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RESonance laser Ionisation techniques for SeparaTors (RESIST)

Deliverable 12.3 - Report on Task 2

ADVANCEMENTS IN EFFICIENCY, SELECTIVITY AND SPECTRAL RESOLUTION

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REFERENCES AND APPLICABLE DOCUMENTS

References have been added within the report at the appropriate sections.

LIST OF ACRONYMS AND ABBREVIATIONS

RESIST	RESonance laser Ionisation techniques for SeparaTors
RILIS	Resonance Ionization 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 deliverable due in month 24 of ENSAR-2. It focuses on the status of Task 2 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 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.

This document herein reports on the second 12 month period of activity of the RESIST partners involved in Task 2. We refer to Deliverable 12.1 (reported at the end of the first 12 month period) for a more detailed background to the different sub-tasks which are discussed independently in the following sections.

SECTION 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

In Deliverable 12.1 we presented a summary of the activities at the velocity filter SHIP at GSI, in which 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) [1].

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 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. 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

GANIL: a new detection system for the coupling of the S^3 mass separator and the IGLIS gas cell

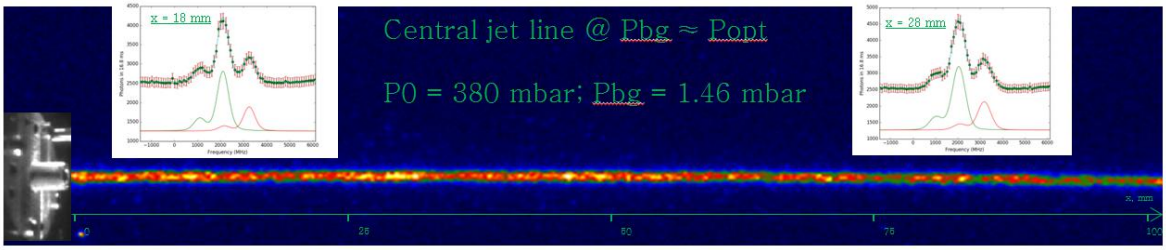
S^3 is the new in-flight Super Separator Spectrometer under construction at GANIL. A new detection system has been designed to efficiently couple S^3 to an IGLIS-type gas cell. The physics output foreseen is very broad, from spectroscopy of heavy and super heavy elements to studies of lighter *N*=*Z* nuclei. This detection system has to provide the incoming ion beam profile and to allow for the optimization of the S^3 settings, i.e. beam-like ion reduction and particle identification. It consists of a Secondary Electron Detector for time-of-flight and position measurement and a Double-Sided Striped Silicon Detector for time-of-flight, position and energy measurement. Alpha/proton correlations with implanted ions are foreseen for ion identification purposes. When the latter is not possible, an additional Coaxial Germanium detector can be used for Gamma-ray detection, thus identification through isomeric decays will be possible. The detection system is under construction and first off-line commissioning is planned by mid-2018.

SECTION 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. In Deliverable 12.1 we presented an extensive summary of studies with stable copper isotopes using a nozzle with a Mach number $\sim$6, via a visualization technique called Planar Laser-Induced Fluorescence (PLIF). We note that a similar design of nozzle was very successfully used in the online experiments to study the nuclear and atomic properties of short-lived ^{214}Ac and ^{215}Ac, reported in Refs. [1, 2].

In these tests, copper atoms seeded in argon are extracted through the nozzle and excited by a laser-sheet beam. The 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 1 illustrates the formation of a 100-mm long and collimated jet under optimized experimental conditions. By means of PLIF spectroscopy we obtained information on the local jet temperature (width of the spectral lines) and jet velocity (Doppler shift) along the jet central line. A total width (FWHM) of 650 (60) MHz was obtained in agreement with the expected performance of the nozzle at the working stagnation temperature.



$$FWHM_{full\ jet} = (650 \pm 60) MHz$$

Figure 1: PLIF image of a 100-mm long jet obtained under optimal conditions with the spectroscopic results corresponding to the local jet areas at x= 18 and 28 mm. Insets highlight the hyperfine spectra obtained from the fluorescence emitted by the copper atoms.

