

GSI Research Proposal

G-22-00052

Experiment title: Follow-Up E142: Laser Excitation of the 229Th Nucleus Using Nuclear Hyperfine Mixing	Proposal type: Standard (ST)
	Scientific College: G-PAC

Abstract:
 The isomer 229mTh with its exceptionally low excitation energy of $E_y=8.28$ eV is a focus of current research. A 229Th-“nuclear clock” is expected to reach 19 digits precision and promises many new applications. 229mTh is also an outstanding candidate for investigations at the interface of atomic and nuclear physics. Here, we propose to investigate a phenomenon that is unique to very highly charged 229Th such as one-electron 229Th89+. In addition to the ordinary hyperfine structure, the strong magnetic of the s-electron mediates a mixing of the $F = 2$ levels of ground state (g.s.) and isomeric state (i.s.). The mixing results in a small energy shift. But more notable, the lifetime of the i.s. decreases by 5-6 orders of magnitude. This accelerated decay implies that the laser excitation is enhanced as well by 10^5 to 10^6 . It is proposed to utilize this hyperfine nuclear mixing to laser-excite the 229Th nucleus and perform precision laser spectroscopy on the isomeric transition at the ESR.

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FAIR Experiment Pillar: APPA	Collaboration: SPARC
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Experiment time requested for		
Target station	Requested experiment time	Link scientist
1.1-E: ESR (Main beam)	54 Shifts	Yury Litvinov

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Follow-Up E142: Laser Excitation of the ^{229}Th Nucleus Using Nuclear Hyperfine Mixing

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Abstract: The “nuclear clock” isomer ^{229m}Th with its exceptionally low excitation energy of $E_\gamma = 8.28$ eV is a focus of current research all over the world. A ^{229}Th -clock based on this nuclear $M1$ transition is expected to reach a more than 19 digits precision and promises many new applications. ^{229m}Th is also an outstanding candidate for investigations at the interface of atomic and nuclear physics. With the present application we propose to investigate a phenomenon that is unique to very highly charged ^{229}Th such as one-electron $^{229}\text{Th}^{89+}$ or three-electron $^{229}\text{Th}^{87+}$. In these thorium charge states, in addition to the ordinary hyperfine structure, the very strong magnetic field of ~ 28 MT (and ~ 3.5 MT, resp.) of the unpaired s -electron mediates a mixing of the $F = 2$ levels of ground state (g.s.) and isomeric state (i.s.). The mixing results in an additional small energy shift. But more notable, the lifetime of the i.s. decreases drastically by 5-6 orders of magnitude, from a few hours down to a few 10 ms. Furthermore, this accelerated decay implies that the excitation probability with a laser is enhanced by these 5-6 orders of magnitude. It is proposed to investigate this hyperfine nuclear mixing using laser spectroscopy at the ESR storage ring. In addition to the measurement of this yet unobserved effect, as a “spin-off” and as important input to the nuclear clock developments we would be (a) the first to excite the 8.28 eV-state with a laser, (b) obtain a significantly improved value for the excitation energy, and (c) measure the γ -lifetime, i.e., the transition strength of the $M1$ -transition between g.s. and i.s.

Requested Shifts:

Preparation/Test beamtime:	12 shifts
Production beamtime (including set-up of ESR):	42 shifts
Requested beam facility:	SIS18, SIS cooler, ESR, ESR cooler
Requested beam:	^{238}U (^{232}Th), $> 2 \cdot 10^9$ ions in SIS18
GSI link scientist:	Yu. A. Litvinov
Collaboration:	SPARC

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The present proposal is the follow-up of experiment E142 (category A) and aims at the investigation of the effect of nuclear hyperfine mixing (NHM) in the case of the ground state (g.s., nuclear spin $I = 5/2^+$) of one-electron $^{229}\text{Th}^{89+}$ and its $I = 3/2^+$ isomeric state (i.s.) using storage-ring laser spectroscopy. Parts of this proposal were already described in an accepted LOI50 from 2011 [1]. In E142 (and in the experiment E128) the foundations were laid for storage-ring laser spectroscopy of artificially in-flight synthesized radioisotopes (RIBs). In E128 the feasibility of such laser experiments with RIBs was nicely demonstrated in a measurement of the hyperfine splitting of 208-Bi. However, in the 229-Th experiment the laser excitation could not be found, yet. Due to rather poor experimental conditions—predominantly from the accelerator side (see details below)—only a small fraction of the allotted beam time (15 – 20%) of the E142 run could be used for the search of the laser transition in $^{229}\text{Th}^{89+}$.

Introduction, motivation, and scientific context

The ‘nuclear clock’ isomer ^{229m}Th :

According to present knowledge, ^{229}Th is the nucleus with the by far lowest nuclear excitation energy of $E_\gamma = 8.28$ eV [2–5] of all nuclei. Since the corresponding $M1$ -transition is in an energy range that can be accessed with present VUV laser systems it is considered as the basis for a nuclear clock [6–12] with a proposed accuracy of 19 decimal places [8]. It is readily imagined that such a precision ‘nuclear clock’ can be exploited in many ways, e.g. for improved length and time standards, more precise GPS signals, but also for very fundamental experiments that question the limits of our present understanding of physics such as a nuclear laser [13] or the time-constance of fundamental constants such as the fine structure constant α or the strong interaction coupling constant Λ_{QCD} [10–12, 14–18] as well as the ability to search for dark matter candidates [10].

Besides the use as a precision clock, ^{229}Th is a unique laboratory at the interface of atomic and nuclear physics. The excited long-lived nuclear level might be selectively populated or depopulated with lasers [3, 6–8, 18, 19], with two photons [20], with X-rays [21] or with electron beams [1]. Uniquely at GSI, investigations on ^{229}Th and its isomer can be performed with highly charged ions which opens up a rich physics portfolio. At GSI, since 2011 an experimental program on ^{229}Th has been envisaged [1] and a dedicated EMMI workshop with many leading scientists from all over the world was conducted in 2012 [22]. Further information and many additional references to ^{229}Th -physics and according experiment proposals can be found in review articles [9–12, 18, 23–25], on the webpages of the European nuClock project [26], the European ThoriumNuclearClock project [27] and in the talks of the EMMI workshop [22].

