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Laser-induced electronic bridge for characterization of the $^{229m}\text{Th} \rightarrow ^{229g}\text{Th}$ nuclear transition with a tunable optical laser

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Abstract

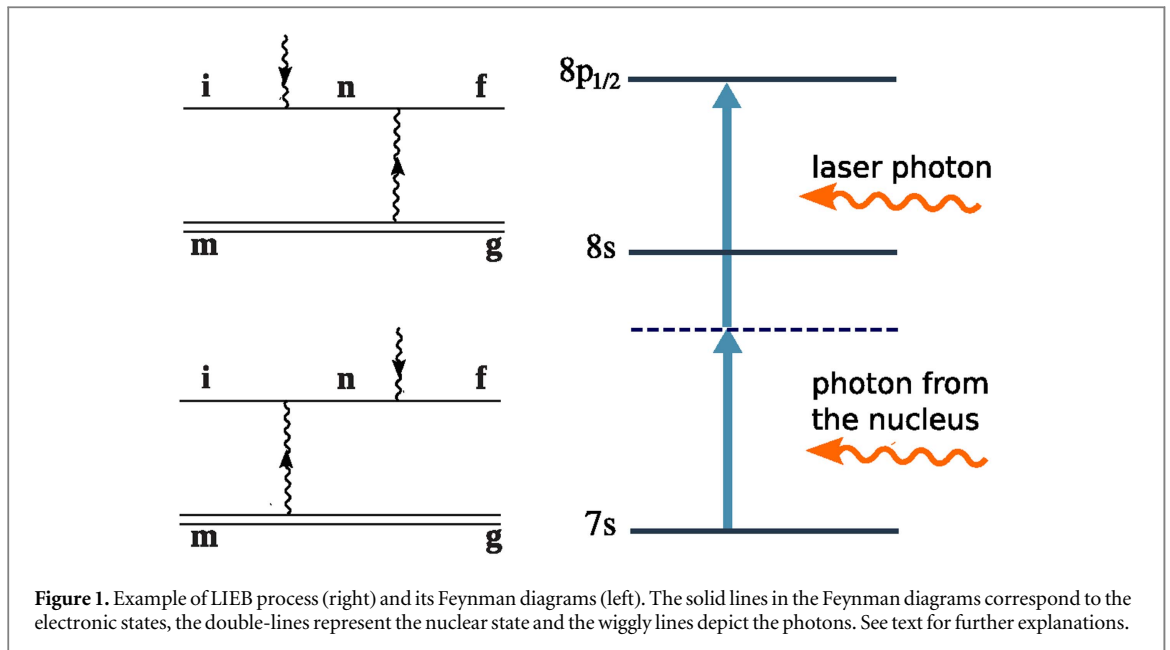
An alternative method to determine the excitation energy of the ^{229m}Th isomer via the laser-induced electronic bridge is investigated theoretically. In the presence of an optical or ultra-violet laser at energies that fulfill a two-photon resonance condition, the excited nuclear state can decay by transferring its energy to the electronic shell. A bound electron is then promoted to an excited state by absorption of a laser photon and simultaneous de-excitation of the nucleus. We present calculated rates for the laser-induced electronic bridge process and discuss the experimental requirements for the corresponding setup. Our results show that depending on the actual value of the nuclear transition energy, the rate can be very high, with an enhancement factor compared to the radiative nuclear decay of up to 10^8 .

1. Introduction

The ^{229}Th isotope has been subject of increased attraction of the scientific community due to its unique long lived excited state ^{229m}Th at an energy lying in the VUV range [1–3]. Such long-lived excited states are termed in nuclear physics *isomers*. The $^{229m}\text{Th} \rightarrow ^{229g}\text{Th}$ transition to the ground state has an extremely small ratio of the radiative width to the energy, estimated at 10^{-19} . This opens the possibility for exciting applications such as a very precise nuclear frequency standard [4–6] or a nuclear laser [7]. The extremely low energy may render nuclear spectroscopy and even coherent control of the nuclear excitation with vacuum ultraviolet (VUV) lasers possible [8, 9]. Moreover, strong coupling of the nuclear transition to the electronic shell in ^{229}Th ions may allow usage of the outer electrons to excite the nuclear isomeric state with optical lasers in a two-photon electronic bridge (EB) process [10].

