EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Study of the radiative decay of the low-energy isomer in ^{229}Th

May 10, 2022

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CERN-INTC-2022-017 / INTC-P-628
10/05/2022

Abstract: A unique feature of ^{229}Th is its isomer with an exceptionally low excitation energy and is proposed as a candidate for future nuclear optical clocks [1]. Development of such a clock is however hindered by the low precision on the excitation energy value and the lifetime of the radiative decay of this isomer. Recently, we observed for the first time the radiative decay of the isomer and determined its energy and lifetime using a novel method to populate the isomer from the $\beta-$decay of ^{229}Ac, embedded in a CaF_2 and MgF_2 crystal [2, 3] at ISOLDE, CERN. In this follow-up proposal we will fully exploit our successful proof-of-principle investigation of this low-energy isomer of ^{229}Th to measure the isomer energy with a precision of 0.1 nm, which is a factor of 30 more precise than the current literature value, and to determine the isomer lifetime in different host crystals more precisely. In addition, the feasibility of the emission

channelling method to characterize the lattice location of actinium and thorium (upon $\beta-$decay) has been shown from the measurements on CaF_2 [3]. Here we propose to perform a systematic measurement campaign studying the lattice incorporation of thorium in crystals with varying band gap, from above to below the isomer's energy. This experiment will lay the foundation for the next generation of ultra stable clocks.
Requested shifts: 28 shifts

1 Physics Motivation

Based on the observation of rotational bands in ^{229}Th populated in the $\alpha-$decay of ^{233}U, a low-lying nuclear state in ^{229}Th with an excitation energy well below 100 eV was proposed nearly half a century ago [4]. Owing to the unique nature of this isomer, numerous experimental campaigns were initiated in an attempt to observe the decay and determine its excitation energy and decay character [5, 6]. Measuring the differences in $\gamma-$transitions feeding the ground state and the isomeric state, an excitation energy of 7.8(5) eV was measured for the isomer, making it accessible to laser excitation [7]. Moreover, an estimated relative radiative decay width of around $\Delta E/E \approx 10^{-19}$ opens up the possibility to develop an optical nuclear clock that could outperform existing atomic clock systems [1, 8]. This can lead to new perspectives in ultra-high precision frequency quantum metrology with implications for both fundamental studies and technology, such as the search for possible time variations of the fundamental constants [5, 9, 10].
Despite numerous efforts, only recently, this isomer has been unambiguously detected by observing the signal induced by the internal conversion (IC) decay of neutral ^{229m}Th atoms [11]: the extracted isomer energy was situated between 6.3 and 18.3 eV while the half-life value for the internal conversion decay channel was reported to be 7(1) μs [12]. Laser spectroscopy studies on $^{229m}Th^{2+}$ ions were performed, and the nuclear magnetic and electric quadrupole moments and the spin were deduced [13]. Precise values for the isomer energy were extracted using two complementary techniques and were reported to be 8.28(17) eV, corresponding to 149.7(31) nm [14], via conversion electron spectroscopy and 8.10(17) eV, corresponding to 153.1(32) nm, via micro-calorimeter $\gamma-$spectroscopy [15]. During our IS658 experiment in the fall of 2021, VUV-photons stemming from the radiative decay of the ^{229m}Th ions that were implanted into CaF_2 and MgF_2 crystals, were observed for the first time. The isomer energy was measured with a precision down to the sub-nm region and the radiative decay lifetime was measured with an anticipated relative uncertainty of 30%. Moreover, emission channelling measurements were successfully performed and allowed the determination of the host position of the implanted ^{231}Th after the implantation of ^{231}Ra/^{231}Fr. These measurements paved the way towards the study of the isomer's properties inside a VUV transparent material.
In this proposal, we aim to perform measurements of the radiative decay of ^{229m}Th using samples of mass A=229 (^{229}Ra, ^{229}Fr and ^{229}Ac) beams implanted into large-band-gap crystals, in order to extract the isomer's lifetime with a relative uncertainty of <10% and energy at a 0.1-nm-precision level, which is a factor of 30 times more precise than the current literature value. In the following sections, a brief introduction to the experimental technique, a status report of the previous measurements followed by the proposed experiments, and a summary of requested shifts will be given.

