EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Quantum emitters in diamond containing octupole-deformed nuclei for electric dipole moment experiments

October 2, 2024

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CERN-INTC-2024-066 / INTC-I-281
02/10/2024

Abstract:

The search for new sources of charge-conjugation-parity (CP) violation is crucial to explain the baryon asymmetry in the universe. One promising approach is the measurement of a permanent electric dipole moment (EDM) in atomic nuclei, particularly using systems sensitive to nuclear Schiff moments. This proposal aims to develop experimental techniques to study CP violation through EDM measurements in solid-state systems. Specifically, we focus on creating and investigating quantum emitters (color centers) in diamond, containing actinide impurities (Pa, Th and Ac) for which octupole-deformed isotopes with large nuclear Schiff moments exist (e.g. ^{229}Pa, ^{229}Th and ^{227}Ac). The proposed experiments consist of studying the structure of the defects using emission channeling and their optical properties with radiotracer photoluminescence, two techniques that are uniquely suited for those purposes and which are unique to ISOLDE. These experiments will serve as the basis for a larger proposal to the INTC and for funding applications dedicated to rare isotope-containing color centers as a new platform for fundamental symmetry tests.

Summary of requested shifts: 7 shifts

1 Physics Motivation

To provide a satisfactory explanation of the observed baryon asymmetry (the excess of matter over antimatter) in the Universe, new sources of charge-conjugation-parity (CP) symmetry violation must exist beyond the mechanisms of the Standard Model [1]. Theories beyond the Standard Model and current experimental constraints indicate that the energy scale for such new physics is beyond the reach of modern particle colliders [2, 3]. The measurement of a permanent electric dipole moment (EDM) within atomic nuclei, induced by the nuclear Schiff moment, has emerged as a potential alternative to probe CP-violation physics [4, 5], complementary to EDM searches in hadrons, leptons, and other approaches [6]. The existence of a permanent EDM implies the breaking of time-reversal symmetry (T) and parity symmetry (P), thus also breaking CP symmetry as a consequency of the CPT theorem [1, 7]. Certain pear-shaped (octupole-deformed) nuclei, such as ^{223}Fr, ^{225}Ra, and ^{229}Pa, show promise for such EDM measurements [4, 5].

The energy shifts caused by CP-odd nuclear moments are amplified in electrically polarized atoms and molecules. This characteristic has prompted various experiments and proposals aimed at investigating nuclear CP violation using polar molecules, whose enormous internal effective electric field can be aligned with well-defined laboratory axes using small external fields [8–12]. Proposals for an alternative experimental approach have also emerged, using atomic ions located in noncentrosymmetric sites within a crystal, as they are highly electrically polarized. Experiments involving nuclei surrounded by polarized ions benefit from increased internal electric fields, similar to those in polar molecules, while achieving greater sensitivity due to the large number of trapped ions that can be interrogated in solid-state samples [13–15].

Experiments with lanthanide ions, such as $^{153}Eu^{3+}$, incorporated in noncentrosymmetric sites within an optical crystal like Y_2SiO_5 (YSO) have been proposed [14], taking advantage of several attractive features of the Eu:YSO system; narrow homogeneous and inhomogeneous linewidths have been observed on the intra-$4f$ ($4f^6\ ^7F_0 \rightarrow 4f^6\ ^5D_0$) transition at 580 nm, the hyperfine and Zeeman interactions of the nuclear sublevels in the $^7F_0$ and $^5D_0$ states have been measured, optical pumping between the nuclear sublevels has been demonstrated, and long-lived coherence between these sublevels has been achieved [14]. On the other hand, actinide isotopes, including ^{229}Pa, ^{229}Th, ^{225}Ac, ^{227}Ac, ^{233}U, ^{235}U, and ^{237}Np, promise even higher EDM sensitivity because the contribution to the atomic EDM due to the Schiff moment strongly scales with Z and A, as seen through the semi-empirical formula [5, 15, 16]:

$$d_{\text{atom}} \propto \frac{\beta_2\,(\beta_3^2)\,Z^3 A^{2/3}}{\Delta E} \tag{1}$$

where β_2 is the quadrupole deformation parameter, β_3 is the octupole deformation parameter, Z is atomic number, A is the mass number, and ΔE is the energy difference between two nuclear levels with same angular momentum and opposite parity.