Recent work has focused on performing PLIF spectroscopy using nozzles made from different materials and surface roughness. First results obtained with brass and stainless steel nozzles following the same contour design do not show significant differences in their flow parameters ruling out possible effects arising from their distinctly different thermal expansion coefficients. Figure 2 shows the results obtained with the highest Mach number obtained so far (M~6) which was achieved using a stainless steel nozzle with an optimized contour roughness.

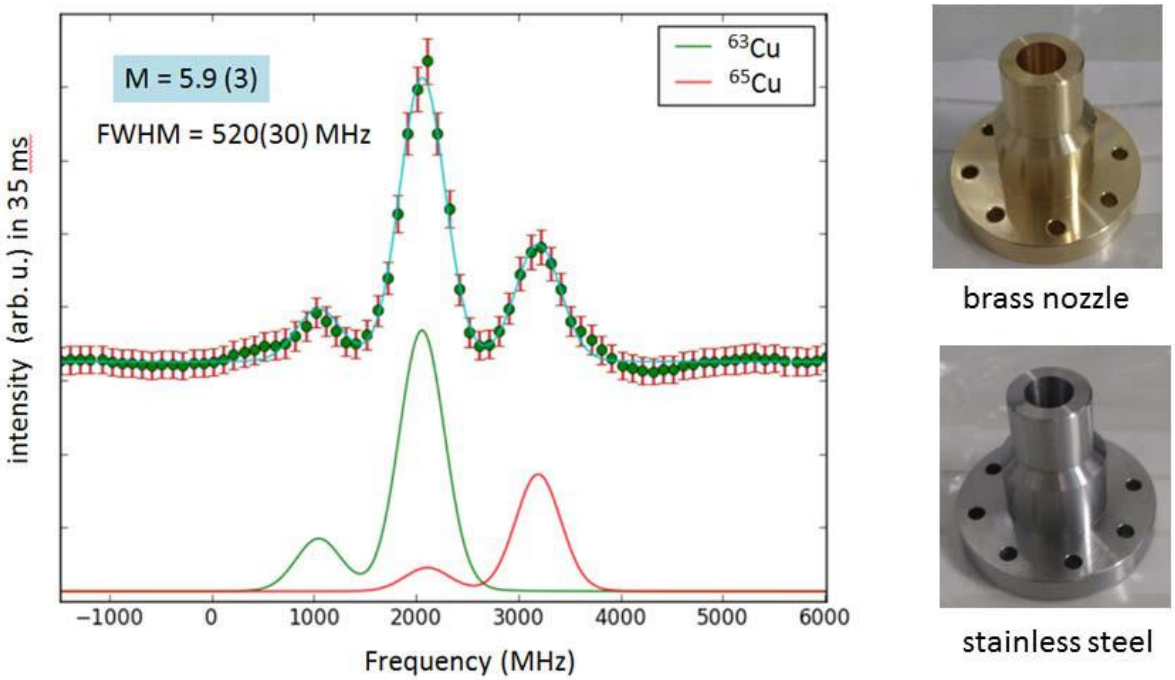


Figure 2: PLIF studies on stable copper atoms using two different nozzles (brass and stainless steel). The hyperfine structure fits for the two isotopes are indicated in the lower part of the figure.

The nozzle temperature was also studied using a mini PT-100 sensor which was separated from the inner contour of the nozzle by a layer only 1 mm thick. As shown in Fig. 3, no significant cooling of the nozzle was seen during working conditions over a time period of around 3 hours.

