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REPORT ON TASK DEVELOPMENTS FROM RESIST

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	Contributors:	N. Lecesne, D. Scarpa, R. Ferrer, M. Block
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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 deliverable summarises the different activities of the three tasks which form the RESIST Joint Research Activity after the first 12 month period of ENSAR2.

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.

RESIST has 8 participants: Jyväskylä (JYU), GANIL, INFN (Legnaro), CERN (ISOLDE), CNRS, GSI, KU Leuven and JOGU Mainz. These partners contribute to three identified tasks which in turn have been split into sub-tasks.

Task 1 focuses on efforts to enhance the ion-beam purity prior to the so-called LIST devices and is led by 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 ionization 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.

Task 2 drives new advances in efficiency, selectivity and spectral resolution and is led by KU Leuven, with contributions from GSI, JYU, GANIL, CERN and JOGU Mainz. RILIS, IGLIS and LIST techniques are already used in combination with hot-cavity targets and/or gas-cell systems to selectively generate intense radioactive ion beams. This task aims to refine these methods for the active and planned ISOL and In-Flight facilities, focusing on the coupling of gas cells to mass separators to improve the selectivity of RIB production, to develop well-collimated gas jets offering unique environments for spectroscopy and to optimize isobaric, isotopic and isomeric selectivity via extensive studies on atomic excitation schemes. Task 2 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.

Task 3 will realise new concepts and developments of laser technologies and is led by JOGU Mainz with contributions from CERN, JYU, CNRS, KU Leuven and INFN. Major advances have been made in recent years in the laser systems used for the selective and efficient production of RIBs, as well as in sensitive in-source spectroscopy for the extraction of fundamental nuclear structure parameters of rare isotopes. New concepts will aim to close the visible gaps in the wavelength spectrum of Ti:sapphire lasers to offer greater coverage of ionization schemes,

new lasers to offer high-power Fourier-limited laser radiation using injection-locking techniques, and optimization of automated wide-range tunability of solid state laser systems for atomic spectroscopy and scheme development. Task 3 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 document herein reports on the first 12 months of activity of the RESIST partners. The status of the aforementioned three key research areas (tasks)are independently discussed in the following sections. These tasks support our aim to address state-of-the-art research in the fields of laser physics and spectroscopy, ion manipulation, materials research and isotope separation, provided by the different partners.

SECTION 1: PRE-LIST TECHNIQUES TO ENHANCE ION BEAM PURITY

This task is led by ISOLDE CERN and has been divided into three sub-tasks. The University of Mainz, KU Leuven, JYU, GANIL and INFN contribute to the different sub-tasks.

Sub-task 1.1: 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-Mainz: Upgrade of the LIST device

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 the LIST's application, electron impact-induced ionisation processes inside the rf structure had been identified as an additional source of contamination. By introducing a second repelling electrode operated on negative potential, electrons from the hot source material were inhibited from entering the LIST volume, therefore ensuring ionisation exclusively by the lasers. Figure 1 shows the different ion counting signals in operation modes with and without additional electron suppression for the case of ^{99}Tc in an off-line measurement campaign at Mainz University [1]. The remaining non-laser related background is reduced to the dark count rate of the ion detector. Results on hyperfine structure data and derived nuclear properties of $^{97-99}$Tc are presented in [2] and are in preparation for journal publication.

To reduce the deposition of radioisotopes inside the rf structure, which was observed to create isotope-dependent contaminant suppression factors in on-line experiments, adaptation of the geometric structure of the LIST has been addressed. Compared to the version used at the experiments at ISOLDE, the LIST 2.0 has been shortened to 45 mm length, and the solid exit electrode has been replaced by a metallic mesh structure, minimising the area of deposition. The quadrupole rods have been reduced in diameter for the same reason. The re-design currently awaits final characterisation at an off-line separator. Compatibility of the design with a time-of-flight laser ion source approach as an alternative concept is foreseen [3]. A fast polarity switch for the atomiser heating current has been constructed at the electronics workshop at Mainz University to enable in-operation mode switching between additional suppression of ions created inside the atomiser and optimum guidance to its exit by the correspondent voltage gradient, respectively.

Additionally, an off-line operation mode with laser irradiation perpendicular to the atom beam effusing in the LIST rf structure has been developed [1]. In addition to its wide spectroscopic advantages for high-resolution investigations, it serves as a powerful tool to characterise the atom beam density along the LIST's central axis. The

geometric atomiser design can therefore be adapted in a systematic way to ensure a well-collimated atomic beam and to aim for maximum efficiency in the atom transport from the atomiser towards the rf structure.

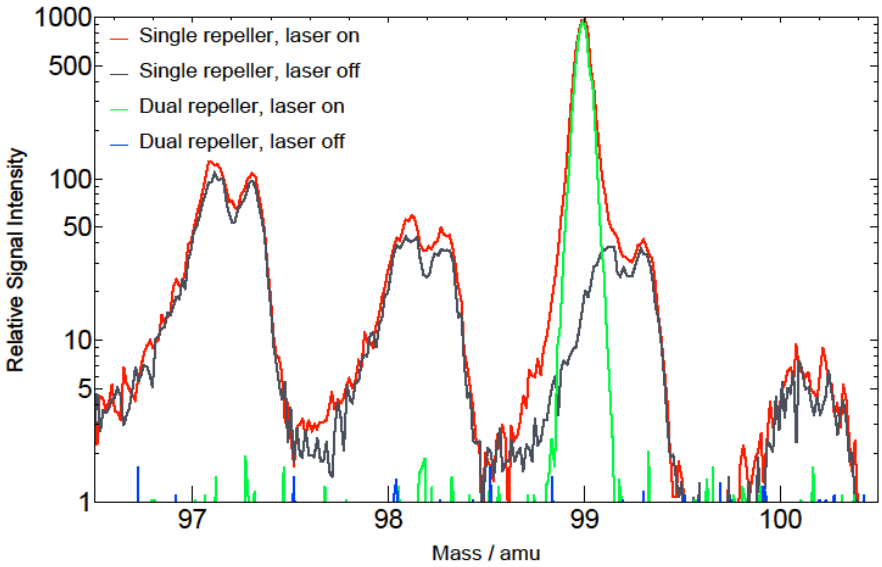

Figure 1: Mass scans in the technetium mass region for comparison between a LIST system using one single ion repeller to the dual repeller configuration. In the latter, background from non-laser related ionisation is completely suppressed [1].

[1] R. Heinke *et al.*, Hyperfine Interact **238** (2016) 6
[2] T. Kron, PhD thesis, Mainz University (2016)
[3] S. Rothe *et al.*, Nucl. Instrum. and Meth. B **376** (2016) 86

<u>Sub-task 1.2:</u> 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.

<u>KU Leuven: A new off-line setup for gas-jet research</u>

Research into optimising the laser-atom interaction region following the extraction of reaction products from a gas cell is being carried out at KU Leuven. A new gas cell with jet extraction has been designed and tested. It includes an S-shaped RFQ ion guide (Fig. 2) which enables ionisation in a crossed-laser beam geometry preventing photo-ionisation in the gas cell and improving spectral resolution (collinear excitation).

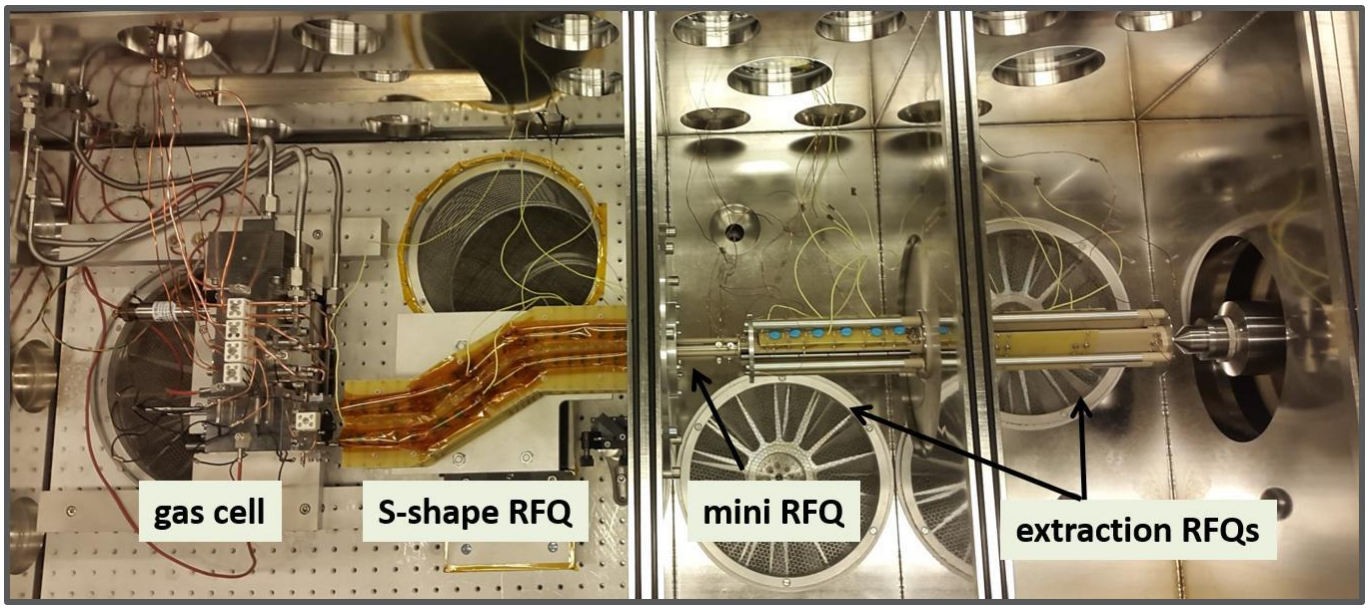

Figure 2: Internal view of the new gas cell during commissioning at KU Leuven.

<u>Sub-task 1.3:</u> 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. GISELE consists of three tuneable Titanium:sapphire (Ti:sa) lasers, a mass spectrometer and a hot-cavity ion source (see Fig. 3). The ion-source body has been designed to be a modular system in order to investigate different experimental approaches by varying the design parameters, to develop the future on-line laser ion source of SPIRAL2.

The efficiency of the ion source for radioactive ion beam production for SPIRAL2-Phase 2 has been tested through the modification of geometry parameters. LISBET (Laser Ion Source Body using Efficient Techniques) is an ion source prototype designed to investigate the geometries of the on-line ion source layout. It consists of two tantalum tubes forming a 90° elbow shape, which is adapted to the Target Ion Source System (TISS) configuration for SPIRAL2 Phase-2 (See Fig. 4). The first tube is used as a transfer/atomiser cavity and the second as 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, to ensure fast desorption of atoms from the surface. A sample of the element of choice (in our case natural Sn) is evaporated in order to produce a flux of neutral atoms into the ionisation tube. The design allows an independent selection of the polarity and the magnitude of the heating currents of the transfer tube and ionisation tube, respectively.