For a long time, the transition energy was considered to be $E_\gamma = 3.5(1.0)$ eV, but in 2007 it was shifted in a microcalorimeter study to a substantially higher value, first to $E_\gamma = 7.6(5)$ eV [37], later corrected by the same group to $E_\gamma = 7.8(5)$ eV [2]. The presently accepted value from conversion-electron spectroscopy is $E_\gamma = 8.28(17)$ eV [3], confirmed in 2019 by a new microcalorimeter study [4] that obtained $E_\gamma = 8.30(92)$ eV. In 2020, in a new high-resolution microcalorimeter measurement, 4 different approaches to extract the isomer energy yielded $E_\gamma = 7.84(29)$ eV, $E_\gamma = 8.1(13)$ eV, $E_\gamma = 7.8(8)$ eV and $E_\gamma = 8.10(17)$ eV, respectively [5]. The combined analysis of these experimental

E_γ [eV]	8.28(17)	[3]
$\tau(\text{neutral})$ [μs]	7(1)	[28]
$B(M1)$ [W.u.]	0.008	[29]
$\mu_{\text{g.s.}}$ [μ_N]	0.360(7)	[30]
$\mu_{\text{i.s.}}$ [μ_N]	-0.37(6)	[31]

Table I. Nuclear input data of ^{229}Th used for the calculation of NHM in $^{229}\text{Th}^{89+}$ (Fig. 1, [32]). For a discussion about E_γ and $B(M1)$, see text. The γ -transition probability is a theoretical value, not confirmed by experiment. Theoretical values for $B(M1)$ range from 0.005 W.u. to 0.060 W.u. [29, 33–36].

data suggests $E_\gamma = 8.12(11)$ eV. Very recently, in a workshop [38] and in a proposal to the CERN ISOLDE beamtime committee [39] the observation of the γ -decay-photons of the isomer in an experiment at CERN ISOLDE has been reported: The results have not been published, yet, but the first preliminary data promise substantially lower error bars than the preceding experiments with a tendency to again somewhat higher transition energies. In [38, 39] it is also indicated that the γ -lifetime of the isomer is well below 1 h.

As a consequence of the shift from 3.5 eV to more than 8 eV, internal conversion (IC) became a possible decay channel; the binding energy of the last electron is $I_b = 6.31$ eV. In fact, IC is by far the dominant decay channel of the isomer leading to a lifetime of neutral ^{229}Th as short as $7(1)\mu\text{s}$ [28]. The utilization of IC lead to a major breakthrough in the field, the direct evidence of the transition by means of conversion electron spectroscopy [23]. Triggered by this observation, in the past 5 years enormous progress in experiment as well as in theory was achieved, cf. [3, 4, 21, 23, 28, 29, 31, 36, 40–46] and references therein. As described above, in a recent workshop presentation the observation of γ 's from the isomer decay has been reported but the results have not been published in a peer-reviewed journal, yet. The range of theoretical estimates for the nuclear transition probability spans more than an order of magnitude, $B(M1) = 0.005 \dots 0.06$ W.u. [29, 33–36]. A recent publication on this subject [29] gives good arguments for $B(M1) \approx 0.008$ W.u. (lifetime 117 min) whereas reference [39] suggests a lifetime of well below 1 h.

Nuclear hyperfine mixing in one-electron $^{229m}\text{Th}^{89+}$:

One of the most interesting features in highly charged thorium is the mutual interplay of tightly bound inner-shell electrons with the nucleus that due to the strong magnetic field induces a sizeable magnetic hyperfine interaction (HFI). The HFI in, e.g., a H-like (one-electron) $^{229}\text{Th}^{89+}$ leads to a mixing of the $F = 2$ levels of g.s. and i.s. This NHM results in small energy shifts for the mixed states. Most notably the mixing dramatically alters the lifetime of the nuclear decay and decreases it by 5 to 6 orders of magnitude compared to a bare nucleus [33, 42, 47–50]¹. The effect is depicted in Fig. 1: On the left, the γ -lifetime of the bare nucleus is displayed which for $B(M1) = 0.008$ W.u. corresponds to roughly 2 hours. In the middle, the ordinary hyperfine splitting of $^{229}\text{Th}^{89+}$ is shown. In the right part of the figure, in addition the hyperfine mixing is “switched on” which slightly alters the energies of both $F = 2$ states and reduces the lifetimes of transitions between the hyperfine states of *different nuclear levels* to a few ten milliseconds. This sensitivity in turn allows for a determination of the $B(M1)$ value from the lifetime of a mixed transition. In two-electron $^{229(m)}\text{Th}^{88+}$ the effective field of the paired electrons at the site of the nucleus is zero and hence the γ -decay-lifetime is the same as for the bare nucleus. Three-electron $^{229}\text{Th}^{87+}$ again shows strongly accelerated decay times but slower (2.5 s to 4 s) [32] than H-like thorium since the field of the electrons at the nucleus is smaller. In essence, depending on the electronic configuration the lifetime of the nucleus can be manipulated on purpose. We would like to emphasize, that this effect is not mediated by internal conversion which is absent in all ions with charge states $q \geq 3+$.

Previous Experiments and Experimental Background: Results from E142 and E128

For easier readability, please find the details on “Previous experiments and experimental background” behind the sections “Presentation of New Approach / Objectives and Expected Results” and “Experimental Design and Methods”.

Presentation of New Approach / Objectives and Expected Results

In ^{229}Th , the mixing of the $F = 2$ states of the two nuclear levels with $I = 5/2^+$ (g.s.) and $I = 3/2^+$ (i.s.) is induced by the magnetic hyperfine interaction (HFI) from the strong magnetic field of an

¹ Please note, the influence of the HFI on nuclear transitions is established in muonic atoms, cf. [51, 52]. For muonic ^{229}Th the effect is calculated in [33, 41].