Precise knowledge of the transition parameters such as energy and γ -decay rate (which in the theoretical treatment can be traced back to the reduced nuclear transition probabilities) is needed for the aforementioned applications. The first direct observation of the ^{229m}Th isomer decay via internal conversion (IC) has been reported recently [1]. IC is a nuclear decay channel open for transition energies exceeding the atomic ionization potential: the excitation energy of the nucleus is transferred to a bound electron with subsequent ionization. As the nuclear excitation energy in ^{229m}Th lies above the first ionization potential of 6.3 eV [11], IC is possible and also very efficient in neutral Th atoms. The nuclear isomeric state ^{229m}Th decays via IC with enhancement factor 10^9 with respect to the γ -decay [1]. The most accepted value of the ^{229m}Th energy $E_m = 7.8 \pm 0.5$ eV [2, 3] could be determined only indirectly in a calorimetric measurement by subtraction of x-ray energy differences between neighboring nuclear levels. We note that for this value IC for Th ions is energetically forbidden as the nuclear excitation energy is lower than the ionization potential of Th^+ , i.e. approx. 12 eV [12].

There are a few reasons today to doubt the precision of the $E_m = 7.8 \pm 0.5$ eV value. In the first place, its extraction from the experimental data in [2, 3] was based on uncertain nuclear branching ratios, making possible an underestimation of the uncertainty ± 0.5 eV [13]. Secondly, recent negative experimental results of two broadband photoexcitation attempts of the isomeric state may indicate that the transition energy lies in a different energy range [14, 15]. Furthermore, the recent results in [16] on the short lifetime of the isomer in Th^+ may be indirect evidence that the IC channel is open also for this charge state, and thus E_m is higher than the



ionization potential of Th^+ at approx. 12 eV. We note that for $E_m = 7.8 \pm 0.5$ eV, energy conservation only allows IC from excited electronic states to occur in Th^+ ions [17].

In this work we propose an alternative to measure the energy of the ^{229}Th nuclear isomeric state at a precision typical to laser atomic spectroscopy, and to improve knowledge of its radiative lifetime. The method is based on nuclear deexcitation by laser-induced electronic bridge (LIEB). For a nuclear transition energy E_m around 12 eV, this process has an especially high rate and could be very efficient to characterize the nuclear transition with the help of a tunable optical laser. The usual EB is a process coupling the nucleus to the atomic shell [18–21]. It occurs when the nuclear transition energy is not sufficient for IC, but is close to an atomic transition energy. In order to fulfill energy conservation, the transfer of the nuclear excitation to a bound electron which undergoes a transition to an excited state is accompanied by the emission of a photon. The EB process might play a significant role in the decay of the isomeric state in Th ions, where the IC channel is energetically closed. Calculations have shown that the EB process can significantly change the isomeric state lifetime if the energy E_m happens to be close to $M1$ transitions of the electronic shells [22, 23].

LIEB is a version of EB process in which the additional photon is not emitted, but absorbed by the electronic shell from an externally applied laser field. The process is thus equivalent to the excitation of the electronic shell with two photons, one of which is provided by the laser source and the other one from the nuclear deexcitation. This process was first considered for atomic ^{229}Th in 1992 by Karpeshin *et al* [24]. It was shown that an enhancement factor of the decay of the isomeric state ^{229m}Th of the order 10^3 can be achieved. We note that at the time, the isomeric energy was believed to lie around 3.5 eV, such that IC of the neutral thorium atom wouldn't have been possible. Meanwhile it is known today that the isomeric nuclear state in a neutral atom decays very fast via IC [1], rendering the scheme proposed in [24] obsolete. We consider LIEB in the $^{229}\text{Th}^{3+}$ ion and show that with up-to-date laser technology the enhancement can reach the order 10^8 if the value E_m is close to the ionization threshold of Th^+ . This would allow for a measurement of the transition energy and decay rate by using a tunable optical laser. $^{229}\text{Th}^{3+}$ ions allow high controllability as far as trapping and cooling are concerned and possess a simple electronic spectrum, rendering a reliable experimental implementation possible.

