Proposal for an experiment to be conducted at FRS/ESR

Measurement of the bound-state beta decay of bare ^{205}Tl ions

Updated from previously accepted proposal E100

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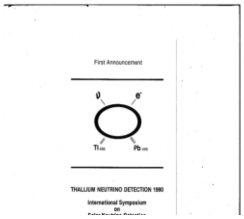
First Announcement

THALLIUM NEUTRINO DETECTION 1990

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1. Prehistory of this proposal (E019, LOI46, E100)

In 1992, immediately after the very first observation of bound-state beta decay (β_b decay) with the use of bare $^{163}Dy^{66+}$ ions stored in the ESR [1], a proposal aiming to measure the half-life of bare $^{205}Tl^{81+}$ ions stored in the ESR was submitted and approved (**E019**).

Like ^{163}Dy, the neutral ^{205}Tl atom is stable in the neutral atomic charge state, but shall decay by β_b decay if all or most of its electrons are stripped-off. Though our proposal has been accepted with "first priority", we were *not* allowed using the stable ^{205}Tl (70% natural abundance) in any of the GSI ion sources for safety reasons (Tl vapour is poisonous), or we had to invest some 700 000 DM for special pumps, glove boxes etc. Hence, at that time we were forced to cancel or at least to shift this experiment for an unknown time span.

In 2008, the proposal was re-evaluated. The physics case was confirmed to be excellent. However, due to the unavailable ^{205}Tl primary beams, the proposal was removed from the backlog and renamed to "Letter of Intent" (**LOI 46**)

In 2010, the proposal was resubmitted. Owing to the continuous development of GSI accelerator facilities over the last two decades, it became feasible to produce sufficient amount of fully ionized ^{205}Tl ions at the *fragment separator* by in-flight fragmentation of a primary ^{206}Pb beam and to transport it into the ESR. The β_b half-life is estimated to be 120 days, with a large error margin [2]. A half-life in the order of 1 year or even longer cannot be excluded [3]. Therefore, at least 10^6 bare ^{205}Tl ions stored in the ESR are needed to get a reliable decay statistics. The intensity of Pb beams, as available in 2010, of the order of $5\text{-}8\cdot 10^8$ particles/spill should be sufficient to successfully conduct the experiment. The physics case was again confirmed to remain excellent. The proposal was approved with the category "A" (**E100**).

In 2015, the proposal to measure half-life of bare $^{205}Tl^{81+}$ ions was evaluated by the European Research Council within the ERC Consolidator Grant application ASTRUm. The physics case was found to be outstanding and the funding was approved.

All preparatory works for running the E100 experiment have been accomplished and documented in PhD Thesis of B. S. Gao [4]. Dedicated cathodes made of enriched ^{206}Pb for the VARIS ion source were purchased and manufactured.

The experiment was considered several times in the preliminary schedules at GSI. However, due to very limited beam time in the past, the experiment could not be run.

2. Physics impact of bound-state beta decay of bare $^{205}Tl^{81+}$

2.1. The flux of solar pp neutrinos

The capture of solar pp-neutrinos ($0 \leq E_\nu \leq 420$ keV) transforms the nucleus ^{205}Tl into ^{205}Pb, where predominantly the first excited state (E^*=2.3 keV, $I^\pi=1/2^-$) is populated [5]:

$$^{205}Tl + \nu_e\ (\geq 52\text{ keV}) \rightarrow {}^{205}Pb^* + e^- \qquad (1)$$

The energy threshold for this reaction amounts to $E_\nu \geq 52$ keV [6], and is the *by far the smallest threshold for any known neutrino-induced nuclear reaction*. The corresponding threshold for capturing solar pp neutrinos by ^{71}Ga nuclei (GALLEX and SAGE experiments) amounts to $E_\nu \geq 233$ keV.