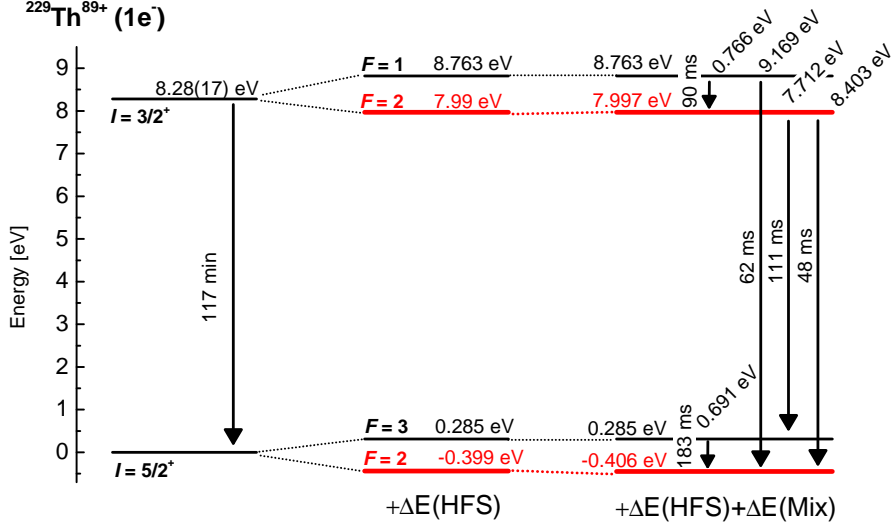


Figure 1. Nuclear mixing of $F = 2$ states of ground state ($I^\pi = 5/2^+$) and isomeric state ($I^\pi = 3/2^+$) in one-electron $^{229}\text{Th}^{89+}$. (Left) Undisturbed isomeric transition in ^{229}Th , e.g. in bare $^{229}\text{Th}^{90+}$. (Center) Ordinary hyperfine splitting in one-electron $^{229}\text{Th}^{89+}$ without nuclear level mixing. (Right) Hyperfine splitting and nuclear mixing effect in one-electron $^{229}\text{Th}^{89+}$. Under the influence of NHM, the isomer decays 5-6 orders of magnitude faster than without. The displayed values are from [32]^a using a similar approach as given in [33, 49, 50, 53]. Data from Table I were used as input parameters. For the sake of simplicity, the additional small energy shift of 40 meV due to different nuclear charge radii of g.s. and i.s. of $\langle r_{229m}^2 \rangle - \langle r_{229}^2 \rangle = 0.012 \text{ fm}^2$ [31] is omitted in this figure.

^a Work performed and published before 2022/02/24

unpaired s -electron in highly charged ^{229}Th ions. Such unpaired electrons can be found, e.g., in one-electron $^{229}\text{Th}^{89+}$. Few-electron ^{229}Th ions are unique candidates for the investigation of this mechanism mainly because of three reasons: (1) The energy splitting of g.s. and i.s. of $E_\gamma = 8.28 \text{ eV}$ is exceptionally low, (2) according to the nuclear spins the isomeric transition is of type $M1$, and (3) due to the large nuclear charge $Z = 90$, the unpaired $1s$ -electron of H-like ^{229}Th produces a very strong magnetic field of $\sim 28 \text{ MT}$ at the site of the nucleus. The combination of these features leads to a sizable mixing of the nuclear states [32, 33, 41, 42, 47–50]. A corresponding mixing effect, also often termed “hyperfine quenching” is well known in the atomic shell where its most notable manifestation is a strong decrease of the lifetimes of atomic metastable states [54–59]. Likewise, for the present case of “nuclear hyperfine quenching” in one-electron $^{229}\text{Th}^{89+}$ the lifetime of the nucleus decreases vastly by 5 to 6 orders of magnitude from about 2 hours to a few ten milliseconds, depending on the actual transition (Fig. 1). As an additional benefit, the shorter lifetime caused by this “nuclear level quenching” also facilitates *excitation* of the nucleus. The faster transitions make a laser excitation 5-6 orders of magnitude more effective than with low-charged ions. At the same time the number of fluorescence photons is enhanced by this value resulting in a total gain of 10 to 12 orders of magnitude. Here, we propose to utilize this NHM-effect and perform laser excitation studies of one-electron $^{229}\text{Th}^{89+}$.

The primary goal of the proposed experiment is to measure the effect of nuclear hyperfine mixing in $^{229}\text{Th}^{89+}$ using laser spectroscopy at the ESR. At present, GSI/FAIR is the only facility world-wide that allows this experiment to be conducted.

An experimental laser spectroscopy set-up is available at the ESR which was successfully used under similar experimental conditions for hyperfine splitting measurements of $^{209}\text{Bi}^{82+}$ and $^{209}\text{Bi}^{80+}$

[60, 61] and very recently, using an updated arrangement with optimizations towards lower photon wavelengths (Figs. 2 and 3), also for a first experiment on ^{229}Th (experiment E142) and with the RIB bismuth isotope $A=208$ (experiment E128, see below).

For the measurement we plan to investigate all three transitions that start from the $^{229}\text{Th}^{89+}$ ($F = 2; I^\pi = 5/2^+$) g.s. This allows us to not only verify the NHM but also to disentangle the individual contributions, to determine the $B(M1)$ value and to obtain substantially improved information on the nuclear parameters of ^{229}Th 's g.s. and i.s. Depending on the particular transition induced by the laser, a cascade of low-energy photons with varying relative intensities is emitted (Fig. 1). We have used $B(M1) = 0.008$ W.u. for the calculations of the NHM-effect as given in Fig. 1. It is noted here that the ‘‘quenched’’ NHM-lifetimes depend almost linearly on $1/B(M1)$ [32]. Hence, a larger $B(M1)$ as suggested by the CERN ISOLDE experiment [38, 39] would also lead to a shorter lifetime of the hyperfine-mixed states and consequently would also enhance laser excitation probability and fluorescence yield.

Highly charged ^{229}Th ions will be preferably produced in-flight from a ^{232}Th or alternatively with reduced performance from a ^{238}U primary beam and will be separated using solely the ESR. This approach that bypasses the FRS has been successfully developed and used for several storage ring experiments of high-intensity beams of artificially synthesized isotopes and also isomers, see [62–65] and below. In extension of the original proposal E142, a further gain in secondary beam intensity in the ESR will be achieved by stochastic-cooling-assisted accumulation of the $^{229}\text{Th}^{89+}$ reaction products.

