Concepts for direct frequency-comb spectroscopy of $^{229m}$Th

Lars von der Wense$^1$ and Chuankun Zhang$^2$

$^1$Ludwig-Maximilians-Universität München, 85748 Garching, Germany.
$^2$JILA and Department of Physics, NIST and University of Colorado, Boulder, CO 80309-0440, USA

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Direct laser spectroscopy of $^{229m}$Th was already discussed back in the 1990s, followed by the proposal for a nuclear optical clock in 2003. However, it was not until very recently that the $^{229m}$Th energy was constrained to sufficient precision in order to determine the laser technology required to drive the transition in the nuclear clock concept. Having this new knowledge at hand, three different concepts for direct frequency-comb spectroscopy of $^{229m}$Th are discussed, making use of the 7th harmonic of an Yb-doped fiber frequency comb, operational at JILA in Boulder, Colorado. The first concept describes the excitation of a large number of surface-bound $^{229}$Th atoms and appears to be promising on a short time-scale. A more advanced scheme of using the same laser to excite the nuclei of multiple laser-cooled $^{229}$Th$^{3+}$ ions in a Paul trap is discussed as a second concept. Finally, the potentials for driving nuclear Rabi oscillations and for the development of an IC-based solid-state nuclear clock are highlighted.

I. INTRODUCTION

The accuracy of time measurements has considerably evolved within the past decades, starting with the development of the first atomic clock by Louis Essen and Jack Parry in 1955 [1], followed by the legal definition of the second by means of the Cesium standard in 1967 [2] and the invention of the fountain clock in the late 1980s [3]. Since the optical frequency comb was invented in 1998 [4] a new technological leap has occurred, allowing for the development of optical atomic clocks, which by today pose the most precise timekeepers [5]. The achieved accuracies of these clocks vary around $10^{-18}$, corresponding to an error of 1 second in 30 billion years, significantly longer than the age of the universe [6-9]. Such accuracy in frequency measurement opens up a plethora of applications, e.g., in satellite-based navigation [10], geodesy [11] and the search for temporal variation of fundamental constants [12, 13].

In recent years, the frequency-comb technology has been drastically extended to higher energies and now also covers wavelengths in the deep to extreme ultra-violet region (see e.g. [14-15]). This is of particular interest as, for the first time, frequency combs exist that cover energy ranges not only of the atomic shell but also of specific extremely low-energy excitations of atomic nuclei [14]. For this reason, this progress opens the door for direct nuclear laser spectroscopy experiments and the development of a nuclear clock that uses an atomic nucleus instead of the atomic shell for time measurement [19]. A nuclear optical clock is expected to outperform even today’s best atomic clocks due to conceptual advantages [20]. In a pictorial understanding, the reason for this expected improved accuracy is that the atomic nucleus is several orders of magnitude smaller than the atomic shell and therefore, due to the related smallness of the nuclear moments, drastically more stable against external influences [21].

Direct laser excitation of a nuclear state is a central requirement for the development of a nuclear optical clock. Currently, there is only one nuclear excited state known, which could serve for a nuclear clock, as it allows for direct nuclear laser spectroscopy due to its extraordinary low excitation energy. This is the first excited state of $^{229}$Th, known as the “thorium isomer”, with an excitation energy that has recently been re-measured to be about 8.3 eV [22]. The state is metastable, with an estimated radiative lifetime of up to $10^4$ seconds [23] and is usually denoted by $^{229m}$Th. The long radiative lifetime leads to a narrow relative linewidth of $\Delta f/f \approx 10^{-20}$ and renders $^{229m}$Th an ideal candidate for a first nuclear clock of unprecedented accuracy [19, 20]. Already the nuclear excited state of second lowest known excitation energy, $^{235}$U, cannot be used for the development of a nuclear clock. Although, with an energy of 76.7 eV [25] it might be in the reach of future frequency combs [14], its radiative lifetime of about $10^7$ s is prohibitively long for efficient direct laser excitation. In the following, the experimental history of $^{229m}$Th will briefly be sketched.

$^{229}$Th was first considered to possess a low-energy nuclear excited state in 1976, when it allowed to explain some peculiarities of the $^{229}$Th nuclear $\gamma$-ray spectrum as observed in the $\alpha$ decay of $^{233}$U, which remained otherwise unexplained [26]. Based on $\gamma$-ray spectroscopy of nuclear rotational states of larger energies, the $^{229m}$Th energy value was constrained to $-1 \pm 4$ eV in 1990 [27] and then further improved to $3.5 \pm 1$ eV in 1994 [28], which was until 2007 the adopted energy value. In 2007, however, a different energy value was measured using a metallic magnetic microcalorimeter with an improved energy resolution leading to $7.6 \pm 0.5$ eV [29], later corrected to $7.8 \pm 0.5$ eV [30].

