

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Characterization of the low-energy $^{229\text{m}}\text{Th}$ isomer

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Abstract

While the existence of a low-energy nuclear isomer in ^{229}Th was suggested from indirect measurements, it is only recently, that the isomer has been firmly identified through the combination of mass/charge separation and the observation of a decay signal. Little is known about the isomer's nuclear properties, even its excitation energy is only estimated from indirect measurements resulting in 7.8(0.5) eV. This uncertainty needs to be reduced to make it suitable for laser excitation experiments. Moreover, for the latter application, a suitable host material needs to be identified that blocks the fast internal conversion electron decay of the isomer.

In most experiments, a long-lived ^{233}U source is used to populate the isomer via alpha decay. This results in a ~2% relative population probability and in many cases radiation-induced background hampering the observation of the isomer's radiative decay.

ISOLDE offers the possibility to populate the isomer from the beta decay of ^{229}Ac ($T_{1/2}=62.7$ m) with a much higher (> 13%) population probability. A new approach is developed that consists in implanting a pure ^{229}Ac beam into large-band gap host materials, using the relatively long half-life to anneal the implantation damage to make sure that the ^{229}Ac and its $^{229\text{m}}\text{Th}$ beta-decay daughter are positioned in a substitutional position and detecting the UV radiation (159 nm) of the radiative decay of the isomer. This should allow, for the first time, to perform a direct measurement of the transition energy as well as of the radiative-transition probability. Further, this might lead to a viable route to study and identify suitable host materials for further applications.



To prepare this project, we request beam time to develop intense and pure ^{229}Ac beams, to prove the substantial population of the ^{229}Th isomer in the beta decay of ^{229}Ac and to study the implantation behavior of these heavy masses in a limited number of host materials.

Requested shifts: 6 shifts, (split into 2 runs over 1 year)

1. Physics background and motivation

Mid '70, a first argument, supporting the existence of an unusually low-lying nuclear state in ^{229}Th , lying only a few eV above the ground state was offered [1]. From that day, a multitude of experiments has searched for this state and attempted to define its properties, without yet settling the issue. Recently, however, the isomer has been unambiguously proven to exist [2]. Although the nuclear origin of this state and the fact that it exists seems to be widely accepted now, the excitation energy from indirect measurements of intra and interband transitions is only known with limited precision and estimated to be 7.8(0.5) eV [3]. This value depends on the assumed branching ratios of the transitions and might, in a worst-case scenario, increase to 10.5 eV [5]. The limits reported in [2] span the region from 6.3 eV to 18.3 eV. Several nuclear properties such as mean-square charge radius, magnetic moment and quadrupole moment, or its radiative decay lifetime are not yet experimentally determined.

The interest in this unique low-lying isomer stems from the fact that it opens up a wide variety of possible applications. The expected long lifetime of $^{229\text{m}}\text{Th}$ results in an exceptionally narrow linewidth $\Delta E/E$ in the order of 10^{-20} . This narrow linewidth and the possibility to drive this nuclear transition with lasers, makes it a possible candidate for a nuclear optical clock. This isomeric nuclear state could also be vital in the development of the first nuclear laser and more fundamentally; the study of the time dependence of fundamental constants of nature [6-10].

Intensive theoretical and experimental work is currently ongoing to characterize the isomer and to exploit the use of its unique properties (see e.g. the NuClock project www.nuclock.eu). An overview of the properties of the ^{229}Th ground and isomeric states is given in Figure 1 (see NNDC [11]).

The current understanding of the nuclear structure of the isomer is that it constitutes the band head of a band built on the $3/2[631]$ Nilsson state while the ground state has a $5/2[633]$ configuration (see Fig. 1), but nuclear-model calculations indicate a certain degree of mixing (see [12]). The decay of the isomer is thus expected to proceed via an M1 transition and values for its radiative M1 decay have been obtained from theoretical calculations and systematic comparison of similar transition in e.g. ^{233}U [11]. The radiative decay lifetime estimates vary over at least one order of magnitude from around 1000 s to 10.000 s [5]. As the low excitation energy is larger than the first ionization potential of neutral thorium (IP = 6.3 eV) the decay of the isomer in neutral ^{229}Th proceeds mainly via internal conversion emitting a very low-energy electron. This decay process was used by the Munich group to firmly identify the existence of the isomer [2] and they recently reported a half-life of 7(1) μs leading to a total conversion coefficient of about 10^9 using an expected radiative half-life of 10^4 s [13].