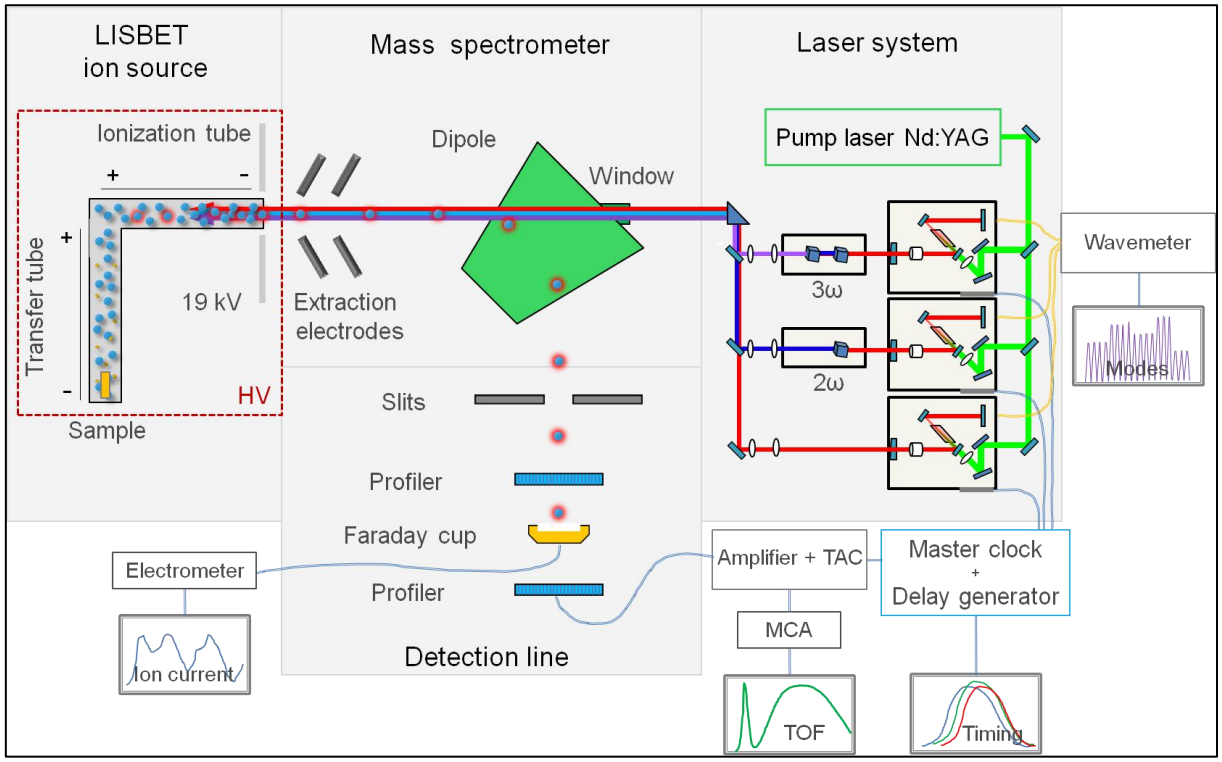

Figure 3: General setup of the GISELE test bench at GANIL.

The efficiency, the emittance and the time structure were analysed with a Sn ion beam as a function of the ioniser tube diameter (3 and 7mm) and length (35 and 60mm). 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.

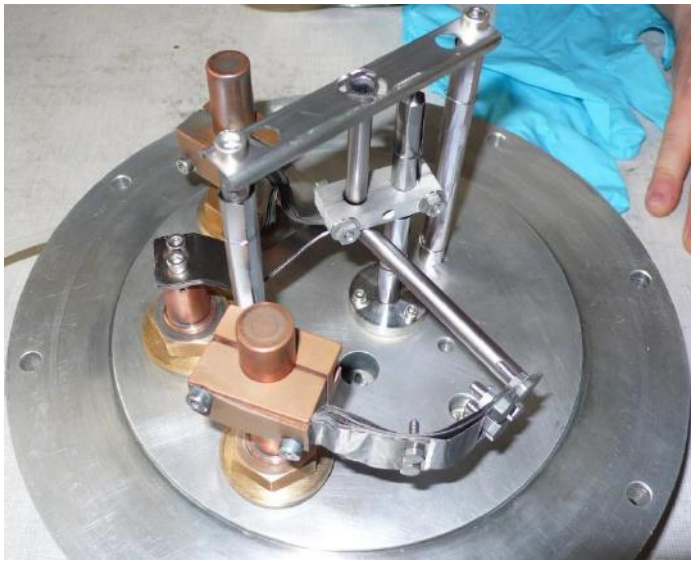

Figure 4: Picture of the LISBET ion source.

In the case of the ioniser diameter, an efficiency of approximately 24% was obtained for 7 mm and 8%

was obtained for 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.

The rms emittance 2σ has been measured for the different LISBET ion source configurations. No significant difference has been observed in the emittance of ionisers with different lengths. In the case of 7 mm ioniser diameters, an emittance of approximately 15.5 π.mm.mrad has been obtained, while 10.5 π.mm.mrad has been deduced for 3 mm ioniser diameters. The emittance values obtained with all the LISBET geometries were small enough to ensure the transport and acceleration of ions in accelerator and mass separator facilities at GANIL without losses.

In conclusion, an enlargement of the ioniser diameter produced an increase of the isotope production efficiency, while the emittance did not increase significantly. An ion source with 7 mm diameter and 35 mm length should provide the best characteristics for the production of short-lived isotopes due to the faster extraction from the ion source (due to the length) and the diminution of the neutralisation within the hot cavity (due to the diameter).

The time structures and energy scans of the produced ions have been measured for all the LISBET configurations. The time structure consists of a narrow peak and a broad main peak that emerges at a certain temperature. The narrow peak is produced by the ions generated in the region of the 19 kV extraction potential inside the ioniser and in the last millimetres at the exit of the ion source. The main peak is caused by the ions coming from the inside of the ioniser. At high temperatures the main peak is getting more pronounced while the narrow peak decreases (see Fig 5.). The increasing number of ions in the main peak at high temperature is explained as a result of the diminution of neutralisation due to the electric potential generated by the heating current. At 1700 K, the use of 7-mm diameter ionisers provided a higher number of ions in the main peak compared to 3-mm diameter ionisers. The fact that the main peak appears at high temperatures raises a question about the reliability of the efficiency measurements because only the ions from the narrow peak are recorded at low temperatures. For that reason, the efficiency measurement should be considered as an average result. A more precise determination of the efficiency would be the one performed directly at 2000 K. However, this sudden increase of temperature will also increase the pressure, with the consequent risk of oxidation of tantalum parts.

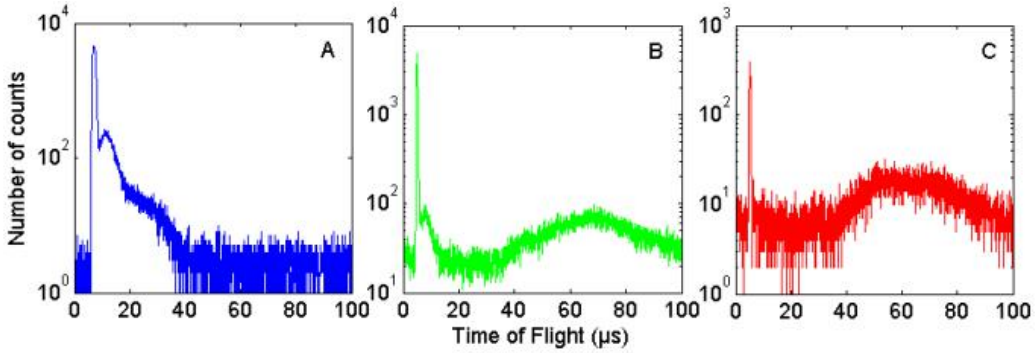

Figure 5: Temporal profile structures of ^{124}Sn measured with LISBET ∅7mm and 60mm length version at (A) 1450 K, (B) 1700 K, and (C) 1900 K.

To fully characterise the ion beam, a technique to measure the convolution of the energy and the spatial

spread generated by the RILIS technique has been developed. An example for the measured energy distribution inside and outside the ioniser is shown in Fig. 6. It was concluded that the narrow peak (seen in the time structure) consists of a broad distribution of energies, which can complicate the isotope selection. The ions coming from inside the ioniser have an estimated energy dispersion of about 5 eV.

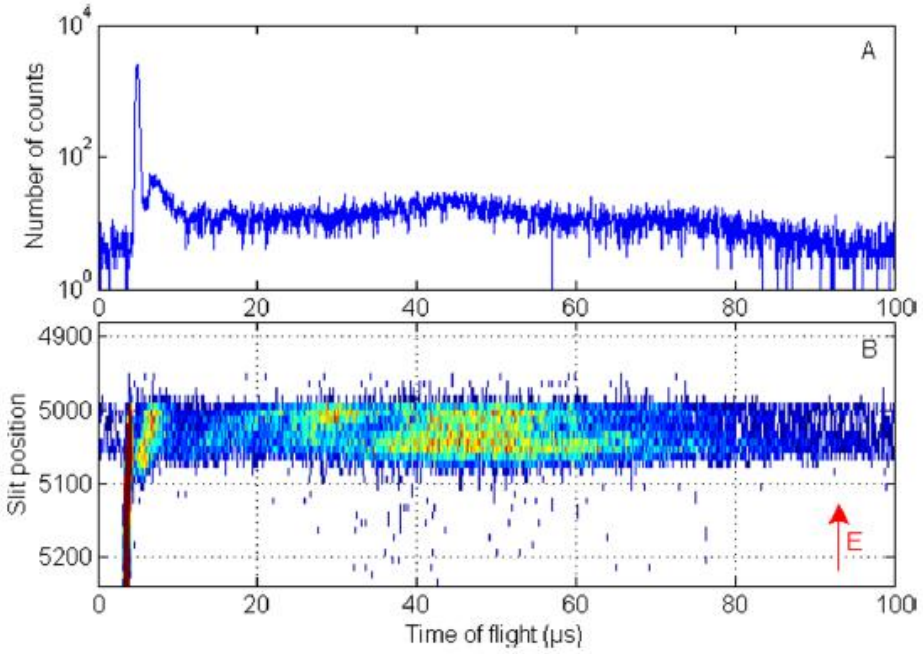

Figure 6: (A) Time profile measurement of ^{124}Sn for LISBET-D7L35 and (B) the energy distribution measured by the magnetic mass spectrometer. The increment of the extraction energy is marked by an arrow.

Additionally, in order to study and optimise the selectivity of the ion production, two methods have been studied in order 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 (see Fig. 3). The option shown in Fig. 3 was found to yield the highest reduction in contamination without compromising the production rate. The second technique utilized 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.

From the point of view of the generation of contamination, it has been observed using the LISBET geometries that an enlargement of the ioniser diameter improved the production of laser-ionised elements. At the same time the production of contaminants was also increased due to the surface ionisation. A comparative study has to be performed connecting the RILIS ion production and contaminants as a function of the geometry and the temperature.

In conclusion, the test bench has been optimised with the optimal extraction and transport parameters and the main objectives of this subtask have been accomplished. At the same time, ionisation schemes of zinc and tin have been chosen which provide the best operation for a Ti:sapphire laser system. Different geometries of the

ion source have been tested, whereby an enlargement of the ioniser diameter increased the isotope production efficiency, with small variations of emittance. The geometry with an ioniser of 7 mm in diameter and 35 mm in length has proven to offer the best performance. Finally a reduction of the contamination was observed by changing the electric field potentials.