The geochemical experiment LOREX (LORandite EXperiment), proposed by Freedman [7], pursues the determination of the long-time average (over ~ 4.3 Ma) of the solar pp-neutrino flux Φ_ν via the neutrino-capture reaction. Lorandite is the Tl-bearing mineral ($TlAsS_2$), which is amply available in the mine of Allchar (FYR Macedonia). The average neutrino flux Φ_ν over the exposure time *a* (age of lorandite since its mineralization) follows from the common activation equation:

$$\Phi_\nu = N^{-1} (T - B) (\sigma\varepsilon)^{-1} \lambda [1 - \exp(-\lambda a)]^{-1} \qquad (2)$$

with N the total number of ^{205}Tl atoms, T the total number of ^{205}Pb atoms, B the background-induced number of ^{205}Pb atoms (mainly via ^{205}Tl(μp,n)^{205}Pb reaction), σ the neutrino capture cross section, ε the overall detection efficiency, λ=4.68·10^{-8}/y the decay constant of ^{205}Pb, and *a*=4.3 Ma the age of lorandite. This renders finally the mean solar pp-neutrino flux, i.e. the mean luminosity of the sun during the last 4.3 million years, the geological age *a* of lorandite.

The LOREX project consists of four distinct challenges:

1) The determination of geological parameters of lorandite ore, like erosion rate and paleodepth. From these studies (see [8]), about 40 atoms of ^{205}Pb in one gram of lorandite are expected. From them, 22 atoms of ^{205}Pb represent contribution of the pp-neutrino capture by ^{205}Tl and 18 atoms are due to muon-induced reactions;

2) The physical extraction of a sufficient amount of clean lorandite. To date, about 700 g of lorandite were separated from about 10.5 tons of ore. See [8] for more details;

3) The determination of the number of ^{205}Pb atoms in the loranidite samples. For this purpose a dedicated experiment is being prepared at the RIKEN facility [9];

4) The determination of the Solar pp-neutrino capture probability transmuting ^{205}Tl into ^{205}Pb, which shall be addressed in this proposal. The nuclear transition matrix element to the first excited state of ^{205}Pb is unknown, but is *the same* as for the β_b decay of ^{205}Tl to this state. The determination of the β_b decay probability of bare ^{205}Tl provides, hence, the *log ft* value for this transition.

Taking into account the present-day state-of-the-art of all the techniques needed to solve the main challenges of LOREX, it seems realistic to expect the first result for the solar pp-neutrino flux averaged over the last 4.3 million years in the foreseeable future.

2.2. ^{205}Pb/^{205}Tl pair as s-process cosmochronometer

The short-lived radioactivities (SLRs) are radioactive nuclei with half-lives in the range 1 Ma to 100 Ma that were alive in the early Solar System. Their abundance in the early Solar System is known from excesses of their daughter isotopes in primitive meteorites that correlate with the abundance of stable isotopes of the parent element. To date, roughly ten such SLRs have been confirmed and their abundances relative to their stable reference isotope inferred [10]. Among these SLRs, ^{205}Pb is unique in that it is the only purely s-process SLR. It provides special insights unavailable from the other SLRs.

The abundance of an SLR relative to a stable reference isotope constrains nucleosynthesis activity just prior to the Sun's birth and the circumstances of Solar System formation [11]. In particular, the crucial quantity is the SLR to reference isotope ratio compared to the value expected from continuous galactic nucleosynthesis. If the ratios agree, then no special circumstances are required to explain the SLR's abundance since the Solar System simply inherited it from the interstellar medium (ISM). If the abundance ratio exceeds the expected value, then input from a special stellar source just prior to the Sun's birth is necessary. This seems to be the case for isotopes like ^{26}Al or ^{60}Fe. If the abundance ratio falls short of the expected value, then Galactic nucleosynthesis of that SLR deviates from the expected continuous picture. This seems to be the case with the r-process SLR ^{129}I but not with the r-process SLR ^{182}Hf, which agrees with expectations from continuous Galactic nucleosynthesis [11]. This may indicate that there are a variety of r processes operating on varying timescales [12].