Most of the experimental work is based on the population of the isomer in the alpha decay of long-lived ^{233}U ($T_{1/2} = 1.6 \cdot 10^5$ years) resulting in an approximate relative population of 2%. At ISOLDE, an alternative way can be followed by using the beta decay of ^{229}Ac ($T_{1/2} = 62.7$ m). Extended beta-decay studies of ^{229}Ac feeding levels in

^{229}Th have already been performed at ISOLDE [12,14] to study, amongst others, octupole deformation in this mass region. Combining this work with other studies of ^{229}Th reveals that the indirect (via higher lying excited states) feeding of the $(3/2^+)$ state at 7.8 eV represents 13.4 % of the total beta feeding of ^{229}Ac [11]. Moreover, a combined 79% feeding to ground state and the state at 7.8 eV is reported [11]. This means that the total isomer population in the beta decay of ^{229}Ac is at least 13.4%. In combination with other advantages, which are discussed in the next section, studying the isomer via the beta decay of ^{229}Ac is complementary with the research focused on the alpha decay of ^{233}U .

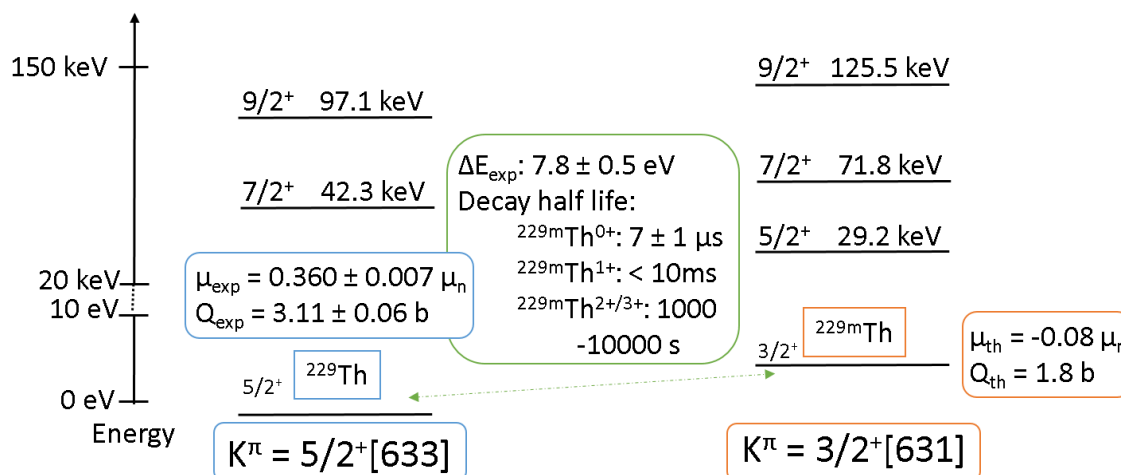


Fig. 1: Partial level scheme of ^{229}Th with the relevant information on the nuclear properties of the ground and low-lying isomeric state.

With this letter of intent, we propose to follow a new way to determine precisely the energy of the isomer and its radiative decay probability by using the opportunities offered by the ^{229}Ac beams from ISOLDE. Further, these studies should allow to answer whether chemical conditions that allow for the suppression of the IC decay channel and therefore for the development of a solid-state nuclear frequency standard can be reached.

2. Methodology

2.a General principles of the approach

The basic idea is to implant the mass separated and pure ^{229}Ac beam with an energy >30 keV in a suited host material. The host material should be chosen in such a way that $^{229\text{m}}\text{Th}$, produced after beta decay, experiences a large enough band gap to prevent the decay via internal conversion and that the material is transparent for VUV radiation to detect the radiative decay of the isomer (7.8 eV radiation corresponds to 159 nm). After an implantation time of about two half-lives, the beta decay of ^{229}Ac will be monitored via a beta/gamma detection system and the VUV light from the decay of the isomer will be detected. If needed, the host material will undergo an in-situ annealing procedure to repair the damage from the implantation process. As the maximum recoil energy of ^{229}Th after the beta decay of ^{229}Ac is 3 eV ($Q_{\text{beta}} = 1.10$ MeV), it is expected that the daughter nucleus will occupy the same position in the host material. The detection of the VUV light from the decay of the isomer will proceed in two stages. An initial VUV detection system whose design will be based on the Munich prototype [15] will be used to prove the observation of VUV radiation from the isomer and to determine a first improved energy value and an estimate of the radiative half-life. This

system is characterized by a good VUV detection efficiency (2%) and a broad spectral observation range (which is needed to make sure that the de-excitation energy is covered). In a second step a VUV spectrometer will be used to determine the energy with an expected precision <0.3 nm, and depending on the value of the radiative half-life, a more precise value will be obtained. Once a clearly identifiable signal has been measured, the radiative half-life can be obtained by carefully monitoring the time behaviour of the radiative decay signal in the VUV detection set-up compared to the time behaviour of the gamma and beta signals from the decay of ^{229}Ac . The sensitivity of this method, however, will depend strongly on the actual half-life of the isomer.

A generic drawing of the set-up is given in Fig. 2 (left).

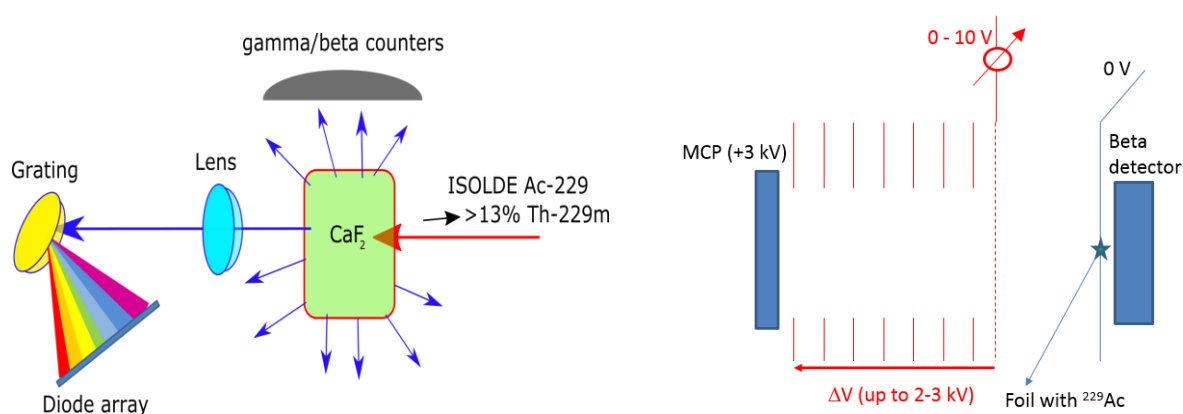


Fig. 2: (left) Schematic drawing of the experimental set-up to be used for the final experimental campaigns. (right) Schematic representation of the detection system used to determine the population of the $^{229\text{m}}\text{Th}$ isomer in the beta decay of ^{229}Ac . The deceleration grid will be removed for the first experiments to prove the observation of the internal conversion electrons. Once that is established, changing the voltage on the grid should allow a rough estimate of the energy of these extremely low-energy electrons.

2.b Experimental issues to be addressed

- Production of ^{229}Ac beams at ISOLDE

These measurements need a high production rate of ^{229}Ac and the beam should be pure in order to minimize the implantation damage in the host material (see further). ^{229}Ac can be produced in two ways. Via the beta-decay chain of ^{229}Fr ($T_{1/2} = 50$ s) \rightarrow ^{229}Ra (4.0 min) \rightarrow ^{229}Ac [12,14]. A yield of $3.8 \cdot 10^4$ atoms per μC for ^{229}Fr is reported from SC-ISOLDE and a yield of $4 \cdot 10^5/\mu\text{C}$ at ISOLDE-PSB with 1.0 GeV protons was measured. Moreover, a cross section gain of a factor 1.5 is expected when going from 1 to 1.4 GeV protons. Alternatively, ^{229}Ac can be produced directly using the RILIS source as efficient ionization schemes for actinium have been developed and used at e.g. LISOL [16, 17]. Based on the cross section the expected in-target production of ^{229}Ac using a UCx target and 1.4 GeV protons is $1.2 \cdot 10^8$ particles per μC . Using a ^{232}Th target will possibly result in a gain in cross section of at least one order of magnitude based on the work of [18] where a factor of 20 gain in cross section was observed for the $^{238}\text{U}(p,2p2n)^{235}\text{Pa}$ compared to the $^{238}\text{U}(p,4p6n)^{229}\text{Ac}$ reaction.

Using RILIS for the production of ^{229}Ac beams should thus result in higher yields compared to production via ^{229}Fr (or ^{229}Ra) and gives an extra control parameter using the laser on/off approach. The latter is essential to control and map out the background signals in the VUV spectroscopy measurements.

- Population of $^{229\text{m}}\text{Th}$ in the beta decay of ^{229}Ac

While detailed information is available from previous spectroscopy work, the $>13\%$ population of the isomer in the beta decay of ^{229}Ac needs to be confirmed before embarking in this project. Initially, during the actinium beam development process, the production of the ^{229}Ac beam and its purity will be determined using standard beta and gamma detectors from the ISOLDE yield station or the ISOLDE Decay Station and an MCP detector to determine the total $A=229$ intensity. The latter is crucial for the project as contaminants can create damage in the host material or create background VUV-signals. After the initial production tests, a ^{229}Ac beam will be implanted in a frozen water sample positioned on a metal foil [19]. After a few half-lives (typically 2 hours), the implantation will be stopped and the foil with the implanted frozen water will be heated to evaporate the water leaving the actinium atoms sticking on the surface. The foil will subsequently be positioned between a plastic scintillator detector to detect the betas from ^{229}Ac , an acceleration stage for the low energy electrons from the internal electron conversion (IC) process and an MCP detector to detect the accelerated conversion electrons. After beta decay, the $^{229\text{m}}\text{Th}$ will reside on the surface, neutralize and decay via IC. This situation is similar to [2, 13], where the IC electrons were detected from $^{229\text{m}}\text{Th}$ deposited close to the surface of an MCP detector via quasi-soft landing. The characteristic decay signal of the isomer will be filtered from the background by delayed beta-electron coincidence making use of the $7\ \mu\text{s}$ half-life of the isomer. A schematic representation of this detection system is shown in Fig. 2 (right). By using the retardation voltage indicated with a grid in Fig. 2 (right) and measuring the transmission of the conversion electrons as a function of retardation voltage, one might obtain a rough estimate of the energy of the conversion electrons and thus, indirectly of the excitation energy of the isomer. However, as the energy of the emitted electrons might depend on the way the thorium atoms are attached to the substrate surface and to surface impurities (most likely forming complexes with oxygen), this can only be considered a very rough estimate.

- Internal electron conversion (IC) decay of the isomer

It has been shown that the $^{229\text{m}}\text{Th}$ in a $2+$ or $3+$ charge state has a long half-life and does not undergo electron conversion [2, 13]. On the other hand, neutral $^{229\text{m}}\text{Th}$ decays by electron conversion with a half-life of $7\ \mu\text{s}$ as the excitation energy is above the first ionization potential of neutral thorium ($\text{IP} = 6.3\ \text{eV}$). While based on the suggested excitation energy of $7.8\ \text{eV}$ it is expected that $^{229\text{m}}\text{Th}^{+1}$ does not decay via IC as the second ionization energy (from Th^{1+} to Th^{2+}) is $12.1(2)\ \text{eV}$ [4], its half-life could possibly be influenced by other processes such as bound-internal conversion or electronic bridge [7]. While an upper limit of the $^{229\text{m}}\text{Th}^{+1}$ half-life of $10\ \text{ms}$ has been reported [13], no direct experimental evidence is available. In order to measure the radiative decay of the isomer, the electron conversion channel should be inhibited. With the host material discussed below, a large enough band gap could be reached to avoid electron conversion of implanted $^{229\text{m}}\text{Th}$.

- Host material

The requirements for the host material are such that it has a large band gap and that the thorium impurity does not create sites that would allow electron conversion to happen. Moreover, the host material should be transparent to VUV radiation. Host materials that

fulfil the requirements have been proposed in the literature (see [20] for a review). CaF₂ [21] is a prime example of materials where the influence of thorium doping on electronic structure has been studied using ab initio atomic calculations. We plan to start our study with CaF₂ as host, but are currently performing initial atomic calculations on these and other materials to identify the optimum probe material [22, 23]. These might include frozen noble gases (like e.g. neon with a band gap of 21.6 eV), that have been studied as hosts for Mössbauer probes ⁵⁷Co, which were implanted at similar energies as envisaged in the present project [24]. In order to check the feasibility of studies to characterize the implantation behaviour of actinium in CaF₂, to study the initial damage from the heavy atom's implantation and to determine the position of actinium after implantation and thorium after beta decay, we propose to perform initial β - emission channeling experiments. A suitable beam is ²³¹Ac ($T_{1/2} = 7.5$ m) since it decays (via β -) to ²³¹Th ($T_{1/2} = 25.5$ h) which also decays via β -. This allows us to study the lattice incorporation of the Ac parent (on-line at the EC-SLI setup [25]) and, afterwards, of the Th daughter (off-line [26]), as a proxy for what occurs with the ²²⁹Ac/²²⁹Th decay. Such experiments would require **yields of the order of 10⁵ atoms/s**. The in-target production of ²³¹Ac on a UC_x target is calculated to be $5.3 \cdot 10^7$ atoms/ μ C. The fluences involved in these experiments are orders of magnitude lower than those expected to produce extended lattice defects. However, given the high mass of Ac ions, local disorder pockets may be formed at the ion end of range, which would locally modify the electronic structure. These β - emission channeling experiments are aimed at investigating precisely that: whether local disorder is formed upon implantation and, if so, under what thermal annealing conditions (temperature and time) is the crystalline order recovered. In addition, these experiments will allow us to identify the eventual population of non-substitutional sites by Th ions (which could create mid-gap levels) and determine how to thermally anneal them. Non-substitutional sites have generally lower thermal stabilities and can therefore be out-diffused by appropriate annealing.

- Radioactive decay-induced fluorescence background and determination of the radiative decay half-life.

A crucial point in the method to detect fluorescence light is the gamma, beta and alpha induced VUV background (see e.g. [27] where Cherenkov radiation induced by β decays is discussed). This has hampered several measurements and has even led to conflicting reports in the literature (see e.g. [28] and the comment in [29]). Quantitative analysis based on the work of [27] of the expected background from the beta and gamma decay of ²²⁹Ac and alpha decay of the ²²⁹Th ($T_{1/2} = 7932$ y) are currently ongoing.

Important to note is the fact that the ²²⁹Ac beta-decay approach represents several advantages compared to the ²³³U case:

- a) higher relative production rate of the isomer
- b) an extra control parameter with lasers on/off measurements to discriminate from background signals induced by A=229 beam related contamination
- c) implanting neighbouring masses with similar decay properties (e.g. ^{230,231}Ac) and the same implantation behaviour but without the presence of the low-lying isomer

Moreover, this experiment will use a thin CaF₂ crystal (<50 nm to stop the 30 keV ²²⁹Ac beam) reducing the radiation- induced background compared to other approaches.

For the final experimental campaign, the well-known beta-decay half-life of ²²⁹Ac ($T_{1/2} = 62.7$ min) should allow determining the time behaviour of the radiative decay of ²²⁹Th^m if the radiative half-life is of the same order or longer compared to 63 minutes.

However, if it is shorter, dedicated implantation/decay cycles with different implantation times have to be used to obtain a value for the radiative decay half-life.

3. Rate estimates and beam-request

3.a Production rate of ^{229}Ac with RILIS

To optimize the laser ionization scheme for actinium 1 shift and for yield and purity measurements of $^{229,231,233}\text{Ac}$ 2 shifts are requested: in total **3 shifts**.

3.b Emission channeling measurements

The feasibility study of the beta-emission channeling measurements requires beam intensities of 10^5 atoms/sec of ^{231}Ac and a beam time estimate of **2 shifts**.

3.c Population of the $^{229\text{m}}\text{Th}$ in the beta decay of ^{229}Ac

A firm proof for the population of the isomer in the beta decay requires a beam intensity of at least 10^5 pps. The overall efficiency of the implantation duty cycle (~50 %), water evaporation and surface sticking efficiency (~50%), installation inside the detection system cycle (~50%) has an estimated overall efficiency of ~10%. The delayed beta – low-energy electron detection system will have an efficiency of 30% (beta efficiency) x 30% (low-energy detection efficiency) x 13%(feeding probability) = 1 %. This will result in 100 coincidences per second in saturation (three half-lives = 3 h). In order to study two different surface materials and the signal as a function of retardation voltage to proof the low-energy of the ICC electrons and possibly obtain a first rough estimate of the excitation energy will require **1 shift** of beam time.

3.d Count rate estimates for the final measurement detecting the radiative (VUV) decay of $^{229\text{m}}\text{Th}$

With a beam intensity of 10^5 pps, a 13% relative population of the isomer, a conservative 10% of the implanted of ^{229}Ac atoms in the correct position of the host material and a conservative 2 % broad-band VUV detection system efficiency one obtains a VUV count rate of 26 counts per second. This should allow to accomplish the first part of the final proposal.

As the efficiency of a VUV spectrometer (see e.g. [30]) reaches only $2 \cdot 10^{-3}$ (1.3% optical efficiency and 15% quantum detection efficiency at 150 nm), this results in 2.6 VUV photons per second in saturation. This efficient VUV spectrometer will have a spectral resolution of 1 nm and an absolute accuracy of +/- 0.3 nm. These estimates need to be compared with the background induced signals. Work along these lines is underway.

Summary of requested shifts

With this LOI, we wish to investigate the possibility of initiating a study of the low-lying nuclear isomer in ^{229}Th populated in the beta decay of ^{229}Ac . This includes the following:

- Yield and purity measurement of ^{229}Ac and ^{231}Ac : 3 shifts
- Feasibility study to perform emission channeling measurements using heavy beams like ^{231}Ac : 2 shifts
- Experimental proof of the population of the isomer by detecting the internal conversion electron decay of $^{229\text{m}}\text{Th}$: 1 shift

In total: 6 shifts are requested for this LOI.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*

Part of the Choose an item.	Availability	Design and manufacturing
SSP-GHM	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used without any modification
EC-SLI on-line	<input checked="" type="checkbox"/> Existing <input type="checkbox"/> New	<input type="checkbox"/> To be used without any modification <input checked="" type="checkbox"/> To be modified in view of possible contamination <input type="checkbox"/> Standard equipment supplied by a manufacturer <input type="checkbox"/> CERN/collaboration responsible for the design and/or manufacturing
MCP based low-energy electron detection system combined with efficient plastic and gamma detectors for the ICC detection from $^{229}\text{Th}^m$	<input type="checkbox"/> Existing <input checked="" type="checkbox"/> New	<input type="checkbox"/> To be used without any modification <input type="checkbox"/> To be modified <input type="checkbox"/> Standard equipment supplied by a manufacturer <input checked="" type="checkbox"/> collaboration responsible for the design and/or manufacturing
[insert lines if needed]		

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazards			
	[Part 1 of the experiment/equipment]	[Part 2 of the experiment/equipment]	[Part 3 of the experiment/equipment]
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum			
Temperature	[temperature] [K]		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	{fluid}, [pressure][Bar], [volume][l]		
Electrical and electromagnetic			
Electricity	[voltage] [V], [current][A]		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	<input type="checkbox"/>		
Capacitors	<input type="checkbox"/>		
Ionizing radiation			
Target material	[material]		
Beam particle type (e, p, ions, etc)			
Beam intensity			
Beam energy			
Cooling liquids	[liquid]		

Gases	[gas]		
Calibration sources:	<input type="checkbox"/>		
• Open source	<input type="checkbox"/>		
• Sealed source	<input type="checkbox"/> [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			
• Description	<input type="checkbox"/>		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
Non-ionizing radiation			
Laser			
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300MHz)			
Chemical			
Toxic	[chemical agent], [quantity]		
Harmful	[chemical agent], [quantity]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	[chemical agent], [quantity]		
Irritant	[chemical agent], [quantity]		
Flammable	[chemical agent], [quantity]		
Oxidizing	[chemical agent], [quantity]		
Explosiveness	[chemical agent], [quantity]		
Asphyxiant	[chemical agent], [quantity]		
Dangerous for the environment	[chemical agent], [quantity]		
Mechanical			
Physical impact or mechanical energy (moving parts)	[location]		
Mechanical properties (Sharp, rough, slippery)	[location]		
Vibration	[location]		
Vehicles and Means of Transport	[location]		
Noise			
Frequency	[frequency],[Hz]		
Intensity			
Physical			
Confined spaces	[location]		
High workplaces	[location]		
Access to high workplaces	[location]		
Obstructions in passageways	[location]		
Manual handling	[location]		
Poor ergonomics	[location]		

0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above):
(make a rough estimate of the total power consumption of the additional equipment used in the experiment)