[1] J.L. Henares, N. Lecesne *et al*., Rev. Sci. Instrum. **87** (2016) 02B701
[2] J.L. Henares, N. Lecesne *et al*., Nuclear Instruments and Methods in Physics Research A **830** (2016) 520
[3] PhD thesis, J.L. Henares, http://hal.in2p3.fr/tel-01314260

LNL-INFN: transfer line studies

In Legnaro, experiments to evaluate the surface ionisation efficiency related to the polarity of the transfer line and ion source power supply have commenced. The way in which the power supply is connected creates an electric field that affects how ions move into the ion source and transfer line, as showed in Fig. 7.

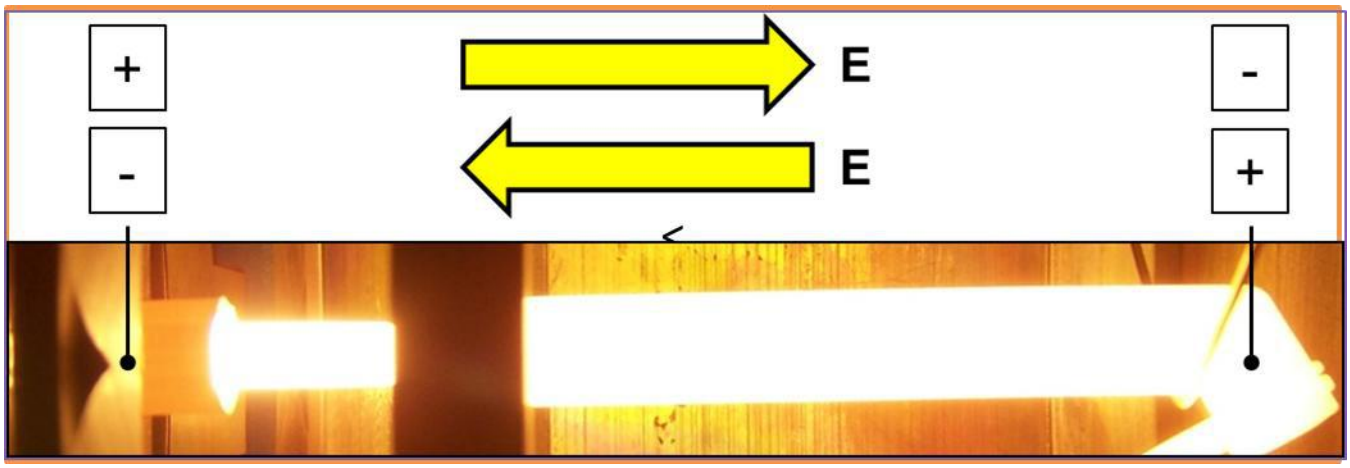

Figure 7: Transfer line and ion source operated at 2000°C and electrical field directions.

Very preliminary results for a tantalum hot cavity heated at 2200 °C are as follows:

- Electrical field acting in the direction of positive ion extraction – efficiency = 40%
- Electrical field acting in the direction opposite to the positive ion extraction – efficiency = 12%

These results will be a starting point for an extensive measurement campaign as soon as a person dedicated to this work is hired.

SECTION 2: ADVANCEMENTS IN EFFICIENCY, SELECTIVITY AND SPECTRAL RESOLUTION

This task is led by KU Leuven and has been divided into three sub-tasks. The University of Mainz, ISOLDE CERN, JYU, GANIL and GSI contribute to the different sub-tasks.

Sub-task 2.1: Optimisation of gas-cell coupling to a mass separator to target selectivity improvements of the future IGLIS technique at in-flight facilities.

GSI: Laser spectroscopy studies on $^{252-254}$No

At the velocity filter SHIP at GSI a tailored setup for in-gas cell laser spectroscopy of trans-fermium elements has been developed. It has recently been used in the first-ever laser spectroscopic investigation of nobelium (No, *Z*=102). In a pioneering experiment, several atomic states have been identified [1]. Among them were several Rydberg states that allowed, for example, the first ionisation potential of nobelium to be determined accurately. In addition, the hyperfine splitting in ^{253}No and the isotope shift of the $^{1}S_0$-$^{1}P_1$ atomic transition in $^{252\text{-}254}$No was measured (see example in Fig. 8). This will provide information on the nuclear spin of ^{253}No and the change in the mean-square charge radii of $^{252\text{-}254}$No to obtain (nuclear-model independent) information of these deformed nuclides around the *N*=152 subshell closure.

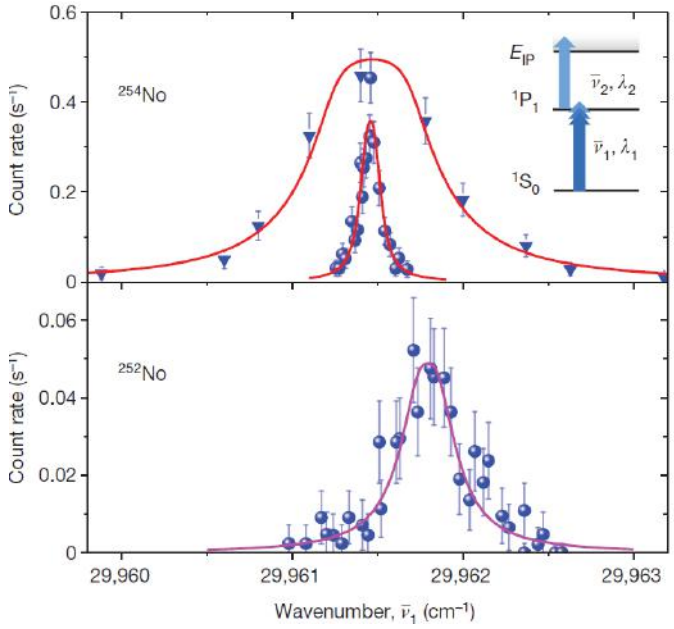

Figure 8: Resonance ionisation signals of nobelium atoms.

The approach utilised a gas cell to thermalise nobelium ions in a 100-mbar argon atmosphere. As most of the thermalised particles remain singly charged a filament-based laser spectroscopy method was developed. To this end the ions are collected on a filament and then re-evaporated as neutral atoms. A high overall efficiency in this approach is crucial for its extension to even heavier elements that are produced in much lower quantities. For nobelium, an overall efficiency of up to about 10% was achieved. However, decay losses occur for shorter-lived nuclides ($T_{1/2}$<1 s) due to the duty cycle.

The overall efficiency also sensitively depends on the filament material and its position within the stopping distribution. The chemical properties of the filament determine the material selection. Ideally they should feature a high thermal stability, a high work function to minimise surface ionisation and not facilitate molecular formation. The stopping distribution of the nobelium ions in the gas can be shaped by the pressure inside the cell and degrader foils that may be used in front of the entrance window.

For nobelium, it was observed that Ta filaments performed well, whereas W filaments did not work at all. In lawrencium (Lr, *Z*=103), initial tests were performed with Ta and Hf filaments. Hafnium lacked mechanical stability at high temperature while Ta may lead to a background from surface ionisation at the high temperature anticipated for the evaporation of Lr atoms. This motivates future studies of composite filaments.

[1] M. Laatiaoui *et al*., Nature **538** (2016) 495.

KU Leuven: In-Gas Laser Ionisation and Spectroscopy (IGLIS) of Ac isotopes in supersonic expansion

The feasibility and impact of the in-gas-jet laser ionisation and spectroscopy method have been demonstrated online by measuring nuclear and atomic properties of the short-lived isotopes ^{214}Ac and ^{215}Ac [2]. A de Laval nozzle installed at the gas cell exit was used in online conditions to produce a collimated supersonic jet of these isotopes seeded in argon buffer gas. In contrast to former in-gas-cell laser-spectroscopy studies the laser beams were overlapped with the gas jet outside the gas cell, leading to the resonant ionisation of actinium in the low-density, low-temperature supersonic gas-jet expansion [3]. The photo-ions were then captured and transported in a radiofrequency ion guide, accelerated, mass separated and subsequently implanted into an alpha-decay detection set-up.

The deduced magnetic and quadrupole moments have been compared to shell-model calculations and witness a stabilising effect of the N = 126 shell up to the actinium isotopes. The obtained efficiency and spectral resolution in these measurements demonstrate that basic ground- and isomeric-state nuclear properties of heavier actinides and eventually super-heavy elements, as well as their atomic properties, can be determined to high precision. The in-gas-jet method, as an extension to the IGLIS technique, has also far reaching consequences for the exploration of the refractory elements, so far hardly accessible to high-resolution laser spectroscopy techniques. In addition, the highly-selective ionisation enables the production of high-quality, high-purity isotopic and isomeric radioactive ion beams that can be used for other applications in nuclear physics, chemistry and astrophysics, as well as in atomic physics.

[2] R. Ferrer *et al*., Nat. Commun. **8** (2017) 14520
[3] S. Raeder *et al*., Nucl. Instrum. Meth. Phys. Res. B **376** (2016) 382

Sub-task 2.2: Development of well-collimated, high Mach number gas jets.

KU Leuven: Planar Laser-Induced Fluorescence for supersonic gas jets at the HELIOS laboratory

In order to achieve the best spectral resolution in IGLIS experiments it is essential to reduce both the flow density and the temperature in the point at which atoms interact with the laser beams. This can be accomplished by applying resonance ionisation in the gas jet rather than within the gas cell. Pressure and temperature in the supersonic gas jet are significantly reduced compared to those in the gas cell. The gas thermal energy is converted to kinetic energy during the gas flow acceleration in a de Laval nozzle. Such a nozzle with a Mach number 5.5 was used in the online experiments reported in Refs. [2, 3] of sub-task 2.1 and is also now used for the first tests of the jet visualisation at the IGLIS laboratory, KU Leuven [1] (see Fig. 9).

In these studies, stagnation temperature and pressure in the gas cell were about 300 K and 300 mbar, respectively. Computer simulations for an optimal background pressure were performed using the COMSOL Multiphysics software. The simulations showed that the supersonic gas jet after exiting the nozzle should have the following characteristics: temperature 27 K, density 0.013 kg/m^3 and pressure 0.7 mbar, which correspond in the case of copper to a Doppler broadening of 420 MHz and a collision broadening of only 40 MHz.

The parameters of the gas cell and of the nozzle should be carefully chosen in order to obtain the maximum benefits of in-gas-jet ionisation and spectroscopy. Visualisation of the supersonic gas jet by the Planar

Laser-Induced Fluorescence (PLIF) technique is used to verify that the parameters of the supersonic gas jet fulfil the optimal experimental conditions for spectroscopy and for the production of short-lived nuclei. In the PLIF technique, copper atoms seeded in argon are extracted through the nozzle and are excited by a laser-sheet beam. The subsequent emitted fluorescence upon de-excitation is recorded by an ICCD camera, making it possible to measure density, velocity and temperature profiles of the gas jet.