The LIEB process can be represented by two Feynman diagrams shown on the left-hand side of figure 1. The states $|m\rangle$ and $|g\rangle$ denote the isomeric and the ground states of the nucleus, respectively. The states $|i\rangle$ and $|f\rangle$ in turn are the initial and the final electronic states, respectively. The intermediate electronic state $|n\rangle$ runs over all the levels allowed by the selection rules (in the whole spectrum, including the continuum). The transition $^{229m}\text{Th} \rightarrow ^{229g}\text{Th}$ has $M1$ multipolarity. We make use of the fact (also considered in [24]) that the coupling of the nuclear transition is maximal to an $M1$ electronic transition between s orbitals and considerable for transitions between $p_{1/2}$ orbitals. We consider therefore the excited $7s$ state as the initial electronic state $|i\rangle$ and the $8p_j$ and $9p_j$ states with the total angular momentum $j \in \{1/2, 3/2\}$ in the role of the state $|f\rangle$. With this choice the channels $7s \rightarrow ns \rightarrow |f\rangle$ with $n = 8, 9$ and $7s \rightarrow 7p_{1/2} \rightarrow |f\rangle$ would then play the dominating role. These two channels are described by the two different Feynman diagrams in figure 1, corresponding to different absorption sequences of the two photons by the electron. A further advantage of using the $7s$ state as the initial state is its

metastability, i.e. very long lifetime of 0.6 s [25]. Note that since $|n\rangle$ is actually a virtual state, the energy of the $M1$ photon does not necessarily have to coincide with the energy of the $M1$ electronic transition, though the overall transition energy $|i\rangle \rightarrow |f\rangle$ has to be equal to the sum of the energies of the two photons (see figure 1).

The nuclear isomer energy E_m can be found via scanning with a tunable laser for a LIEB resonance, i.e., for the population and decay of the upper state $|f\rangle$. The decay observation of the $|f\rangle$ state would correspond to the sum of the nuclear excitation energy and the energy of the laser photon equaling the overall electronic transition energy. If the resonant laser photon frequency is ω , then E_m can be calculated as

$$E_m = E_f - E_i - \hbar\omega, \quad (1)$$

where E_i and E_f are the energies of the electronic state $|i\rangle$ and $|f\rangle$ respectively and \hbar is Planck constant. The height of the measured LIEB resonances would provide information about the magnitude of the nuclear γ -decay rate.

2. Theory

We calculate the LIEB rate W_{LIEB} by relating to the rate of the inverse spontaneous process W^{spont} via the expression [26]

$$W_{\text{LIEB}} = W^{\text{spont}} \frac{4\pi^3 c^2}{\hbar\omega^3} P_\omega \delta, \quad (2)$$

where ω and P_ω are the frequency and the spectral intensity of the laser field, c is the speed of light and

$$\delta = \frac{(2I_g + 1)(2J_f + 1)}{(2I_m + 1)(2J_i + 1)} \quad (3)$$

denotes the ratio of magnetic quantum number degeneracies of the initial states in the case of the direct and the inverse processes. These states are characterized by the nuclear spin I and the total angular momentum of the electronic shell J . For calculation of W^{spont} we adopt the formalism developed in [22] for the traditional EB scenario, taking into account the different direction of the nuclear process, i.e. excitation instead of deexcitation. The Feynman diagrams of this process can be obtained from the diagrams in figure 1 by mirror reflection and inversion of the photon lines. Based on the corresponding amplitudes, one can derive the rate of the decay W^{spont} , which after averaging over the initial states, summation over the final states and application of Wigner-Eckart theorem [27] can be rewritten with the help of the isomer γ -decay rate Γ_γ as

$$W^{\text{spont}} = \left(\frac{\hbar\omega}{E_m} \right)^3 \frac{G_1 + G_{12} + G_2}{3(2J_f + 1)} \cdot \frac{2I_m + 1}{2I_g + 1} \cdot \Gamma_\gamma. \quad (4)$$