Although, direct laser spectroscopy of $^{229m}$Th was discussed already in the 1990s [31, 32], followed by the proposal for a nuclear clock in 2003 [19], these goals have not been achieved until today. A central challenge, which has hindered the development of a nuclear clock
within the past decade, was that the $^{229m}$Th energy was not sufficiently well constrained to allow for direct laser spectroscopy of the nucleus. This has led to a multitude of efforts to pin down the isomeric energy value to higher precision (a recent review can be found in Ref. [33]). It would be ideal to measure the energy via spectroscopy of photons directly emitted in the isomer’s ground-state decay. However, despite worldwide efforts, until today no unambiguous signal of photons emitted in the isomeric decay has been observed, potentially pointing toward a significant non-radiative decay channel [34–37].

In 2016, the direct observation of electrons emitted in the internal conversion (IC) decay channel of the isomeric state opened a new path for exploration of the isomer’s properties [38]. In the internal conversion decay, the nucleus couples to the atomic shell and transfers its energy to an electron, which is subsequently ejected. Importantly, in the IC decay channel the nucleus couples directly to the atomic shell and the process does not correspond to a $\gamma$ decay followed by photo-electron emission. For this reason the lifetime of the nuclear excited state shortens by the amount of the conversion emission. For this reason the lifetime of the nuclear $\gamma$ decay followed by photo-electron emission. For this reason the lifetime of the nuclear excited state shortens by the amount of the conversion emission. For this reason the lifetime of the nuclear excited state shortens by the amount of the conversion emission. For this reason the lifetime of the nuclear excited state shortens by the amount of the conversion emission.

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Most importantly, in 2019, the $^{229m}$Th energy was measured to higher precision via direct spectroscopy of the IC electrons emitted in the isomeric decay, obtaining a value of $8.28 \pm 0.17$ eV (corresponding to $149.7 \pm 3.2$ nm) [22]. This new energy value allows for the first time to determine the laser technology required for driving the transition in the nuclear clock concept. The isomeric energy is matching with already existing frequency-comb technology: the 7th harmonic of an Yb-doped fiber laser [14, 15], generated via high-harmonic generation (HHG) in a noble gas could provide an ideal tool for driving the transition. The 5th harmonic of a Ti:Sapphire laser, could be an alternative [18].

Until recently, there was the hope that the $^{229m}$Th nuclear excitation energy would be sufficiently low to allow for spectroscopy via continuous-wave (cw) laser technology. This would have been advantageous, as such systems are typically more robust, compact and easy to operate than laser systems based on HHG. Using KB$_2$BO$_3$F$_2$ (KBBF) as a non-linear crystal for frequency doubling, the shortest wavelength that is currently obtainable with this technology is, however, around 165 nm [44], which is not sufficiently short in order to drive the nuclear transition. For this reason, there is currently no alternative to the operation of a nuclear clock based on direct frequency-comb spectroscopy using a vacuum ultra-violet (VUV) frequency comb generated via HHG.

Here it is proposed to use an already existing Yb-doped fiber laser system [14, 17], operational at the JILA laboratory in Boulder, Colorado, for first time frequency-comb-based direct laser excitation of $^{229m}$Th. The laser system operational at JILA is the most powerful laser of this technology in the world [17] and therefore ideally suited for this purpose. Three different concepts to use this laser for nuclear spectroscopy are proposed: In the first approach the irradiation of more than $10^{14}$ $^{229}$Th atoms on a surface is discussed, which can be considered as the simplest method and would allow for a high-precision $^{229m}$Th energy determination and the development of an IC-based solid-state nuclear clock. In the second concept, nuclear frequency-comb spectroscopy of multiple, laser-cooled $^{229}$Th ions in a Paul trap is considered. For the third method, it is assumed that the bandwidth of an individual comb mode is narrowed down to the Hz-range as would be ideal for clock operation. In this case it is found that the laser intensity would be sufficient to drive nuclear Rabi oscillations.

II. THEORETICAL BACKGROUND

In the following, the nuclear excitation probability as a function of time under resonant laser irradiation will be modeled. The system of nuclear ground- and excited state is approximated as a pure nuclear two-level system without taking magnetic sub-states into consideration. Under this simplification, the time-dependent nuclear excitation probability $\rho_{exc}(t)$ for a single nucleus under resonant irradiation is given by Torrey’s solution of the optical Bloch equations ($\rho_{exc} = 1/2 (1 - w)$ and $w$ taken from Eq. (16) of Ref. [45] was used):

$$\rho_{exc}(t) = \frac{\Omega^2}{2 (\Gamma \bar{\Gamma} + \Omega^2)} \left[ 1 - e^{-\frac{1}{2}(\Gamma + \bar{\Gamma}) t} \left( \cos(\lambda t) + \frac{\Gamma + \bar{\Gamma}}{2\lambda} \sin(\lambda t) \right) \right].$$