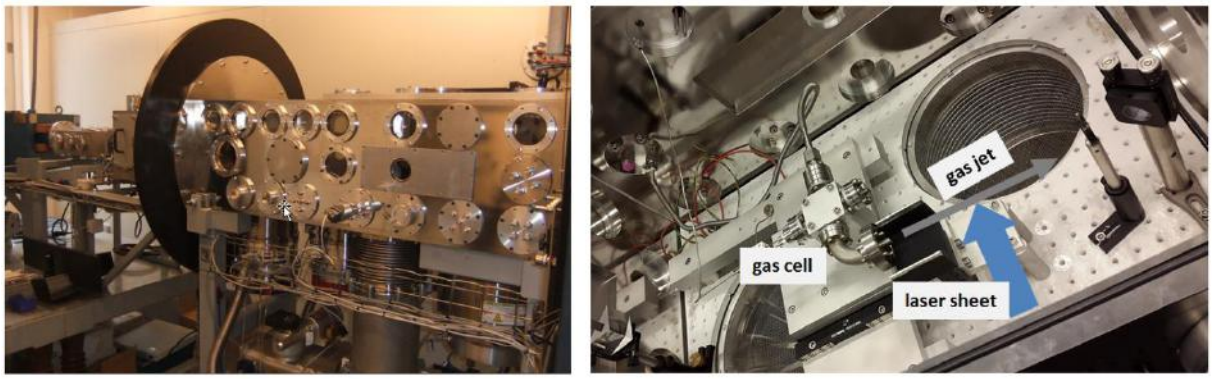

Figure 9: (Left) Front end of the IGLIS beam line comprising the gas cell chamber with three inner sections, one for the gas cell and the jet formation and two for differential pumping. Also visible is part of the high-voltage platform (still under construction) with the dipole magnet at the far left. (Right) Interior of the first section of the gas cell chamber housing the gas cell used for the visualisation of the jets by planar laser-induced fluorescence.

The excitation of copper seed in argon as buffer gas is carried out by one of the dye lasers or by the amplified light of the cw diode laser in a pulse dye amplifier (for broad- or narrowband scans, respectively) after frequency doubling in a non-linear BBO crystal for the generation of UV radiation at 327.49 nm. The average energy per pulse of the UV light is about 0.8 mJ, and the pulse duration is 7 ns. The combination of a spherical lens of 1000 mm focal length and a cylindrical telescope (consisting of a plano-convex and -concave lenses with a focal length of 500 mm and 25 mm, respectively) installed on the laser beam path, forms the laser beam into a thin sheet. The spherical lens focuses the laser sheet to a 1 mm thickness at the region of the intersection with the supersonic gas jet, while the cylindrical telescope expands the laser beam to form a sheet 55 mm wide. After excitation of the copper atoms at 327.49 nm a small fraction of them (1.2 %) de-excite to a metastable state emitting fluorescence at 578 nm. The fluorescence light is collected with a Thorlabs Camera Lens (effective focal length 8 mm) and detected by the ANDOR iStar ICCD camera.

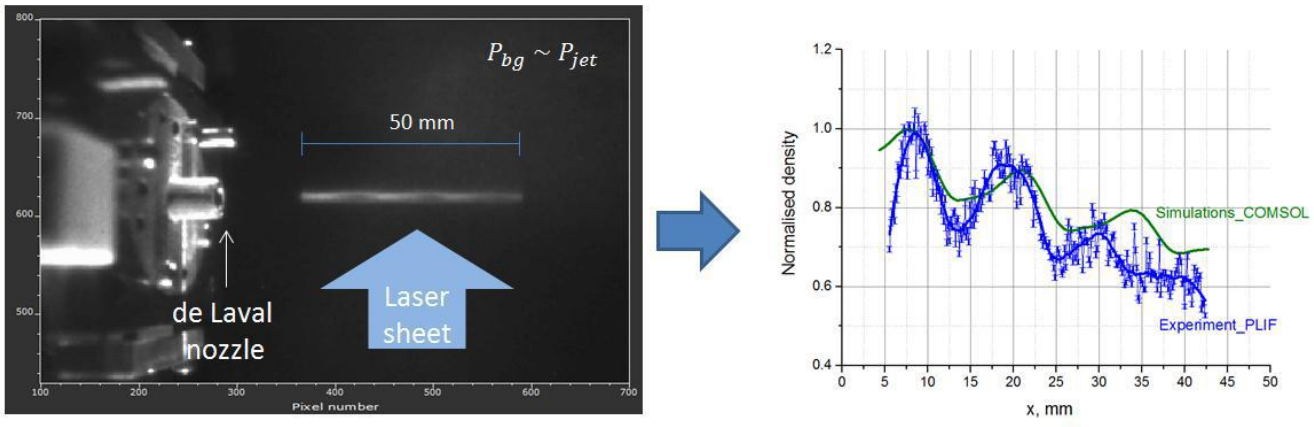

Figure 10: (Left) Visualisation of the gas jet under optimal conditions of background-to-jet pressures (P_{bg}/P_{jet}~1). (Right) Comparison between the experimental density distribution in the jet as a function of the distance from the nozzle exit and that obtained from COMSOL simulations.

In typical PLIF spectroscopy studies the copper fluorescence signal was collected by gating the ICCD photocathode with 50 ns pulses synchronously with the laser operated at a repetition rate of 5 kHz. The fluorescence signal was then accumulated for a total exposition time of 1 s and the cycle was repeated for 100 s at each wavelength. Results of these tests on the visualisation and properties of the supersonic gas jet using copper are shown in Fig. 10. Fair agreement between simulations and the first experimental data can be observed in the right panel of the figure.

The results of fluorescence single-mode PLIF spectroscopy experiments on ^{63,65}Cu (with a resulting 750 MHz FWHM) at Mach ~6 and using 4×4 pixel area (pixel size 13 µm) are shown in Fig. 11. The low-energy hyperfine transitions were visible for the natural isotopic admixture, although the individual transitions for each isotope could not be resolved. Further reduction of the linewidth will be possible after reducing the power broadening contribution and jet divergence, and by using a nozzle with a greater Mach number. In this way, extraction of accurate information on the gas jet (density, temperature and velocity distributions) will be possible by performing PLIF with narrowband laser radiation around the absorption line of each specific copper isotope.

In order to check the predictive power of our simulation tool COMSOL and to study the effect of higher jet Mach numbers on the achievable spectral resolution, we performed a series of measurements using a freely expanding jet. The free jet is characterised by highly inhomogeneous density and temperature areas in contrast to the jets produced by a de Laval nozzle under optimal conditions. In the free jets it was possible to perform spectroscopy in local (small) areas with high Mach numbers (M ~13). Results on the position of the Mach disk and on the density distribution were well reproduced by the simulations.

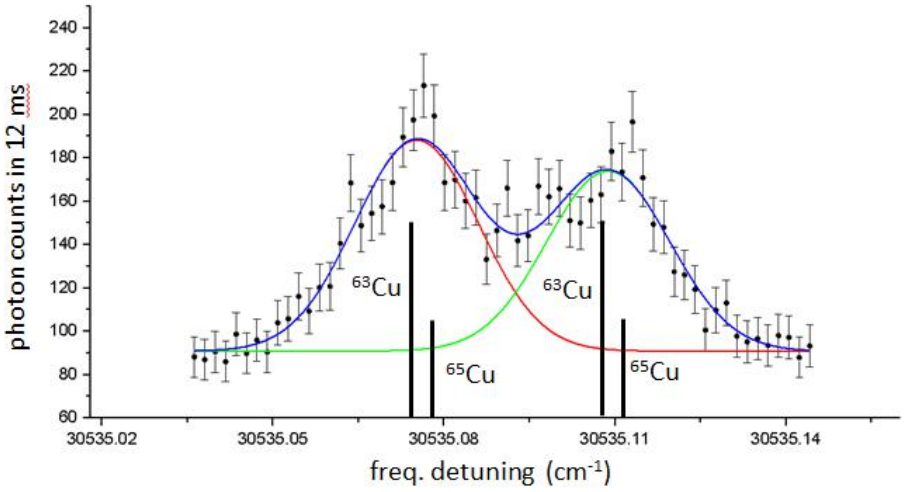

Figure 11: Single-mode PLIF spectroscopy experiments on ^{63,65}Cu (750 MHz FWHM) at Mach M~6.

The results of PLIF spectroscopy in two different areas of the free jet, indicated by circles of different colour, are shown in Fig. 12. Fully resolved hyperfine transitions of the copper isotopes can be seen in the partial hyperfine structure spectra.

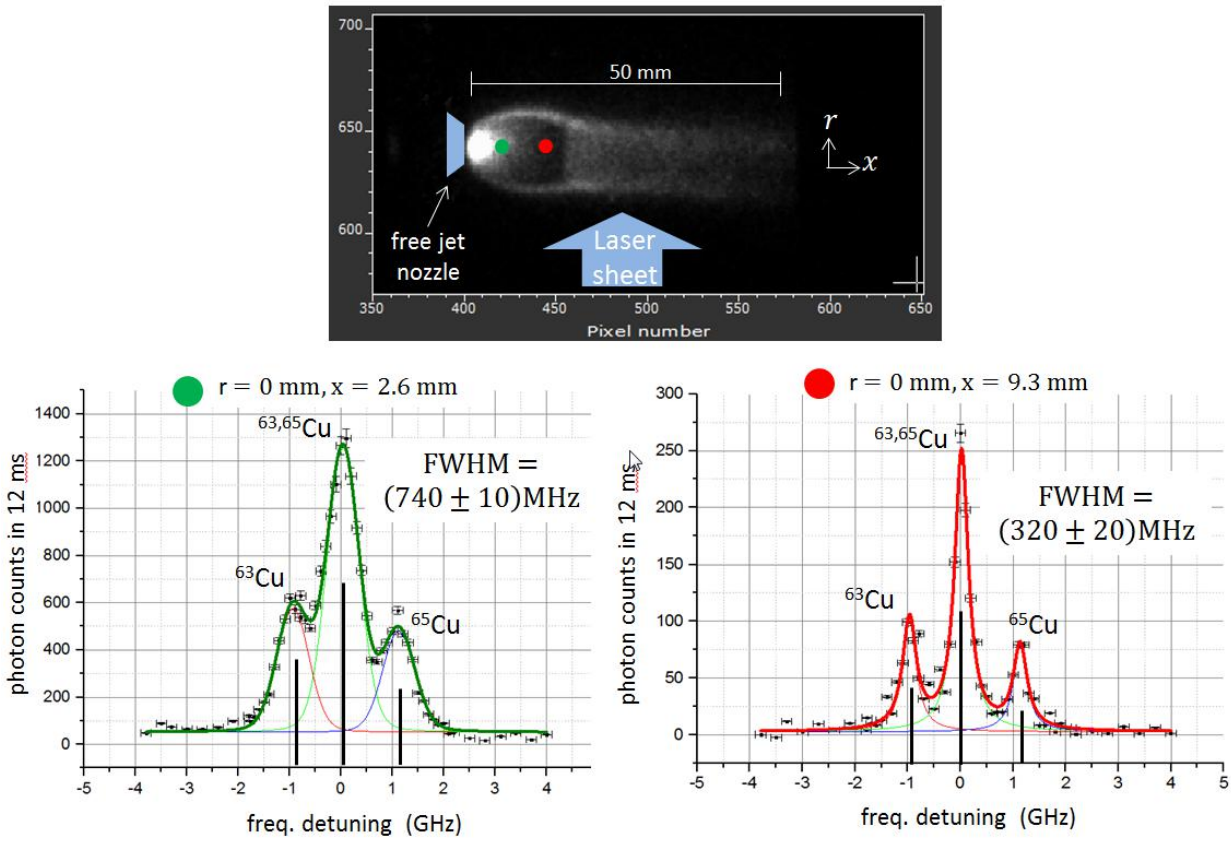

Figure 12: PLIF image of copper isotopes in free jet expansion (top). PLIF spectroscopy in two different areas of the free jet indicated by green (lower left) and red (lower right) circles.

[1] Yu. Kudryavtsev *et al.*, Nucl. Instr. Meth. B **376** (2016) 345

<u>Sub-task 2.3:</u> Extensive ionisation scheme development to optimise isobaric, isotopic and isomeric selectivity.