Here, the ratio $\frac{2I_m + 1}{2I_g + 1}$ takes into account different initial nuclear states for the considered spontaneous process and γ -decay. The quantities G_i with $i = 1, 2$ corresponding to the two Feynman diagrams and their interference can be written with the help of the reduced matrix elements of the electric D and magnetic T dipole operators [28] of the valence electron giving the following expressions (in atomic units)

$$G_1 = \sum_n \frac{1}{2J_n + 1} \times \left| \sum_{\gamma_k} \frac{\langle \gamma_i J_i \| D \| \gamma_k J_n \rangle \langle \gamma_k J_n \| T \| \gamma_f J_f \rangle}{E_f - E_k - E_m} \right|^2, \quad (5)$$

$$G_{12} = 2 \sum_{J_n} (-1)^{J_i + J_n} \begin{Bmatrix} J_f & J_t & 1 \\ J_i & J_n & 1 \end{Bmatrix} \times \sum_{\gamma_k} \frac{\langle \gamma_i J_i \| D \| \gamma_k J_n \rangle \langle \gamma_k J_n \| T \| \gamma_f J_f \rangle}{E_f - E_k - E_m} \times \sum_{\gamma_s} \frac{\langle \gamma_i J_i \| T \| \gamma_s J_t \rangle \langle \gamma_s J_t \| D \| \gamma_f J_f \rangle}{E_i - E_s + E_m}, \quad (6)$$

$$G_2 = \sum_{J_n} \frac{1}{2J_n + 1} \times \left| \sum_{\gamma_k} \frac{\langle \gamma_i J_i \| T \| \gamma_k J_n \rangle \langle \gamma_k J_n \| D \| \gamma_f J_f \rangle}{E_i - E_k + E_m} \right|^2. \quad (7)$$

The sums are carried out over the total angular momenta of the intermediate states J_n and J_i and over all other electronic quantum numbers denoted by the generic indices γ_k and γ_s . The notation $\langle \| \cdot \| \rangle$ stands for the reduced matrix elements after application of the Wigner–Eckart theorem [27].

3. Numerical results

For numerical evaluation of W_{LIEB} we require the nuclear γ -decay rate, the spectrum of the valence electron and the $E1$ and $M1$ matrix elements between the states $|i\rangle$ and $|n\rangle$ and the states $|f\rangle$ and $|n\rangle$ for all $|n\rangle$ allowed by the selection rules (including the continuum states). We consider the value $P_\omega = 1 \text{ W m}^{-2} \text{ Hz}^{-1}$ for the optical laser spectral intensity, as typical for a continuous laser that emits 1 W of output power within a 1 MHz linewidth and focused to a 1 mm² focus area. This can be achieved with modern tunable lasers in the range from 560 to 1000 nm. We assume $\Gamma_\gamma = 10^{-4} \text{ s}^{-1}$ corresponding to the recent theoretical value for the reduced matrix element of transition from the nuclear isomeric state to the nuclear ground state $B_1 = 0.0076 \text{ W.u.}$ [29]. This theoretical value is smaller than previously estimated [30, 31] and provides therefore a more pessimistic result for the isomer radiative decay rate. However, recent experimental results [16] confirm the value $\Gamma_\gamma = 10^{-4} \text{ s}^{-1}$ and support the theoretical prediction in [29].