Beyond the primary goal to probe the NHM-effect several other outstanding physical and technical aspects will be addressed:

- (1) The experiment will be the first to excite the ^{229}Th nucleus with a laser. The detection scheme directly registers the hyperfine mixed 8.28-eV- γ -fluorescence photon (in neutral ^{229}Th the primary decay channel is internal conversion). In doing so, we will be able to (a) determine the hyperfine mixed transition probability and, using this value, derive the unaltered nuclear transition probability, i.e., the important γ -lifetime, (b) give up to 3 orders of magnitude more precise values for E_γ and for the hyperfine splittings of g.s. and i.s.
- (2) The experiment will provide the basis for a future research program with ^{229}Th at the ESR but also at the attached ‘‘precision facilities’’ CRYRING and HITRAP.
- (3) The in-ring isotope separation technique, and the accumulation of the so-produced isotopes in the storage ring and according experimental procedures are also the basis for many other present and future experiments at the ESR/CRYRING/HITRAP complex, e.g. dielectronic-recombination-assisted laser spectroscopy of ^{208}Bi or as outlined in the CRYRING physics book [66].

Experimental Design and Methods

Laser set-up and fluorescence detection:

For the laser excitation of NHM transitions in $^{229}\text{Th}^{89+}$ according to the presently established nuclear values, laser energies of more than 8.4 eV and more than 9.2 eV (Fig. 1) are needed. Such energies allow us to laser-excite $^{229}\text{Th}^{89+}$ from the hyperfine and nuclear ground state ($F = 2$, g.s.) to the lower ($F = 2$) hyperfine level of the i.s. or to the uppermost ($F = 1$) level of the i.s., respectively. At the ESR a pulsed (30 Hz) laser system with pulse lengths of 7 ns and pulse energies of up to 100 mJ (in the visible range) is available that is used to pump a dye laser system in order to produce second-harmonic pulses at a wavelength of $\lambda_{\text{Laser}} = 314$ nm (3.95 eV) (Fig. 2). The fast ions at the ESR moving at up to about 72.5% of the speed of light in combination with a counter-propagating laser beam experience a Doppler shift of more than a factor of 2.5 and

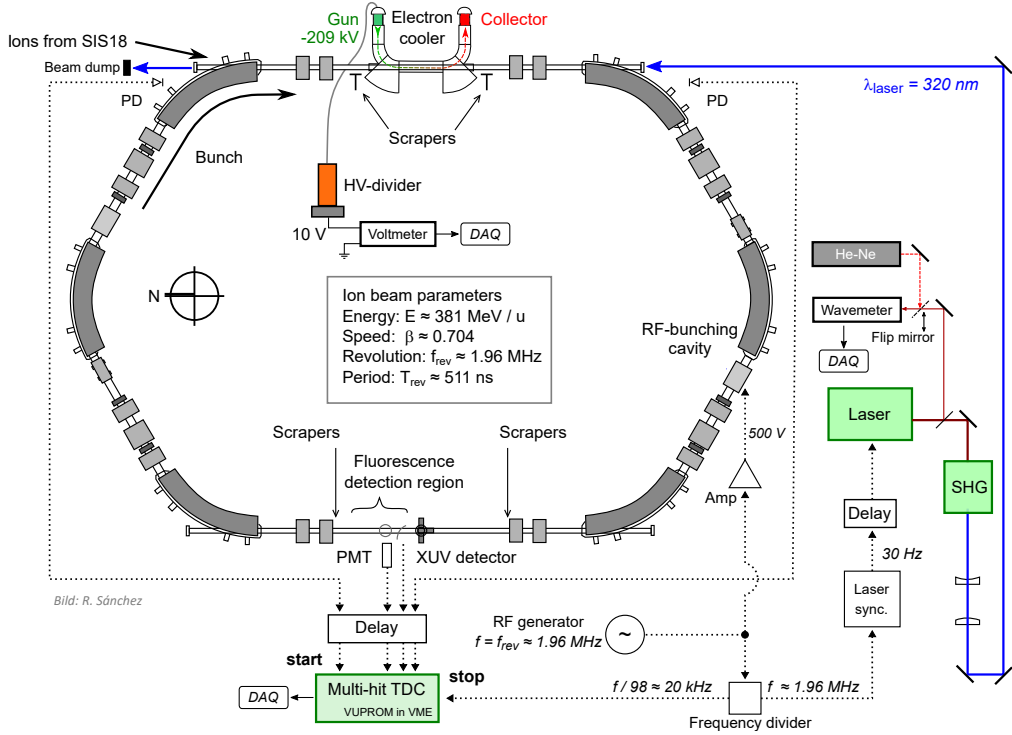


Figure 2. Laser set-up at the ESR storage ring as used in the E142 ^{229}Th run. The laser system and optics operate at a wavelength of $\lambda_{\text{Laser}} = 320 \text{ nm}$ in the laboratory frame. With a Doppler boost of up to a factor of 2.5 ($E_{\text{ion}} = 420 \text{ MeV/u}$; $U_{\text{Cooler}} = 230 \text{ kV}$) in the cm-frame energies close to 10 eV can be accessed. Options to go even up to 11 eV are available but at a lower intensity. A similar set-up has been successfully and routinely operated in many beamtimes on hyperfine spectroscopy of highly charged ions (e.g. [60, 61, 67]) and recently also with RIBs, cf. Fig. 4.

according photon energies in the center-of-momentum(cm)-frame of 10 eV. If it turns out that even higher photon energies would be needed switching to a different laser dye that yields a wavelength of 283 nm (4.38 eV) is also possible. The photon energy in the cm-frame is scanned with the laser in finer steps or by variation of the ion energy. Electron and ion energy can be determined very precisely using a high-voltage divider operated by the Physikalisch Technische Bundesanstalt (PTB) [69] in combination with Schottky spectroscopy [60, 67, 70–73]. For spectroscopy of the lower hyperfine transition the photon energy of 0.691 eV can be achieved with the same laser but ion and laser beam co-propagating. Details can be found in [60, 61, 67].