<u>JYFL: Pu ionisation scheme development</u>

Recently, a new programme of heavy element research has been initiated at the IGISOL facility, initially focusing on long-lived actinide isotopes for which sufficiently large (ng) sample sizes may be produced in research reactors. The first element of interest has been Pu, whereby samples of $^{238\text{-}240,242}$Pu and ^{244}Pu have been evaporated from Ta filaments, laser ionised, mass-separated and delivered to the collinear laser spectroscopy station for high-resolution studies. These samples were prepared by the Institut für Kernchemie of the University of Mainz.

Initially, a three-step ionisation scheme using laser radiation at wavelengths of 420.76, 847.26 and 750.24 nm was chosen based on earlier trace analysis studies by Raeder *et al*.. In that work, each transition was fully saturated ensuring a high efficiency of the ionisation process. In the gas cell at IGISOL, little or no effect of the IR transitions was seen and it was suspected that preferential ionisation occurred via a Rydberg state populated by a photon from the first step excitation - subsequently ionised via buffer gas collisions.

A second step was therefore introduced operating in the blue wavelength regime, the wavelength optimised to a resonance observed at 422.53 nm. Further studies showed that both transitions were able to ionise independently from one another, which may be explained if the second step is driving population from a low-lying metastable population, as summarised in Fig. 13. Population of such an excited state was initially a surprise however could be explained if the state was available due to the temperature of the filament (1000 - 1200 °C). Further details of the gas cell ionisation may be found in [1], however explanation of the ionisation scheme required further work and thus use of a new frequency-doubled grating laser (explained in more detail in Section 3, sub-task 3.1).

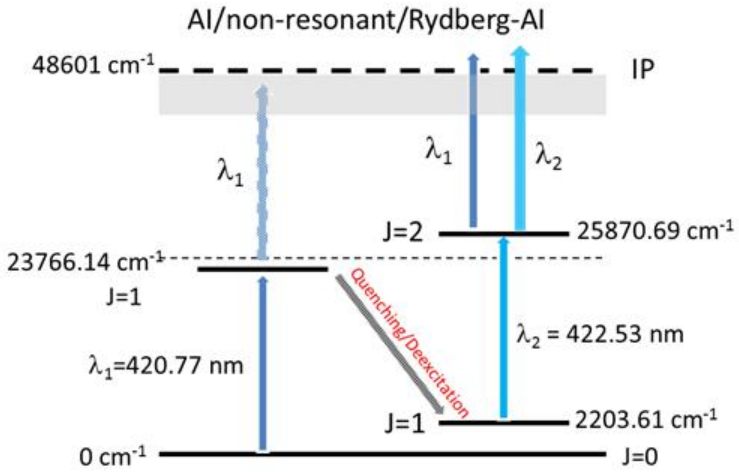

Figure 13: The postulated ionisation scheme used for in-gas-cell laser ionisation of Pu [1].

Following successful characterisation of the grating laser supplied by the University of Nagoya, Japan, in November 2016, the laser was used in a more detailed study of the laser ionisation of Pu in a gas cell at the IGISOL facility. The analysis is currently underway. The initial scheme presented in Fig. 13 was confirmed with the addition of the grating laser, and was also expanded to include new levels (schematically illustrated in Fig. 14). This work will be combined with the general characterisation of the laser in a manuscript to be submitted.

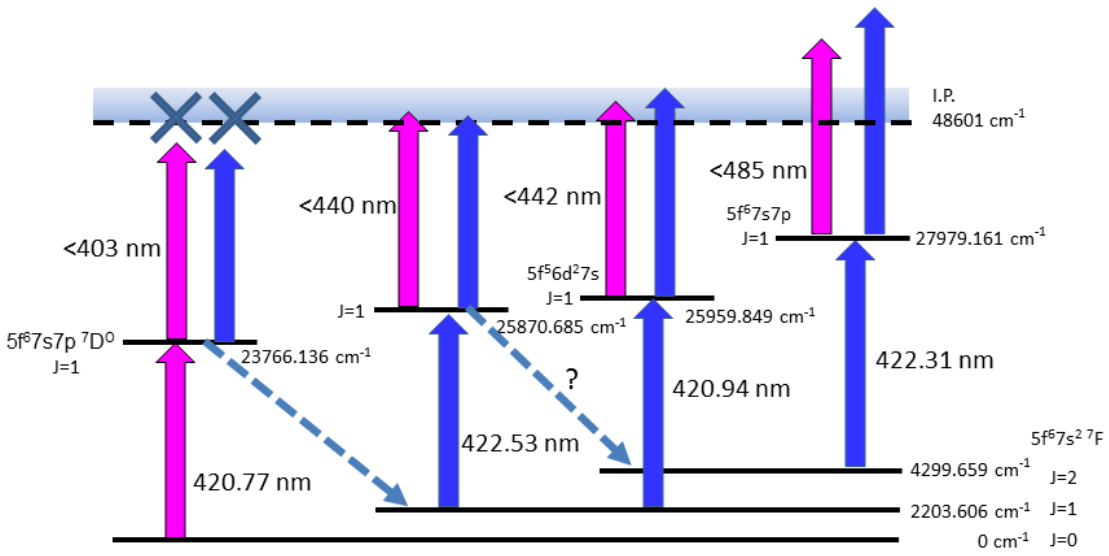

Figure 14: Following the commissioning and testing of the grating laser operating in the second harmonic, the ionisation scheme of Fig. 13 was confirmed and extended. The analysis of the data is underway.

[1] I. Pohjalainen, I.D. Moore et al., Nucl. Instrum. and Meth. B **376** (2016) 233.

JGU Mainz: Excitation scheme development in Dy, Er and Lu

With regards to both the exciting nuclear physics in the rare earth region of the nuclear chart as well as the growing interest in the production of medical radioisotopes at RIB facilities [1], efficient excitation schemes for the lanthanide elements Dy, Lu and Er, were developed at JGU Mainz (Fig. 15).

RIS studies in Dy were carried out for both two- and three-step excitation schemes at the atomic beam unit MABU at JGU Mainz, resulting in a re-determination of the first ionisation potential of the dysprosium atom and the development of an efficient three-step excitation scheme, aiming for an auto-ionising state (404.71 nm – 802.72 nm – 837.67 nm). An outstanding overall efficiency of 25(4) % could be demonstrated at the RISIKO off-line mass separator at JGU Mainz [2].

For the element lutetium, several two-step excitation schemes for Ti:sapphire lasers were tested with respect to relative ion beam intensities. The most promising scheme features a transition in the ultraviolet regime (298.93 nm), which was accessed via third harmonic generation of the fundamental Ti:sapphire radiation, and a second step in the fundamental wavelength regime (883.37 nm). Both transitions could be saturated with the available output powers of approximately 20 mW and 2.5 W, respectively [3]. Efficiency measurements are currently evaluated and will be published soon.

The RIS measurements in Er carried out at JGU Mainz in 2015 were primarily dedicated to a re-determination of the first ionisation potential, whereby the precision of the current literature value could be improved by two orders of magnitude [4]. In the course of this work a multitude of auto-ionising states was found. Aiming for the high-intensity transition to the auto-ionising state at 49279.8 cm^{-1}, erbium can be resonantly ionised by using two frequency-doubled Ti:sapphire lasers (400.80 nm – 410.91 nm). This scheme will be tested and compared to a three-step dye laser scheme (622.1 nm – 645.2 nm – 564.9 nm) [5].

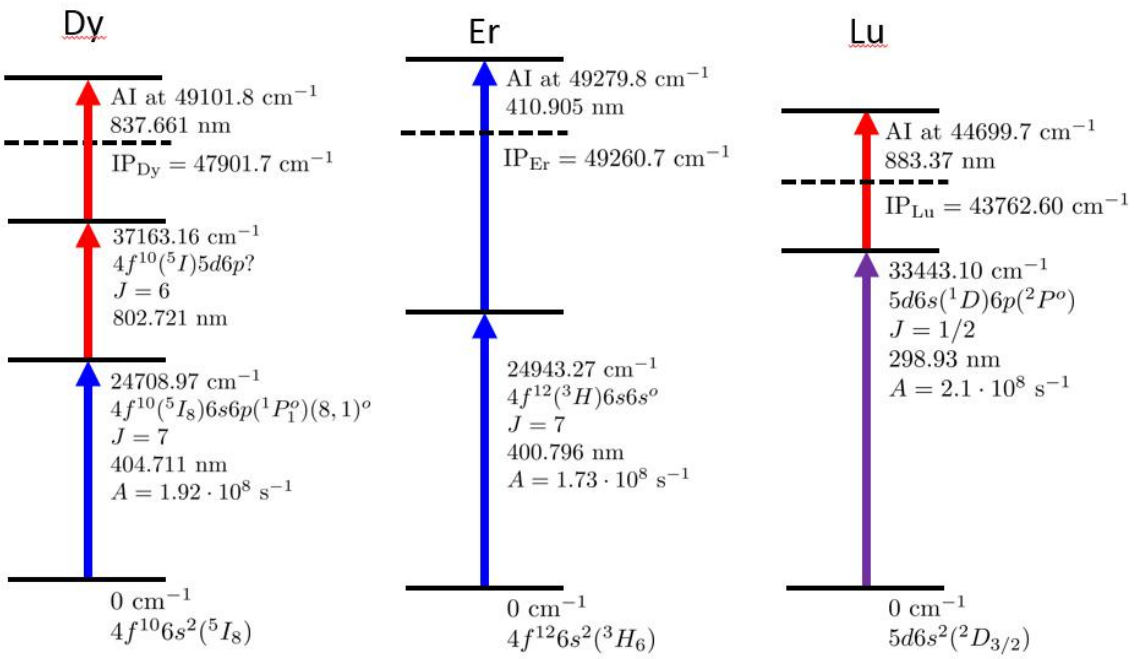

Figure 15: Excitation schemes for Dy, Er and Lu, as developed at JGU Mainz.

[1] R. M. dos Santos Augusto *et al.*, Appl. Sci. **4** (2014) 265
[2] D. Studer *et al.*, Hyperfine Interact **238** (2017) 8
[3] V. Gadelshin *et al.*, Hyperfine Interact (2017) (accepted, to be published)
[4] D. Studer, diploma thesis, JGU Mainz (2015)
[5] R. Formento Cavaier *et al.*, CERN-INTC-2017-023; INTC-I-175 (2017)

CERN-ISOLDE: Excitation scheme development

The Resonance Ionisation Laser Ion Source (RILIS) is the principal ion source of the CERN-ISOLDE facility. Its unmatched combination of selectivity and efficiency, enabling intense and pure beams of a chosen isotope, is a crucial requirement of the majority of experiments. In 2016 for example, more than 75% of ISOLDE experiments requested RILIS-produced ions.

