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Deliverable 12.7 – Final report on Task 3

NEW CONCEPTS AND DEVELOPMENT OF LASER TECHNOLOGIES

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Disclaimer

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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 final deliverable due in month 48 of ENSAR-2. It focuses on the status of Task 3 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 3 realises 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 total period of activity of the RESIST partners involved in Task 3. 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 AUTOMATED WIDE RANGE TUNABILITY OF SOLID-STATE LASER SYSTEMS FOR ATOMIC SPECTROSCOPY AND IONISATION SCHEME DEVELOPMENT

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

Wide range tunability is of highest relevance for comprehensive spectroscopy in unknown atomic systems, as e.g. found in Pm, Pa (U-Mainz), and in addition, permits for fast switching between different elements with rather similar spectral features, e.g. different lanthanides or actinides. Indeed, 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. In some cases it may well be useful to use a simple two-step ionisation scheme with the final transition to a very broad auto-ionisation (AI) state. To find such schemes, tunable frequency-doubling techniques are required, and one key goal has been to develop intra-cavity frequency doubling of a grating-based Ti:sapphire laser.

In November 2016, a team from the University of Nagoya, Japan, came to JYFL to commission such a laser. The cavity design is almost identical to the standard grating laser 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. The laser was successfully used in the study of the ionization scheme of Pu discussed in Deliverable 12.1 [1]. In collaboration with colleagues in Japan, the laser was subsequently used in the development of an efficient two-step ionization scheme for thorium [2]. Similar upgrades to the grating-based laser resonators are underway at U-Mainz where a demonstration measurement using two of these lasers to study different rare earth elements in sequence has been made.

More recently, progress continues in the development of grating-based lasers. At JYFL, a series of systematic studies have been performed using a self-seeding grating-based Ti:sapphire laser. The principal is rather simple, highlighted in the schematic of Fig. 1. A partially reflective (PR) mirror is installed inside the cavity of the traditional grating laser, resulting in two oscillators. Most of the laser radiation build-up is retro-reflected by the PR mirror forming a broadband resonator. A smaller amount of radiation leaks through the mirror towards the diffraction grating, effectively forming a second cavity which acts as a seed for the first resonator. The motivation behind this work is to address the shortcomings of the classic grating-based pulsed Ti:sapphire, which generally has lower output compared to the standard pulsed cavities in use. This is because of the limited reflectivity of the coating on the grating, local deformation and heating at high intra-cavity power. In the future, it is planned to extend this self-seeded cavity to also include intra-cavity frequency doubling.

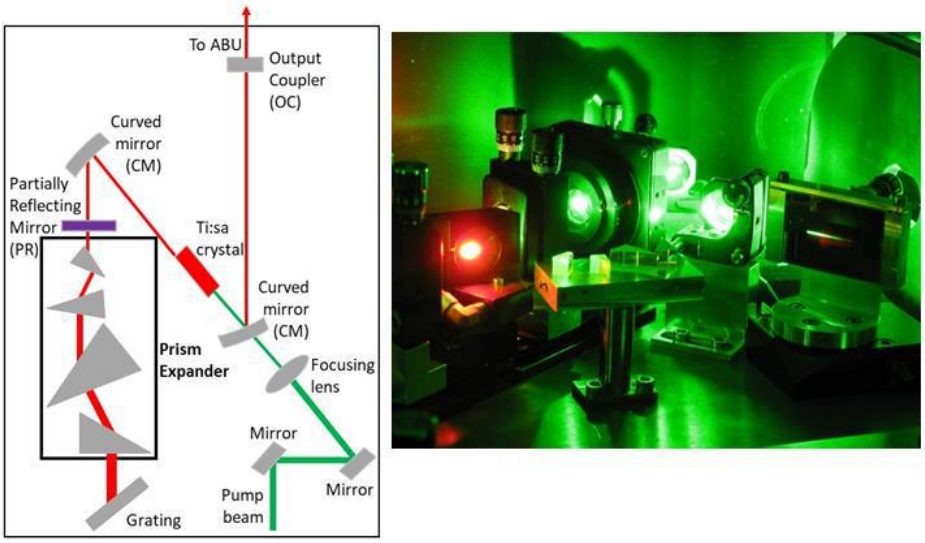


Figure 1: (Left) schematic of the self-seeding grating-based Ti:sapphire laser. (Right), photograph of the standard grating-based Ti:sapphire laser in use.