Also the available fluorescence detection system (Fig. 3) with three separate detectors optimized for different energy ranges is ideally suited for the proposed experiment. The option to register the low-energy hyperfine photons ($\sim 0.7 - 0.8 \text{ eV}$) independently from photons that involve a nuclear transition (about 7.7 eV to 9.2 eV) is a valuable asset for the experiment.

The aluminium fluorescence mirror system was completely redesigned and the new mirrors—in a collaboration with the Fraunhofer IOF Jena— were coated with MgF_2 to allow for registration of short-wavelength photons down to about 110 nm. In addition, the UHV viewports were exchanged against MgF_2 viewports, and small-band solar-blind PMTs optimized for 110 nm - 180 nm were installed. The PMTs were aerated with N_2 in order to avoid the absorption of the VUV photons in air/oxygen.

In the scope of experiment E142 and of experiment E128, that was performed subsequent to the first run with ^{229}Th , we could demonstrate that the new fluorescence detection section performs as devised and allowed for the first successful storage ring laser spectroscopy experiment (E128) with an artificially in-flight synthesized radioisotope, namely $^{208}\text{Bi}^{82+}$ (Fig. 4), see below.

The whole laser set-up is well established and has produced many high-quality publications in the

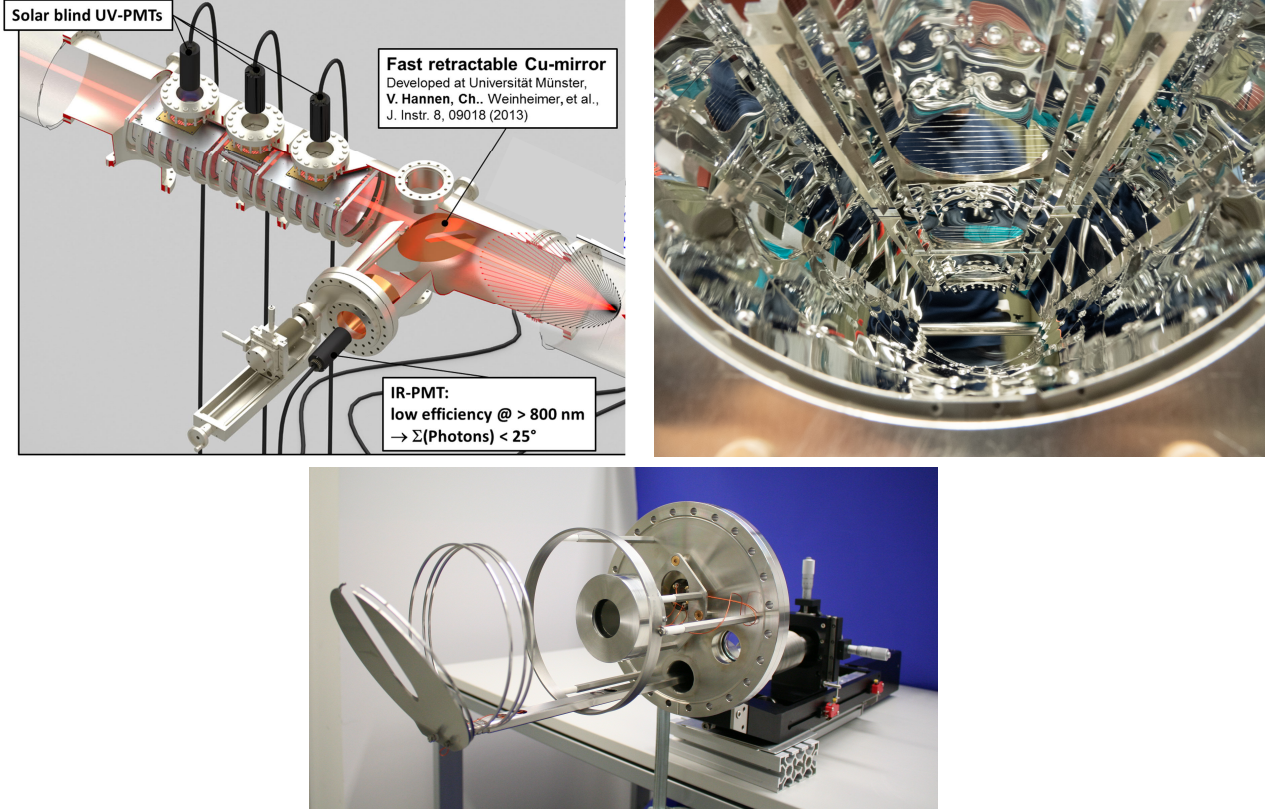


Figure 3. (Top Left) Fluorescence detection section at the ESR storage ring as it was used for the bismuth hyperfine laser spectroscopy [60]. The fluorescence detection section consists of two parts: one system with a copper mirror for lower photon energies around 1 eV, and a second one with an aluminium mirror system and solar-blind photomultipliers for UV to VUV photons. For the ^{229}Th -experiment the mirror system has been re-designed and coated with MgF_2 to be able to detect photons down to ~ 110 nm (top right, view into the beam pipe with mirror system). (Bottom) A third detection system that consists of a CsI-plated slotted disk with secondary electron detection for higher photon energies (XUV, soft x-rays) has recently been successfully commissioned [68]. The combined use allows us to simultaneously detect the low-energy photons of the hyperfine transitions and the higher-energy i.s. \rightarrow g.s. photons, thus significantly enhancing the detection efficiency and information on the decay cascade.

field of laser spectroscopy of hyperfine transitions in very heavy ions such as H-like $^{209}\text{Bi}^{82+}$ and Li-like $^{209}\text{Bi}^{80+}$. These measurements covered the transition energies [60] as well as the lifetimes of the HF states [61]. The energy accuracy that is obtained in these experiments is of the order of 0.1 meV which is more than 3 orders of magnitude smaller than the presently smallest error bar for the isomeric transition. In the present case of NHM in $^{229}\text{Th}^{89+}$ the transition energies stem from different effects (nuclear transition, HF splitting, NHM) that need to be measured independently and need to be disentangled. Therefore, we plan to measure all three excitations that are possible from the g.s. (Fig. 1), i.e., $F = 2(\text{g.s.}) \rightarrow F = 3(\text{g.s.})$; $F = 2(\text{g.s.}) \rightarrow F = 2(\text{i.s.})$ and $F = 2(\text{g.s.}) \rightarrow F = 1(\text{i.s.})$.