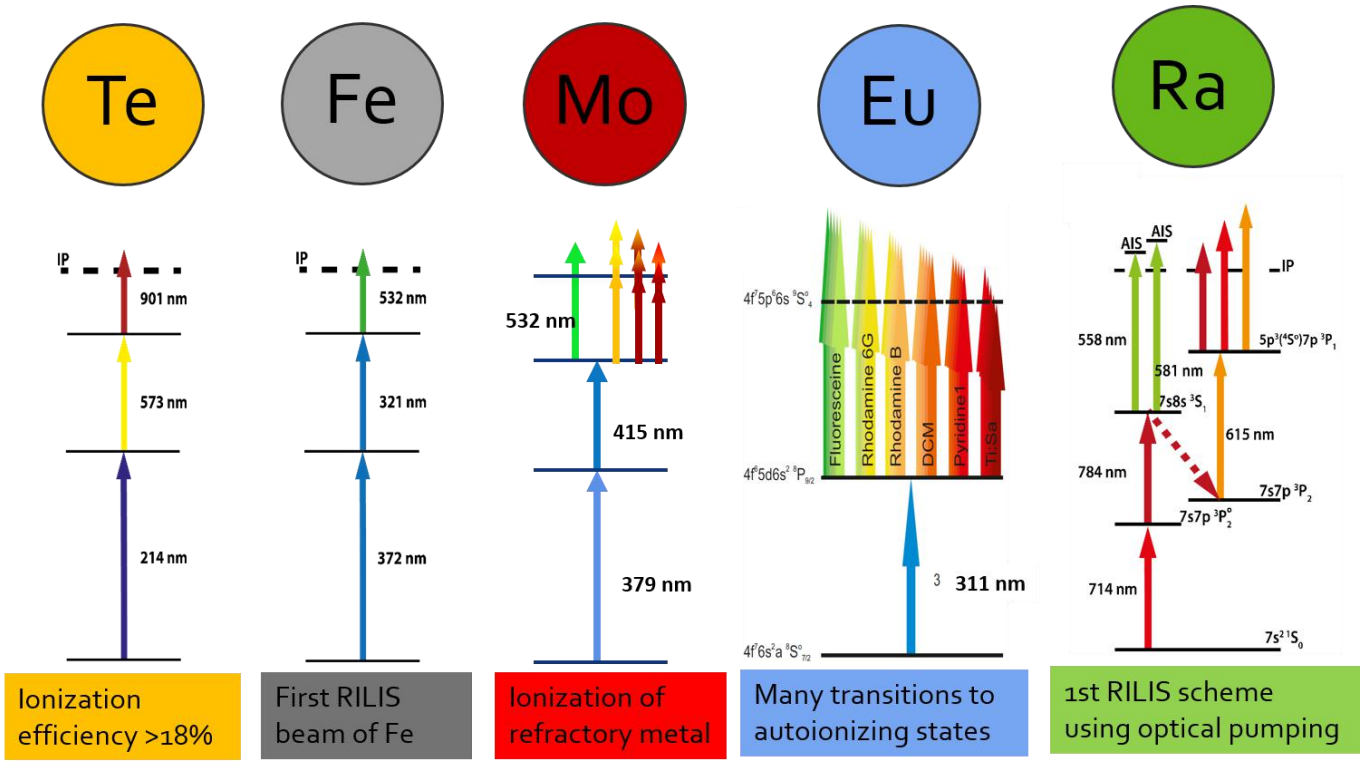

Figure 16: Resonance ionisation schemes for ISOLDE/RILIS developed at CERN.

In parallel to the operation, an extensive work of development of new schemes of resonance ionisation was continued. The following results have been achieved and illustrated in Fig. 16:

- Measurements of RILIS ionisation efficiency using the scheme developed in 2015 [1].
- Development of the iron ionisation scheme and first extraction of radioactive Fe isotopes produced in a UCx target and ionised using RILIS.
- On-line development of radium ionisation schemes. First-time observation of efficient ionisation involving transitions populated by optical pumping in the hot cavity ion source. Application of the newly found scheme for production and study of Ra isotopes using the Collinear Resonance Ionisation Spectroscopy (CRIS) method.
- Development of europium ionisation schemes using the off-line Photo-Ionisation Spectroscopy Apparatus (PISA). The multitude of discovered strong transitions to auto-ionising states offers a broad choice for optimisation of Eu ion production. Measurements of Rydberg series in europium atoms produced data for a precise determination of the ionisation potential of the europium atom [2].
- Development of an ionisation scheme for molybdenum. Laser ionisation of this refractory element was detected in the Versatile Arc Discharge and Laser Ion Source (VADLIS) [3] equipped with a molybdenum anode.

[1] T. Day Goodacre *et al.*, *Nucl. Instrum. and Meth. Phys. Research* A **830** (2016) 510
[2] K. Chrysalidis, *PhD thesis*, Mainz University (2016)
[3] T. Day Goodacre *et al., Nucl. Instrum. and Meth. Phys. Research* B **376** (2016)39

SECTION 3: NEW CONCEPTS AND DEVELOPMENT OF LASER TECHNOLOGIES

This task is led by the University of Mainz and has been divided into three sub-tasks. KU Leuven, JYU, ISOLDE CERN, CNRS and INFN contribute to the different sub-tasks.

Sub-task 3.1: Optimisation of automated wide range tunability of solid-state laser systems for atomic spectroscopy and ionisation scheme development.

JYFL: Characterisation and exploitation of a frequency-doubled grating-based laser

For many elements there is a lack of tabulated spectroscopic data in the region of higher-lying excited states. This becomes increasingly problematic for heavier elements which have complicated electronic structure and for which the computation of energy levels to the required precision for laser excitation of <1 cm^{-1} is thus far not possible. Therefore, the only reliable way to obtain spectroscopic information is by direct measurement. While the standard Ti:sapphire lasers used at ISOL and IGISOL-type facilities for efficient laser excitation and ionisation have a sufficiently wide tuning range, the scanning of the wavelength with a combination of etalon and birefringent filter often leads to mode jumps and a strong output power variations and dependence on the wavelength. On the other hand, using a grating for wavelength tuning allows for scanning over wide wavelength ranges.

At JYFL a grating-based laser was designed and constructed for operation in the fundamental wave length regime. The design of the resonator is shown in Fig. 17. The setup uses a similar resonator geometry to that of the standard Z-shaped design used in many facilities, however the end mirror of the cavity has been replaced with a pulse compression grating (>90% efficiency from 700-900 nm, Spectrogon, 1800 lines/mm). The grating is used close to the Littrow configuration and allows for a continuous frequency tuning of the laser by changing the incident angle. In order to achieve a desired bandwidth below ∼10 GHz and to reduce possible damage due to high power densities, a four-prism beam expander is inserted into the cavity arm in front of the grating. The grating itself is mounted on a computer-controlled stepping motor rotation stage. Further technical details and characterisation of the laser used at JYFL can be found in Ref. [1] in which the laser was successfully used in a search for auto-ionisation states of Sm, an element exhibiting seven naturally occurring isotopes and for which a suitable ionisation scheme had thus far been missing for Ti:sapphire lasers.

For accurate spectroscopy results in a multi-step RIS scheme the structure of high-lying states can prove critical. To avoid misinterpretation of results it may well be useful to use a simple two-step ionisation scheme with the final transition to a very broad auto-ionisation (AI) sate. To find such schemes, frequency-doubling techniques are required, and one key goal in this Task is to develop intra-cavity frequency doubling of a grating-based Ti:sapphire laser.

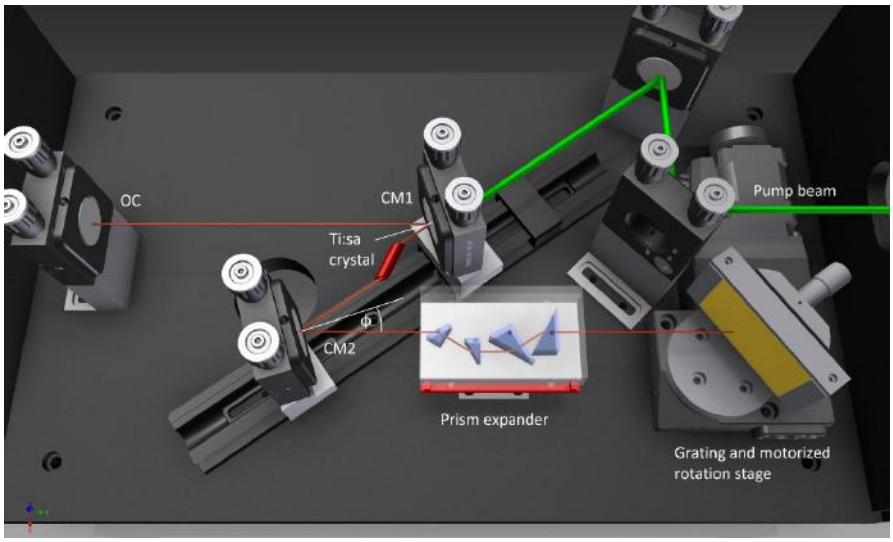

Figure 17: A 3D CAD design of the grating-based Ti:sapphire laser at JYFL [1]. The water-cooled crystal mount and the Nd:YAG pump lens have been omitted for clarity.

In November 2016, a team from the University of Nagoya, Japan, came to JYFL to commission a new design of intra-cavity frequency doubled grating-based Ti:sapphire laser. The visit also allowed for a comparison of the fundamental characteristics of the Nagoya and JYFL lasers, and a full characterisation of the second harmonic (output power versus wavelength, pointing stability of the beam and so on). A schematic diagram showing the layout of the Nagoya laser for second harmonic operation is shown in Fig. 18. The cavity design is almost identical to the standard grating laser (operated in JYFL and elsewhere), however a frequency-doubling crystal (BBO) is mounted onto a motorised stage into the opposite resonator arm to the prism expander. Via computer control, the grating angle can be tuned and synchronised to the crystal angle to maintain the optimal frequency doubled power.

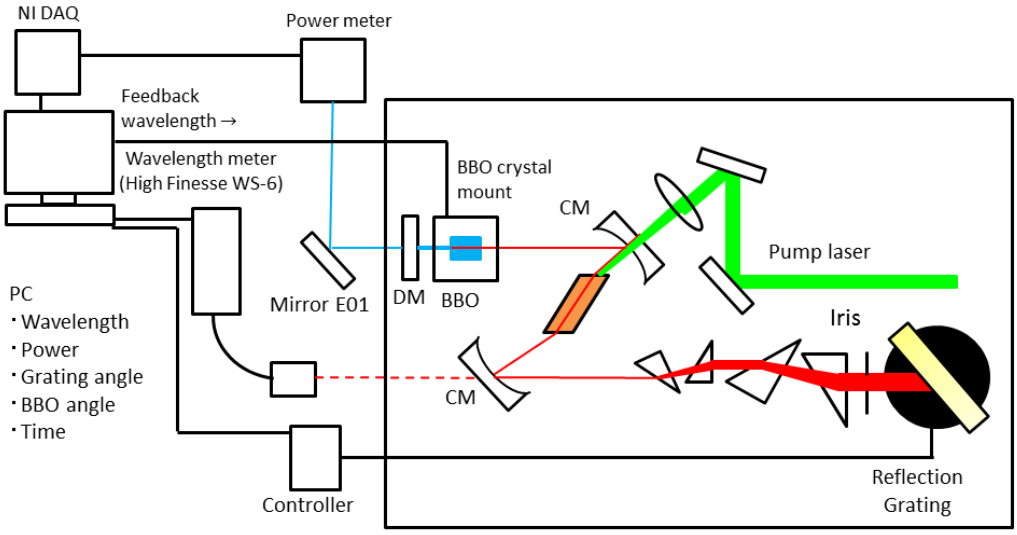

Figure 18: Schematic diagram of the grating laser setup for frequency doubling.

