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RESonance laser Ionisation techniques for SeparaTors (RESIST)

Deliverable 12.5 – Final Report on Task 1

PRE-LIST TECHNIQUES TO ENHANCE ION BEAM PURITY

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LIST OF ACRONYMS AND ABBREVIATIONS

RESIST	RESonance laser Ionisation techniques for SeparaTors
RILIS	Resonance Ionisation Laser Ion Source
IGLIS	In-Gas Laser Ionisation and Spectroscopy
LIST	Laser Ion Source and Trap
RIB	Radioactive Ion Beam

ISOL	Isotope Separator On-Line
RIS	Resonance Ionisation Spectroscopy

EXECUTIVE SUMMARY

This report summarises the final deliverable due in month 48 of ENSAR-2. It focuses on the status of Task 1 of the RESIST Joint Research Activity.

INTRODUCTION

The RESIST (RESonance laser Ionisation techniques for SeparaTors) Joint Research Activity of ENSAR2 aims to refine the highly successful Resonance Ionisation Laser Ion Source (RILIS), the In-Gas Laser Ionisation and Spectroscopy (IGLIS) and Laser Ion Source Trap (LIST) technologies. These techniques are coupled to both approaches currently in use, i.e. hot cavities and gas cells at on-line facilities, for the production and study of radioactive ion beams (RIBs).

Our goal is to obtain RIBs of the highest purity regarding selection of isobaric as well as isomeric species, which otherwise are indistinguishable for conventional mass spectrometers. We propose to implement, for the first time, laser techniques not only at « traditional » ISOL facilities, but also at existing and upcoming in-flight facilities. This approach will enable dedicated studies on the shortest-lived exotic and specifically refractory isotopes far from stability and will result in spectroscopy of exceptional sensitivity and significance, aiming at the coupling of new laser systems (of dramatically reduced linewidth) which are tailored to exploit optimised experimental environments. Radionuclides produced in the rarest quantities will thus be accessed, extending and complementing earlier nuclear and laser spectroscopic techniques.

Task 1 focuses on efforts to enhance the ion-beam purity prior to the so-called LIST devices and is led by ISOLDE CERN, with contributions from JOGU Mainz, KU Leuven, GANIL and INFN. This task aims to find solutions for the optimal configuration of electrical fields to reduce non-resonant ionisation processes when coupling the LIST with the target, both in hot-cavity ion sources as well as gas-cell based sources, and to investigate low work-function materials resistive to the high-temperature conditions. Task 1 has three deliverables, namely a series of reports due at the end of the first 12 months, after 24 months and finally after 48 months. This task is divided into three subtasks.

This document herein reports on the total period of activity of the RESIST partners involved in Task 1. We focus on those results from the task deemed most important in terms of application to current and/or future Transnational Access facilities. We refer to Deliverable 12.1 (reported at the end of the first 12-month period) for a more detailed background to the different subtasks involving all partners.

SUBTASK 1: A REDUCTION OF SECONDARY ELECTRON IONISATION PROCESSES PRE-RF STRUCTURE. MINIMISATION OF RADIOISOTOPE DEPOSITION ON THE RF STRUCTURE WHICH LEADS TO ISOTOPE-DEPENDENT IONISATION MECHANISMS

<u>JOGU Mainz: Implementation of Fast Beam Gating and the Perpendicular-Illuminated Laser Ion Source and Trap (PI-LIST)</u>

The Laser Ion Source and Trap (LIST) is a tailored upgrade of the well-established, highly element-selective resonance ionisation laser ion source RILIS. It comprises additional suppression of non-laser ionised contaminants produced in the hot atomisation cavity immediately at its exit by an electrostatic repelling electrode, providing a clean laser – atom interaction volume within a radially confining rf quadrupole structure. During previous application of the LIST, electron impact-induced ionisation processes inside the rf structure had been identified as an additional source of contamination. In Deliverable 12.1, we highlighted the effect of introducing a second repelling electrode operated on negative potential to suppress electrons from entering the LIST, which are created by the hot source material [1].

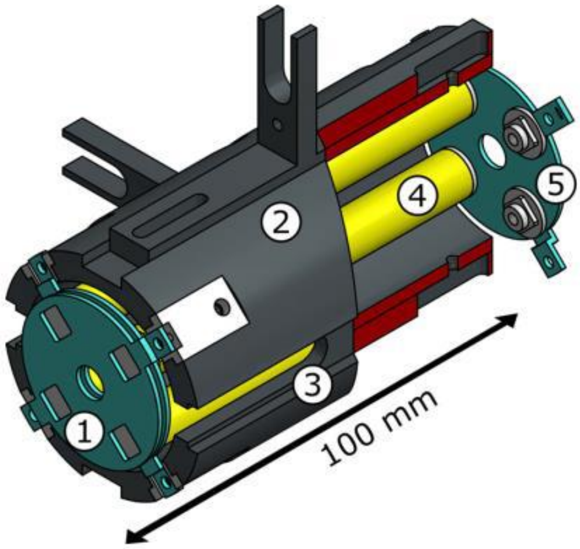

Figure 1: Technical drawing of the newly developed Laser Ion Source and Trap comprising two repelling electrodes (1), housing (2), side cuts for perpendicular illuminations (3), four rods forming the rf-quadrupole structure (4), and exit electrode (5) [1].

On-line operation of such a LIST unit, additionally comprising reduced surface deposition by a reduced overall length, was foreseen at CERN-ISOLDE during the physics campaign in 2018 for the production of pure Mg RIBs. However, in the preliminary tests of the version of shortened LIST the efficiency of ionisation was an order of magnitude lower than expected. Therefore, a double-repeller LIST with usual length of quadrupole electrodes (see Fig. 1) was applied in the on-line experiment IS614 for measurement of the super-allowed branching ratio of ^{22}Mg. The experiment was successful due to the high isobaric purity of the ion beam ensured by the LIST (see Fig. 2). The work on LIST development is continuing toward improving its reliability. In particular, a new design of insulators for fixing the LIST electrodes is being implemented at ISOLDE.

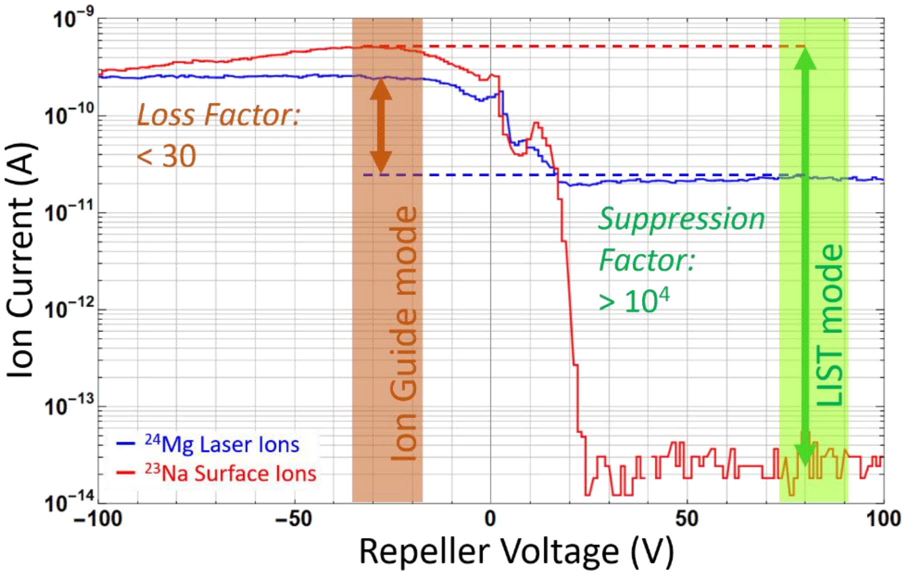

Figure 2: Ion currents of laser-ionised ^{24}Mg and surface-ionised ^{23}Na versus the repeller voltage measured with the LIST unit used for the IS614 experiment in June 2018 at ISOLDE.

For further surface ion suppression and preservation of the desired nuclides laser-ionised in the hot cavity before entering the LIST structure, which would otherwise be lost at the repelling electrode, a fast polarity switch to control the heating current applied to the atomiser is currently under development [2]. The voltage gradient induced by the heating current may then either guide ions out towards the extraction or back into the source, where they can re-neutralise on the walls. Therefore, four high current IGBT switches in an H-bridge configuration operated by home-built TTL-controllable electronics allow in-situ polarity switching of up to 100 A on a sub-µs timescale. Adaption of this system to higher currents used at on-line facilities is ongoing. Later on, this development will be utilised to improve the performance of time-of-flight technique-based laser-ion-source concepts [3].