(1)
Here \( t \) denotes the time since the start of the irradiation, \( \Omega \) is the Rabi frequency, \( \Gamma = (1 + \alpha_{\text{IC}}) \Gamma_\gamma \) is the total nuclear decay rate including \( \gamma \) decay and internal conversion (IC), with \( \alpha_{\text{IC}} \) representing the IC coefficient (the ratio of IC compared to \( \gamma \) decay) and \( \Gamma_\gamma \), the radiative decay rate. \( \tilde{\Gamma} = (\Gamma + \Gamma_L)/2 \) denotes the total decay rate of the coherences, with \( \Gamma_L \) the bandwidth of the laser light used for excitation. The parameter \( \lambda \) is defined as \( \lambda = |\Omega^2 - (\tilde{\Gamma} - \Gamma_\gamma^2)/4|^{1/2} \). For \( \Omega < |\tilde{\Gamma} - \Gamma_\gamma^2|/2 \), the sin and cos functions have to be exchanged by sinh and cosh, respectively. The Rabi frequency of the system can be calculated as

\[
\Omega = \sqrt{\frac{2\pi c^2 I \Gamma_\gamma}{h\omega_0}},
\]

with \( I \) the intensity of the laser light, \( \omega_0 \) the angular frequency of the nuclear transition, \( \Gamma_\gamma \) the radiative decay rate of the nuclear transition, \( c \) the speed of light and \( h \) the reduced Planck constant.

Note, that Eq. (1) remains valid also if the bandwidth of the laser light used for irradiation, \( \Gamma_L \), is significantly broader than the total linewidth \( \Gamma \) of the nuclear transition, as the definition of the decay rate of the coherences \( \tilde{\Gamma} \) takes respect for that. The number of excited nuclei \( N_{\text{exc}} \) is obtained from Eq. (1) by multiplication with the number of irradiated nuclei \( N_0 \). Throughout the paper Eq. (1) will be used to model the time-dependent nuclear excitation probability.

### III. CONCEPT 1: SURFACE EXCITATION

In this concept the 7th harmonic of the Yb-doped fiber frequency comb, operational at JILA [14, 17], is used to irradiate a solid sample of \( ^{229}\text{Th} \) atoms deposited as a thin layer on a surface. The presented concept is similar, but not identical to a proposal presented in Refs. [40, 47], where it was shown that broadband VUV laser light, generated via four-wave mixing in a noble gas, could be used for \( ^{229m}\text{Th} \) laser excitation. Here, the concept is adapted to a source of narrowband laser light as offered by the VUV frequency comb. The advantages of the presented scheme compared to the scheme proposed in Refs. [40, 47] are that the isomeric energy could be constrained to higher precision and that the same laser system could also be used as a driving laser in the nuclear-clock concept.

For the purpose of laser excitation of the \( ^{229}\text{Th} \) nuclei, it is assumed that the laser light is gated, with gate pulses of 100 \( \mu \)s duration and a gate frequency of 5 kHz and a wavelength tunable between 143 and 157 nm, are generated from the existing JILA frequency-comb system [14, 17] either with the help of a mechanical shutter [48] or with an acousto-optical modulator. In case that an individual mode of the frequency comb is tuned to the nuclear resonance energy, a fraction of the nuclei is excited into the isomeric state during a single gate pulse. After the end of the laser gate pulse the excited nuclei will decay with about 10 \( \mu \)s lifetime via internal conversion (IC) [38, 39]. The low-energy electrons emitted in the IC decay will be guided by magnetic fields and detected with the help of a micro-channel plate (MCP) detector [49] in order to probe the nuclear excitation. In this way it can be inferred if the laser was tuned to the nuclear resonance. A schematic of the experimental setup is shown in Fig. 1. The advantages, compared to other potential nuclear laser excitation schemes, which make use of charged \( ^{229}\text{Th} \) ions in a Paul trap (discussed in Sects. IV and V), are: a huge number of irradiated atoms of up to \( 10^{14} \), an IC-broadened nuclear transition linewidth of 15.9 kHz and a 9 orders of magnitude shortened lifetime of about 10 \( \mu \)s, which allows for laser-triggered isomeric decay detection.

![FIG. 1. Schematic of the experimental setup proposed for frequency-comb-based direct nuclear laser excitation of \( ^{229m}\text{Th} \). A vacuum ultra-violet (VUV) frequency comb is generated via cavity-enhanced high-harmonic generation (HHG) from an Yb-doped-fiber frequency comb that has been amplified with three amplification stages [14, 17]. Gate pulses of 100 \( \mu \)s duration with 5 kHz gate frequency are produced with the help of a mechanical shutter [48]. A solid sample surface of \( ^{229}\text{Th} \) is irradiated with the laser light. In case of resonance, the nuclei isomeric state is excited and will decay within about 10 \( \mu \)s lifetime via internal conversion (IC) with emission of an electron [39]. The electrons can be guided with the help of magnetic fields and detected by a micro-channel plate (MCP) detector [46, 47].](image-url)
The concept requires three ingredients as visualized in Fig. 1: (1) A target consisting of a thin surface of $^{229}\text{Th}$ (2) A gated laser system used for target irradiation and (3) a detection system that will allow for the detection of the low-energy IC electrons with high efficiency. Each part will be discussed individually in the following.