Thus far the first tests of the frequency-doubled grating laser have been performed at JYFL. One example of the tuning curve compared to the fundamental wavelength tuning is shown in Fig. 19. The tuning range in this test was from 360 nm to 445 nm, the lower limit determined by the specifications of the tuning crystal. Further work is required to minimise the fluctuations seen in the second harmonic power which result from the optimisation of the crystal angle. Finally, the laser was then transported to the IGISOL target chamber and the pointing stability was tested as a function of wavelength. In combination with a standard broadband intra-cavity doubled Ti:sapphire laser, the grating laser was subsequently used in an important study of the ionisation scheme of Pu in a gas cell, discussed in more detail in Section 2, sub-task 2.3.

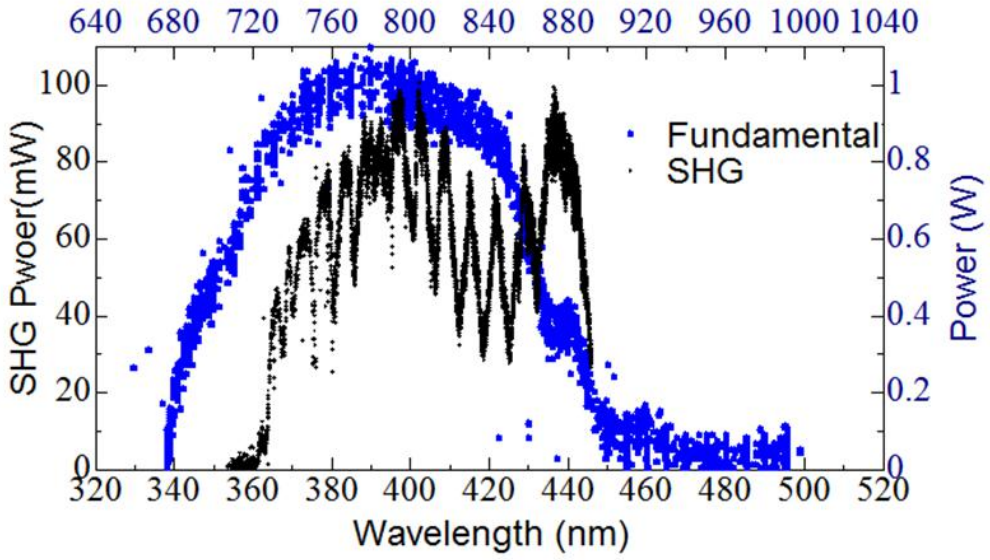

Figure 19: A wavelength scan of the grating laser showing the output power as a function of wavelength both in the fundamental and in the second harmonic.

[1] V. Sonnenschein, PhD thesis, University of Jyväskylä (2014)

<u>Sub-task 3.2:</u> Generation of high power Fourier-limited laser radiation using injection-locking techniques with a narrow bandwidth cw laser and pulsed dye amplification of a cw diode laser for in-gas-jet spectroscopy.

<u>JYFL: An injection-locked Ti:sapphire laser</u>

The relatively recent implementation of resonance ionisation spectroscopy (RIS) in a supersonic gas jet using a narrowband first resonant excitation step [1] has initiated considerable interest within the nuclear-atomic physics community. Typical pulsed laser systems whose goal is the efficient and selective ionisation of radioactive atoms at on-line facilities possess spectral linewidths of several GHz, suitable for matching the atomic absorption line profiles which are broadened in hot cavity (ISOL) or gas cell-based (IGISOL) environments. The realisation that spectroscopy may be performed in the cold environment of an expanding supersonic gas jet, immediately downstream from a gas stopping cell, necessitates a considerable reduction in pulsed laser linewidth in order to fully exploit the gas jet environment. Indeed, novel in-gas-jet spectroscopy may provide a means of performing high precision studies of the ground- and isomeric-state properties of short-lived exotic nuclei, produced far from stability in minute quantities. Optical spectroscopy yields model-independent information of fundamental nuclear

properties including direct measurements of the nuclear spin, electromagnetic moments and changes in mean-square charge radii between isotopes.

In close collaboration with the University of Mainz, a team from JYFL has developed a 10 kHz repetition rate pulsed Ti:sapphire laser which has been locked to a continuous wave (CW) seed laser [2]. The pulsed amplification of a narrow-band CW laser may be realised either by dye or Ti:sapphire technology. At JYFL, the input seed radiation is generated by a commercial Ti:sapphire laser (Sirah Matisse TS). Figure 20 provides an overview of the injection-locked laser and the set-up used for locking and subsequent spectral analysis. Briefly, the CW laser is injected into a pulsed pumped bow-tie cavity via an output coupler. The cavity is locked to the CW laser via a commercial stabilisation unit (TEM LaseLock). By dithering the length of the bow-tie cavity using a piezo-actuated mirror and through phase sensitive detection (PSD), a regulating loop can be constructed to keep the cavity in resonance with the Matisse laser. By measuring the laser linewidth using a Fabry-Pérot Interferometer, the laser linewidth has been measured on several occasions (and indeed at several different facilities where it has been used in a number of experiments to date) to be below 20 MHz. This is an impressive factor of approximately 200× lower than the pulsed lasers used in standard RILIS applications.

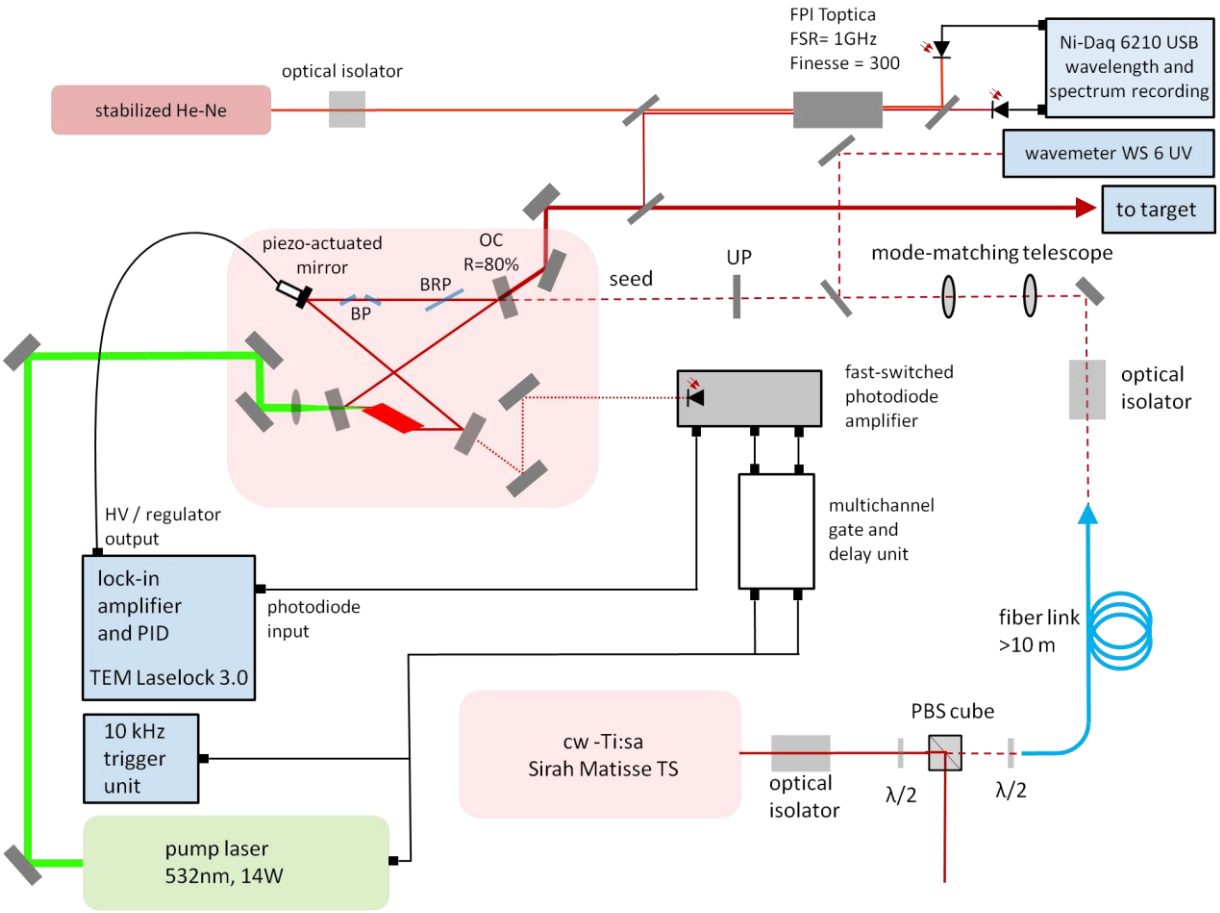

Figure 20: Overview of the setup of the injection-locked Ti:sapphire laser system at JYFL [2].

To test the capability of the injection-locked Ti:sapphire laser for high-resolution measurements, the laser was first commissioned at JYFL in a study of the hyperfine structure of stable copper isotopes [3]. Figure 21 illustrates a frequency scan of the hyperfine structure associated with the hyperfine $F_g = 1 \rightarrow F_e = 2$ component of

the 244-nm ground-state transition. This measurement was performed in an atomic beam unit and directly compared with laser scans using a standard broadband tuneable laser with either one or two etalons inserted into the resonator, respectively. For the excited state hyperfine coupling constant in ^{63}Cu a reduction factor of ten in the resulting error compared to previous literature was achieved. A further improvement in resolution was obtained by delaying the ionisation of the long-lived excited state, also indicated in Fig. 21 [4].

The laser cavity was then shipped to the University of Mainz where it was used for crossed-beam resonance ionisation spectroscopy of ^{227}Ac and in a study of long-lived Pu isotopes, namely ^{238,239,241,242,244}Pu. For actinium, several ionisation schemes were compared and hyperfine parameters extracted with a precision of a few MHz. Later, the results obtained for ^{227}Ac were used as reference data in a pioneering in-gas-jet spectroscopy experiment of $^{214\text{-}215}$Ac at the LISOL facility, Belgium. In order to test the reliability of the crossed-beams technique used in the Pu experiment, this work has recently been compared with high resolution collinear laser spectroscopy at the IGISOL facility, JYFL, and the combined data set has been sent for publication [5].

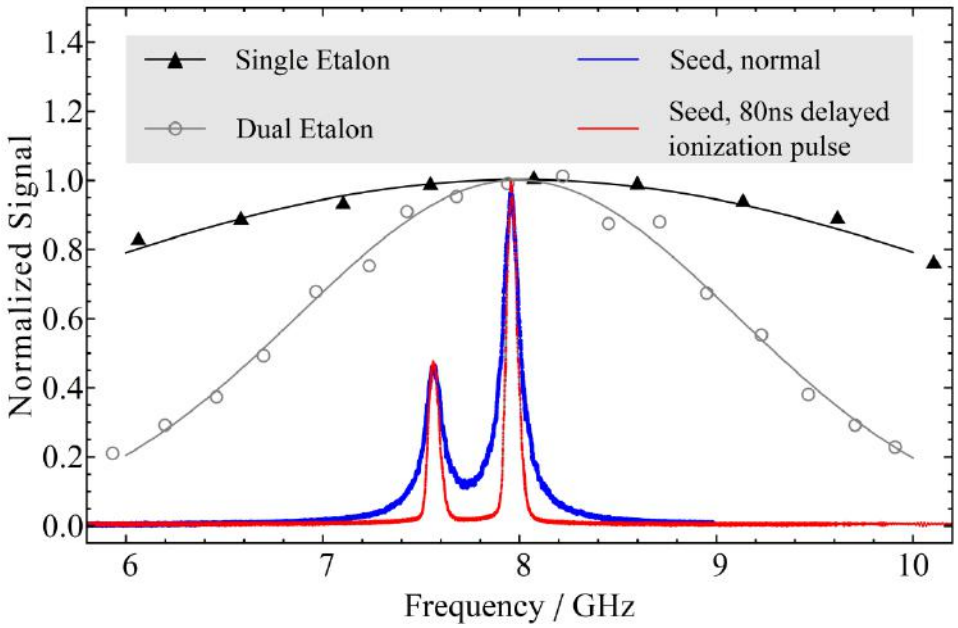

Figure 21: Ti:sapphire frequency scans of select hyperfine components of the 244-nm transition in stable ^{63,65}Cu. The improvement in resolution due to the reduction in laser linewidth from using a single- to double etalon Ti:sapphire laser, and using the injection-locked laser with and without delayed ionisation is highlighted.