More recently, a significant reduction of beam contamination from interfering species in the Laser Ion Source as well as the LIST unit has been obtained by rapid switching of a beam gate in perfect synchronisation with the sequence of laser pulses. This technology was demonstrated in holmium to enhance the suppression of dominant stable isotopes and long-lived isomers with respect to a desired radioisotope. The pronounced tailing of the mass peak towards lower mass, caused by ionisation processes taking place inside the acceleration field and leading to interfering contributions on lower masses, as visible for a sample of ^{163,166,166m}Ho in Fig. 3 (red line), could be strongly reduced (green line). A gain in selectivity and corresponding ion-beam purity by up to two orders of magnitude was achieved depending on the load of higher mass ions escaping from the hot cavity and LIST structure.

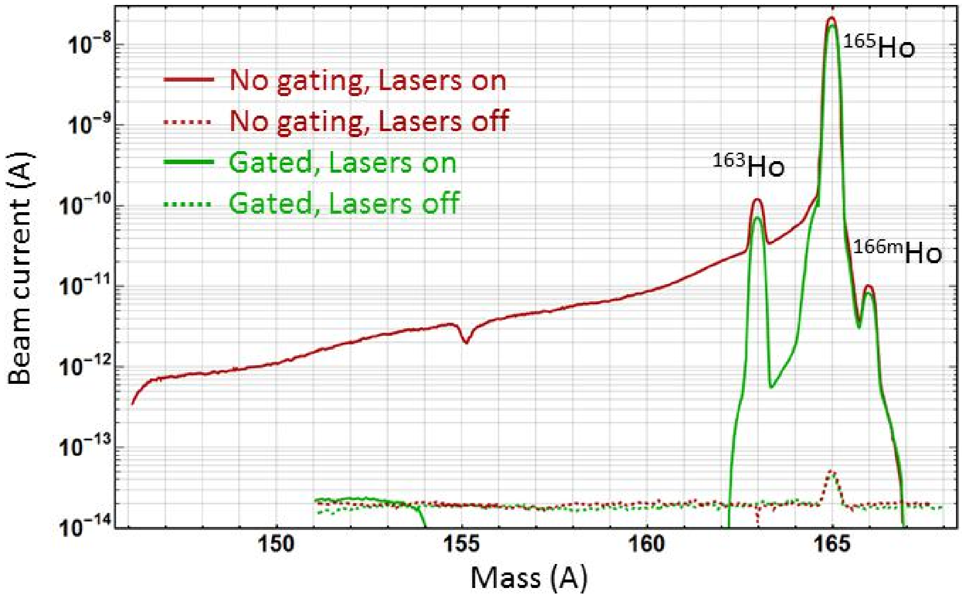

Figure 3: Mass scans in the holmium mass region illustrating the standard operation (red line) and fast ion-beam gating (green line) on a sample of ^{163,165,166m}Ho. Non-specific background at the lower mass side stemming from ionisation in the acceleration region is clearly suppressed (dotted lines show negligible surface-ionisation background).

[1] R. Heinke *et al*., Hyperfine Interact. **238** (2017) 6
[2] S. Rothe *et al*., Nucl. Instr. Meth. B **376** (2016) 86
[3] V.I. Mishin *et al*., AIP Conference Proceedings **1104** (2009) 207

SUBTASK 2: OPTIMISATION OF LASER-IONISATION GEOMETRY IN THE GAS JET TO MINIMISE OR EVEN PRECLUDE PHOTO-ION CREATION IN THE GAS CELL VOLUME AND MINIMISING THE DEPOSITION OF RADIOISOTOPES ON THE RF STRUCTURE

We refer to Deliverable 12.1 for the introduction to this topic. This work is related to Subtask 1.2 and the development of an off-line setup for gas-jet research in the IGLIS laboratory of KU Leuven. In a former report (Deliverable 12.2, Section 2), we studied the longitudinal ion-beam emittance, energy spread and transport efficiency after ion extraction from the radiofrequency quadrupole (RFQ) ion guides used in the IGLIS laboratory. The RFQ ion guides are important devices to transport efficiently the laser-ionised species in the gas jet at a pressure $p \approx 10^{-1}$ mbar, up to the acceleration region at a pressure $p \approx 10^{-5}$ mbar. A maximum transmission efficiency of 91(7)% has been achieved and a longitudinal energy spread of 1.9(8) eV was reported [1]. Owing to ion-atom collisions within the first RFQ structure, the original ion-beam distribution is cooled down improving thus the transverse ion-beam emittance ε. In recent studies, presented here, we were able to determine ε and fully characterise the optical properties of the extracted ion beams.

Figure 1 shows schematically the gas cell and the electrode configuration in the IGLIS beam line used in these studies. Laser-ionised copper atoms were extracted from the gas cell and transported through the S-shaped (S)-RFQ. This is followed by the differential pumping (DP)-RFQ and finally the last ion-guide (IG)-RFQ structure. Leaving the IG-RFQ, the ions were accelerated to an energy of 40 keV and focused by means of an Einzel lens onto an MCP detector inserted in the path of the beam.

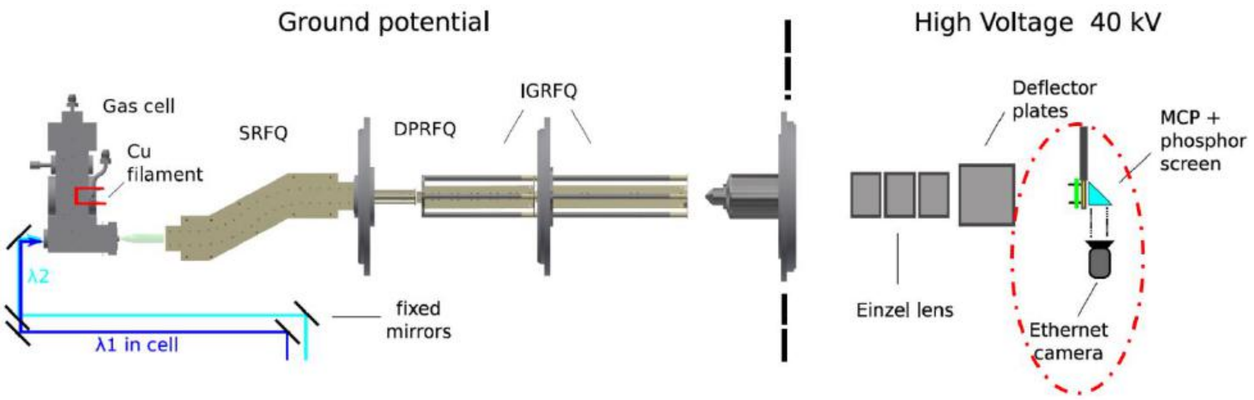

Figure 4: Schematic representation of the IGLIS setup at KU Leuven used to determine the transverse emittance of the ion beam extracted from the RFQ ion guides.

A phosphor screen, serving as the MCP anode, was used to produce an image of the ion beam, which was recorded by a CCD camera. From a series of recorded beam images (see Fig. 5, left), each of them for a different voltage at the Einzel lens, the spatial distribution of the beam was obtained. Fitting the spatial distributions as a function of the voltage at the Einzel lens (see Fig. 5, right), along with the results obtained in simulations of the focusing properties of the Einzel lens, the ion beam emittance could be determined, as explained in [2]. From these studies the beam emittance was found to be ε_x= 2.5(2) π mm mrad for the horizontal component and ε_y= 2.8(2) π mm mrad for the vertical component.

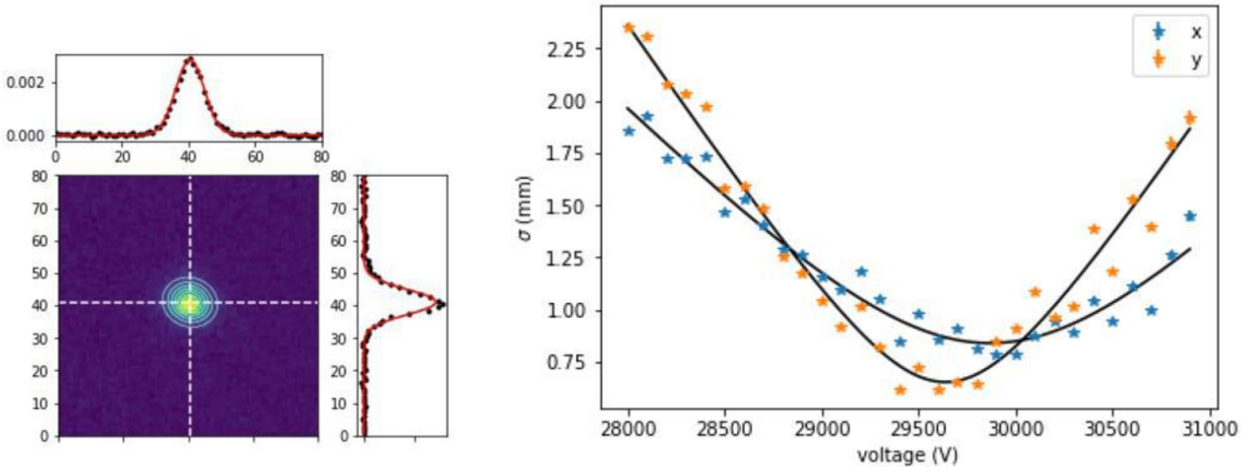

Figure 5: (Left) Image of the beam spot on the phosphor screen recorded by a CCD camera and the corresponding fit of the Gaussian intensity distributions in the vertical and horizontal dimensions. (Right) Spatial width of the beam spots for the vertical and horizontal components as a function of the Einzel lens voltage.

[1] S. Sels *et al*., Nucl. Instrum. and Meth. **B 463** (2020) 148

[2] R. Ferrar et al., NIM **A 735** (2014) 382

SUBTASK 3: HIGH TEMPERATURE MATERIALS RESEARCH FOR THE TRANSFER LINE AND THE LASER IONISATION CAVITY FOR SURFACE ION SUPPRESSION

<u>GANIL: Studies at the GISELE test bench</u>

The off-line RILIS test bench at GANIL, named GISELE (GANIL Ion Source using Electron Laser Excitation) was built to study ionisation processes and to optimise ion source parameters, in particular for surface ion suppression. A new ion source prototype, LISBET (Laser Ion Source Body using Efficient Techniques) has been designed to be a modular system in order to investigate different experimental approaches by varying the design parameters. It consists of two tantalum tubes forming a 90° elbow shape, used as a transfer/atomiser cavity and an ionisation tube where the laser light interacts with the atomic vapour. Both tubes are resistively heated by an electrical current to high temperatures of about 2000 K. The design allows an independent selection of the polarity and the magnitude of the heating currents.

The efficiency, the emittance and the time structure were analysed with a Sn ion beam as a function of the ioniser tube diameter and length. An increase of the ioniser diameter substantially improved the ion beam production. The electric potential gradient along the tube, generated by the heating current, together with the reduction of neutralisation due to the decrease of the contact surface, produced an increase of the ion extraction. In the case of the ioniser diameter, an efficiency of approximately 24% and 8% was obtained for 7 mm and 3-mm diameter ionisers, respectively. The simulations showed that an ioniser with a diameter of 7 mm should provide the best transport efficiency. An increase in the ioniser length was expected to ameliorate the efficiency, however, no improvement was observed.

In order to study and optimise the selectivity of the ion production, two methods have been studied to decrease the production of contaminants (due to the surface ionisation) with an alkali marker (Rb). The first technique is based on changing the electric field potential directions in the transfer tube and/or the ionisation tube. The option in which the electrical potentials are directed towards the entrance aperture for the transfer tube and towards the exit aperture for the ionisation tube was found to yield the highest reduction in contamination without compromising the production rate. The second technique utilised the insertion of ZrC ceramics into the ioniser tube. Further reduction of contamination at high temperatures was expected in this case. As described in the literature, ZrC materials could have a low work function property that diminishes the alkali contamination. However, no significant improvement was found with the inserted material configuration. Preliminary work on the microstructure of ZrC has to be carried out in order to reduce the work function before new investigations continue.

LNL-INFN: Cavity development for isobaric suppression

Taking as a reference the techniques developed at RILIS (ISOLDE), the team of engineers in charge of developing the ionisation source for SPES, Legnaro, recently started a dedicated study based on Finite Element Methods (FEM) simulations, in order to optimise the ionisation efficiency of the SPES RIB. In particular, the study of the temperature distribution inside the laser cavity should be uniform along the cavity line. This aspect is paramount to avoid the presence of cold zones, which become condensation surfaces (traps) for the isotopes coming from the production target.

U-Mainz: Optimisation of temperature distribution and reliability of the laser-ion-source hot cavity

For thoroughly optimising design details, materials and heat distribution in the hot-cavity ion source, heat transport and dissipation was analysed by a finite-element code (Fig. 6a). This refinement led to a further gain in reliability and reproducibility of the hot-cavity laser-ion-source unit. Holmium was used to test the system, an element which is of particular interest as the radioactive isotope ^{163}Ho is a prime candidate for calorimetric measurements of the electron-neutrino rest mass. Initial measurements performed using stable ^{165}Ho at the RISIKO separator have been reported [1]. More recently, the previously measured overall ionisation efficiency (from 2015) of 41(6) %, (Fig. 6b), has been verified.

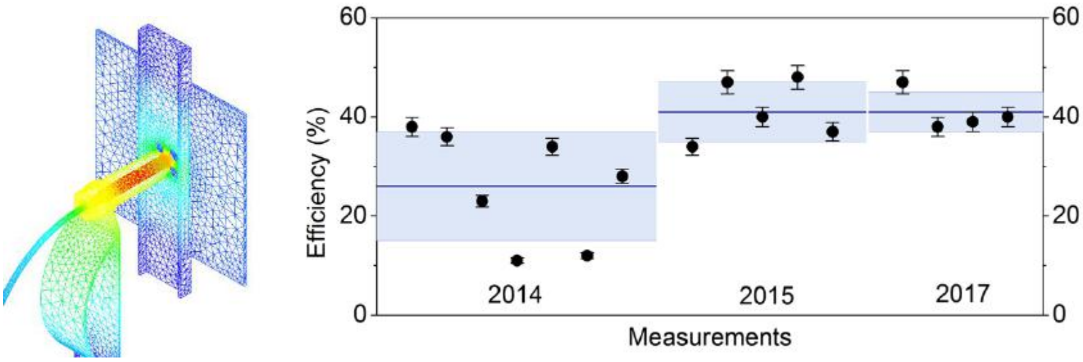

Figure 6: (Left figure) Finite-element simulation of the hot-cavity laser-ion source; (right figure) development of experimental ionisation efficiencies for holmium with reproducibility achieved in recent years (grey range).

[1] F. Schneider *et al.*, Nucl. Instrum. and Meth. **B 376** (2016) 388

CERN-ISOLDE: Low-work-function materials

At CERN, studies of ionisation using low-work-function materials were performed in the context of negative-ion-beam developments. A promising candidate for the MK4 negative-ion source is strontium vanadate with a predicted work function of about 1.79 eV. Different phases of $SrVO_3$ were synthesised in-house, identified via X-ray diffraction and tested against an ioniser made of LaB_6 at the ion source test stand. In the tests, LaB_6 was shown to perform better than $SrVO_3$. This may be due to the used material being in powder form and therefore having different crystal orientations, or a possibility that residuals of other compounds affected the measurement, i.e. the work function was higher than expected for pure crystalline form. Further tests comparing powders to crystals are planned.

As an alternative material, GdB_6 was tested, performing much worse than predicted by its literature work function of 1.5 eV. Electron emission measurements at the ion source working conditions typical for ISOLDE targets confirmed a lower value of the work function of LaB_6 relative to GdB_6. Therefore, to date, LaB_6 is the preferred ioniser due to its better stability and performance. Based on these findings, a target unit with a transfer-line insert made of LaB_6 was applied in an experiment with negative ions. This material is also expected to be efficient in suppression of positive surface ions when used in combination with the resonance laser-ionisation method.

Conclusion

Resonant laser ionisation enjoys a dominant position as the most fruitful and in-demand ionisation mechanism for the production of radioactive ion beams worldwide. The partners within the RESIST collaboration bring complementary expertise to the forefront of this field, developing advanced techniques not only to improve the efficiency and selectivity of RIB production, but also to use state-of-the-art laser systems for the study of exotic nuclei produced at the source for nuclear structure studies. The objectives of RESIST aim to support the Transnational Access Facilities of ENSAR2, while in parallel provide a rich research programme of development and spectroscopy. This ensures that Europe continues to lead the field in resonant laser ionisation and spectroscopy.

As highlighted in this report, the partners of RESIST have made excellent progress in the development of techniques to enhance radioactive ion beam purity.