Excitation of the $^{229}\text{Th}$ nucleus via a two-photon electronic transition

Robert A. Müller,1,2,* Andrey V. Volotka,3,4 and Andrey Surzhykov1,2

1 Physikalisches-Technische Bundesanstalt, D-38116 Braunschweig, Germany
2 Technische Universität Braunschweig, D-38106 Braunschweig, Germany
3 Helmholtz Insitute Jena, D-07743 Jena, Germany
4 GSI Helmholtzzentrum für Schwerionenforschung, D-64291 Darmstadt, Germany

We investigate the process of nuclear excitation via a two-photon electron transition (NETP) for the case of the doubly charged thorium. The theory of the NETP process has been devised originally for heavy helium like ions. In this work we study this process in the nuclear clock isotope $^{229}\text{Th}$ in the 2+ charge state. For this purpose we employ a combination of configuration interaction and many-body perturbation theory to calculate the probability of NETP in resonance approximation. The experimental scenario we propose for the excitation of the low lying isomeric state in $^{229}\text{Th}$ is a circular process starting with a two-step pumping stage followed by NETP. The ideal intermediate steps in this process depend on the supposed energy $\hbar\omega_N$ of the nuclear isomeric state. For each of these energies the best initial state for NETP is calculated. Special focus is put on the most recent experimental results for $\hbar\omega_N$.

I. INTRODUCTION

Currently all experiments aiming for a precise determination of the second are based on electronic transitions in atoms and ions. Fifteen years ago it has been proposed by Peik and coworkers to build a clock based on a nuclear transition [1]. The most suitable of such transitions is found in the thorium isotope with mass number $A = 229$, nowadays sometimes referred to as nuclear clock isomer. Therefore intense research, theoretically and experimentally, has been performed on $^{229}\text{Th}$ and especially the nucleus in its first excited state, the isomer $^{229m}\text{Th}$ [2–5]. Recently, for example, the nuclear moments of $^{229m}\text{Th}$ have been determined [6,7], which may give insight into the energy of the nuclear isomeric state [8]. Moreover the emission of internal conversion electrons from the $^{229m}\text{Th} \rightarrow ^{229}\text{Th}$ transition has been observed [9]. However a controlled excitation of the nuclear isomer has not been achieved yet [10].

A large number of different processes have been proposed to produce the $^{229m}\text{Th}$ nuclear isomer ranging from direct laser excitation to the interaction with hot plasmas [1–4]. Out of these the excitation of nuclei by the energy excess from electronic processes appears to be very efficient and largely outnumbers, e.g. direct laser excitation [6,13,14]. However all such electronic bridge processes come with a major challenge: The electronic transition needs to be in resonance with the transition between the nuclear ground and the low lying isomeric state of $^{229}\text{Th}$, i.e. both the electronic and the nuclear transition need to have the same energy. This challenge can be overcome by using two-photon transitions in the electron shell [13]. In such a transition one, virtual, photon excites the nucleus while the other is emitted as a real photon. The energy share between both photons is continuous and, thus, the resonance condition does not apply here. This so called nuclear excitation by a two-photon electron transition (NETP) has been introduced for heavy highly charged ions, to access nuclear excited states in the keV regime [17].

In this work we want to investigate NETP in $^{229m}\text{Th}$. In contrast to other nuclear levels, the $^{229m}\text{Th}$ isomeric state is found only about 8 eV above the $^{229}\text{Th}$ ground state. Therefore the electronic transition needs to be in the same energy range. Consequently lower charge states, especially $^{229}\text{Th}^{2+}$, are promising candidates to observe NETP in thorium.

In contrast to the scenario discussed in Ref. [17] for helium like ions, $\text{Th}^{2+}$ has many real intermediate resonances between the upper and the final state of the NETP process, provided by the rich level structure of the thorium ion. Ideally such a resonance is close to the nuclear excitation energy, thus enhancing the probability of the NETP process. The location and number of the resonances, however, strongly depends on the initially pumped upper state. Therefore the upper state which offers the highest probability for NETP depends on the energy of the nuclear isomeric state. In this paper we therefore provide detailed calculations for NETP in $^{229}\text{Th}^{2+}$ and give clear recommendations for the levels to excite, depending on the energy range in which the isomer is searched.

Hartree atomic units ($\hbar = m_e = e = 1$) are used throughout this paper unless stated otherwise.