Following the experiments in Mainz, the JYFL laser cavity was then shipped to the LISOL facility, Belgium. As already noted, a pioneering experiment was performed which demonstrated the first online application of atomic resonance ionisation spectroscopy in a supersonic gas jet. The technique was characterised in a measurement of ^{214,215}Ac around the N=126 neutron shell closure. Here, the injection laser was used in combination with seeding from a diode laser, and allowed a full exploitation of the cold gas environment to be made [6].

Most recently, the laser has been in use at ISOLDE in connection with the Collinear Resonance Ionisation Spectroscopy (CRIS) beamline for a successful campaign of high resolution optical spectroscopy experiments on

neutron-rich copper isotopes. Currently, a new injection-seeded laser is under development at JYFL and will be used for gas-jet spectroscopy at the MARA recoil separator as well as the future S^3 separator, SPIRAL-2, GANIL.

[1] Yu. Kudryavtsev *et al.*, Nucl. Instrum. and Meth. B **297** (2013) 7
[2] V. Sonnenschein, PhD thesis, University of Jyväskylä (2014)
[3] V. Sonnenschein *et al.*, to be submitted (2017)
[4] R.P. de Groote *et al.*, accepted for publication in Phys. Rev. A (2016)
[5] A. Voss *et al.*, submitted to Phys. Rev. A (2016)
[6] R. Ferrer *et al.*, Nat. Commun. **8** (2017) 14520

Sub-task 3.3: Investigation of new laser concepts for future RIB facilities as well as difference frequency mixing and Raman shifting to produce visible radiation from Ti:sapphire lasers.

JGU Mainz: Difference frequency generation and spectroscopy of sodium

A proof-of-concept for the generation of visible radiation in the missing wavelength gap of 480 to 680 nm of the Ti:sapphire lasers was provided by using difference frequency generation (DFG). The demonstration of the usability of this technology was done by three-step resonance ionisation spectroscopy in sodium, performed at JGU Mainz [1]. The excitation scheme included the well-known D_1 and D_2 transition doublet from the ground state with wavelengths of 589.76 nm and 589.16 nm for the first excitation steps, which were accessed via DFG. For this purpose two Ti:sapphire lasers were operated at wavelengths of 920 nm (idler - fundamental) and 718 nm (pump - frequency doubled) with the latter being frequency-doubled to generate a wavelength of 359 nm. The pump and the idler beam were overlapped in a BBO crystal to generate the visible radiation at 589 nm. An average power of well above 10 mW was obtained, which is by far enough for saturation of strong ground state transitions. The value corresponds to a conversion efficiency of $I_{DFG}/(I_{idler} + I_{pump}) = 0.3$ %, which may be improved in the future.

Using two further excitation steps in the fundamental Ti:sapphire wavelength regime for resonance ionisation, spectroscopy of odd-parity Rydberg levels was performed. A corresponding spectrum for Rydberg excitation in the range of n = 12 to 60 is given in Fig. 22, precisely delivering the convergence to the ionisation potential, indicated by "IP" in the figure. The level positions and the extracted value for the first ionisation potential are in perfect agreement with the very precise data from literature, which was used as an additional calibration for the wavelength measurement equipment. Data have been published in Ref. [1].

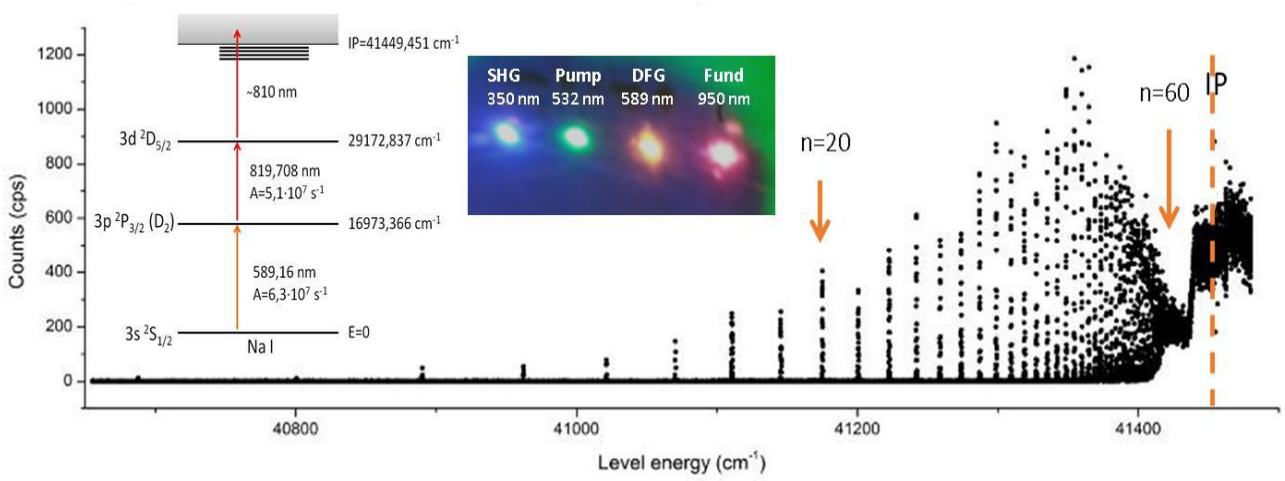

Figure 22: Spectrum of the third step excitation in Na I with Rydberg levels between n = 12 and 60. The complete excitation ladder and a photo of the four laser beams used for DFG and the IP are indicated in the inset [1].

[1] P. Naubereit et al., Phys. Rev. A **93** (2016) 052518

LNL-INFN: the off-line SPES laboratory and a time-of-flight mass spectrometer

An upgrade phase for a time-of-flight (ToF) mass spectrometer has been started in order to have a better instrument to analyse the laser resonant ionisation process and related efficiency. The aim is to have a calibrated evaporation source to provide atoms for laser resonant ionisation. To date, atoms are produced by laser ablation. A thermo-electro-mechanical (TEM) finite element analysis simulation of the oven has been performed and a first prototype is under testing to benchmark simulations, as illustrated in Fig. 23.

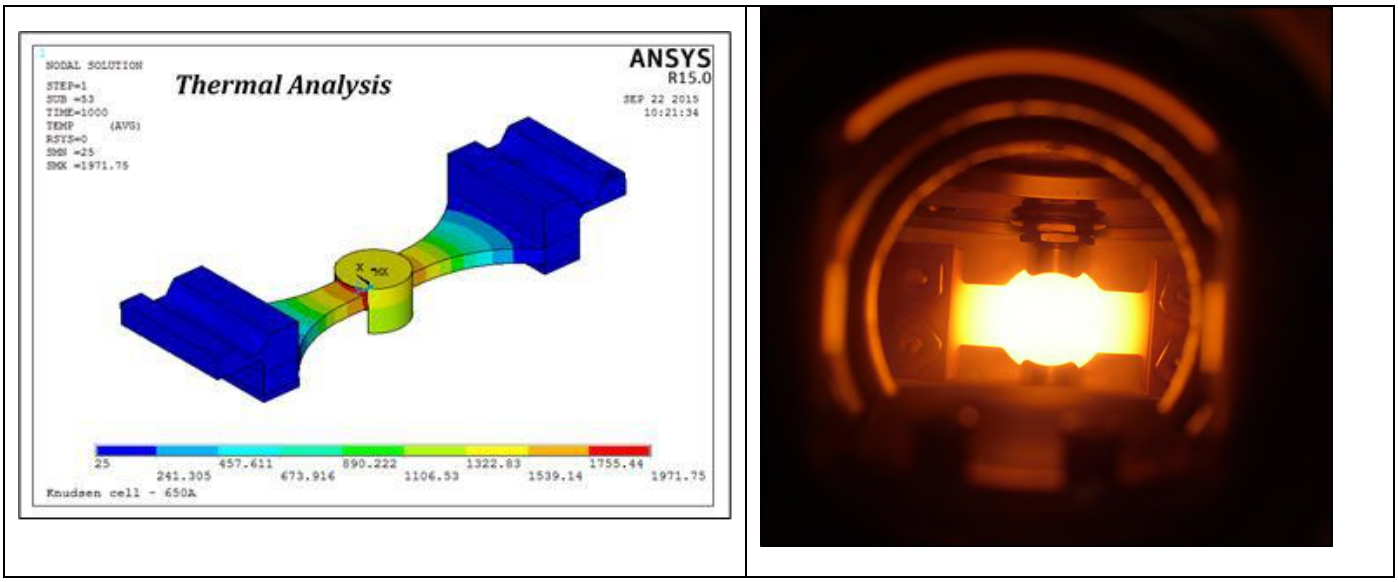

Figure 23: FEM oven simulation (left) and first heating tests (right).

The first assembly of the oven system in the ToF chamber (Fig. 24) is expected in the second half of 2017, and tests with the laser are foreseen at the end of 2017.

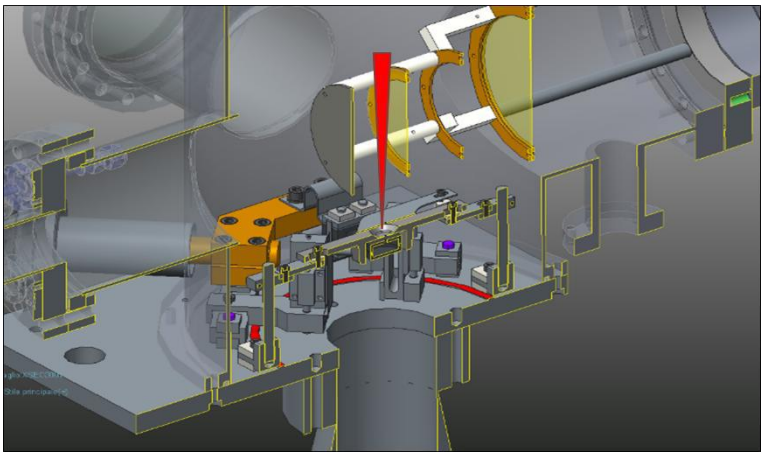

Figure 24: ToF chamber in the off-line SPES laser laboratory (left); 3D design of the installed oven (right).

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 which will ensure that Europe continues to lead the field in resonant laser ionisation and spectroscopy.

As highlighted in this report, the partners of RESIST are making good progress in all tasks and sub-tasks. Indeed, as reflected by the number of references, many of the developments have already been met and in several cases, exploited for physics.