For radioactive ^{205}Pb and stable ^{204}Pb, which are both secondary nucleosynthetic species, the expected ratio in the ISM is [10]:

$$N_{205}/N_{204}=(k+2)\ P_{205}/P_{204}\ \tau_{205}/T, \tag{4}$$

where N_{205} and N_{204} are the ISM abundances of ^{205}Pb and ^{204}Pb, respectively, P_{205}/P_{204} is the production ratio of the two species at their stellar source, τ_{205} is the mean life of ^{205}Pb, and T is the age of the Galactic disk (~8.5 billion years). The parameter k is the infall parameter in Clayton's standard Galactic chemical evolution model [13]. A typical value for k is in the range 1 to 3. Clearly the ISM abundance ratio is proportional to the production ratio in the s process.

The abundance ratio in the molecular cloud that formed the Sun is modified by mixing from the hot ISM into which stellar outflows are ejected into the colder ISM phases in which stars form [13]. For realistic mixing times, this tends to reduce N_{205}/N_{204} by a factor of 2 to 4. We thus have a good estimate for the ^{205}Pb abundance in the early Solar System, if we can estimate P_{205}/P_{204} in the s-process.

We expect P_{205}/P_{204} to be of order unity. In this case, the expected N_{205}/N_{204} in the early Solar System is ~0.0025. This agrees reasonably well with the value $(1\pm0.4)\times10^{-3}$ measured in carbonaceous chondrites [14]. A production ratio reduced by a factor of ~2-3 might be preferred for better concordance between the expected and meteoritical values.

The concern for the ^{205}Pb chronometer is that P_{205}/P_{204} might be strongly affected by electron capture from the 2.3 keV first excited state of ^{205}Pb which could dramatically reduce the production of ^{205}Pb in the s process [15]. On the other hand, Yokoi et al. [2] pointed out that bound-state beta decay of ^{205}Tl could counter balance the ^{205}Pb electron capture and keep the ^{205}Pb production high. Current s-process models use theoretical estimates for the rate of ^{205}Tl bound-state beta decay [2] and do give P_{205}/P_{204} values near unity [16]. Because of the importance of the ^{205}Pb chronometer, however, an experimentally determined value for the rate of ^{205}Tl bound-state beta decay is crucial to clarify the plausibility for the source of the live ^{205}Pb in the early Solar System.

As mentioned above certain isotopes require recent stellar production and injection into the early Solar cloud. The currently favored models are injection from a massive star or from an AGB star [16]. Injection from AGB stars can marginally explain the Solar System's ^{205}Pb/^{204}Pb ratio [14] given current s-process calculations that find $P_{205}/P_{204} \approx 1$. However, the same production ratio seems to lead to the conclusion that continuous Galactic chemical evolution can explain the early Solar System's ^{205}Pb. A reduction of P_{205}/P_{204} by a factor of several due to ^{205}Pb electron capture may rule out injection from

AGB stars as the source of the early Solar System's ^{205}Pb and might bring the value from continuous Galactic chemical evolution into better agreement with the meteoritic value. A reduction of P_{205}/P_{204} by a large factor (>10) may present a significant challenge.

3. Proposed experiment at FRS/ESR

3.1. Rate estimate for the ^{205}Tl bound state beta decay

The aim of this proposal is to determine the lifetime of bare $^{205}\text{Tl}^{81+}$ ions, stored and cooled in the ESR. Neutral ^{205}Tl atoms are stable. If they are highly ionized (at least one vacancy in the K shell, i.e. hydrogen-like or bare ^{205}Tl) the bound-state beta decay (β_b decay) process from the ^{205}Tl g.s. ($I^{\pi} = 1/2^{+}$) with an almost 100% branch to the first excited state of ^{205}Pb (E^{*} = 2.3 keV, $I^{\pi} = 1/2^{-}$) and with the created electron bound in the K shell of ^{205}Pb becomes possible (see figure 1). The Q-value for this transition amounts, for *bare* $^{205}\text{Tl}^{81+}$ parent ions, to [1]:

$$Q_{\beta b}(\text{bare}\rightarrow\text{K}, E^{*}) = - Q_{EC} - |\Delta B_e| - |E^{*}| + |B_K|,$$

where Q_{EC} is the Q-value for electron capture (EC) from the g.s. of neutral ^{205}Pb to the g.s. of neutral ^{205}Tl, ΔB_e the difference of the sum of the binding energies of all electrons in neutral ^{205}Tl and ^{205}Pb, respectively, E^{*} the excitation energy of 2.3 keV of the ^{205}Pb nucleus, and B_K the K binding energy of the created electron in the hydrogen-like ^{205}Pb daughter ion. With Q_{EC} = 50.5(5) keV [6], ΔB_e = 17.35 keV [17], B_K = 101.32 keV [18] and E^{*} = 2.329 keV [6] one obtains:

$$Q_{\beta b}(\text{bare} \rightarrow \text{K}, E^{*}) = + 31.14 \text{ keV}$$

The *log ft* value for the basically unknown nuclear matrix element of this transition was estimated to [2]:

$$\log ft \text{ (g.s. of } ^{205}\text{Tl} \rightarrow {}^{205}\text{Pb}^{*} \text{ (2.3 keV))} = 5.4,$$

where a large error margin has to be taken into account. Hence, one derives for *bare* $^{205}\text{Tl}^{81+}$ ions a half-life of about $T_{1/2}$ (β_b) = 120 days, or a decay probability in the ^{205}Tl rest frame of $\lambda_{CM}(\beta_b) = \ln 2/T_{1/2} = 6.7 \cdot 10^{-8}\ \text{s}^{-1}$.

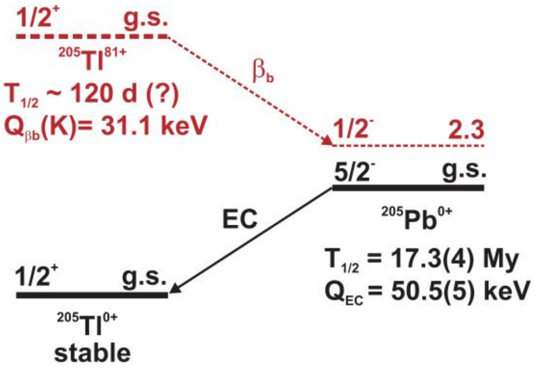

Figure 1. Decay scheme of neutral ^{205}Pb atoms (black) and of bare $^{205}\text{Tl}^{81+}$ ions (red). Neutral ^{205}Pb atoms decay by unique first-forbidden orbital electron capture (EC) from the L- and higher electron shells to stable neutral ^{205}Tl atoms with a half-life of 17.3 Ma and a Q value Q_{EC} = 50.5 keV. Bare $^{205}\text{Tl}^{81+}$ (or H-like $^{205}\text{Tl}^{80+}$) ions can decay to almost 100 % by β_b decay to the first excited state of $^{205}\text{Pb}^{81+}$ at E^{*} = 2.3 keV with the created electron captured into the K shell.

3.1. Experiment

The experimental procedure to detect the β_b decay of bare $^{205}\text{Tl}^{81+}$ ions follows the same steps as applied in the half-life determination of bare ^{163}Dy- [1] or bare ^{187}Re-ions [19], with the only exception that the ^{205}Tl ions are provided by the FRS and not by the SIS as in the former cases. After rf-stacking in the ESR, which was demonstrated recently by accumulating ~$5 \cdot 10^{6}$ radioactive ^{56}Ni ions in the ESR [20], about 10^{6} bare ^{205}Tl ions, will be stored and continuously electron-cooled in the preserved atomic charge state for different times from 1 to 5 hours, where some of them may decay by β_b decay to hydrogen-like ^{205}Pb with the generated electron bound in the K shell of $^{205}\mathrm{Pb}^{81+}$. Due to the small $Q_{\beta b}$(K) value of only 31 keV, the frequency traces of the β_b daughters cannot be resolved from the corresponding traces of the parent ions, but remain "hidden" in a common Schottky frequency signal (same as in the measurements for ^{163}Dy and ^{187}Re).

