Alternative proposals. Left: Application of Coulex as generator of the ^{229}Th isomeric state. High energy ^{229}Th recoils are slowed down in a stopper layer and are caught by the host crystal. Right: Experimental approach to study the ground state transition in $^{235\mathrm{m}}$U and its use for optical laser stabilization. A pulsed fs laser is used to produce high harmonics. A second narrow-band YAG 532 nm laser can be used to produce and precisely change new harmonic frequencies and can be stabilized by the detection of IC electrons from the ^{235}U target.

Laser excitation of 73 eV level in ^{235}U. ^{229}Th is very difficult to produce and an inconvenient material; so alternative nuclear transitions in optical and UV ranges have to be found and studied. A second candidate after ^{229}Th is ^{235}U with a 73 eV state. Pulse femtosecond (fs) laser due to nonlinear effects can produce enough high harmonics (≈ 30) for a population of this level. We propose to use a second narrow-band YAG 532 nm laser for the production of additional harmonics between the harmonics of the fs-laser. The second optical laser can be tuned in a ±250 GHz range and would allow to precisely change the harmonic frequency. After measurements of the transition energy its frequency can be stabilized by the detection of IC electrons from ^{235}U target (Fig. 3 (right)).

For the metrological applications, we propose to make efforts to search for other unknown nuclear low-energy transitions. Candidates for such transitions cannot only populate the ground but also isomeric excited nuclear states. It seems that the highest probability to find most such transitions is among heavy odd–odd nuclei.

5. Summary

A short review of available experimental data on the energy splitting of low-energy levels in ^{229}Th and the lifetime of the isomeric level is given.

Available projects on the investigation and use of this transition for the creation of a frequency reference are discussed.

Alternative ways to populate isomeric levels and other candidates to be used as reference are proposed for discussion.

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