The $^{229}\text{Th}$ target consists of a thin ($\sim 10$ nm thick) $^{229}\text{Th}$ layer deposited as a round surface with 1 mm diameter onto a gold substrate. The useful thickness is limited by the mean free path length of electrons in the target material [50]. With a density of 11.57 g/cm$^3$, the number of $^{229}\text{Th}$ atoms in the target amounts to $2.4 \cdot 10^{14}$. As the $^{229}\text{Th}$ lifetime is about 7900 years, this corresponds to an activity of $\sim 660$ Bq. Due to the comparatively short lifetimes of the daughter isotopes, the total activity of the target will soon grow by a factor of about 10, leading to a total activity of estimated 6.6 kBq. Targets of this type have already been produced at the Institute for Radiochemistry of the University of Mainz via a newly developed drop-on-demand technique [51].

The laser system is an ytterbium-doped fiber-based frequency comb operating at a wavelength around 1070 nm. The system is already operational at JILA in Boulder, Colorado [14, 17]. With the help of three amplification stages and an enhancement cavity, a high power of 10 kW is produced. Focusing this laser power to a helium-xenon gas-jet leads to the generation of high harmonics (HHG). The seventh harmonic spans an energy range of 0.038 eV (9.2 THz) and tunability between 7.9 and 8.7 eV (143 to 157 nm) is planned to be provided. It consists of about $1.2 \cdot 10^5$ comb modes, each of them with a bandwidth of about 490 Hz and a time-averaged power of about 100 nW. The mode spacing is 77 MHz. In the considered concept the laser beam is gated either with the help of a fast rotating shutter system [48] or with an acousto-optical modulator to provide laser gate pulses of 100 $\mu$s pulse duration at 5 kHz gate frequency. Important parameters of the concept are visualized in Fig. 2.

The values of variables used for the quantitative analysis according to Eq. (1) are listed in Tab. 1. The resulting number of nuclear excitations as a function of time is shown in Fig. 3. The estimated number of excited nuclei after 100 $\mu$s irradiation time amounts to $N_{\text{exc}} = 782$, which is close to the maximum number as obtained in saturation of 792 excited nuclei.

When using this laser system for surface irradiation, the potential for ablation requires consideration. The frequency comb has a repetition rate of 77 MHz and a fourier-transform limited pulse duration of about 50 fs of the 7th harmonic. Considering that all $1.2 \cdot 10^5$ comb modes will irradiate the target in parallel with a well defined phase relation to create a pulse train, the peak intensity of light amounts to $4.1 \cdot 10^8$ W/cm$^2$, which is still by eight orders of magnitude below the typical laser ablation threshold for femtosecond laser pulses [52]. For this reason no significant laser ablation of the target is expected to occur.

![FIG. 2. Sketch of the proposed experimental concept together with important parameters. a) A VUV frequency comb with $1.2 \cdot 10^5$ comb modes of 490 Hz mode-width (100 nW power per mode) and 77 MHz mode spacing, tunable between 7.9 and 8.7 eV, is used to search for the nuclear resonance. b) Laser excitation is achieved during gate pulses of 100 $\mu$s duration (5 kHz gate frequency). The isomeric decay is observed shortly after the end of each laser pulse. A 5 $\mu$s detection window is assumed for a signal-to-background estimate.](image-url)
TABLE I. Values of variables used for the calculation of the number of excited nuclei per laser pulse based on Eq. (1).

| Variable | Description | Value | Comment |
|----------|-------------|-------|---------|
| \(I\)    | Laser intensity | \(1.3 \cdot 10^{-5}\) W/cm\(^2\) | Single-mode (100 nW) focused to \(\varnothing\) 1 mm |
| \(\Gamma_L\) | Laser bandwidth | \(2\pi \cdot 400\) Hz | Smaller than IC-broadened nuclear linewidth |
| \(\omega_0\) | Angular frequency | \(2\pi \cdot 2.0\) PHz | Corresponding to 8.3 eV energy [22] |
| \(\Gamma_\gamma\) | Radiative decay rate | \(10^{-4}\) Hz | Estimated from theory [23] |
| \(\alpha_{ic}\) | IC coefficient | \(10^9\) | Based on \(\Gamma_\gamma\) and the IC lifetime [39] |
| \(N_0\) | Irradiated atoms | \(2.4 \cdot 10^{14}\) | For 10 nm layer thickness and 1 mm diameter |

![Figure 3](image_url)

FIG. 3. Expected number of excited nuclei as a function of time if \(2.4 \cdot 10^{14}\) \(^{229}\)Th atoms on a surface are irradiated in parallel.