2 Experimental Technique and setup

This proposal includes two different methodologies:

1. **Vacuum-ultraviolet spectroscopy of the radiative decay of ^{229m}Th**: A tailored VUV grating spectrometer (VM180, Resonance Ltd, Canada) coupled to a windmill system that hosts the different crystals and beam diagnostics for beam tuning was successfully commissioned during our 2021 beam time at CERN [3]. A beam of mass A=229 (^{229}Fr and ^{229}Ra) was implanted for a varying implantation time, ranging from 2 h down to 30 mins, into commercially available CaF_2 and MgF_2 crystals, CaF_2 epitaxial thin films from IMEC (see section 3). The sample was subsequently transferred under vacuum in front of the entrance slits of a VUV-spectrometer, where photons from the radiative decay of the isomer, populated in the $\beta-$decay of ^{229}Ac, were detected. An overall efficiency of the spectrometer about 0.001% was extracted.

   Based on the experience of the previous beam time, an upgrade of the moving mechanism for precise crystal positioning has been undertaken. This moving system will be controlled by a stepper motor and will reduce the time between the end of implantation and the start of the VUV spectroscopy measurement to less than 30 seconds. A robust energy calibration procedure to reduce the systematic uncertainty using different VUV sources is being commissioned, and development of a new DAQ system that combines the read-out of the Photo-multiplier tube (PMT) countrate, the spectrometer motor position and the gamma-ray spectra, is being installed.

2. **Emission channelling study of the thorium lattice position**: The lattice location of impurities in single crystals can be probed with the emission channelling (EC) technique, detecting charged particles emitted by a radioactive isotope of the dopant element under study. These isotropically emitted charged particles undergo anisotropic scattering determined by the screened Coulomb potential of atomic rows and planes in the host crystal. This anisotropic scattering results in typical channelling and blocking effects that are measured with a position-sensitive detector [16].

   To ensure the suppression of the internal conversion decay channel of the isomer in a crystal environment, the thorium ion must occupy specific lattice positions. Using the emission channelling technique with ^{229}Ac and ^{231}Ac/^{231}Th, the actinium and thorium lattice sites will be studied, allowing to optimize their incorporation in a variety of suitable hosts.

3 Status report from IS-658

VUV spectroscopy: Previous experiments at ISOLDE in the framework of IS-658 allowed us to perform vacuum-ultraviolet spectroscopy on the ^{229}Th isomer for the first time [3]. Despite the lack of a pure laser-ionized ^{229}Ac beam from ISOLDE, species of mass A=229 (^{229}Fr and ^{229}Ra) were produced from the uranium carbide target and surface ionized. Using germanium and $LaBr_3$ detectors, the mass A=229 beam composition was determined to be ^{229}Ra (about 10^6 pps) and ^{229}Fr (about 10^5 pps). The ^{229}Ra and ^{229}Fr beams were implanted at 30 keV into CaF_2 and MgF_2 crystals with an estimated beam spot diameter of 6 mm. The mean implantation depth was calculated with SRIM [17] to be about 17 nm with a full-width half-maximum spread of 7 nm. ^{229}Fr $\beta-$decays into ^{229}Ra, which in turn $\beta-$decays to ^{229}Ac. Both ^{229}Ra and ^{229}Fr are short lived with a half-life of $T_{1/2}(^{229}\text{Ra})$ = 4 mins and $T_{1/2}(^{229}\text{Fr})$ = 50 s. After 2 h implantations, on average 5×10^9 ^{229}Ac ($T_{1/2}$ = 62.7 min) nuclei were implanted in the crystal. The crystals initially used were commercially available CaF_2 and MgF_2 crystals. These crystals are 12.5 mm in diameter and 5 mm thick. Figure 1 shows a typical spectrum obtained in our measurements. The VUV photon count rate is plotted against the grating motor position and shows the first observation of the radiative decay of the ^{229}Th isomer. This measurement was

performed with a slit width of 3 mm and has a resolution of 6.5 nm, which becomes better for narrower slit widths. The background is dominated by the Cherenkov radiation induced by $\beta-$decays of ^{229}Fr, ^{229}Ra, and ^{229}Ac. Reduction of this Cherenkov background was achieved through the use of a 50-nm thick CaF_2 film, epitaxially grown on a silicon substrate, and obtained from our collaborator C. Merckling from IMEC (Belgium). Spectra were measured for different crystals and varying implantation times. The isomer energy determined after implantation in MgF_2 or CaF_2 crystals agrees within 0.2 nm. Measurement of the lifetime of the isomer was performed by studying the time evolution of the VUV signal using a shorter implantation time (about 1 h) as it increased sensitivity to the isomer's lifetime. A lifetime of about 10 mins, not corrected for the refractive-index of the host crystal, was measured for the ^{229}Th isomer implanted in a MgF_2 crystal with an uncertainty of about 30%. A similar spectroscopy measurement was also performed by implanting mass A=230 (^{230}Fr and ^{230}Ra), which both $\beta-$decay into ^{230}Ac which in turn decays into ^{230}Th. In this case, no VUV peak was observed.

All these arguments lead us to conclude that we have observed the radiative decay of the ^{229m}Th embedded in MgF_2 and CaF_2 crystals. While the analysis is still ongoing, the energy will be determined with a precision better than 0.5-nm and the half-life with a precision of 30%, because of the non-optimized implantation/decay cycles. This result is compatible with the current knowledge of the isomer's properties and the energy is a factor of 6 more precise.

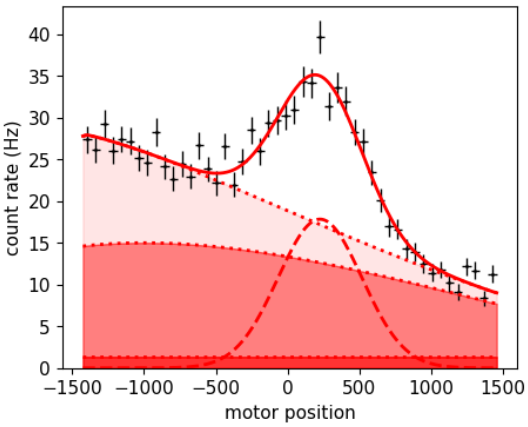

Figure 1: VUV signal of the ^{229}Th isomer. The measurements were performed on a MgF_2 crystal with a slit width of 3 mm. The signal was fit using a Gaussian with a polynomial background which accounts for the Cherenkov contributions from ^{229}Ra (light red area), ^{229}Ac (darker red area), and a constant background stemming from the dark current in the PMT (darkest red area).

Emission channelling: Figure 2 shows two-dimensional patterns measured after the $\beta-$decay of different isotopes in CaF_2 and MgF_2 crystals. Theoretical electron emission channelling patterns, for probes occupying possible lattice sites, are calculated using the *many beam* formalism [18]. The site occupancy is quantified by fitting the experimental patterns with linear combinations of theoretical ones. Different isotopes of the same element are expected to occupy the same lattice positions due to their chemical similarity.

VUV grade CaF_2 crystals were implanted with ions of mass A=231 (^{231}Fr and ^{231}Ra) and A=229 (^{229}Fr and ^{229}Ra) and $\beta-$ emission-channelling patterns were measured upon decay of the ^{231}Th and ^{229}Ac daughter nuclei. The analysis of the ^{231}Th patterns shows that a fraction of 0.76±0.05 of the thorium atoms are in a calcium substitutional position with no indication that there is a second lattice site present. This substitutional fraction still needs to be corrected for backscattered electrons, a correction factor typically on the order of 1.4, which brings the substitutional fraction close to 1. In addition, fitted displacements from the substitutional site are in agreement with theoretical displacements caused by equilibrium charge compensation mechanisms. Therefore, a large fraction of the thorium dopants is expected to be in a local atomic configuration that blocks the internal conversion decay channel of the ^{229}Th isomeric state.

Since the recoil energy of the ^{229}Th daughter nucleus from the decay of ^{229}Ac is very small (<6 eV), it is expected that ^{229}Th stays in the same lattice position as ^{229}Ac. Similar to the ^{231}Th data, ^{229}Ac has a large calcium substitutional fraction. However, in this case fitting results show that 10–20% of the ^{229}Ac probes are in an interstitial lattice site, possibly causing a similar fraction of the ^{229}Th daughter nuclei to not be in an environment that blocks internal conversion. Another possibility is that the actinium $\beta-$decay provides enough recoil energy to the daughter nucleus to move from the interstitial site to a calcium substitutional site. Preliminary ^{229}Ac emission channelling patterns were also measured in MgF_2 (see Fig.2-right). Although the analysis is still ongoing, these measurements demonstrated the feasibility of such studies.

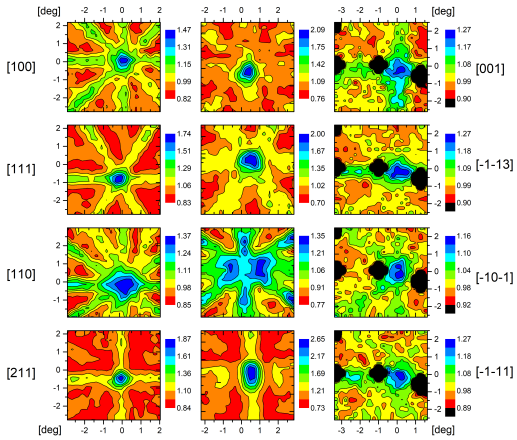

Figure 2: Normalized experimental $\beta-$emission channelling patterns in the vicinity of different crystal directions after room temperature implantations. Left: ^{229}Ac in CaF_2. Center: ^{231}Th in CaF_2. Right: ^{229}Ac in MgF_2.

4 Proposed Experiment

We propose to improve the uncertainty on the lifetime and the energy of the radiative decay of the ^{229}Th isomer in CaF_2 and MgF_2 crystals.

4.1 VUV spectroscopy

Energy measurement: Laser-ionized actinium beam produced from a uranium carbide target will be implanted at 30 keV into a 50-nm thick CaF_2 film grown on a silicon substrate and into commercially available MgF_2 crystal. During the previous beam time in 2021, the isomer energy was, in most cases, measured with a relatively large slit width of 0.5 mm or larger. The systematic uncertainty in isomer energy determination is dominated mainly by the distribution of the beam over the implanted source in front of the entrance slit of the spectrometer and the calibration procedure. In the proposed experiment we aim to measure the isomer energy with a slit size of <0.5 mm. This will reduce the systematic effect from the distribution of the implanted beam. A reduced slit size leads to a loss of the signal strength of the VUV photons and needs to be compensated for by a larger production rate using laser-ionized ^{229}Ac from RILIS. Preliminary analysis of the emission channelling measurements shows that after implantation at room temperature every second ^{229}Ac ion occupies a favourable position in the CaF_2 crystal lattice, thus blocking the internal conversion decay channel of ^{229m}Th. This fraction was enhanced to about 90% by annealing the crystal at 600 K. For the VUV spectroscopy measurement the crystals were not annealed during the 2021 beam time but the set-up has been adapted to be able to anneal crystals during and after implantation. Thus, to enhance the VUV signal we also propose to anneal the crystals. Moreover, offline measurements with different VUV sources and polished crystals, to obtain secondary scattering sources, are being developed to optimize the energy calibration procedure. With these refined experimental procedures, and the upgrades mentioned in section 2, we will be able to reach the instrument's spectral uncertainty limit of 0.1 nm.

Lifetime measurement: We aim to measure the lifetime of the ^{229}Th isomer more precisely, by

varying the implantation/decay cycle using different crystals for which a VUV signal can be observed (MgF_2, CaF_2, etc.). Since the half-life of the feeding ^{229}Ac $\beta-$decaying nucleus ($\sim$1 h) is long than the isomer's lifetime, long implantation cycles brought the thorium fraction to near saturation, diminishing the sensitivity to the isomer's lifetime. Thus, shorter implantation times, 1 h or 30 mins, will be used to get a better sensitivity to the isomer's lifetime. However, the loss in signal, due to shorter implantation time, needs to be compensated by a larger production rate using laser ionized ^{229}Ac from RILIS, which in turn might request a higher target temperature to increase the release of actinium. Moreover, by measuring the lifetime of the isomer in MgF_2 and CaF_2 crystals precisely (<10% relative uncertainty) we will be able to check the lifetime dependence on the refractive index [19, 20, 21].
Ratio of VUV photons to implanted ions: We propose to measure the ratio of the implanted ^{229}Ac to the number of VUV photons detected to determine the fraction that radiates VUV photons. This will be compared with the relative occupancy of lattice sites, as determined by channelling measurements, to increase our understanding not only of the lattice location, but also of the local chemical environment. An issue here is the determination of the total efficiency of the spectrometer as well as the ^{229}Ac $\beta-$branching feeding the isomer. A relative measurement of the ratio between the implanted ^{229}Ac to the detected VUV photons for different crystals will be independent of the above mentioned factors. Moreover, during the 2021 beam time, measurements were performed by monitoring the implanted beam particles, with germanium and $LaBr_3$ detectors, and investigating the decay of the VUV signal as a function of time. However, as the beam distribution on the crystal and the fraction that sits in front of the entrance slit were not determined properly, the total $\beta-$decay branching ratio into the isomer and the spectrometer efficiency were to a large extent correlated and could not be determined with sufficient accuracy. Using short implantation time and investigating the Cherenkov background in our measurement will enable the extraction of the spectrometer efficiency more precisely, and this will in turn lead to a more precise value of the $\beta-$decay branching ratio. With the planned upgrade of the spectrometer and the use of shorter implantation cycles, we will be able to determine these parameters at higher precision.

4.2 Emission channelling

An important remark for our emission channelling results is that the uncertainties obtained for the ^{229}Ac results are larger than the ^{231}Th uncertainties due to systematics stemming from a combination of surface charging effects, a higher $\beta-$energy spectrum, and a larger $\gamma-$radiation background. In addition, the mass A=231 decay chain provides direct measurements of the thorium lattice location. On the other hand, the mass A=229 decay chain is the same as in the VUV spectroscopy experiments; moreover, due to the higher yield and the shorter half-life, data can be accumulated at a much higher rate than for mass A=231, which allows covering a larger phase space (implantation temperature etc.). Therefore, in order to take advantage of this complementarity, we are requesting both ^{229}Ac and ^{231}Ac beams. In our previous experiments, we measured the isomeric decay in CaF_2 and MgF_2 crystals. To further investigate the impact of the lattice location on the possibility to block the isomer's internal conversion decay channel we propose to further study emission channelling in MgF_2 and in thin film CaF_2. This includes measuring after room temperature implantations and optimizing the lattice position by studying the effect of annealing treatment and higher temperature implantation.

4.3 Alternative crystals for VUV spectroscopy

In addition to the main aim, we intend to investigate other crystals with band gaps smaller than the isomer's energy, to investigate the effect of the band gap on the radiative decay probability. On one hand, this will help us understand the interaction between the decaying nucleus and the electrons in the crystal; in particular, it will allow us to test whether or not using a crystal host with a *band gap larger than the isomer's energy* is a necessary condition for blocking internal conversion. On the other hand, these measurements constitute a first test towards application of these crystals for future development of a solid state nuclear clock. Two sets of materials will be considered: CdF_2 (band gap of 8.0 eV [22]) and PbF_2 (5.9 eV [23]); MgO (7.8 eV) and AlN (6.0 eV). Priority will be given to VUV spectroscopy experiments on CdF_2 and PbF_2, since they will provide the intended band gap variation while keeping as many parameters as possible unchanged with respect to our previous work on CaF_2 (CdF_2 and PbF_2 have the same crystal structure as CaF_2). However, if during sample preparation and pre-characterization, issues such as high Cherenkov background or limited availability of high-quality single crystals would emerge, other hosts with a similar variation in band gap, namely MgO and AlN, will be considered.

Summary of requested shifts:

- For RILIS optimization of ^{229}Ac, the optimization of extraction of ^{229}Ac and ^{231}Ac and for purity checks, a total of 3 shifts is requested.
- **For VUV spectroscopy**: A full implantation/measurement cycle of the VUV spectroscopy starts with changing the beam to LA1 which takes about 30 mins followed by an implantation cycle varying from 30 mins to 2 h. This is followed by a measurement cycle lasting about 3-5 h, during which the beam can be given to another user. This corresponds to about 1 implantation/measurement cycle per shift during which we request typically 3 hours of beam.

  For the isomer energy measurement in CaF_2 and MgF_2, 10 implantations are required. For the lifetime measurement in CaF_2 and MgF_2 another 10 implantations are required. 5 additional implantations are required for VUV spectroscopy in CdF_2 and PbF_2 crystals. In the 2021 beam time, we performed 23 implantations during a time period corresponding to 7 days where, other users were using the radioactive beam while we were performing the VUV spectrometry. Taking into consideration a longer implantation/measurement cycle for lifetime measurement a total of 25 implantations for the proposed measurements would correspond to 25 shift, of which about 3 h radioactive beam time per shift is used.
- **Emission channelling**: Implantations for the emission channelling measurements will be done during the measurement cycle of the VUV spectroscopy. Measurements with ^{229}Ac and ^{231}Th in CaF_2 thin films and MgF_2 crystals, at room temperature and after annealing treatments, 15 shifts are requested.

For the VUV spectroscopy of the radiative decay of thorium isomer and the study of lattice incorporation of thorium in different crystals, which can be seen as a key measurements towards a solid state nuclear clock, we request a total of **28 shifts**.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The setup comprises: the EC-SLI online setup, two offline EC setups dedicated to emission channeling experiments and the travelling VUV spectroscopy setup.

Part of the	Availability	Design and manufacturing
Emission Channeling EC-SLI	☒ Existing	☒ To be used without any modification (EC-SLI setup at b.170-GHM)
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Emission Channeling Offline	☒ Existing	☒ To be used without any modification (offline EC setups in b.508-R-008) ☐ To be modified and/or manufacturing
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
VUV Spectroscopy	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

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 experiment/ equipment]	[Part 2 of experiment/ equipment]	[Part 3 of experiment/ equipment]
Thermodynamic and fluidic			
Pressure	NA	NA	NA
Vacuum	1×10^{-6}		
Temperature	NA	NA	NA
Heat transfer	NA	NA	NA
Thermal properties of materials	NA	NA	NA
Cryogenic fluid	NA	NA	NA
Electrical and electromagnetic			
Electricity	230 V, 15 A	230 V, 15 A	230 V, 15 A, 3.5 kV (HPGe detector)
Static electricity	NA	NA	NA
Magnetic field	NA	NA	NA
Batteries	☐	☐	☐
Capacitors	☐	☐	☐

Ionizing radiation			
Target material [material]	CaF_2, MgF_2, CdF_2, PbF_2, MgO, AlN	CaF_2, MgF_2, CdF_2, PbF_2, MgO, AlN	CaF_2, MgF_2, CdF_2, PbF_2, MgO, AlN
Beam particle type (e, p, ions, etc)	ions	ions	ions
Beam intensity	$<5x10^7$	NA	$<5x10^7$
Beam energy	30-60 kV	NA	30 kV
Cooling liquids	NA	NA	LN_2 (HPGe detector)
Gases	NA	NA	N_2
Calibration sources:	☐	☐	☐
• Open source	☐	☐	☐
• Sealed source	☐	☐	☒
• Isotope	NA	NA	^{152}Eu
• Activity	NA	NA	20 kBq
Use of activated material:			
• Description	☐	☐	☐
• Dose rate on contact and in 10 cm distance	NA	NA	NA
• Isotope	NA	NA	NA
• Activity	NA	NA	NA
Non-ionizing radiation			
Laser	NA	NA	NA
UV light	NA	NA	120-400 nm (deuterium lamp)
Microwaves (300MHz-30 GHz)	NA	NA	NA
Radiofrequency (1-300 MHz)	NA	NA	NA
Chemical			
Toxic	NA	NA	NA
Harmful	NA	NA	NA
CMR (carcinogens, mutagens and substances toxic to reproduction)	NA	NA	NA
Corrosive	NA	NA	NA
Irritant	NA	NA	NA
Flammable	NA	NA	NA
Oxidizing	NA	NA	NA
Explosiveness	NA	NA	NA
Asphyxiant	NA	NA	NA
Dangerous for the environment	NA	NA	NA
Mechanical			
Physical impact or mechanical energy (moving parts)	NA	NA	NA
Mechanical properties (Sharp, rough, slippery)	NA	NA	NA
Vibration	NA	NA	NA
Vehicles and Means of Transport	NA	NA	NA
Noise			

Frequency	NA	NA	NA
Intensity	NA	NA	NA
Physical			
Confined spaces	NA	NA	NA
High workplaces	NA	NA	NA
Access to high workplaces	NA	NA	NA
Obstructions in passage-ways	NA	NA	NA
Manual handling	NA	NA	NA
Poor ergonomics	NA	NA	NA

Hazard identification:

Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): < 3kW