[1] I. Pohjalainen et al., Nucl. Instrum. and Meth. **B 376** (2016) 233.
[2] H. Tomita et al., Prog. in Nucl. Science and Technology **5** (2018) 97.

SECTION 2: 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

JYFL and Mainz: An injection-locked Ti:sapphire laser

An extensive introduction to the use of injection-locking techniques for linewidth narrowing of pulsed lasers with a narrowband cw laser has been presented in Deliverable D12.1. In the following a short summary is given.

The recent implementation of resonance ionisation spectroscopy (RIS) in a supersonic gas jet using a narrowband first resonant excitation step 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. In order to fully exploit the cold environment of an expanding supersonic gas jet, a considerable reduction in pulsed laser linewidth is required. Novel in-gas-jet spectroscopy offers 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.

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 [1]. 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. Figure 2 provides an overview of the injection-locked laser and the set-up used for locking and subsequent spectral analysis. The laser linewidth has been measured to be ∼20 MHz using a Fabry-Pérot Interferometer. This is a factor of approximately 200× lower than the pulsed lasers used in standard RILIS applications. The laser cavity has been used in a number of measurements at different radioactive beam facilities, summarized in [1], including the pioneering on-line application of the first in-gas-jet resonance ionization spectroscopy (at Louvain-la-Neuve) [2].

In Mainz, the injection locking of a Ti:sapphire laser by a continuous wave narrow bandwidth diode laser has shown its suitability for high resolution spectroscopy in various elements, e.g. Rb, Tc, Ac and Pm. To prevent the dependence on the availability of numerous individual single mode laser diodes, each delivering only a very limited spectral band, injection locking using a cw single-mode Ti:sapphire ring laser is presently under development, which will cover the whole accessible Ti:sapphire amplification range.

Separately, the development of an unseeded single-mode laser is underway. Compared to the standard Ti:sapphire resonator design, this laser features an additional air-spaced etalon in order to achieve single-mode operation. Currently the laser is operated in pulsed mode, which prevents continuous stabilization of the resonator. However already without active stabilization of the cavity length a linewidth of ∼600 MHz can be achieved by fast scanning of a piezo cavity mirror, thus effectively averaging over fast frequency jitter. An example of the laser in operation for isotope shift studies in dysprosium is shown in Fig. 3. Further suppression of side modes is currently under investigation in order to narrow down the effective linewidth.

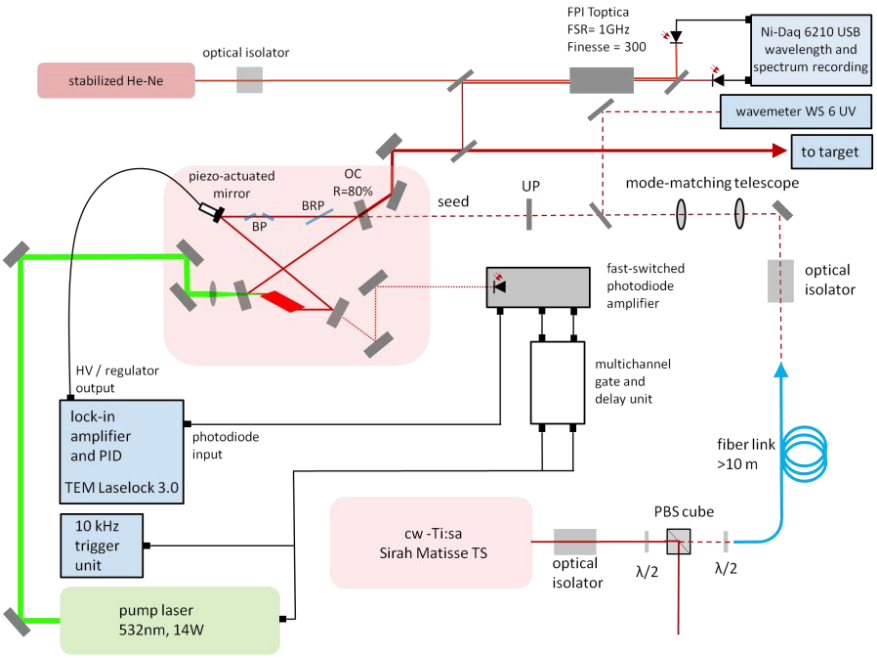


Figure 2: Overview of the setup of the injection-locked Ti:sapphire laser system at JYFL [1].