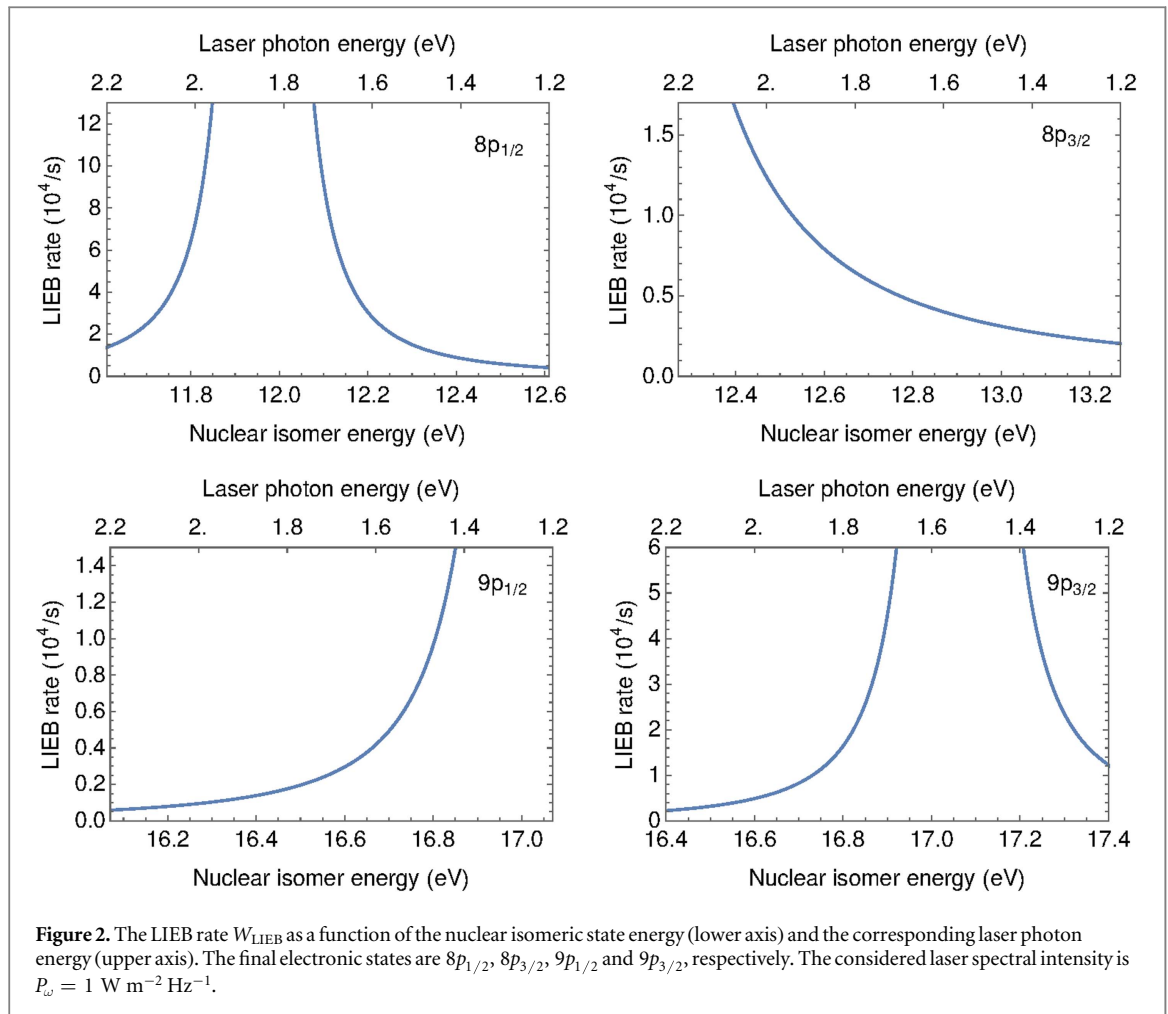
The spectrum of the valence electron is taken from the database in [32]. The electronic matrix elements are calculated based on *ab initio* wave-functions evaluated employing the following procedure: first we calculate a few low-lying states of the valence electron using Dirac–Hartree–Fock method in the frozen-core approximation. These serve as initial estimates for more accurate wave functions calculated by considering the ion to be placed into a cavity of the radius $R = 60 \text{ a.u.}$ and building virtual orbitals 1–20s, 2–20p, 3–20d, 4–25f, 5–18g via the expansion in a B -spline basis [28]. Due to the large size of the cavity, this procedure does not affect the matrix elements considerably, but allows us to work with a discrete spectrum at positive energies. In order to benchmark our method we have constructed the same basis set as in [22] and have reproduced numerical results presented therein. Our results show good agreement with [22] confirming the reliability of the computed electronic wave functions and of our numerical results.