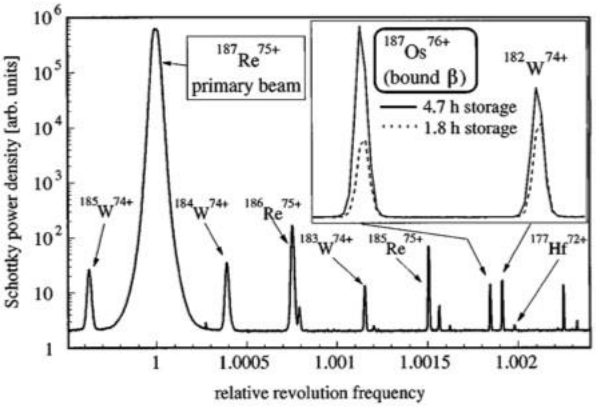

Figure 2. Schottky frequency spectrum after a storage time of 1.8 h and after the interaction of the coasting beam, consisting of bare $^{187}\mathrm{Re}^{75+}$ parents and hydrogen-like $^{187}\mathrm{Os}^{75+}$ β_b daughters, with an Ar gas jet. The revolution frequency is a linear function of the charge-to-mass ratio. Besides nuclides produced in nuclear reactions with the nuclei of the gas jet, the β_b decay daughters, bare $^{187}\mathrm{Os}^{76+}$ ions are seen, after the electron created in the β_b decay has been stripped-off in the interaction with the gas jet. The inset demonstrates that the intensity of the $^{187}\mathrm{Os}^{76+}$ line grows significantly, if the storage time is increased from 1.8 h (dashed line) to 4.7 h (full drawn) in contrast to that from the nuclear reaction product $^{182}\mathrm{W}^{74+}$.

Therefore, to reveal the creation of hydrogen-like β_b daughters, $^{205}\mathrm{Pb}^{81+}$, the same technique as in the former cases [1,19] has to be applied: After a given storage time, a strong argon gas jet (about 10^{13} argon atoms/cm^2) will be turned-on for about two minutes, stripping-off the electron in the K-shell of the $^{205}\mathrm{Pb}^{81+}$ ions and transforming the hydrogen-like $^{205}\mathrm{Pb}^{81+}$ ions to bare $^{205}\mathrm{Pb}^{82+}$ ions. This change of the atomic charge state causes a significant alteration of the trajectory and hence the revolution frequency, which is directly measured by the time-resolved Schottky spectroscopy [4]. Figure 2 shows the Schottky-noise spectra taken for the β_b decay of ^{187}Re [19] after the gas jet was turned-off. All lines in this spectrum can be assigned to nuclei produced by reactions of ^{187}Re with nuclei in the gas jet (mainly loss of a few nucleons) except for the bare $^{187}\mathrm{Os}^{76+}$ ions, originating from the β_b decay of $^{187}\mathrm{Re}^{75+}$. Only the latter Schottky peak grows linearly with the storage time, as demonstrated in the inset of figure 2, proving its origin from β_b decay of $^{187}\mathrm{Re}^{75+}$. The spectrum for the case ^{205}Tl/^{205}Pb will be just analogous, except for the fact that the performance of the Schottky spectrometry has been improved in the last years by several orders of magnitude and is now sensitive to single stored ions [21,22].