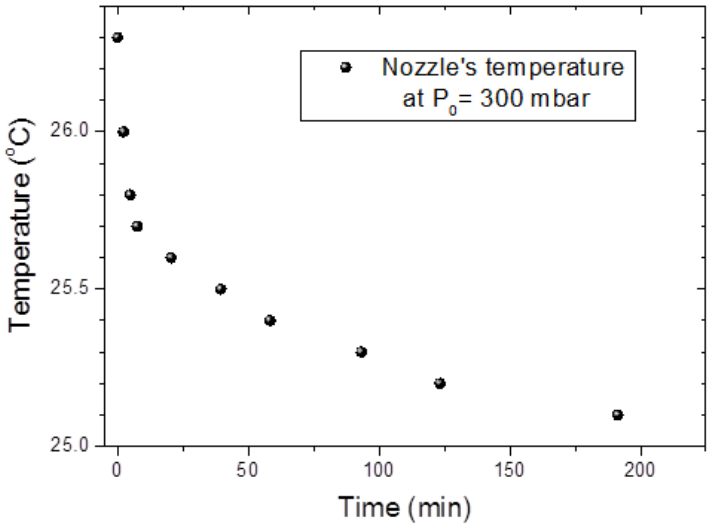


Figure 3: Nozzle temperature as a function of time.

[1] R. Ferrer *et al*., Nat. Commun. 8 (2017) 14520
[2] C. Granados *et al*., Phys. Rev. C 96 (2017) 054331

SECTION 3: EXTENSIVE IONISATION SCHEME DEVELOPMENT TO OPTIMISE ISOBARIC, ISOTOPIC AND ISOMERIC SELECTIVITY

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.

In Deliverable D12.1 we discussed the extensive work for the development of new schemes of resonance ionization and highlighted a number of results. In 2017 this work continued and Fig. 4 presents an updated picture of the schemes which have been investigated during the period of ENSAR2. The new results include:

- Laser ionization of selenium has been achieved for the first time. Using a transition from a rather weakly populated atomic level the ionization efficiency in a VADLIS ion source was measured to be 1.2%.
- Ionisation schemes of titanium, scandium and samarium have been developed and tested on-line.

U-Mainz: Excitation scheme development

Ionisation scheme development at Mainz University focussed on two aspects: (1) as novel technology, high-resolution two-photon spectroscopy, applied either longitudinally in-source inside the hot cavity (by a rear mirror) or in the perpendicular-illuminated LIST by a retro-reflecting mirror was tested. Comparison was made in the spectrum of rubidium using the $s_{1/2}$-$d_{5/2}$ two-photon transition with the D2 $s_{1/2}$-$p_{3/2}$-$d_{5/2}$ two-step excitation process. The hyperfine structure of the $s_{1/2}$ ground and the first excited $p_{3/2}$ state were clearly resolved with linewidths in the order of 60 MHz, illustrated in Fig. 5. Further analyses of the two-photon excitation regarding resolution and sensitivity are in progress.