The graphs in figure 2 show the LIEB rate W_{LIEB} as a function of the nuclear isomer energy for different final electronic states. We depict also the energy of the resonant laser photon $\hbar\omega$, considering values in the range from 1.2 to 2.2 eV available with modern high power tunable lasers. If the energy E_m is close to the electronic $M1$ transition, then a large enhancement $W_{\text{LIEB}}/\Gamma_\gamma$ up to 10^8 can be achieved. The large peak appearing in all the four cases corresponds to the resonance in the lower diagram in figure 1 for the $M1$ electronic transition between the initial state $|i\rangle = |7s\rangle$ and the intermediate state $|n\rangle = |8s\rangle$ or $|n\rangle = |9s\rangle$. We note however that this substantial enhancement would only be present if the isomer energy lies around 12 or 17 eV.

In case E_m is closer to the presently accepted value of $7.8 \pm 0.5 \text{ eV}$ [2, 3], a laser with higher photon energy is required. In figure 3 we show the LIEB rate W_{LIEB} calculated using the value $P_\omega = 10^{-2} \text{ W m}^{-2} \text{ Hz}^{-1}$ for laser photon energies higher than 2.2 eV, as obtainable with an average laser power of 0.01 W in a 1 MHz linewidth and a 1 mm² focus area. This might be a too conservative estimate for 2.5 eV, but simultaneously already demanding for 4.5 eV photon energies, respectively. We observe new peaks, which correspond to the case of resonances in the upper Feynman diagram in figure 1, when E_m coincides with the energy between the final state and an intermediate state among $7p_j$ and $8p_j$ with $j \in \{1/2, 3/2\}$. In this case the strongest peaks occur for electronic $M1$ transitions between $p_{1/2}$ states. Provided the spectral intensity $P_\omega = 10^{-2} \text{ W m}^{-2} \text{ Hz}^{-1}$ in the depicted region of laser photon energy can be reached, the described method would be applicable for lower values of E_m as low as 8 eV under conditions of good detection efficiency.