The detection system consists of a micro-channel plate (MCP) detector [49] providing a high detection efficiency of up to 50% for electrons when post-accelerated to about 300 eV [53]. The detector is placed inside a solenoid coil, which produces guiding fields in order to guide only electrons of below \(\sim 1\) keV kinetic energy to the MCP detector. In this way the detector can be placed in a relatively large distance (50 cm or more) to the \(^{229}\)Th target and only a minor amount of high-energy \(\alpha\) and \(\beta\) particles, as produced in the radioactive decays, will reach the MCP detector, as these particles are not guided by the magnetic fields. Time-dependent electric fields of a few volts can be applied in order to repel photoelectrons generated during the gate pulses and in order to accelerate the electrons emitted in the internal conversion process towards the MCP detector [47]. A similar detection technique was used for the energy determination of the isomeric state via IC electron spectroscopy [22] [54].

Based on the \(^{229}\)Th target- and laser parameters stated above, a direct calculation reveals that about 780 nuclei can be expected to be excited into the isomeric state during each laser gate pulse. These excited nuclei will decay into the nuclear ground state with about 10 \(\mu\)s lifetime via the emission of low-energy internal conversion electrons [39]. Given the gate frequency of 5 kHz, up to 3.9 million nuclear excitations are expected to occur per second in case of resonance. It was successfully shown that time gating is sufficient for the exclusion of photoelectrons as a background, if a time window from 5 to 10 \(\mu\)s after the end of each laser pulse is chosen for IC electron detection [47]. After \(\sim 5\) \(\mu\)s, the background will instead be dominated by the radioactive decays, which are intrinsic to the \(^{229}\)Th target [46]. Assuming a total target activity of 6.6 kBq, about \(1.3 \cdot 10^3\) low-energy electrons are expected to be emitted per second due to shell-reorganization after \(\alpha\) decay [55]. On average, this corresponds to 0.07 emitted electrons in the 5 \(\mu\)s detection window. The number of expected isomeric decays in the same time window amounts to \(0.24 \cdot N_{exc} \approx 190\). Even when assuming a factor of 10 of electron losses in the target, the estimated signal-to-background ratio is 270 in case of resonance.

During the search for the isomeric excitation, the frequency comb has to be tuned. As the scan is performed with all \(1.2 \cdot 10^5\) comb modes in parallel, it is sufficient to bridge the mode spacing of 77 MHz of two consecutive comb modes in order to probe the total bandwidth of the frequency comb of 0.038 eV (9.2 THz). Considering the IC-broadened nuclear transition linewidth of 15.9 kHz, about 5000 scan steps will therefore be required. Assuming that 100 gate pulses per scan step are used, the total time required for scanning of the 0.038 eV energy range amounts to 100 seconds. In order to cover the 0.34 eV energy range corresponding to the 1 \(\sigma\) uncertainty interval of the currently best energy constraint of 8.28 \(\pm\) 0.17 eV [22], 9 of such scans would have to be performed leading to a minimum total scanning time of 900 s. The main parameters of the proposed method are shown in Tab. I[3].

IV. CONCEPT 2: MULTIPLE IONS IN A PAUL TRAP

For the development of a trap-based nuclear optical clock, direct nuclear laser excitation of individual \(^{229}\)Th\(^{3+}\) ions in a Paul trap is required. In this section it is assumed that the laser system already presented in Sect. II[3] is used to irradiate 10 laser-cooled \(^{229}\)Th\(^{3+}\) ions in a Paul trap. Laser cooling could be achieved either directly [50] or via...
sympathetic cooling [57]. Further, it is assumed that the laser light is focused to a spot of 10 μm diameter in order to irradiate the chain of ions. In this concept, the successful laser excitation is probed via exploiting the change of the hyperfine structure of the electronic shell, induced by the different spins of nuclear ground- and excited state, which is known as the double-resonance method [19].

In 229Th ions the internal conversion decay channel is energetically suppressed and α_{ic} = 0 holds. For this reason the isomeric lifetime is by nine orders of magnitude prolonged and may approach the radiative lifetime \( \tau = 1/\Gamma_\gamma \) of up to \( 10^4 \) s. The parameters used to calculate the nuclear excitation probability via Eq. (1) are listed in Tab. III.

TABLE III. List of input values used in Eq. (1) for calculation of concept 2.

| Variable     | Value | Comment                        |
|--------------|-------|--------------------------------|
| \( I \)      | 0.13 W/cm² | Single-mode focused to \( \varnothing 10 \) μm |
| \( \Gamma_L \) | 2 \( \pi \cdot 490 \) Hz | Larger than nuclear linewidth |
| \( \omega_0 \) | 2 \( \pi \cdot 2.0 \) PHz | Corresponding to 8.3 eV energy |
| \( \Gamma_\gamma \) | \( 10^{-4} \) Hz | Estimated from theory |
| \( \alpha_{ic} \) | 0 | IC suppressed for Th^{3+} ions |
| \( N_0 \) | 10 | Multiple laser-cooled ions |

Under these considerations the time required to reach saturation is about 20 s as shown in Fig. 4. The excitation probability amounts to 50%, resulting in 5 excited nuclei in case of resonance. In the search for the nuclear excitation the mode spacing of 77 MHz has to be bridged. With a mode bandwidth of 490 Hz, this leads to a 1.57 \( \cdot 10^5 \) scan steps. Assuming that each scan step takes a time of 20 s, the time required to bridge the mode spacing amounts to 3.14 \( \cdot 10^6 \) s or 36.4 days. As all 1.2 \( \cdot 10^5 \) comb modes are used in parallel, the energy range scanned in this way equals 9.2 THz or 0.038 eV. With a 1 \( \sigma \) uncertainty range of ±0.17 eV, at least 0.34 eV would have to be covered, which would require nine shifts of the center wavelength of the frequency comb, leading to a total net scanning time of 328 days. For this reason it appears favorable to constrain the energy via concept 1 or the scheme presented in Refs. [46, 47] prior to direct laser excitation of 229mTh ions in a Paul trap. The main parameters of concept 2 are listed in Tab. IV.