II. SCENARIO

A sketch of the scheme we propose for the excitation of the low lying isomeric state in $^{229}\text{Th}$ can be seen in Fig. 1. First the electron shell of the thorium ion is excited to an upper state. From this upper state the NETP process occurs, where the nuclear excitation energy either corresponds to the energy splitting between the upper and the intermediate (left panel) or the intermediate and the lower state. Due to atomic selec-

* robert.mueller@ptb.de
We propose for the excitation of the excited state via two-step laser excitation. Generally the intermediate state can be virtually an, thus, we have to sum over the entire spectrum $|\gamma_n J_n \mu_n\rangle$, where we assume that the continuous spectrum can be neglected. Note that in Eq. (1) we have omitted the width of the nuclear excited state, since it is much narrower than the electronic states.

$$M_{fi} = \sum_{\gamma_n J_n \mu_n} \left( \langle \gamma_f J_f \mu_f, I_e M_e | \alpha \cdot u_\lambda e^{ik \cdot r} | \gamma_n J_n \mu_n, I_e M_e \rangle \right) \left( \langle \gamma_n J_n \mu_n, I_e M_e | H_{int} | \gamma_i J_i \mu_i, I_g M_g \rangle \right)$$

$$+ \sum_{\gamma_n J_n \mu_n} \left( \langle \gamma_f J_f \mu_f, I_e M_e | H_{int} | \gamma_n J_n \mu_n, I_g M_g \rangle \right) \left( \langle \gamma_n J_n \mu_n, I_g M_g | \alpha \cdot u_\lambda e^{ik \cdot r} | \gamma_i J_i \mu_i, I_g M_g \rangle \right)$$

(1)

where $\omega_N$ is the frequency of the nuclear transition and $\alpha$ denotes the vector of Dirac matrices. State energies and widths are denoted by $\epsilon$ and $\Gamma$, respectively, while the subscripts $i, n$ and $f$ specify the initial, intermediate and final states. Generally the intermediate state can be virtual an, thus, we have to sum over the entire spectrum $|\gamma_n J_n \mu_n\rangle$, where we assume that the continuous spectrum can be neglected. Note that in Eq. (1) we have omitted the width of the nuclear excited state, since it is much narrower than the electronic states.

Both terms in Eq. (1) each split into two matrix elements of the operators $H_{int}$ and $\alpha \cdot u_\lambda e^{ik \cdot r}$. The latter is the usual interaction of the electron shell with a plane-wave photon with momentum $k$ polarized along $u_\lambda$, where $\lambda$ is the helicity. The interaction Hamiltonian $H_{int}$ mediates the interaction between the electron shell and the nucleus, thus acting on both electronic and nuclear degrees of freedom.

To obtain the probability of the NETP process we can...
where \([k] = 2k + 1\), \(\alpha\) is the fine structure constant and \(\omega\) the frequency of the real, emitted, photon. In Eq. (2) we average over \(M_q\) and \(\mu_i\), assuming that the initial electronic and nuclear states are unpolarized. Moreover neither \(\mu_f\) and \(M_e\) nor the emission direction of the real photon is observed, thus we sum over the magnetic quantum numbers of the final states and integrate over \(\Omega_k\).

To express Eq. (2) in a more convenient way the photon emission operator \(\alpha \cdot \mathbf{u}_\lambda e^{ikr}\) is readily expanded into electric \((p = 1)\) and magnetic \((p = 0)\) multipoles \(L\) with magnetic quantum number \(M\) [18, 19]:

\[
\mathbf{u}_\lambda e^{ikr} = \sqrt{2\pi} \sum_{LMp} i^L [L]^\frac{1}{2} (i\lambda)^p D_{LM}^L (\phi_k, \theta_k, 0) \mathbf{a}^{(p)}_{LM},
\]

where \(D_{LM}^L (\phi_k, \theta_k, 0)\) is the Wigner-D matrix and \(\mathbf{a}^{(p)}_{LM}\) are irreducible tensors of rank \(L\) resembling the multipole fields.