4. Discussion

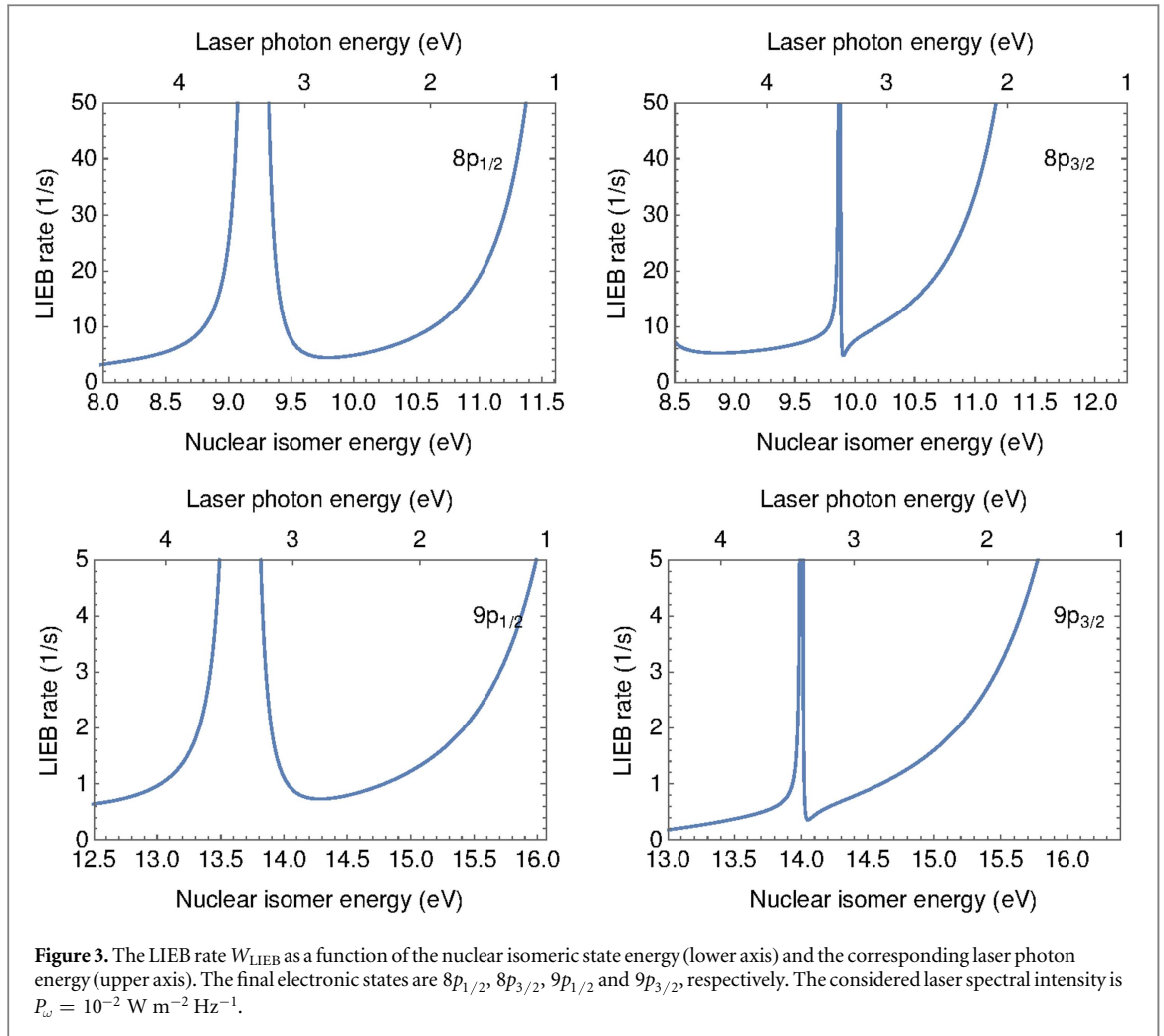
Using the described scheme the energy of the VUV nuclear transition $^{229\text{m}}\text{Th} \rightarrow ^{229\text{g}}\text{Th}$ can be determined using a tunable optical laser. The rate of the LIEB process can be measured by means of detection of the spontaneous decay photons of the final electronic state $|f\rangle$. Comparing the value of the resonant LIEB rate to the theoretical predictions, it is possible to find Γ_γ . Note that among various charge states of thorium, the monovalent $^{229}\text{Th}^{3+}$ ion allows particularly accurate *ab initio* calculations for the electronic shell. The $^{229}\text{Th}^{2+}$ ions which would also fulfill this requirement cannot be used for the described method, as no metastable states with either s - or $p_{1/2}$ -electrons involved are available. The described scheme in $^{229}\text{Th}^{3+}$ is therefore a unique reliable way for



determination of the Th nuclear isomeric state parameters via laser-induced nuclear deexcitation involving the electronic shell.