V. CONCEPT 3: NUCLEAR RABI OSCILLATIONS

For the single-ion nuclear clock, it is the goal to irradiate an individual, laser-cooled 229Th^{3+} ion with a single mode of a frequency comb with extraordinarily narrow bandwidth [20]. Ideally, Rabi oscillations should be generated, in order to allow for the Ramsey interrogation scheme in the nuclear clock concept [5]. For the following it is assumed that the same frequency comb already discussed in the previous sections is used and focused to a diameter of 10 μm to irradiate a single ion, however, the bandwidth of an individual comb-mode is narrowed down by a factor of \( 500 \) to 1 Hz. In this case it is shown that nuclear Rabi oscillations could be generated. The input values for Eq. (1) are listed in Tab. V.

The nuclear excitation probability as a function of time if 10 229Th^{3+} ions in a Paul trap are irradiated in parallel.

FIG. 4. Expected number of excited nuclei as a function of time if \( 10^{229}\text{Th}^{3+} \) ions in a Paul trap are irradiated in parallel.

TABLE IV. Main parameters of concept 2.

| Description                                      | Value   |
|--------------------------------------------------|---------|
| Time per scan step                               | 20 s    |
| Number of excited nuclei per scan step           | 5       |
| Number of scan steps for 0.34 eV interval        | \( 1.41 \cdot 10^6 \) |
| Time required to scan 0.34 eV                    | 328 days |

TABLE V. Main parameters of concept 3.

| Variable     | Value | Comment                        |
|--------------|-------|--------------------------------|
| \( I \)      | 0.13 W/cm² | Single-mode focused to \( \varnothing 10 \) μm |
| \( \Gamma_L \) | 2 \( \pi \cdot 490 \) Hz | Larger than nuclear linewidth |
| \( \omega_0 \) | 2 \( \pi \cdot 2.0 \) PHz | Corresponding to 8.3 eV energy |
| \( \Gamma_\gamma \) | \( 10^{-4} \) Hz | Estimated from theory |
| \( \alpha_{ic} \) | 0 | IC suppressed for Th^{3+} ions |
| \( N_0 \) | 10 | Multiple laser-cooled ions |
TABLE V. Values of variables used for the calculation of concept 3.

| Variable | Value | Comment |
|----------|-------|---------|
| $I$ | 0.13 W/cm$^2$ | Single-mode focused to $\varnothing$ 10 µm |
| $\Gamma_L$ | $2\pi \cdot 1$ Hz | Larger than nuclear linewidth |
| $\omega_0$ | $2\pi \cdot 2.0$ PHz | Corresponding to 8.3 eV energy |
| $\Gamma_\gamma$ | $10^{-4}$ Hz | Estimated from theory |
| $\alpha_{ic}$ | 0 | IC suppressed for Th$^{3+}$ ions |
| $N_0$ | 1 | Individual laser-cooled ion |

FIG. 5. Nuclear excitation probability as a function of time for a single $^{229}$Th$^{3+}$ ion in a Paul trap. Nuclear Rabi oscillations with a frequency of 5.9 Hz become visible.

of scan steps amounts to $7.7 \cdot 10^7$. This corresponds to 2.4 years scanning time in order to cover the energy range of 0.038 eV. Again, 9 shifts of the comb center would be required to cover the current 1 $\sigma$ uncertainty interval of 0.34 eV. For this reason, driving nuclear Rabi oscillations, although realistic, will require a drastic improvement of the isomeric energy value, e.g., by concept 1 or concept 2. The main parameters of concept 3 are listed in Tab. VI.

TABLE VI. Main parameters of concept 3.

| Description | Value |
|-------------|-------|
| $\pi$-pulse duration | $\sim$170 ms |
| Excitation probability | $\sim$ 90% |
| Time per scan step | 1 s |
| Number of scan steps for 0.34 eV interval | $6.9 \cdot 10^8$ |
| Time required to scan 0.34 eV | 21.9 years |

VI. A NEW SOLID-STATE NUCLEAR CLOCK CONCEPT

The development of a nuclear optical clock based on individual $^{229}$Th$^{3+}$ ions in a Paul trap has been extensively discussed in literature \[19\] [21]. Such a clock is expected to achieve an extraordinary high accuracy approaching $10^{-19}$ \[20\]. Although expected to be less accurate \[58\], also a nuclear clock that makes use of a large number of $^{229}$Th ions embedded in a crystal-lattice environment has attracted significant attention, as it may pose a practical tool for time measurement \[19\] [58] [59]. Here, a new solid-state nuclear clock concept is proposed, which is based on the laser irradiation of a thin $^{229}$Th layer deposited on a surface, as described in Sect. III. It is shown, that this concept might be advantageous compared to the crystal-lattice clock approach \[58\] [59].