The absolute number of bare $^{205}\mathrm{Pb}^{82+}$ ions, originating from the β_b decay of bare $^{205}\mathrm{Tl}^{81+}$, is extracted in a standard way [21] from the areas of the corresponding Schottky frequency peaks, after correcting for the electron-stripping efficiency of the gas jet. The latter will be determined from the corresponding number of helium-like $^{205}\mathrm{Pb}^{80+}$ ions, which are generated by the interaction with the gas jet and will be counted by a particle detector positioned behind the gas jet in the outer part of the aperture. In case of the ^{187}Re experiment a ratio R = 0.2 of electron capture and electron stripping (from the K shell) in the gas jet was found. For the proposed ^{205}Tl experiment, this fraction should be comparable. The amount of bare ^{205}Pb ions, generated by nuclear reactions in the gas jet, will be determined in runs with very short storage times.

For the design value of 10^6 stored bare ^{205}Tl ions and a very cautiously estimated β_b half-life of 1 year, corresponding to a decay probability in the ^{205}Tl rest frame of $\lambda_{\beta b}$(c.m.) =

$2.2 \cdot 10^{-8}$ s^{-1}, we expect a number of about 40 β_b decays within a storage time of 1 hour. For this estimate, the Lorentz factor $\gamma = 1.43$ (corresponding to the kinetic energy of 400 MeV/u of the stored ions) as well as a half-life (lab.) of the stored ions with respect to ring losses of $T_{1/2} = 40$ min was taken into account. This half-life has been measured in many runs at the ESR for stored bare Pb ions at 400 MeV/u, an electron-cooler current of 50 mA, and at standard conditions concerning the mean pressure ($2 \cdot 10^{-11}$ mbar) as well as the standard composition of the residual gas (see [21] for more details).

It is emphasized that the number of about 40 created β_b daughter ions within a storage time of 1 hour is calculated carefully, but with rather pessimistic assumptions of a rather long half-life of 1 year for bare ^{205}Tl^{81+}.

4. Beam-time request

We ask for a total beam time of **21 shifts (à 8 h)** (9 for beam setting and 12 shifts for the measurements) for the realization of our proposal. We emphasize that our experiment requests - after the optimization of the beam parameters - injections from SIS only at intervals of (several) hours. During the time between these injections the ESR will operate in a stand-alone mode, i.e., in an extreme **"parasitic" mode.**

In detail, our request of beam parameters and beam time is:

^{206}Pb from the VARIS source (enriched to 99 % cathodes are purchased and prepared);

$1 \cdot 10^9$ ^{206}Pb ions / spill is in accordance with beam intensities expected for 2018/19 [22];

E ≈ 500 A MeV after (fast) extraction from SIS and transfer to the target at the FRS

Production and transfer of bare ^{205}Tl to the ESR at 400 A MeV

Stochastic pre-cooling and rf-stacking at the ESR to reach **$1 \cdot 10^6$ ^{205}Tl^{81+} ions**

We estimate a need of **9 shifts** for the setting up the accelerator chain, including the production, transfer, storage, stacking and cooling of bare ^{205}Tl in the ESR.

For measurements of the β_b half-life of bare ^{205}Tl (6 cycles of 12 hour-runs with different storage times of 1, 2...5 h) and for auxiliary measurements (e.g. ratio of the probability of electron capture and electron stripping in the gas jet) we estimate a need of **12 shifts.**

4. Statement of the uniqueness and relation to FAIR

Outside of GSI there is no facility worldwide where the proposed measurements can be performed! This physics case is unique for the SIS-FRS-ESR complex at GSI or FAIR. For the latter, however, the experiment may (possibly) be feasible if the entire complicated chain of facilities SIS18-SIS100-Super-FRS-CR-HESR is employed.

In the present experiment we will employ several detectors (including the corresponding data acquisitions and analysis methods), that are being developed for ILIMA and SPARC storage-ring experiments at FAIR. We will use new resonant Schottky detectors [20,24] as well as the CsISiPHOS (CsI–Silicon Particle detector for Heavy ions Orbiting in Storage rings) particle detectors [25]. The testing of these detectors with beam is indispensable for ILIMA, which is emphasized in the ILIMA FAIR-0 strategy paper [26]. Furthermore, we will use the --constantly upgraded by the SPARC collaboration-- internal gas-jet target and its optical diagnostics as well as particle detectors.

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