In-flight production and separation of $^{229}\text{Th}^{89+}$ using the ESR:

Almost all the realized and planned experimental scenarios are based on neutral or low-charged ^{229}Th . Uniquely at GSI, investigations on ^{229}Th and its isomer can be performed with in-flight synthesized fast beams of highly charged ions which opens up a rich physics portfolio [1, 32, 49, 50]. The in-flight production and separation which leads to a single isotope, single charge state, phase-space cooled fast ion beam has the additional benefit of very clean experimental conditions. The basis of the proposed ESR measurement is the efficient production and separation of ^{229}Th . This

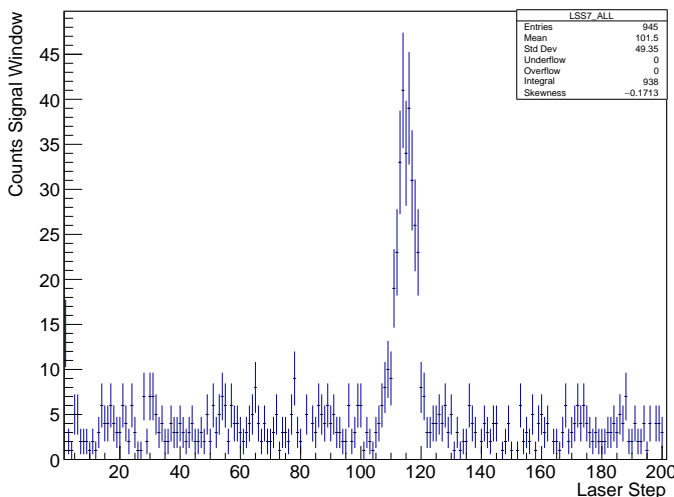


Figure 4. Result from the 2022 laser beamtime E128: Laser resonance of the hyperfine transition in the artificially synthesized (“secondary”) H-like $^{208}\text{Bi}^{82+}$. The spectrum comprises 2 – 3 hours of data taking with $\sim 1 - 2 \cdot 10^5$ separated $^{208}\text{Bi}^{82+}$ ions per injection. The ions were produced from a ^{209}Bi primary beam in the SIS and separated in the ESR. The experimental setup and measurement scheme were the same as for the ^{229}Th experiment.

can be accomplished with a 1 cm thick (1.85 g/cm^2) Be-stripper target in the transfer beamline between SIS18 and ESR for production and the ESR utilized as an isotope separator. For the present experiment the $^{229}\text{Th}^{89+}$ will be produced from a ^{238}U or a ^{232}Th primary beam. Using this scenario, the thorium experiments can run in *in parallel* to FRS operation. Our very efficient approach for production and separation of long-lived artificial isotopes has been successfully demonstrated for several in-flight synthesized isotopes [62–64], and even for isomer production [65]. In 2022, in experiments E142 and E128 (cf. next section) this isotope-separation technique using the ESR could be successfully recommissioned for $^{229}\text{Th}^{89+}$ (from ^{238}U primary beam) and for $^{208}\text{Bi}^{82+}$ (from ^{209}Bi), respectively.

Previous Experiments and Experimental Background: Results from E142 and E128

In 2022, a first experimental run (E142) was conducted with the primary goals to (a) re-establish the isotope separation technique, (b) to lay the foundations for storage-ring laser spectroscopy with radioisotopes, and, to (c) search for the NHM laser transition(s) in $^{229}\text{Th}^{89+}$. These goals could only be partially achieved, mainly due to severe restrictions from the accelerator side, namely, availability of beam, interference with parallel experiments, issues with the new FAIR control system, with accelerator integrated devices such as scrapers and pneumatic detector drives and moderate vacuum conditions (factor of 2 - 3 worse than usually). In addition, one of our new (initially tested and accepted by the GSI UHV group) MgF_2 UHV-viewports got leaky during the final vacuum baking of the fluorescence section, had to be sealed with “Vacseal” and could therefore not be used in the experiment. The initial and time-consuming (1 to 2 days) set-up of the storage ring, first with primary beam, then with secondary beam, isotope identification, separation cycle, subsequent optimizations and laser ion-beam alignments (temporal and spatial) had to be redone after a major failure of the SIS18, after which the beam properties *from* the SIS had changed considerably. In essence, we had “beam on target” only about 30 – 35 % of the time, and thus, together with the subpar experimental conditions, we could only cover $\sim 10\%$ of the planned energy search range. Disregarding these set-backs from the accelerator side we could substantially progress with our experimental program and could fulfill goals (a) and (b) but, in the limited scanning time, the laser resonances could not be found. The main achievements are summarized in the following list:

- The production and separation of $^{229}\text{Th}^{89+}$ from a ^{238}U was established. The secondary-beam intensity that was finally available for the laser experiment was optimized from initially a few 10^3 to more than $2 \cdot 10^4$ ions. In the subsequent E128 experiment with ^{208}Bi further improvements

were achieved that, applied to $^{229}\text{Th}^{89+}$, would translate to another factor of 2 to 3. For the present proposal, based on these first results and discussions with specialists from the ESR group (M. Steck et al.) an accumulation scheme using stochastic cooling in the ESR is envisaged to further increase the number of $^{229}\text{Th}^{89+}$ ions by one order of magnitude.