Clock performance is generally expressed via two parameters: accuracy and stability. The stability of a clock corresponds to the statistical uncertainty of a frequency measurement and depends on the measurement time. Opposed to that, the accuracy (or more precisely: the systematic frequency uncertainty) of a clock also takes all systematic uncertainties of a frequency comparison between different clocks into consideration. In order to achieve a high clock performance, it is important that the statistical uncertainty approaches the systematic uncertainty within short averaging times. On the other hand, any improvement of the statistical uncertainty of the frequency measurement significantly below the value of the systematic frequency uncertainty will not improve the clock performance, as this will be limited by other systematic effects.

It was shown in Ref. \[59\] that the stability of a $^{229}$Th-based solid-state nuclear clock can be estimated for the realistic case of low laser intensities and short interrogation times ($T < 1/\Delta \omega_0$) via a shot-noise limited Allan deviation $\sigma^2_T(\tau)$

$$\sigma^2_T(\tau) \approx \frac{1}{\omega_0 T} \frac{\tilde{\Gamma}}{\Gamma} \frac{1}{\sqrt{R N_{\text{eff}} \tau}}. \quad (3)$$

Here $T$ denotes the interrogation time, which should not exceed the isomeric lifetime, $\tilde{\Gamma}$ is the decay rate of the coherences and $\Gamma = \Gamma_\gamma + \Gamma_{ic}$ the total decay rate of the transition, including non-radiative decay channels. $N_{\text{eff}}$ denotes the effective number of irradiated nuclei defined as $N_{\text{eff}} \approx \Gamma_\gamma / \Gamma \cdot kS/(4\pi) \cdot N_0$, with $k$ being the quantum efficiency, $S$ the effective solid angle covered by the detector and $N_0$ the actual number of irradiated nuclei. $\tau$ is the averaging time, which has to be equal or larger than the time $T$ chosen for interrogation and $R$ is the nuclear excitation rate, which can be estimated for the considered case of low laser intensities using $\Omega$ from Eq. \[2\] as

$$R \approx \frac{\Omega^2}{2T} = \frac{\pi c^2 I \Gamma_\gamma}{\hbar \omega_0^2 \Gamma}. \quad (4)$$

1 The equation is obtained by inserting Eq. (37) of Ref. \[59\] into Eq. (48) of the same paper and using that by definition $\tau = 47$, where $\tilde{\tau}$ denotes the time for a complete interrogation cycle used in Ref. \[59\].
In the following, the crystal-lattice approach as proposed in Refs. [19, 58] is discussed. Assuming purely radiative decay (Γ = Γ_{ic}) and an interrogation time of T = 10^4 s, corresponding to the expected radiative lifetime, as well as a decay rate of the coherences of \( \Gamma \approx 1 \text{ kHz} \) due to coupling to the crystal-lattice environment (see Ref. [59] for details), a straightforward calculation reveals a high stability of \( \sigma_{\text{ic}}(\tau) \approx 1.9 \cdot 10^{-17}/\sqrt{\tau} \) for a solid state nuclear optical clock [59]. Here, values of \( N_0 = 10^{14}, k = 0.1, S = 4\pi/10 \text{ and } f = 1.3 \cdot 10^{-5} \text{ W/cm}^2 \) (corresponding to 100 nW of laser power irradiating a crystal of 1 mm diameter) were used. Thus, after a short averaging time of \( \tau = 10^4 \text{ s} \), a stability in the lower 10^{-19} range appears to be achievable. However, the systematic frequency uncertainty of the clock is expected to be limited by other effects, most importantly temperature shifts of the crystal, to a value of \( 2 \cdot 10^{-16} \) [58]. For this reason only limited advantage will be gained from the high stability as long as other systematic shifts will not be sufficiently well controlled.