The optical properties of the intra-$5f$ transitions of actinides are poorly known, and nothing necessarily suggests that they would be as suitable as the intra-$4f$ transitions of some lanthanides. A more recent proposal [17] explores an alternative approach, based on transitions between (ground and excited) electronic states associated with a crystal defect that contains the actinide atom, instead of relying on intra-f transitions (similar to those of the free atoms/ions). Defects exhibiting optical transitions between localized

electronic states that lie within the bandgap of the host crystal, in particular those with high quantum efficiencies, are known as color centers. The proposal outlined in [17] aims particularly at color centers in diamond involving ^{229}Pa. This approach offers several advantages, including high number densities, efficient optical probing, and large internal electric fields for oriented non-inversion symmetric crystal defects in optical crystals [17].

Diamond is a particularly attractive host material for EDM-sensitive isotopes: its high radiation hardness makes it resistant to damage from implantation and the decay of incorporated radioactive isotopes [18, 19]; the wide bandgap (5.5 eV) enhances the likelihood that defect states emerge within the gap, resulting in thousands of optically active defects in diamond [20]; synthetic diamond can be produced free of nuclear spins by using precursors enriched with ^{12}C for chemical vapor deposition growth, thus eliminating an important source of spin decoherence [21, 22]; in situ co-magnetometry in the EDM experiment can be provided by nitrogen-vacancy centers, a well-estabished, highly sensitive quantum magnetometer [23]. The density functional theory (DFT) calculations presented in [17] indicate that ^{229}Pa can form optically active color centers in diamond, specifically in the form of PaV_2 defects. Depending on the Fermi level (E_F), and considering the usual E_F range in diamond samples, these PaV_2 defects may be found in one of two stable charge states, -1 and -2. For the -2 charge state of the PaV_2 defect, the zero-phonon line (ZPL) is predicted to be in the visible range, around 553 nm, while for the -1 charge state, the ZPL is predicted to be in the infrared range, around 1294 nm. These ZPL values correspond to transitions between specific spin states that are favorable for the search of a nuclear Schiff moment, based on RF spectroscopy, applying well-developed quantum control schemes [17].

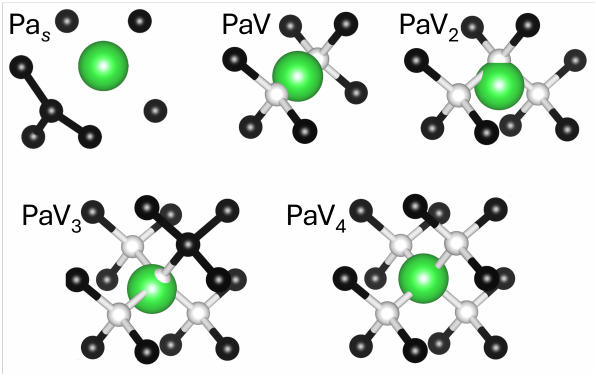

Figure 1: Defect structures simulated in [17]: substitutional Pa without neighboring vacancies (Pa_s) as well as with one (PaV), two (PaV_2), three (PaV_3) and four (PaV_4) neighboring vacancies. The green and black atom correspond to Pa and C, respectively. The white atoms represent vacancies. The apparent extra vacancy is due to the Pa atom not being in a substitutional site.

2 Experimental Technique and setup

The next crucial steps in the development of this approach to EDM search are (i) to investigate if and how these PaV_2 defects can be created, in particular, by ^{229}Pa ion implantation into diamond, and (ii) to investigate if the optical transitions predicted in [17] exist and how they can be optimized for the purpose of the EDM measurements (e.g. via post-implantation annealing for optical activation, electrical co-doping for charge-state stabilization, etc.). Here we propose to do so using emission channeling (EC) and radiotracer photoluminescence (rPL), two techniques that are uniquely suited for that

purpose and which are unique to ISOLDE.

Emission channeling is a powerful technique to directly and quantitatively determine which lattice sites are occupied by implanted impurities. This is particularly critical in this context, because only noncentrosymmetric sites, as in the case of the PaV_2 defect according to the DFT calculations in [17] (fig. 1), are suitable for EDM-sensitive experiments. However, such defects have never been identified in diamond. The defects that are commonly studied in the context of color centers in diamond contain only one vacancy, e.g. NV, MgV, GeV, SnV centers. However, for most impurities, e.g. as we have shown for SnV [24], GeV [25] and MgV [26] and as predicted for Pa in [17], when the defect contains only one vacancy, the impurity relaxes to the center of the bond, occupying the so-called bond-centered (BC) side, and the defect becomes centrosymmetric. Higher order defects such as PaV_2 have never been reported. In fact, arguably only EC would be able to detect such defects, especially if coexisting with other configurations such as Pa_s and PaV. This is perfectly illustrated by measurements that we recently performed as a feasibility test for the present LOI: a brief EC experiment on the lattice location of ^{229}Ac in diamond, which we could perform with minimal effort with a few-minute implantation during a run dedicated to experiment IS715, where this beam is used to study the nuclear clock ^{229m}Th isomer. These EC experiments reveal the coexistence of various sites, namely S (11%), BC (23%), near-CO (33%) and near-ABO (33%) (fig. 2), which can be attributed to Pa_s/PaV_4, PaV, PaV_2 and PaV_3, respectively, based on the DFT calculations in [17]. This is quite a remarkable result on its own, considering that despite our extensive EC studies of numerous impurities in diamond over the last years in the context of experiment IS668 (Na, Mg, Ca, Sr, Ni, Ga, As, He, Ne, Kr, Xe, Ge, Sn and Pb), this is the first time we clearly identify the occupation of a lattice site other than the substitutional and bond-centered (i.e. with zero and one vancancy). Interpreting these results for Ac in the context of our previous work on diamond and the DFC calculations in [17] suggests that the formation of defects with more than one vacancy is favored for impurities that are heavy (high atomic number) and consist of a transition element. These results are thus highly encouraging for prospects of creating non-centrosymmetric defects in diamond containing actinide impurities (heavy

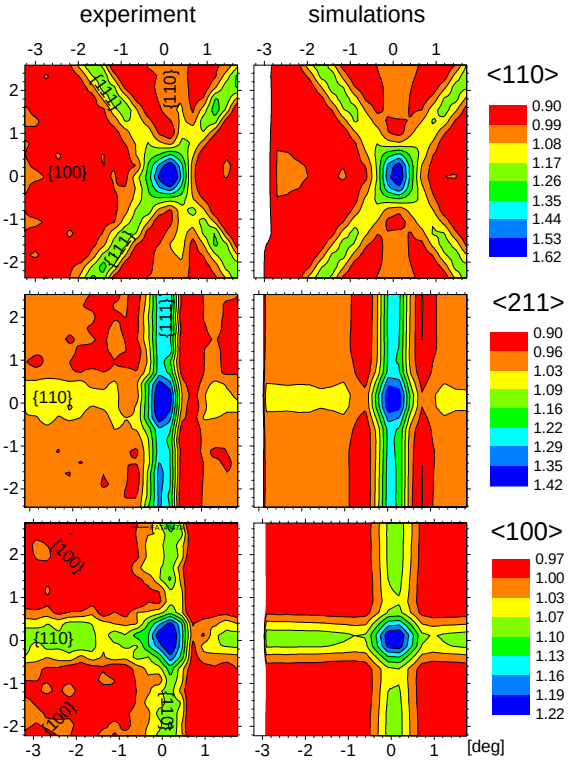

Figure 2: (*Left*) Normalized experimental β^- emission channeling patterns of ^{229}Ac in diamond, following annealing at 900 °C, in the vicinity of three major crystallographic axes: ⟨110⟩, ⟨211⟩, and ⟨100⟩. (*Right*) Best fits with simulated patterns corresponding to a combination of S (11%), BC (23%), near-CO (33%) and near-ABO (33%) sites.

transition elements) for EDM experiments.

Forming the non-centrosymmetric defects is however not sufficient. They must also be optically active, i.e. exhibit photoluminescence lines such as those predicted for PaV_2 in [17]. Investigating these effects is, however, also far from trivial. Not only are these samples radioactive, posing safety and radioprotection constraints, ion implantation can lead to the appearance of optically defects (i.e. of PL lines) that do not involve the implanted impurities themselves (e.g. by creating or activating other defects and impurities). Radiotracer PL is specifically designed to overcome this limitation of standard PL: using a radioactive isotope and recording the time dependence of the intensity of the PL lines allows to correlate them with the (half-life of the) parent or daughter isotopes. The intensity of PL lines from defects that do involve parent/daughter isotopes decreases/increases according to the half-life; defects that do not involve the probe isotope (and are thus not relevant for the purpose of the experiment) do not exhibit a time dependence. In the context of experiment IS668, we have recently completed the construction of an rPL setup specifically optimized for studying color centers in diamond, located at the newly created *Quantum Photonics Lab* in Building 508.

3 Proposed Experiment

We propose to use $A = 229$ and $A = 231$ beams (surface-ionized $^{229}Ra^+$, $^{229}Fr^+$, $^{231}Ra^+$, $^{231}Fr^+$ from a ThC_x target) to perform an exploratory study on the formation of color centers in diamond containing Pa, Th, and Ac impurities, in particular studying the structure of the defects with emission channeling and their optical properties with radiotracer PL. These results would serve as the basis for a larger proposal to the INTC and for funding proposals dedicated to further developing a method or methods to study *CP*-violation physics using these color centers as a platform.

^{229}Pa is an important candidate isotope, predicted to provide over six orders of magnitude higher sensitivity than the current experimental limit on EDM measurements taken with ^{199}Hg [15, 27]. However, the limited global production of ^{229}Pa constitutes a practical challenge. The newly opened Facility for Rare Isotope Beams (FRIBs) is expected to produce a significant amount of ^{229}Pa within the next ten years [28], and (molecular) beams of ^{229}Pa will also be developed in the coming years at ISOLDE within the project "Fundamental Physics Research with Radioactive Molecules", funded via CERN's Physics Beyond Colliders (PBC) group. Still, the only advantage of ^{229}Pa over other actinide isotopes such as ^{229}Th, ^{227}Ac and ^{225}Ac is the expected higher sensitivity (one order of magnitude) assuming that a level of opposite parity and the same angular momentum exists very close to the ground state (i.e. an exceptionally small $\Delta E = 60$ eV in equation 1) exists. The latest measurement yielded a position of this level at 60 ± 50 eV, but with this large uncertainty the existence of the parity doublet is not certain [29]. As long as we do not know if the parity doublet exists or has such a low ΔE, and if suitable beams can be produced, several other actinides such as ^{229}Th, ^{227}Ac and ^{225}Ac are also highly promising. Therefore, this proposal deals with color centers involving not only Pa, but also Th and Ac impurities. In fact, ^{229}Th ($t_{1/2} = 7880$ y), ^{227}Ac ($t_{1/2} = 22$ y) and ^{225}Ac ($t_{1/2} = 10$ d) offer an important advantage of their own that may even compensate for an eventually lower EDM senstivity: they are all longer lived than ^{229}Pa, whose half-life of 1.5 days poses strong constraints on the time available to prepare the samples and carry out

the measurements. The extremely long half-lives of ^{229}Th and ^{227}Ac would allow for the implantation to be performed at a RIB facility such as ISOLDE and post-processed and measured elsewhere. For example, high-temperature and high-pressure post-annealing will likely be required to produce the best optical properties of the actinide-implanted diamond samples [30, 31], which requires specialized equipment and expertise. Moreover, the ability to ship samples for EDM measurements elsewhere would more easily allow for different groups/collaborations to apply different variations of the color-center-based EDM experiment; ^{229}Pa ($t_{1/2}$ = 1.5 d) would require those experiments to be located at or very close to the RIB facility. The extremely long half-lives also imply much lower activity, which is also convenient in terms of safety and radioprotection constraints. ^{229}Th, ^{227}Ac and ^{225}Ac also have a very practical advantage: they can already be produced at ISOLDE. In addition, the EC and rPL experiments proposed here rely on ^{229}Th, ^{229}Ac, ^{231}Pa, and ^{231}Th, all of which can be obtained in sufficient yields. In fact, the experimental run for this LOI could be seamlessly combined with the upcoming campaign in 2025 for experiment IS715 (dedicated to the nuclear clock ^{229m}Th isomer), using exactly the same beams and experimental conditions, i.e. implanting surface-ionized ^{229}Ra$^+$, ^{229}Fr$^+$, ^{231}Ra$^+$ and ^{231}Fr$^+$, obtained from a ThC$_x$ target, which decay to the desired isotopes. Finally, if the experiments proposed in this LOI are successful, the ensuing proposal could even survey other actinides such as U and Np, since ^{233}U, ^{235}U, and ^{237}Np are predicted to lead to equally large EDMs, have even longer half-lives than ^{229}Th, and can be easily produced at ISOLDE.

Concretely, the experiments would consist of:

- **EC experiments using $A = 229$ beam:** After the implantation in the online EC setup (EC-SLI in GHM), we wait for the decay of ^{229}Fr ($t_{1/2}$ = 50 s) and ^{229}Ra ($t_{1/2}$ = 4 min), at least 5 half-lives of ^{229}Ra (20 minutes), and then measure EC patterns from the β^- particles emitted upon decay of ^{229}Ac ($t_{1/2}$ = 63 min), i.e. yielding the lattice location of Ac.

- **EC experiments using $A = 231$ beam:** After the implantation using the collection chamber in GLM, we wait for the decay of ^{231}Fr ($t_{1/2}$ = 18 s), ^{231}Ra ($t_{1/2}$ = 104 s), and ^{231}Ac ($t_{1/2}$ = 7.5 min), at least 5 half-lives of ^{231}Ac (38 minutes), and then measure EC patterns from the β^- and conversion electrons emitted upon decay of ^{231}Th. The β^- component yields the lattice location of ^{231}Th, whereas the CE component yields the lattice location of ^{231}Pa.

- **rPL experiments using $A = 229$ beam:** After the implantation using the collection chamber in GLM, we wait for the decay of ^{229}Fr, ^{229}Ra, and ^{229}Ac, until the activity is sufficiently low for the sample to be safely processed and measured offline, at which stage the PL measurements will yield PL lines associated with eventual color centers involving ^{229}Th. Color centers involving ^{229}Ac (and their decaying intensity) may still be detectable then, despite the waiting time and the time required for sample processing before the PL measurement.

- **rPL experiments using $A = 231$ beam:** After the implantation using the collection chamber in GLM, we wait for the decay of ^{231}Fr, ^{231}Ra, and ^{231}Ac, until the activity is sufficiently low for the sample to be safely processed and measured offline, at which stage the PL measurements will yield PL lines associated with eventual

color centers involving both ^{231}Th and ^{231}Pa. The time dependence of the intensity of the observed PL lines indicates if the respective color center involves Th (if decreasing), Pa (if increasing) or neither (if constant).

This measurement plan is similar to that used for experiment IS715 (nuclear clock ^{229m}Th isomer); those experiments allowed us to carefully optimize the finer details (implantation times, waiting times for decay of the percursor isotopes, etc.). The main differences are: (i) this LOI involves implantation into diamond (similar to experiment IS668 on color centers in diamond), whereas IS715 dealt with crystals with larger bandgaps (e.g. CaF_2 and MgF_2); (ii) this LOI uses rPL instead of VUV spectroscopy.

These measurements will be performed as a function of annealing temperature, which is one of the optimization variables. A higher annealing temperature normally improves the optical properties (increasing the activation and brightness of the color centers). However, it is possible that the defect(s) most suitable for the EDM experiments, i.e. the non-centrosymmetric configurations (with two and three vacancies), are less stable against annealing than the symmetric ones (with zero or one vacancy). In such a scenario, the optimal annealing temperature would correspond to a compromise between the stability of the desired defect and its optical performance.

Summary of requested shifts:

- ***A* = 229 beam.** Two EC experiments on the lattice location of ^{229}Ac will be performed in the EC-SLI (online) setup, in the 1×10^{12} and 1×10^{13} cm^{-2} fluence ranges, in the as-implanted state and after annealing at various temperatures (up to 900 °C *in-situ*, up to 1500 °C *ex-situ*). For each experiment, this corresponds to about 6 implantation/annealing/measurement cycles of approximately 4 hours each: 1-2 hours for implantation, wait for decay, and annealing; 2-3 hours for measurement. These two online experiments thus add up to about **48 hours, 6 shifts**. The other experiments described in the following will use the beam while it is not used by these online EC experiments. With mass 229, these correspond to short collections for the rPL experiments on ^{229}Ac/^{229}Th.

- ***A* = 231 beam.** Two EC experiments on the lattice location of ^{231}Th and ^{231}Pa will be performed offline, in the 1×10^{12} and 5×10^{12} cm^{-2} fluence ranges, in the as-implanted state and after annealing at various temperatures (up to 1500 °C). These collections will be performed incrementally between the EC-SLI experiments described above, and will take up to 15 hours. Two additional samples will be implanted under the same conditions for the rPL experiments on ^{231}Th/^{231}Pa. This results in a total collection time of 30 hours, which approximately matches the amount of time available between the EC-SLI experiments. In experiment IS715 (dedicated to the nuclear clock ^{229m}Th isomer), we have successfully used a similar workflow, that perfectly optimizes the use of the protons, i.e. the radioative beam is used almost without interruptions.

- **Stable beam.** We request **1 shift** of stable ^{138}Ba beam to optimize the transmission through a 1 mm collimator, both at GHM and GLM, as required from the proposed experiments.

We therefore request a total of **7 shifts**.

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4 Details for the Technical Advisory Committee

3.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

⊠ Permanent ISOLDE setup: *EC-SLI and Quantum Photonics Lab*

⊠ To be used without any modification

☐ To be modified: *Short description of required modifications.*

☐ Travelling setup (*Contact the ISOLDE physics coordinator with details.*)

☐ Existing setup, used previously at ISOLDE: *Specify name and IS-number(s)*

☐ Existing setup, not yet used at ISOLDE: *Short description*

☐ New setup: *Short description*

3.2 Beam production

For any inquiries related to this matter, reach out to the target team and/or RILIS (please do not wait until the last minute!). For Letters of Intent focusing on element (or isotope) specific beam development, this section can be filled in more loosely.

- Requested beams:

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
229Fr/229Ra 231Fr/231Ra	1E+8 1E+6	2E+7 2E+5	50 s / 4 min 18 s / 104 s

- Full reference of yield information:

  We successfully used exactly the same conditions in the last two runs of experiment IS715.

- Target - ion source combination:

ThC target, surface ionization MK1 Ta

- RILIS?

  Not necessary, but it can be scheduled together with an experiment (e.g. for IS715) that requests RILIS.

  ☐ Special requirements: none

- Additional features?

  ☐ Neutron converter: no

  ☐ Other: no

- Expected contaminants: *Isotopes and yields*

  From previous runs of IS715, we are aware of the level of contamination with lower masses (down to mass 227), and these are acceptable

- Acceptable level of contaminants:

  From previous runs of IS715, we are aware of the level of contamination with lower masses (down to mass 227), and these are acceptable

- Can the experiment accept molecular beams?

  Yes

- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of?

  Yes, this experiment can and should be scheduled together with IS715 runs.

3.3 HIE-ISOLDE

For any inquiries related to this matter, reach out to the ISOLDE machine supervisors (please do not wait until the last minute!).

- HIE ISOLDE Energy: *(MeV/u); (exact energy or acceptable energy range)*

  ☐ Precise energy determination required

  ☐ Requires stable beam from REX-EBIS for calibration/setup? *Isotope?*

- REX-EBIS timing

  ☐ Slow extraction

  ☐ Other timing requests

- Which beam diagnostics are available in the setup?
- What is the vacuum level achievable in your setup?

3.4 Shift breakdown

The beam request only includes the shifts requiring radioactive beam, but, for practical purposes, an overview of all the shifts is requested here. Don't forget to include:

- Isotopes/isomers for which the yield need to be determined
- Shifts requiring stable beam (indicate which isotopes, if important) for setup, calibration, etc. Also include if stable beam from the REX-EBIS is required.

Summary of requested shifts:

With protons	Requested shifts
Data taking with 229Fr/229Ra beam Data taking with 231Fr/231Ra beam	3 3
Without protons	Requested shifts
Stable beam using ^{138}Ba	1

3.5 Health, Safety and Environmental aspects

3.5.1 Radiation Protection

- If radioactive sources are required:

  (none required)

  - Purpose?
  - Isotopic composition?
  - Activity?

- Sealed/unsealed?

- For collections:
  - Number of samples?

    2 samples implanted with mass 229, for rPL offline

    4 samples implanted with mass 231: 2 for offline EC, 2 for offline rPL

  - Activity/atoms implanted per sample?

    1 sample with 1E10 atoms of ^{229}Ac (<2 MBq)

    1 sample with 1E9 atoms of ^{229}Ac (<0.2 MBq)

    2 samples with 5E10 atoms of ^{231}Th (< 0.5 MBq)

    2 samples with 1E10 atoms of ^{231}Th (< 0.1 MBq)

  - Post-collection activities? (*handling, measurements, shipping, etc.*)

    The post-collection activities for EC and PL experiments have been documented during previous beam times and can be accessed via EDMS:
    https://edms.cern.ch/nav/P:CERN-0000159974:V0/P:CERN-0000255393:V0

    The handling and operation of EC-SLi are comprehensively detailed here:
    https://edms.cern.ch/nav/P:CERN-0000159974:V0/P:CERN-0000159974:V0

    For QPL handling and operation, refer to:
    https://edms.cern.ch/nav/P:CERN-0000159971:V0/P:CERN-0000255421:V0

    All samples mentioned in this LOI are solid diamond single crystals, making their collection and handling straightforward and safe. Before each beam time, all steps are thoroughly reviewed and detailed with RP and safety contacts at ISOLDE as part of the IMPACT authorization process.

3.5.2 Only for traveling setups

(no traveling setups will be used)