The LIEB scheme requires production and trapping of $^{229\text{m}}\text{Th}$ in the $3+$ ionization state and the excitation of the electronic shell to the $7s$ initial state. Regarding the production of the isomeric state, the direct nuclear photoexcitation of the ground-state thorium has failed so far due to the poor knowledge of the transition energy. Instead, one typically takes advantage of the 2% branching ratio to the low-energy isomeric state [33] in the process of α -decay of ^{233}U and subsequent gamma-ray de-excitation. Inevitably a mixture of mostly ground state and just few isomeric state $^{229}\text{Th}^{3+}$ ions would then be loaded in the ion trap. We anticipate that 10^4 – 10^5 ions could be produced and trapped at the Munich setup [1, 16] where a cryogenic Paul trap is being built [34]. This setup will keep $^{229}\text{Th}^{3+}$ ions in the absence of charge exchange and chemical reactions for a storage time longer than the time of isomer radiative decay. The electronic shell can be effectively transferred from the ground state $5f_{5/2}$ to the $7s$ state making use of the stimulated Raman adiabatic passage (STIRAP) method [35] via the intermediate state $6d_{3/2}$. Note that the second transition in the STIRAP scheme is an electric quadrupole ($E2$) transition and thus requires higher driving laser intensity. Once reached, the $7s$ state has a long lifetime (0.6 s) allowing for applying the LIEB scheme.

Upon interaction with the tunable optical or ultra-violet laser, the isomeric state which otherwise has a long radiative lifetime of approx. 10^4 s may decay via the LIEB process. Population and radiative decay of the upper electronic states np_j with $n = 8, 9$ and $j \in \{1/2, 3/2\}$ could be observed by detection of fluorescence photons. We present the calculated radiative decay rates for the electric dipole channels together with experimental photon energy values from [32] in tables 1–4. Since the energy of the emitted photon in the dominant decay channels is mostly in the range 15–20 eV, i.e. significantly higher than that of the laser photons that are used for the excitation, the selection of a suitable photocathode material (like CsI) for photoelectron production inside the vacuum system will ensure a detection that is free from background of stray laser light. The background due to the absorption of the laser radiation by the electron in thorium ions with the nucleus in the ground state is expected to be small since the envisaged electronic transitions are energetically much higher than the laser photon energy. A direct comparison with the case of ^{232}Th which does not possess any nuclear states at optical energies should help to identify the signal stemming from the isomeric state decay.

**Table 1.** Radiative decay channels of the $8p_{1/2}$ state.

Final state	Photon energy (eV)	Rate (10^8 s^{-1})
$7s_{1/2}$	13.8	0.26
$8s_{1/2}$	1.85	4.96
$6d_{3/2}$	15.5	19.4
$7d_{3/2}$	1.84	5.81

Table 2. Radiative decay channels of the $8p_{3/2}$ state.

Final state	Photon energy (eV)	Rate (10^8 s^{-1})
$7s_{1/2}$	14.5	5.05
$8s_{1/2}$	2.51	11.3
$6d_{3/2}$	16.2	3.12
$6d_{5/2}$	15.6	24.9
$7d_{3/2}$	2.50	1.15
$7d_{5/2}$	2.29	8.76

Table 3. Radiative decay channels of the $9p_{1/2}$ state.

Final state	Photon energy (eV)	Rate (10^8 s^{-1})
$7s_{1/2}$	18.3	0.57
$8s_{1/2}$	6.31	0.045
$9s_{1/2}$	1.21	3.30
$6d_{3/2}$	20.0	11.5
$7d_{3/2}$	6.30	1.36
$8d_{3/2}$	1.06	3.13

Table 4. Radiative decay channels of the $9p_{3/2}$ state.

Final state	Photon energy (eV)	Rate (10^8 s^{-1})
$7s_{1/2}$	18.6	1.76
$8s_{1/2}$	6.64	1.30
$9s_{1/2}$	1.54	6.01
$6d_{3/2}$	20.3	1.80
$6d_{5/2}$	19.68	14.5
$7d_{3/2}$	6.63	0.42
$7d_{5/2}$	6.42	3.02
$8d_{3/2}$	1.39	0.56
$8d_{5/2}$	1.32	4.73

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