- A successful commissioning of the new redesigned experimental set-up was performed:
 - * For the MgF_2 -coated fluorescence detection mirror system (Fig. 3) we could verify the expected performance at low wavelengths (due to unfortunate strong ion-bunch related photon signals from ion-residual-gas collisions, i.e., H Ly- α photons at 121.6 nm).
 - * Reliable 30 Hz laser operation at around 320 nm (Doppler-boosted photon energies of up to 9.3 eV).
 - * New "background-free" low-beam-intensity bunch-length measurements using recombination/electron capture in the electron cooler and subsequent registration of the recombined ions in fast plastic scintillation counters with a detection efficiency of 100% and virtually no background counts.
 - * From experiment detailing (signal/background), improvements of the DAQ and of the 1st level analysis we could achieve a roughly 10-fold sensitivity enhancement. Here, in particular, the ratio of the very short ion bunches ($\sim 5 - 10$ ns) of the low intensity ion beam compared to the revolution time of the ions in the ESR of about 500 ns allowed for refined timing, easier background subtraction and a much better signal-to-background ratio.
 - * Flat background due to optimized ion beam lifetime enabled $h = 1$ measurement, i.e., with a single bunch as opposed to the previously used $h = 2$ scheme with one signal and one background bunch. As a consequence, twice as many ions could be excited with the laser.
 - * Separate from the moderate vacuum conditions, we could establish a very low experimental background (cf. Fig. 4).
- It is noted here, that for these initial optimizations of intensity, alignment and background, as well as the ideal adaptation of the setup to the low beam intensities, in the original E142 proposal and already in a granted but not scheduled precursor experiment (E139, category A, not scheduled in 2020 due to Corona) 9-12 shifts (3-4 days) were foreseen.
- The potential of the new set-up and the general feasibility of the approach could be clearly demonstrated in experiment E128 which was performed subsequent to the E142 run. E128 used the same experimental setup and measurement procedures developed and optimized in the E142 run. In E128, for the first time in an ESR experiment, the laser excitation of a secondary ion, here $^{208}\text{Bi}^{82+}$, could be measured (Fig. 4). In contrast to ^{229m}Th , the ^{208}Bi benefitted from a well known nuclear magnetic moment and a correspondingly good initial prediction for the hyperfine splitting / resonance energy, and hence a considerably smaller energy search band that had to be scanned.

Scope of the experiment and justification of beamtime request

The most important aspect for the present experiment but also for other experiments at the ESR/CRYRING/HITRAP complex is the production and subsequent separation of intense beams of in-flight produced artificially synthesized isotopes, in our case of highly charged ^{229}Th .

In E142 and E128 (with ^{208}Bi) we could re-establish the isotope production and separation scheme using the ESR, and we expect an intensity of $3 - 4 \cdot 10^4$ $^{229}\text{Th}^{89+}$ per 10^9 ^{238}U primary beam ions in SIS18 for the present experiment. This is roughly in accordance with simulations using the tool LISE++ [74] that predicts $7 \cdot 10^4 - 8 \cdot 10^4$ $^{229}\text{Th}^{89+}$ ions per 10^9 SIS ions and taking into account

that LISE++ underestimates the initial width of the momentum distribution due to the substantial recoil in the nuclear production reaction.

Based on these initial achievements together with the ESR storage-ring team headed by M. Steck, a future intensity upgrade path was devised: We plan to additionally increase the number of $^{229}\text{Th}^{89+}$ ions employing an accumulation scheme in the ESR assisted by stochastic cooling. Such stochastic-cooling-aided accumulation has been done in the scope of experiment E127 at the ESR, but is not part of the routine operation. We therefore explicitly ask for an extended 4 days (12 shifts) initial beam preparation and test beam time together with the ESR accelerator crew. The outcome of this joint machine and experiment shifts will also be of great benefit for other experiments at the ESR/HITRAP/CRYING facilities that plan to use RIBs.

With the expected number of $^{229}\text{Th}^{89+}$ ions (few 10^5) produced by using a ^{238}U primary beam, the present experiment will be feasible, yet, we ask to investigate if a ^{232}Th primary beam can be made available for the production run since we expect an additional gain of about an order of magnitude.

Based on the long-time experience resulting from the $^{209}\text{Bi}^{82+}$ and $^{209}\text{Bi}^{80+}$ laser spectroscopy experiments at the ESR and in particular from the recent first ESR laser experiment with the $^{208}\text{Bi}^{82+}$ radioisotope beam (Fig. 4), we estimate that with few 10^5 $^{229}\text{Th}^{89+}$ ions we need a time of about 3 - 4 minutes per laser scanning point. The bandwidth of the frequency-doubled laser can be adjusted in the range of $\sqrt{2}(2\text{ GHz}-6\text{ GHz})$, whereof for the broad-band energy scan in E142 the 6-GHz setting was used.

For the main experiment we ask for a total of 14 days (42 shifts) of beamtime to evidence the nuclear hyperfine mixing (NHM) effect. This number assumes that the $^{229}\text{Th}^{89+}$ secondary beam is already stored in the ESR, ready for set-up of the experiment. For the initial set-up of the experiment, i.e., laser set-up and alignment (spatial alignment and temporal alignment of laser pulse and ion bunch), initial determination of the ion energies and space charge potentials of the cooler (which sensitively enters the ion energy) we foresee 6 shifts. For the data taking we request 12 days (36 shifts). The 12 days are justified given the boundary condition that the energy range to be scanned with the laser system is rather broad. The required search interval is dominated by the uncertainty of the nuclear excitation energy and by the uncertainty in the nuclear magnetic moment of the isomer. The four relevant existing values for the undisturbed nuclear transition energy of $E_\gamma = 7.8(5)$ eV [2], $E_\gamma = 8.28(17)$ eV [3], $E_\gamma = 8.30(92)$ eV [4] and $E_\gamma = 8.10(17)$ eV [5], all error bars 1σ , suggest an energy range that needs to be covered by laser scanning of about 500 meV.

Yet, it is assumed that by the time the experiment will be carried out, first results with smaller error bars from the ISOLDE decay measurement [38] will be available and hence will narrow down the uncertainty due the transition energy to less than ± 100 meV.

In $^{229}\text{Th}^{89+}$ an additional energy uncertainty of ~ 65 meV arises for the 8.4 eV-transition and of ~ 130 meV for the 9.2 eV-excitation, respectively, arises from the 16% error of the nuclear magnetic moments μ_n of the i.s. that enters the HF splitting (cf. Fig. 1). The uncertainties of ± 100 meV from the excitation energy and of ± 65 meV from the HFS, both 1σ error bars, suggest an approximate scan range of $\pm(100 - 150)$ meV. For the scan we plan a stepwidth of 0.1 meV. With 3000 steps with a width of 0.1 meV cover a range of 300 meV. Assuming 5 mins per laser step this yields a total laser scanning time of about 10.4 days. This time already includes the duty cycle caused by beam preparation time of 5 to 7 min compared with the pure measurement time of 30 to 45 min. For maintenance (e.g. change of laser dye, change of ion source) and accelerator interruption due to set-up of parallel experiments we include an overhead of 1.5 days in our beamtime request. Thus, the requested 12 days of production run would allow us to cover an energy range of about 250-300 meV.