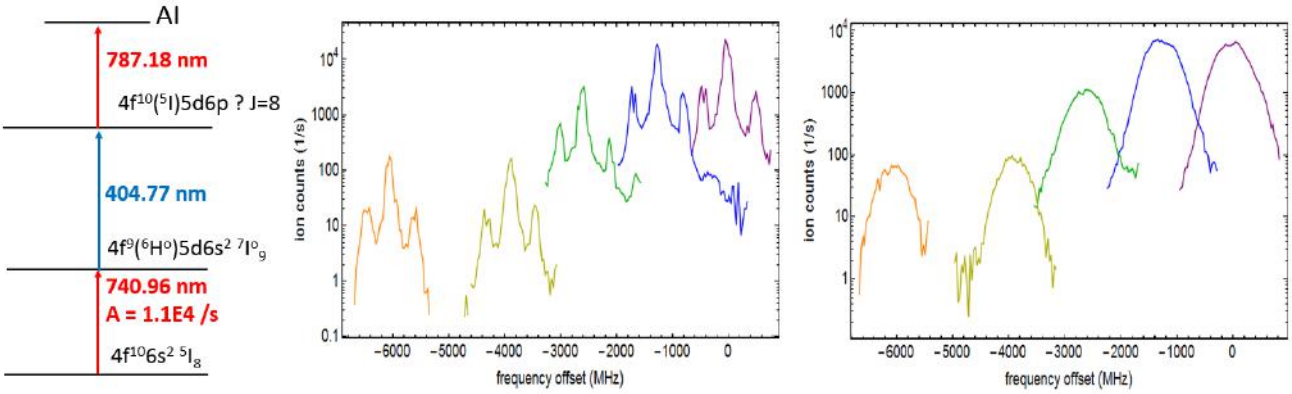


Figure 3: Isotope shifts of the 741-nm ground state transition in stable, zero-spin dysprosium isotopes. The unstabilized laser operation leads to strong frequency jitter and visible side bands at ~450 MHz, suppressed by approximately one order of magnitude (left). By fast scanning of a piezo-mounted cavity mirror a stable linewidth of ~600 MHz is achieved (right).

Currently, a new injection-seeded laser has been characterized at JYFL, highlighted in Fig. 4, and will be used for gas-jet spectroscopy at the MARA recoil separator. A similar injection-seeded laser cavity will be in operation at the S^3 separator, SPIRAL-2, GANIL.

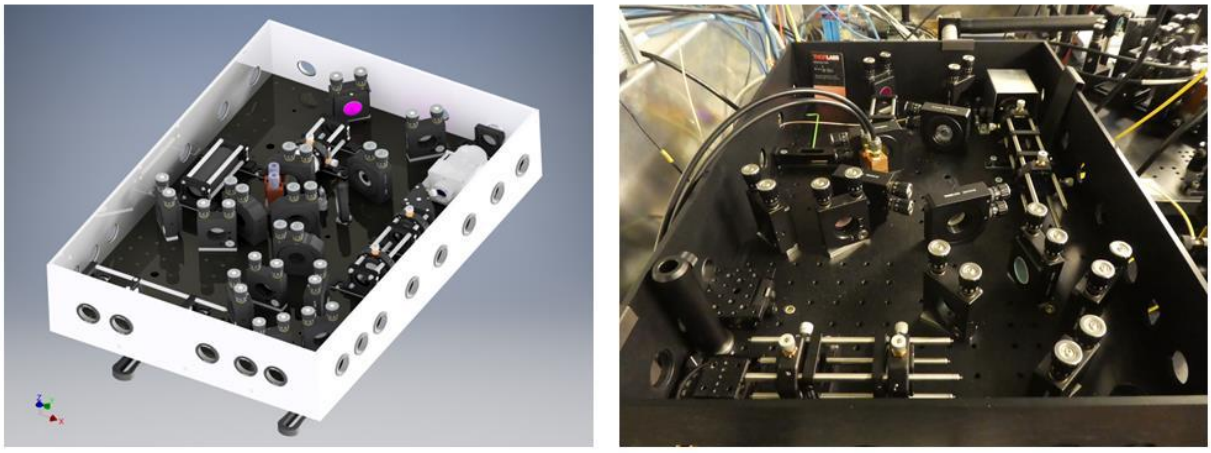


Figure 4: (Left) 3-D design of a new injection-locked Ti:sapphire laser cavity for use at the future MARA-LEB facility. (Right) Photo of the finished laser resonator.

[1] V. Sonnenschein *et al*., Laser Phys. 27 (2017) 085701
[2] R. Ferrer *et al*., Nat. Commun. **8** (2017) 14520

SECTION 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

In Deliverable 12.1 we highlighted 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 using difference frequency generation (DFG). The demonstration of the usability of this technology was done by three-step resonance ionization spectroscopy in sodium, performed at JGU Mainz [1]. Similar work was performed in parallel at JYFL.

An alternative way for producing light in the 480-680 nm wavelength range is through so-called Raman shifting. Instead of frequency mixing light of different wavelengths, the output light of a single laser can be shifted to higher wavelengths inside of a Raman medium. Since the shifted light can be reabsorbed inside the medium and shifted multiple times more ("cascaded"), one distinguishes between different "Stokes orders". This method enables a large spectral range to be spanned. Raman lasers are already commonly used for telecommunication applications and show great promise to become an efficient, reliable and easy-to-use alternative to frequency mixing in the visible wavelength range. A multitude of materials are available for Raman shifting, with diamond showing the best characteristics for the UV to IR spectral range, possessing a high damage threshold and conversion efficiency. The first demonstration for converting light from a Ti:sapphire laser with diamond as the Raman medium has been published in [2] and was selected as Editor's pick. A schematic for the laser is shown in Fig. 5.

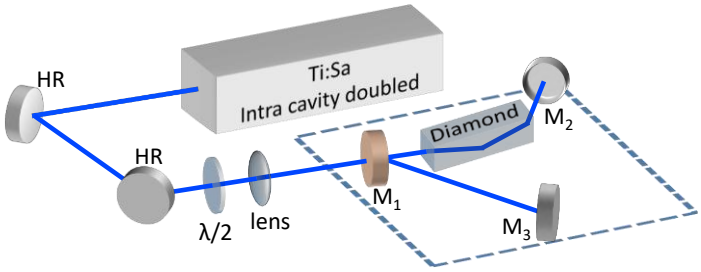


Figure 5: Layout of the first external Raman laser, which was described in detail in [2].

This work has shown that the typical linewidth of the Ti:sapphire lasers used throughout the RESIST network (typically several GHz) can be conserved during the Raman process. This is especially important for the application of resonance laser ionization, in which the hyperfine structure and isotope shifts have to be covered for efficient excitation. Since the Raman conversion is wavelength independent, only the output wavelength of the Ti:sapphire laser needs to be adjusted, making changes in wavelength faster and easier than with frequency mixing. A more thorough investigation into the cascading of the Stokes orders has been launched and first spectroscopy experiments using the Raman-shifted light have been conducted [3]. Figure 6 highlights different Stokes orders which can be generated. Currently, two different approaches for building a Raman laser are under investigation: 1) an external conversion unit, which is transportable, that can be used in a "plug-and-play" manner and pumped by any given laser; 2) intra-cavity Raman conversion inside the "home-built" Ti:sapphire cavities for enhanced tunability and additional access to deep infra-red wavelengths (960-1080nm).

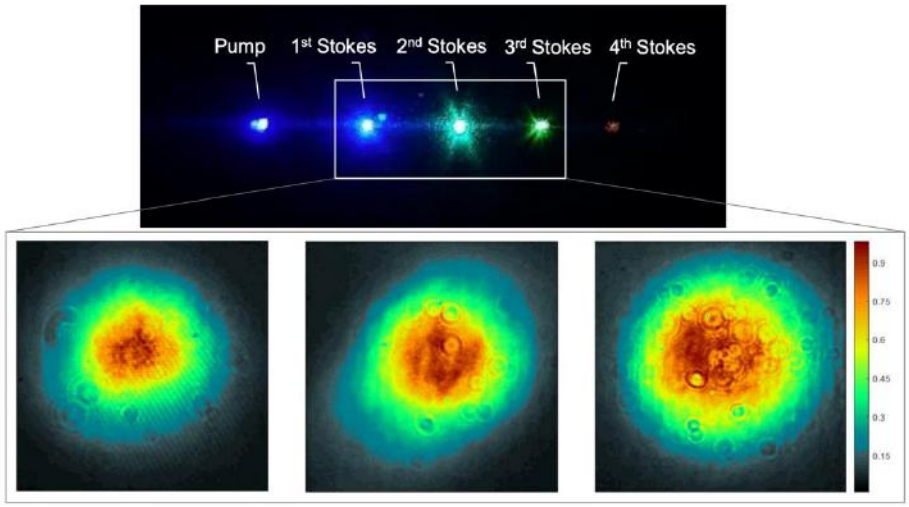
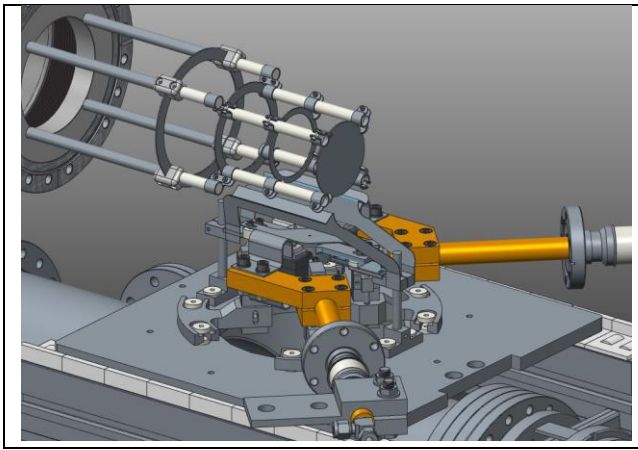


Figure 6: Different Stokes orders which can be generated with laser light at 450 nm (top) and the corresponding laser beam profiles (bottom). A more detailed investigation into the cascading effect was launched and details will be published in [3].

[1] P. Naubereit *et al.*, Phys. Rev. A **93** (2016) 052518.
[2] K. Chrysalidis *et al.*, Opt. Lett. **44,** 3924-3927 (2019)
[3] D. T. Echarri *et al., submitted to Optics Express*

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

Most of the preparatory activities performed in the task of investigation of new laser concepts for the future RIB facility, SPES, have been focused on the study and development of a new solid state laser system for photo-ionisation together with a company that will build the system. Other preparatory activities involve the design and construction of a home-made Time-of-Flight Mass Spectrometer (Fig. 7) as well as the design and the test of an evaporation atomic source.



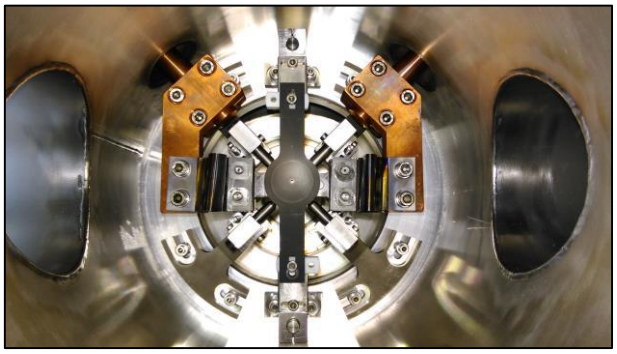


Figure 7: 3D drawings of the evaporation oven (left) and its preliminary installation (right).

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 have made excellent progress the development of new laser concepts and new technologies which will continue beyond the scope of ENSAR2.