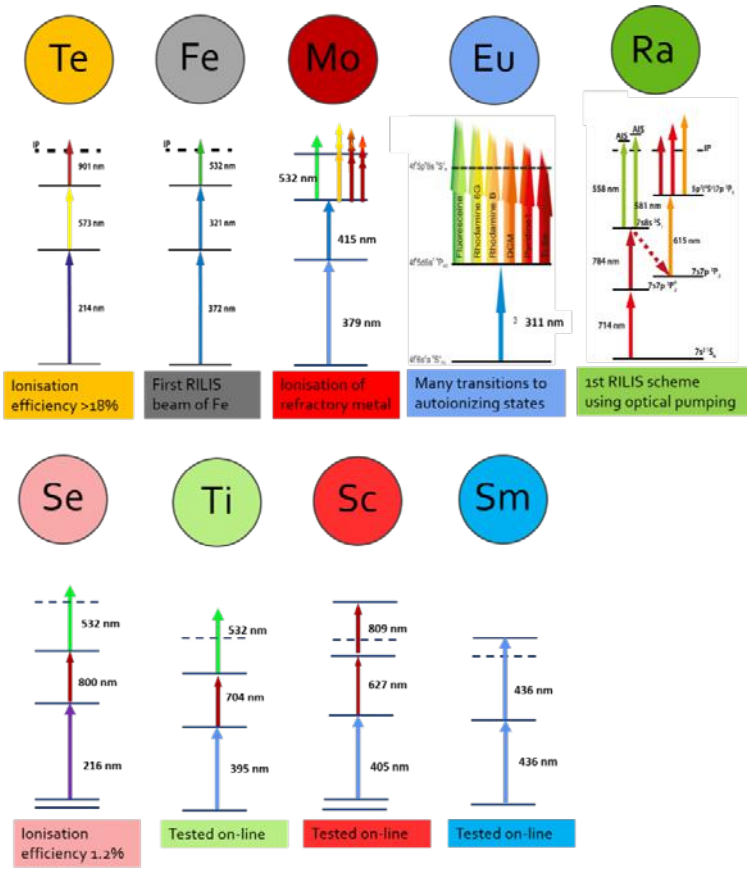


Figure 4: Resonance ionisation schemes for ISOLDE/RILIS developed at CERN.

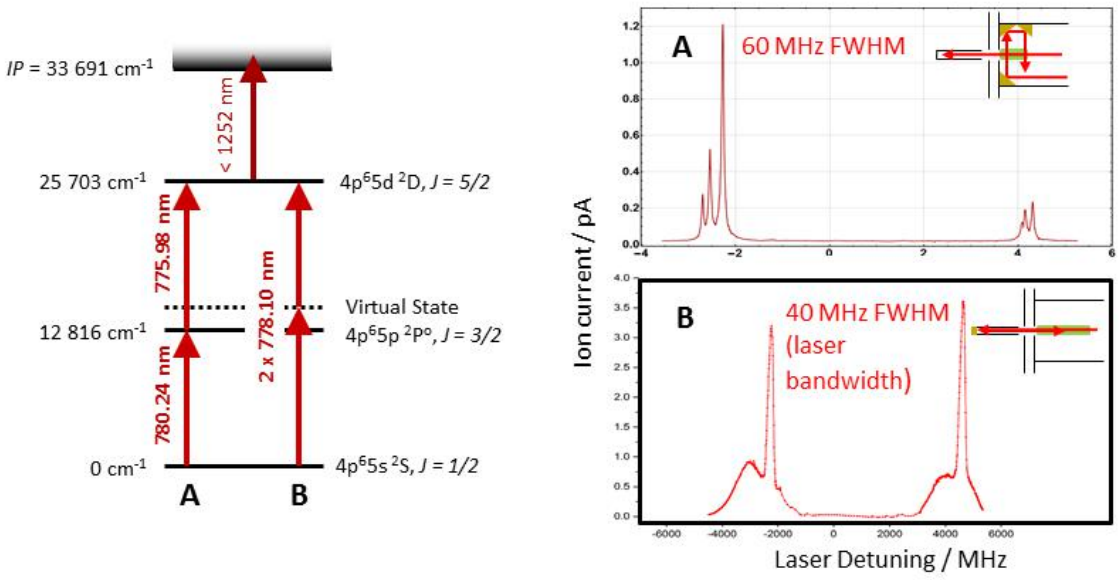


Figure 5: High resolution spectroscopy of Rb in the LIST. The two schemes shown on the left were tested: scheme A is a single-photon transition done in a perpendicular geometry (see inset of spectra A); scheme B is a two-photon transition performed in a collinear geometry. Residual Doppler broadening under the Doppler-free peaks can be seen in scheme B.

(2) Medium-resolution laser spectroscopy for scheme development and the precise determination of unknown ionization potentials were carried out on the long-lived isotopes ^{147}Pm and ^{231}Pa of the all-radioactive elements promethium (Z = 61) and protactinium (Z = 91). For both elements, over 1000 new transitions were studied covering the frequency doubled and fundamental ranges of 410 – 460 nm and 800 – 920 nm, respectively. Various 2-step and 3-step resonance ionization schemes were developed. The determination of ionization potentials for both elements is significantly complicated by strong configuration interactions of the various atomic levels and corresponding appearance of quantum chaos, particularly in the higher energy parts of the spectra. This so far has prevented a conclusive determination of an IP value for Pa while the precision in the determination for IP_{Pm} in the order of well below 1 cm^{-1} is envisaged from the spectra obtained so far. The excitation scheme and a spectrum of the third excitation step just below the predicted IP value is given in Fig 6.

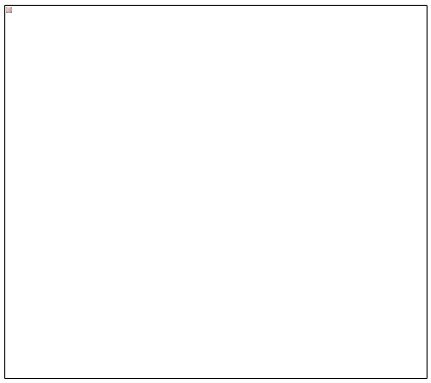


Figure 6: Ionization scheme and spectrum of the third excitation step in Pa including the predicted position of the IP with the error highlighted by the green band.

In a second high resolution campaign the hyperfine structure in Pm was analyzed using the PI-LIST approach in combination with a narrow bandwidth injection-locked Ti:sapphire laser. Experimental linewidths of about 120 MHz were achieved and the hyperfine structure of the two lower excitation steps could well be resolved (Fig. 7).

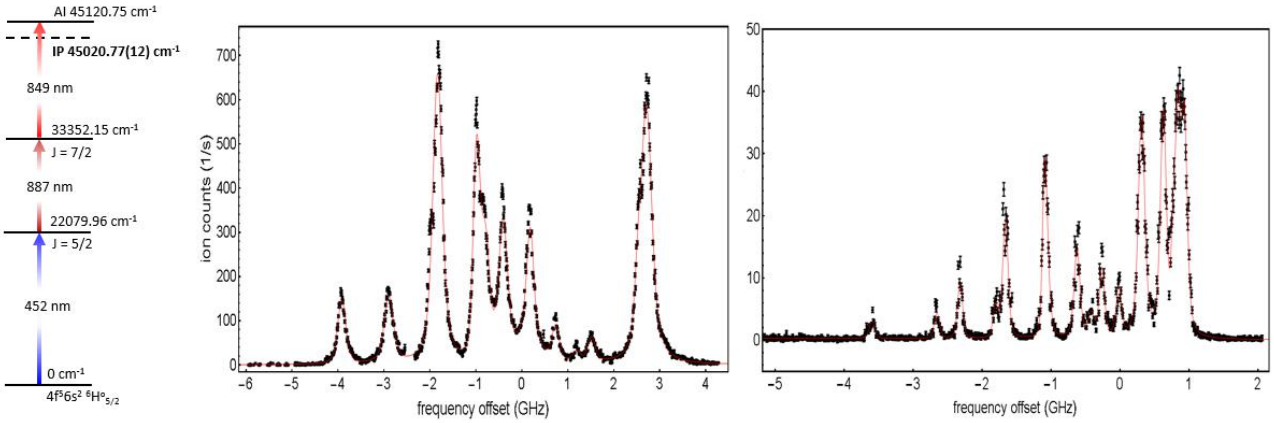


Figure 7: Ionization scheme and hyperfine structures of the first two excitation steps in Pm. The resulting spectral linewidth of the 452-nm transition was 180 MHz and that of the 887-nm transition, 115 MHz.

(3) Development of one-color and two-color excitation schemes for Tb and Nd. In order to allow for fast switching of the laser ionization schemes between different elements, new one- and two color excitation schemes

were developed at JGU Mainz. Similar to the one-color ionization scheme of Sm, which was successfully tested on-line at ISOLDE in 2017, the new schemes for Nd and Tb allow a simple laser setup and thus fast switching between elements, which can be done within minutes. At the RISIKO mass separator at JGU Mainz, terbium ionization efficiencies of 53 % and 33 % could be demonstrated for the two-color and one-color schemes, respectively. For Nd the spectroscopic data is currently under evaluation, efficiency measurements will be carried out in summer 2018.

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 with the development of techniques to advance the efficiency and selectivity of the laser ion sources, as well as the resulting spectral resolution in optical spectroscopic studies.