Similar to the photon interaction operator, the electron-nucleus interaction \(\hat{H}_{int}\) can be expanded into

\[
\hat{H}_{int} = \sum_{qr} \hat{T}_{qr} \hat{M}_{qr},
\]

where is important to note that for each multipole the operator \(\hat{H}_{int}\) splits into the hyperfine interaction operators \(\hat{T}_{qr}\) acting only on electronic degrees of freedom and \(\hat{M}_{qr}\) interacting with the nuclear part of the wave function. That way we can find the NETP probability for each multipolarity \(q\) of the nuclear transition and electronic transitions \(L\) and \(p\).

\[
\mathcal{W}_{fi}^{(Lpq)} = \frac{8\pi \alpha^3 \omega}{|J_i| |I_2|} \sum_{Lpq} \frac{|\langle I_e| \hat{M}_{pq}|I_g\rangle|^2}{|q|} \times \left( G_1^{(Lpq)} + G_2^{(Lpq)} + G_1^{(Lpq)} \right),
\]

where the total probability of the process would be the sum over all possible \(L\), \(p\) and \(q\) and

\[
G_1^{(Lpq)} = \sum_{J_n} \frac{1}{|J_n|} \left| \sum_{\gamma_n} \langle \gamma_f J_f | [\alpha \cdot \mathbf{a}_L^{(p)}] | \gamma_n J_n \rangle \langle \gamma_n J_n | \hat{T}_q | \gamma_i J_i \rangle \right|^2,
\]

\[
G_2^{(Lpq)} = \sum_{J_n} \frac{1}{|J_n|} \left| \sum_{\gamma_n} \left\langle \gamma_f J_f \right| \hat{T}_q \left| \gamma_n J_n \right\rangle \left\langle \gamma_n J_n \right| [\alpha \cdot \mathbf{a}_L^{(p)}] \left| \gamma_i J_i \right\rangle \right|^2,
\]

\[
G_{12}^{(Lpq)} = 2(-1)^{q+L} \sum_{\gamma_n \gamma_n'} (-1)^{J_n + J_n'} \left\langle J_i q J_f L J_n' \right\rangle \Re \left( \frac{\langle \gamma_f J_f | [\alpha \cdot \mathbf{a}_L^{(p)}] | \gamma_n J_n \rangle \langle \gamma_n J_n | \hat{T}_q | \gamma_i J_i \rangle}{\epsilon_i - \omega_N - \epsilon_n - i\frac{\Gamma_n}{2}} \right) \times \frac{\langle \gamma_f J_f | [\alpha \cdot \mathbf{a}_L^{(p)}] | \gamma_n J_n \rangle \langle \gamma_n J_n | [\alpha \cdot \mathbf{a}_L^{(p)}] | \gamma_i J_i \rangle^*}{\epsilon_f + \omega_N - \epsilon_n' + i\frac{\Gamma_n}{2}}.
\]

The equations above show that the NETP probability for each multipole splits into three parts proportional to the amplitudes \(G_1^{(Lpq)}\). The first two amplitudes \(G_1^{(Lpq)}\) and \(G_2^{(Lpq)}\) correspond here to the cases illustrated in Fig. 1 where the real photon is emitted either due to the transition between the initial and the intermediate state or the intermediate and the final state. The last amplitude \(G_{12}^{(Lpq)}\) covers the interference between these two coherent processes.
B. Resonance Approximation

For our specific case of the probability [5] can be further simplified. In contrast to the very simple electronic structure of helium-like systems, for which NET has been first discussed [17], Th$^{2+}$ has a rich and dense level structure. Therefore it is safe to assume that only the closest resonance will contribute to the NET probability. This allows for the application of the so-called resonance approximation. In this approximation all interference terms vanish, thus, $c_{12}^{pp}$ can be neglected and the terms $G_{1}^{pp}$ and $G_{2}^{pp}$ become:

\[
G_{1}^{(Lpq)} \approx \sum_{\gamma_{n}J_{n}} \frac{1}{|J_{n}|} \times \left| \frac{\langle \gamma_{f}J_{f}||\alpha \cdot a_{L}^{(p)}||\gamma_{n}J_{n}\rangle}{\epsilon_{f} - \omega_{N} - \epsilon_{n} - i\frac{1}{2}} \langle \gamma_{n}J_{n}||\tilde{T}_{q}||\gamma_{i}J_{i}\rangle \right|^{2},
\]

\[
G_{2}^{(Lpq)} \approx \sum_{\gamma_{n}J_{n}} \frac{1}{|J_{n}|} \frac{\Gamma_{i} + \Gamma_{n}}{\Gamma_{n}} \times \left| \frac{\langle \gamma_{f}J_{f}||\tilde{T}_{q}||\gamma_{n}J_{n}\rangle}{\epsilon_{f} + \omega_{N} - \epsilon_{n} - i\frac{1}{2}} \langle \gamma_{n}J_{n}||\alpha \cdot a_{L}^{(p)}||\gamma_{i}J_{i}\rangle \right|^{2},
\]

where we could incorporate the width of the initial state in resonance approximation following Ref. 22.