It is noted, that after the initial set-up, the experiment requires an injection into the ESR and thus a SIS pulse typically only every 30-45 min and is thus ideally suited for parallel operation with other

experiments.

Our beamtime request is summarized in the following table:

Task	Shifts
Preparation/test beamtime:	
ESR stochastic cooling accumulation and ^{229}Th isotope separation in the ESR, intensity optimizations for ^{229}Th experiment	12
Production run:	
(including 6 shifts for initial set-up of the experiment at the ESR for spatial laser alignment, temporal laser-pulse – ion-bunch alignment and precise energy calibration of the cooler using Schottky methods)	42
Total (setup + production run)	12 + 42 shifts

3 Publications of Spokesperson

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3. V. M. Shabaev, D. A. Glazov, A. M. Ryzhkov, C. Brandau, et al., Phys. Rev. Lett. **128**, 043001 (2022).

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Technical requirements ESR

G-22-00052-1.1-E

Target station ESR	Requested experiment time: 54 Shifts	Link scientist: Yury Litvinov
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Mode of operation: Main beam

Comments, e.g. on n° of runs: 12 shifts stochastic-cooling assisted accumulation of 229Th in ESR (joint accelerator and experiment), 42 shifts experiment.

N° of days for set-up and disassembling w/o beam (if > 2 days) [days]

Ion Beam Specifications (for parasitic mode please enter 'none' or '0' in the obligatory fields):

Ion Species and Isotope (e.g. 197-Au) 232-Th (preferred) / 238-U in SIS

Enriched? Yes No

Charge State (e.g. 67) []

Energy (e.g. 1250 MeV/u) 400 [MeV/u]

Intensity [particle nA, ions/s] e.g. 1e11 ions/s 2e9 per spill

Pulse Duration 0 [ns]

Duty Cycle (e.g. 5 Hz) [Hz]

On SIS18 slow extraction fast extraction

Extraction time needed? (e.g. 10 s) [s]

Special requests on beam properties SIS cooler, ESR cooler, ESR stochastic cooling, thick Be-stripper in SIS-ESR transfer beamline

Additional information

Use of ... an existing setup a new setup

Detector(s) used in experiment ESR laser spectroscopy, ESR laser fluorescence setup

1. General Safety

Do you use combustable or hazardous gases within your experiment (e.g. gas target, gas detectors) ?

Yes No

If yes, what sort of gases?

Which quantities or flow rates?

(A flow scheme and description of the safety concepts have to be submitted to the Safety Engineers at GSI)

Upload safety concept

Do you use any other dangerous (e.g. toxic, inflammable, biologically hazardous, etc.) materials / chemicals within your experiment?

Yes No

(Note: Only biological material of biological safety level 1 must be irradiated at GSI)

If yes, what sort of materials/chemicals?

Which quantities?

Is your vacuum setup equipped with fragile parts like thin glass or foil windows, etc. (danger of implosion)?

Yes No

Is it intended to move heavy parts for setting up your equipment or during the experiment?

Yes No

If yes, brief description of the equipment and working procedure:

2. Radiation Safety

Do you use radioactive sources or materials onsite?

Yes No

If yes, which isotopes/type?

Which activities [Bq]?

Do you use a target?

Yes No

If yes, position:

Indicate thickness of target [mm] or [g/cm²], and Interaction probability [%] with primary beam:

Material:

Do you use a secondary target/degrader?

Yes No

If yes, position: SIS18-ESR transfer beamline

Indicate thickness of target [mm] or [g/cm²]/ and Interaction probability [%] with primary/secondary beam:

thick Be stripper 1850mg/cm² (as installed in the stripper section)

Material: Be

Do you use a beam stop for primary/secondary beam?

Yes No

If yes, position:

3. Electrical / Laser Safety

Do you use electrical instruments that you bring on site?

Yes No

If yes, please describe devices above 1kV, self-made equipment etc. Photomultipliers, Multi Channel Plates, HV Amplifiers

Do you use high-intensity radio frequency (rf) sources onsite?

Yes No

If yes, frequency region/power:

Brief description of the rf sources:

Do you use lasers in your equipment?

Yes No

If yes, laser-type(s): Pulsed Nd-YAG, Dye, SHG (second harmonic generation)

Max. power/energy: 1.4 J@1064 nm, 800 mJ@532 nm, 150 mJ@650 nm, 25 mJ@325 nm

Class: 4

Repetition rate: 30 Hz

4. Special Safety

Is there any other special safety aspect to be considered in connection with your proposal?

Yes No

If yes, brief description:

The timely knowledge on requirements of host lab resources by our users permits a solid in-house planning and allocation of respective resources. Please indicate here roughly, what you will need, and discuss details with the respective department later, if beamtime is granted. You might discuss your entries here with your link scientist before submission of your proposal.

Target Laboratory

Do you need targets from the department Target Laboratory?

Yes No

If yes, please specify targets:

Detector Laboratory

Do you need support from the Detector Laboratory?

Yes No

If yes, please specify:

Experiment Electronics

Do you need support from the Experiment Electronics department?

Yes No

If yes, please specify:

IT Department

Do you need resources from the IT department?

Yes No

Needed data storage:

Computing requirements:

Indicate further requirements here:

Vacuum Systems

Do you need support from the department Vacuum Systems?

Yes No

If yes, please specify:

Transport and Installation

Do you need support from the department Transport & Installation for transporting or installing heavy equipment? (formerly "Großraummontage")

Yes No

If yes, please specify:

Mechanical Workshop

Do you need resources from the department Mechanical Workshop?

Yes No

If yes, please specify:

Other Host Departments

Do you need resources from other host departments?

Yes No

If yes, please specify: