Report on new ion manipulation methods, spectroscopic techniques and novel instrumentation

This final report summarizes the activities of Task 2 of the PREMAS Joint Research Activity. Much of the material has already been presented within the individual milestones and where appropriate, we refer to these as attachments within this deliverable. In the following we present a more general summary.

Task 2, advanced ion manipulation, spectroscopic techniques and instrumentation (led by JYFL) is sub-divided into 5 sub-tasks:

Subtask 2.1 (KU-Leuven): Investigation of optimized trap structures and development of related dedicated novel detection setups for decay spectroscopy with ion traps.

A full report on this activity may be found in the updated attached milestone report MS102 – new trap spectrometer characterized and experiment performed. This new compact beta spectrometer has been designed for precision beta spectrum shape measurements and completed in collaboration with the University of Krakow, Poland.

Subtask 2.2 (Manchester): Development and demonstration of collinear resonance ionization spectroscopy coupled to a gas-filled RF cooler facility.

Collinear resonance ionisation spectroscopy (CRIS) combines the advantages of resonance ionisation spectroscopy (a detection efficiency of up to 50%, high selectivity and ultra-low background) with collinear laser spectroscopy of atoms (high resolution due to the Doppler compression of the spectral linewidth). A dedicated CRIS beam line is now under operation at ISOLDE, CERN, profiting from the availability of bunched beams from the ISCOOL cooler-buncher.

Figure 1 illustrates the experimental layout and serves to highlight the method. Ions from the target ion source are mass separated, cooled and released as bunches from ISCOOL, following which they are neutralised in a charge-exchange cell. The non-neutralised component is deflected in a differential pumping region while the atom bunches proceed to a UHV region where they are resonantly ionised in a multi-step laser ionisation scheme. The capability to bunch the radioactive ion beam enables a 100% temporal overlap with synchronised pulsed lasers, alleviating the losses due to the few-percent duty cycle which limited earlier pre-cooler-buncher work. Resonantly produced ions are either deflected towards a multi-channel plate detector or implanted into a C foil for charged-particle decay spectroscopy.

In a recent experimental campaign, isotope shifts and magnetic moments of neutron-deficient Fr isotopes were measured with half-lives as short as 300 ms and production rates at a level of <100 atoms/s [1]. In that work, a total experimental efficiency of ~1% has been demonstrated. A background pressure in the laser-atom

interaction region of 8×10^{-9} mbar resulted in contamination from non-resonant and collisional ionisation of below one ion in 10^5 beam particles.