In the following, the same stability estimate is applied to a new nuclear clock concept, which is based on the observation of internal conversion electrons following the laser irradiation of neutral \(^{229}\text{Th}\) atoms on a surface. In this case, the nuclear decay rate will be dominated by the short IC lifetime of about 10 \( \mu \text{s} \) [39]; \( \Gamma \approx \Gamma_{ic} \approx 10^5 \text{ Hz} \). Further, as the laser used for irradiation is narrowband, the decay rate of the coherences will be dominated by the nuclear decay rate: \( \Gamma \approx \Gamma_{ic}/2 \approx \Gamma_{ic}/2 \). Inserting these values, together with a short interrogation time of \( T = 10 \mu \text{s} \), into Eq. (3), a stability of \( \sigma_{\text{ic}}(\tau) \approx 3 \cdot 10^{-15}/\sqrt{\tau} \) is achieved. Here it was assumed that \( N_0 = 10^{14} \) atoms are irradiated with a laser of intensity \( I = 1.3 \cdot 10^{-5} \text{ W/cm}^2 \). Further, \( N_{\text{eff}} = kS/(4\pi) \) was used, taking into account that no competing decay channels exist, with \( k = 0.1 \) as before, but \( S = 4\pi/2 \), as the emitted electrons can be attracted towards the detector. Obviously, the calculated stability performance is lower than the best that could be achieved with a crystal-lattice nuclear clock. Importantly, however, due to the short interrogation time of \( T = 10 \mu \text{s} \), the averaging time \( \tau \) can be chosen nearly arbitrarily short and after about 250 \( s \) of averaging the stability has surpassed the expected systematic frequency uncertainty, for which a value of \( 2 \cdot 10^{-16} \), like for the crystal-lattice approach, is assumed. Of course there is the option to choose shorter interrogation times also in the crystal-lattice clock approach. This will, however, significantly affect the clock’s stability performance, as it scales proportional to \( 1/T \) (e.g., using \( T = 100 \text{ s} \) interrogation time a stability of \( 1.9 \cdot 10^{-15}/\sqrt{\tau} \) is obtained, comparable to the expected stability of an IC-based nuclear clock).

From the above considerations it is evident that the stabilities of both, a crystal-lattice nuclear clock as well as an IC-based nuclear clock, should be sufficient to approach a statistical uncertainty of the frequency measurement of about \( 10^{-16} \) after a few hundred seconds of averaging. Longer measurement times are not expected to improve clock performance, which will most likely be limited to a level of \( 10^{-16} \) by systematic frequency uncertainties [58]. However, it may be seen as a big advantage of the IC-based nuclear clock approach that no competing decay channels exist next to internal conversion, which has already been experimentally observed and is known to be the dominant decay channel for the considered experimental conditions. Opposed to that, the crystal-lattice nuclear clock makes important use of the \(^{229}\text{mTh}\) \( \gamma \) decay, which requires suppression of the IC decay channel by nine orders of magnitude in a solid-state environment. Even when achieved, non-radiative decay via bound internal conversion (BIC) and electronic bridge (EB) mechanisms might be present, reducing the isomeric lifetime and the fraction of observable \( \gamma \) decays.

VII. CONCLUSION AND OUTLOOK

Three different concepts for direct frequency-comb spectroscopy of \(^{229}\text{mTh}\) were discussed. The first concept proposes to use the 7th harmonic of a Yb-doped fiber frequency comb to perform direct nuclear laser excitation of \(^{229}\text{Th}\) deposited as a thin layer on a surface. The successful excitation of the isomeric nuclear state is probed via the detection of internal conversion electrons emitted in the isomer’s decay. It is the first time that such an experiment has been proposed and it appears to be advantageous compared to direct nuclear laser spectroscopy of \(^{229}\text{Th}\) ions in a Paul trap in terms of measurement time and absolute number of nuclear excitations. The mode spacing of the frequency comb of 77 MHz could be scanned within 100 \( s \), thereby effectively probing an energy range of 9.2 THz (0.038 eV), as all 1.2 \( \times 10^5 \) comb modes are used in parallel in the search for the nuclear excitation. Further, in case of success, the concept could be applied for the development of an IC-based solid-state nuclear clock in a straightforward manner.

The proposed experiment is similar to a direct laser excitation scheme described in Refs. [46, 47], where laser excitation of \(^{229}\text{mTh}\) is envisaged using a broadband (10 GHz) widely-tunable and pulsed laser system as obtained via four-wave mixing in an argon gas jet. The advantage of this scheme compared to the one described in Refs. [46, 47] is that here laser excitation is performed via a single mode of a frequency comb offering a bandwidth of 490 Hz. Therefore, any energy determination of the isomeric excitation will be limited by the IC-broadened natural transition linewidth of 15.9 kHz.

Two further experiments using the same frequency comb for excitation of laser-cooled \(^{229}\text{mTh}\) ions were discussed: Concept 2 describes the case where multiple \(^{229}\text{mTh}\) ions in a Paul trap are irradiated. Although definitely realistic, long required scanning times let a further energy constraint appear favorable prior to...
experimental realization. For concept 3 it is assumed that a single mode of the frequency comb is narrowed down to a bandwidth of 1 Hz as would be advantageous for a single-ion nuclear clock. Although this concept requires significantly more effort and much improved constraints on the isomeric energy value, it is shown that in principle it should be possible to drive nuclear Rabi oscillations with the laser system under consideration. Therefore, the same frequency comb used for concept 1 could also be applied for nuclear laser spectroscopy in a Paul trap and thus for the development of a single-ion nuclear clock, as soon as the energy of $^{229m}$Th has been sufficiently constrained. These experiments can be envisaged as follow-up investigations to concept 1.

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