Now, the remaining task to calculate the NET probability [5] in resonance approximation is the evaluation of the reduced nuclear and electronic matrix elements. The nuclear transition amplitudes $\langle I_{e}||M_{q}||I_{g}\rangle$ are known from elaborate nuclear calculations, e.g., by Minkov and Pálfy 23, where previous estimates by Tkalya et al. 24 have been refined.

C. Enhancement Factor $\beta$

Due to the complexity of nuclear calculations, the nuclear amplitudes provided e.g. in Ref. 23 are a major source of uncertainty in our calculations of the NET probability [5]. To circumvent these uncertainties one can define the enhancement factor $\beta$ (cf. 21) which is independent on the nuclear transition probability:

\[
\beta^{(Lpq)} = \frac{\mathcal{M}_{q}^{(Lpq)}}{\Gamma_{q}},
\]

where the nuclear decay width $\Gamma_{q}$ is defined by:

\[
\Gamma_{q} = \frac{8\pi(q + 1)}{q(2q + 1)!} \left( \frac{\omega_{N}}{I_{q}} \right)^{2q + 1} \langle I_{e}||M_{q}||I_{g}\rangle^{2}.
\]

The enhancement factor [8] is defined in analogy to Refs. 20, 21 and given here mainly to make a connection to these works and to test our theory with respect to effects coming from the electronic structure of Th$^{2+}$.

Specifically for the case of $^{229}$Th, the leading multipole of the nuclear transition are $M1$ and $E2$, so that $q$ is either 1 or 2. From now on we will assume that all radiative electronic transitions are of $E1$ type, so that $L = 1$ and $p = 1$. Therefore, in resonance approximation, the enhancement factors of interest are

\[
\beta^{(111)} = 3\omega_{N} \frac{1}{2\omega_{N}^{2}} \left( G_{1}^{(111)} + G_{2}^{(111)} \right),
\]

\[
\beta^{(112)} = 30\omega_{N} \frac{1}{\alpha^{2}\omega_{N}^{2}} \left( G_{1}^{(112)} + G_{2}^{(112)} \right).
\]

IV. NUMERICAL DETAILS

Up to now we have shown how the NET process may be discussed by taking the nuclear transition amplitude from the literature or by investigating the enhancement factor instead. Now we will briefly sketch the evaluation of the electronic matrix elements. To calculate these, we apply a combination of configuration interaction (CI) and many-body perturbation theory (MBPT), that has been described in detail in Refs. 25–27. In particular we used the package assembled by Kozlov et al. 28. The CI+MBPT method is a powerful method to calculate reliable transition matrix elements. Level energies, however, especially for complicated systems like Th$^{2+}$ are determined more accurately in experiments. Because the exact position of the resonances is important to determine the NET probability accurately, we take the experimental values for all level energies instead of the theoretical ones. We will discuss the importance of this step in the section below.

V. RESULTS AND DISCUSSION

Before we discuss the the probability of the NET process in Th$^{2+}$, we will have a brief look on the enhancement factor $\beta^{111}$ [cf. Eq. (5)]. In particular we want to investigate how the replacement of the calculated level energies by the experimental ones influences the results. Therefore we performed calculations shown in for $\beta^{(111)}$ as a function of the nuclear excitation energy $\omega_{N}$ using both. The results of these calculations are shown in Fig. 2 the theoretical (black solid line) and the experimental (red dashed line) level energies. The first feature we notice in Fig. 2 is the large number of available decay paths to the 6d$^{2}$(J = 2) : 63 cm$^{-1}$ state from each of these upper states. While for $J_{i} = 4$ and a $E1$ radiative transition, the intermediate state must have $J_{f} = 3$, for $J_{i} = 2$ there are three possible $J_{n}$ and, therefore, more intermediate resonances available. But there are two more important things to notice. Foremost we see that the high energy cutoff of $\beta$ is reduced for the case of the experimental
level energies. Therefore we note, that it is very important to take the energy splitting between the initial and final electronic state accurately into account. Moreover we see that the replacement of the energies of the intermediate states to their experimental values does not change the qualitative behaviour of $\beta^{111}$ and, thus, can be safely done to achieve accurate results.

The primary aim of this paper is to provide information about the most promising excitation paths to observe the NETP process in $^{229}$Th$^{2+}$. Therefore we assume according to available experimental setups that the exciting lasers are tunable between 3.45 eV and 5.25 eV \cite{31} (cf. Fig. 1). With such lasers 19 possible upper states $|\gamma_iJ_i\rangle$ can be pumped. This number reduces to 16, if we fix the final state to be the level $6d^{2}(J = 2) : 63$ cm$^{-1}$, in order to be able to cycle through the process multiple times. For each these 16 possible upper states we calculated the NETP probability \cite{32} sumning over $1 \leq q \leq 2$ in order to account for both the $M1$ and $E2$ nuclear transition channels. This step is necessary because it has been shown recently that both, the $M1$ and the $E2$ channel, may contribute equally to the NETP probability \cite{32}. Similar to Fig. 2 we display the NETP probability $W_{fi} = \sum_{q=1}^{2} W_{fi}^{(E1,q)}$ as a function of the nuclear excitation energy $\omega_N$. This data, however, is not very conclusive. Thus it needed to be processed, which is illustrated in Fig. 3. In the upper panel of this figure we display the NETP probability for four upper states as a function of the nuclear excitation energy $\omega_N$. To get our final result we take the envelope of this family of curves as shown in the bottom panel of Fig. 3. Moreover we omit resonance peaks narrower than 0.1 eV for it would make the figure impractical to use, especially at higher $\omega_N$, where the resonances get more dense. Note also that we do not show the NETP probability for those of the 16 possible upper states that do not contribute to the envelope. The vertical lines in Fig. 3 denote the most recent values for the energy of the nuclear isomer state, ranging from 7.1 eV to 7.6 eV and 7.8 eV \cite{24,24,30}. The grey shaded area denotes the combined errorbars of all three measurements and, thus, a recommended initial search area for the nuclear isomer.

With the preparation of the data explained above we are now able to generate the main result of this work. In Fig. 3 we show, which is the ideal upper state to observe the NETP process in $^{229}$Th$^{2+}$ as a function of $\omega_N$. It can be seen that also for the entire energy range between 6.5 eV and 10.5 eV only 5 of the possible 16 upper states need to be considered for a possible experiment. Again the vertical lines and the grey area in Fig. 3 mark the recommended initial search area for the nuclear isomer state. We have to mention that all we find for all these three energies a resonance closely. Except for the highest experimental value of $\omega_N$ these resonances are narrower than 0.1 eV and, thus, are not shown in the figure but might be used for the excitation of the nucleus. For higher energies the resonances get so dense that basically the whole energy range is covered.

Let us finally discuss how the excitation of the nucleus could be monitored in the experiment we propose. Recently the hyperfine structure of the electronic levels in Th$^{2+}$ has proven to be a good indicator of whether the nucleus is in its ground or first excited state \cite{6}. This would be as well possible in the scenario proposed in the present work by either applying an additional laser or observing the fluorescence from one of the pumping stages. Another common option would be to observe the time delayed photoemission from the nuclear decay. This, however, would not be recommended for the scenario proposed here, because we could cycle through the process, no matter if the upper state decayed via NETP or the more likely two-photon cascade. This allows for a good statistics an does not require a shot-by-shot analysis of the data with accurate timing.
FIG. 3. Probability of NETP for different upper states (top panel) and the envelope of these probabilities (bottom panel), where contributions narrower than 0.1 eV are neglected. The colors are distributed as follows: blue dotted line: $5f^8s(J = 2): 74,644$ cm$^{-1}$, green dashed line: $5f^7d(J = 2): 79,916$ cm$^{-1}$, turquoise dash-dotted line: $5f^7d(J = 2): 83,237$ cm$^{-1}$, red solid line: $5f^7d(J = 3): 84,374$ cm$^{-1}$. The black vertical lines show the supposed energies of the low lying isomeric state according to Refs. [24, 29, 30] with the corresponding uncertainty interval shown by the grey-shaded area.

VI. CONCLUDING REMARKS

The NETP process has been shown to be a promising candidate to investigate the nuclear structure of highly charged ions [17]. In the present work this process is discussed for many electron systems within the resonance approximation. To excite the $^{229}$Th nucleus, we propose a combination of a two-step pumping of an upper state from which the NETP process occurs. To overcome the difficulty of a small branching ratio between NETP and a generic radiative two-step decay of the upper state, the proposed process can be cycled independent on the way the ion decays.

A disadvantage of many proposed electronic bridge processes for the excitation of the $^{229}$Th nucleus is the requirement of a continuous scanning with a tunable laser. This disadvantage does not apply in the scenario described in this paper, where the lasers are adjusted only once to ensure the most efficient pumping of the upper state. For a first test of our theory we recom-
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