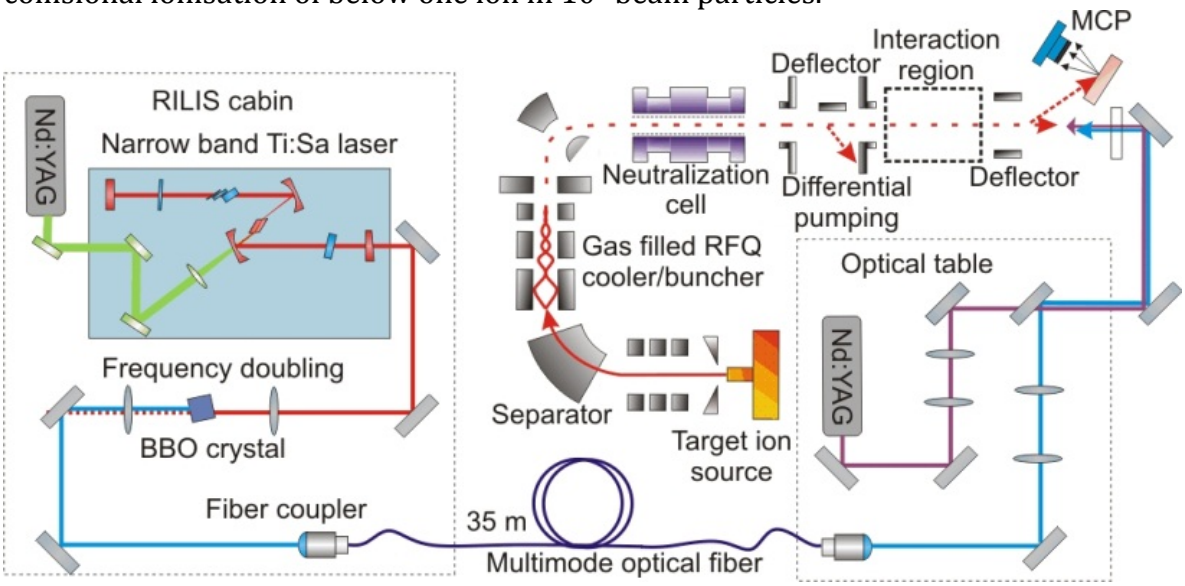

Fig. 1: Schematic representation of the CRIS experimental layout.

The CRIS technique not only provides access to the study of hyperfine structure and isotopes shifts of nuclei at the limits of stability, but offers a powerful means to selectively purify an ion beam suffering from isobaric contamination, including separation of nuclear isomeric from ground states. Such laser-assisted separation of nuclear states is of particular interest to nuclear spectroscopy studies and thus has resulted in a decay spectroscopy station (DSS) being built at the end of the CRIS beam line (downstream from the MCP illustrated in Fig. 1).

The DSS consists of a rotating wheel C foil implantation system and passivated implanted planar Si detectors for charged-particle detection. Gamma rays may be detected by up to three high-purity germanium detectors placed around the implantation site [2]. In a first application of decay-assisted laser spectroscopy the DSS was used to identify components in the hyperfine spectra associated with low-lying nuclear states in neutron-deficient ^{202,204}Fr. An alpha decay measurement as a function of laser frequency, illustrated in Fig. 2, led to the determination of nuclear observables from the three low-lying isomeric states in the decay of ^{204m2}Fr [3].

In the most recent publication, the CRIS collaboration compared changes in mean-square charge radii of neutron-rich Fr isotopes (^{218m,219,229,231}Fr) with re-evaluated Ra data in a region which lies on the border associated with reflection-asymmetric shapes [4].

The current limitation in the application of CRIS is related to the linewidth of the tunable pulsed lasers used in the spectroscopy, namely a dual-etalon Ti:sapphire laser pumped with a 10 kHz diode-pumped solid state laser. As discussed in ref. [5], the laser is optimized for in-source spectroscopy and has a linewidth of ~800 MHz. In the Fr work the spectral resolution was thus not sufficient to resolve the splitting of the excited $^8P_{3/2}$ state, and therefore it was not possible to determine the nuclear spin or quadrupole moments. In the fall of 2014, a new technique was used, namely delayed resonance

ionization spectroscopy. Recent simulations suggest that the linewidth of a transition is affected by the power broadening caused by the laser driving the ionization process [6]. A delay of the ionization step with respect to the first step excitation is expected to minimize this broadening mechanism. At JYFL, this was recently demonstrated on stable Cu isotopes wherein a very clear reduction in the Lorentzian linewidth of the 244-nm transition was seen as a function of delay of the ionization laser [7]. Importantly, no obvious systematic trends were observed in the extraction of the hyperfine *A* parameters for the ground and excited state in ^{63,65}Cu. At ISOLDE, the most recent CRIS beam time demonstrated an on-line linewidth of <20 MHz for Fr using the delayed-ionization technique, enabling a search for ^{201}Fr as well as the isomeric state in ^{203}Fr. The results are currently under analysis.

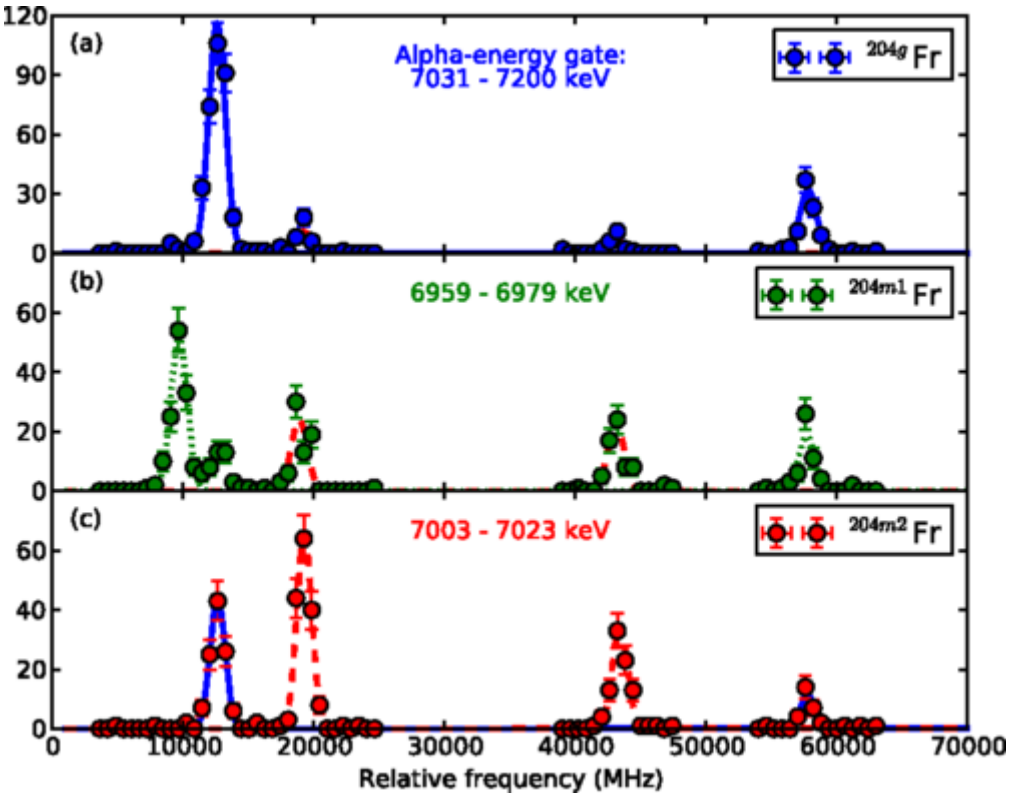

Fig. 2: Illustration of the use of decay-assisted laser spectroscopy at the CRIS beam line, ISOLDE. Alpha-tagged hyperfine structure of low-lying states in neutron-deficient ^{204}Fr. Figure from [3].

Milestone reports MS103 and MS104 are attached to this document.

[1]. K.T Flanagan, K.M. Lynch *et al.*, Phys. Rev. Lett. **111** (2013) 212501
[2]. M. Rajabali, K. Lynch, T. Cocolios *et al.*, Nucl. Instrum. and Meth. in Phys. Res. A **707** (2013) 35
[3] K.M. Lynch *et al.*, Phys. Rev. X **4** (2014) 011055
[4] I. Budinčević *et al.*, Phys. Rev C **90** (2014) 014317
[5] V. Sonnenschein, I.D. Moore *et al.*, Hyp. Int. **227** (2014) 113

[6] R. de Groote, Master's thesis, KU Leuven, 2013
[7] V. Sonnenschein, Ph.D. thesis, University of Jyväskylä, 2015.

Subtask 2.3 (JYU-JYFL): In-source spectroscopy in a gas-cell environment and development of laser spectroscopy in a gas-jet environment.

2.3.1 In-source spectroscopy in a gas cell

With the high efficiency afforded by resonance ionization spectroscopy (RIS) inside a buffer gas-filled ion guide, one has a very sensitive tool for a first (low-resolution) determination of isotope shifts and hyperfine structure of exotic nuclei. Experiments conducted so far at the Leuven isotope separator on-line (LISOL) facility have focused on neutron-deficient Cu and Ag isotopes, providing important information on changes in mean-square charge radii, and single-particle properties of ground- and isomeric states, namely nuclear spins and magnetic moments. Recently these studies were extended to the heavy-mass region, by resolving hyperfine structure of neutron-deficient $^{212-215}$Ac isotopes. Demonstration of in-gas-cell spectroscopy has been highlighted in the attached milestone reports MS105 and MS106. In the following we shortly discuss the developments in the laser technology required to realize appropriate linewidths.

At several institutes participating in the PREMAS activity solid-state Ti:sapphire laser systems are in use, with typical laser linewidths of 5 GHz in the fundamental. For spectroscopy purposes, this linewidth in many cases is the dominant spectral line broadening effect. In a gas-cell environment with typical operating pressures of several hundreds of mbar, a laser system with a medium resolution of about 1 GHz would be most suitable for in-source spectroscopy. At JYFL, Mainz and ISOLDE this has been realised in a relatively simple modification to the existing Ti:sapphire resonator design, namely the addition of a second etalon into the cavity. The first spectroscopy experiments of the dual-etalon laser were performed on stable copper at JYFL, resulting in a reduction of a factor of three in the spectral linewidth in measurements of the 327-nm transition.

The construction of a scanning plane-parallel Fabry-Pérot interferometer (FPI) allowed a direct study of the mode structure of the laser as shown in Fig. 3. Based on the number of modes seen in the spectrum (black solid line) and an approximate FWHM of the distribution, the linewidth of the dual-etalon resonator configuration has been estimated to be 0.85(10) GHz [1]. This value is similar to that reported by the RILIS collaboration, ISOLDE [2].

In collaboration with Mainz and Leuven, an exploratory laser spectroscopy study on heavy elements has begun at JYFL with long-lived isotopes of plutonium prepared with support from the Nuclear Chemistry department of Mainz using the Mainz research reactor. The goal is to use in-gas-cell laser ionization to prepare ion beams of Pu isotopes for high-resolution collinear laser spectroscopy. Plutonium would be so far the heaviest nucleus attempted with the fast beams collinear method and, as with Ac (discussed above), be a milestone towards spectroscopy of the superheavy elements. Figure 4 illustrates mass scans over the Pu^+ mass region following laser ionization of ^{240,242,244}Pu. Impurities mainly from the gas cell result in the formation of oxides as well as subsequent hydrate addition. The analysis of the data from this first test is underway.

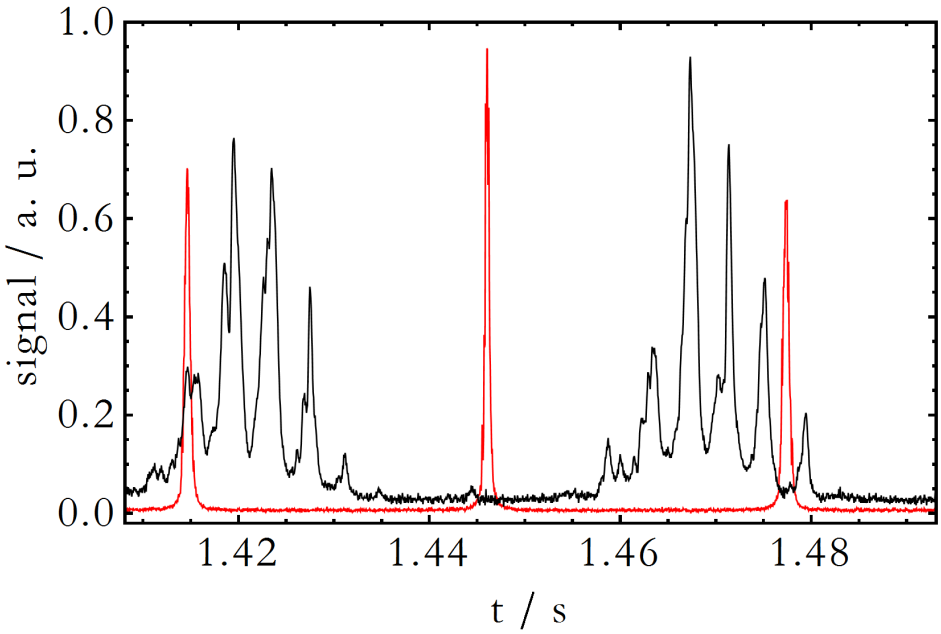

Fig. 3: A scan of the Fabry-Pérot Interferometer showing the multi-mode structure of the dual-etalon Ti:sapphire laser (black) and the reference fringes of the HeNe laser (red). A simple Gaussian approximation to the mode structure resulted in a FWHM of 0.85(10) GHz [1].

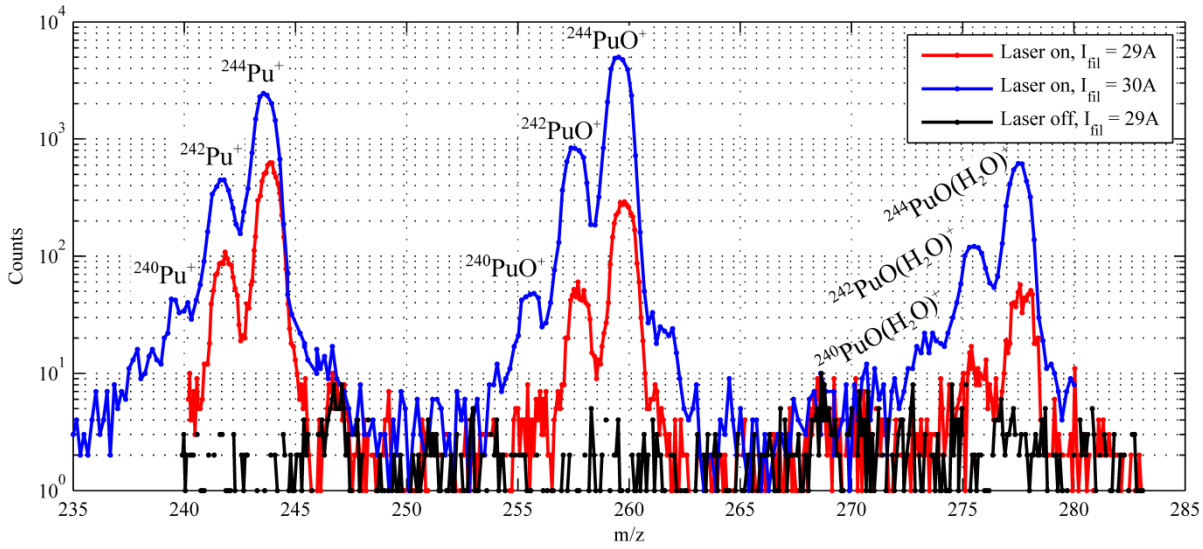

Fig. 3: Mass scan following laser ionization of Pu isotopes in a gas cell at JYFL.

[1]. V. Sonnenschein, I.D. Moore *et al.*, Hyp. Int. **227** (2014) 113
[2]. S. Rothe *et al.*, Nucl. Instrum. and Meth. in Phys. Res. B **317** (2013) 561

2.3.2 In-source spectroscopy in a gas jet

The spectral resolution of resonance ionization spectroscopy in a gas cell is limited by the collisional broadening contribution to the atomic linewidth. A novel approach to address such limitation is to perform spectroscopy in the expanding gas jet immediately

after the exit nozzle of a gas cell [1]. A combination of low temperature and low atom density in the supersonic jet suppresses the collisional broadening mechanism and substantially reduces the Doppler contribution, improving the spectral resolution of in-gas-cell spectroscopy by one order of magnitude.

To fully exploit the gas-jet environment, narrow bandwidth, high power, tunable laser sources are required. Recently this has been realized with the development of an injection-locked Ti:sapphire laser [2]. Using either a diode laser or cw Ti:sapphire laser acting as a Master laser, a bow-tie ring cavity is stabilized to a multiple of the Master wavelength. Tests of the new laser showed a reduction in linewidth to ~20 MHz, with an output power of close to 4W, achieved with only a few mW of injected cw laser power. The impressive reduction in linewidth is immediately evident in Fig. 4 which illustrates Ti:sapphire frequency scans of the $F_g=1 \rightarrow F_u=2$ hyperfine components of the 244-nm transition in ^{63,65}Cu using either a single etalon in the resonator, a double etalon (the so-called dual-etalon laser discussed previously) and the injection-locked laser, with and without the effect of delaying the ionization step (as discussed in this report in relation to the CRIS technique).

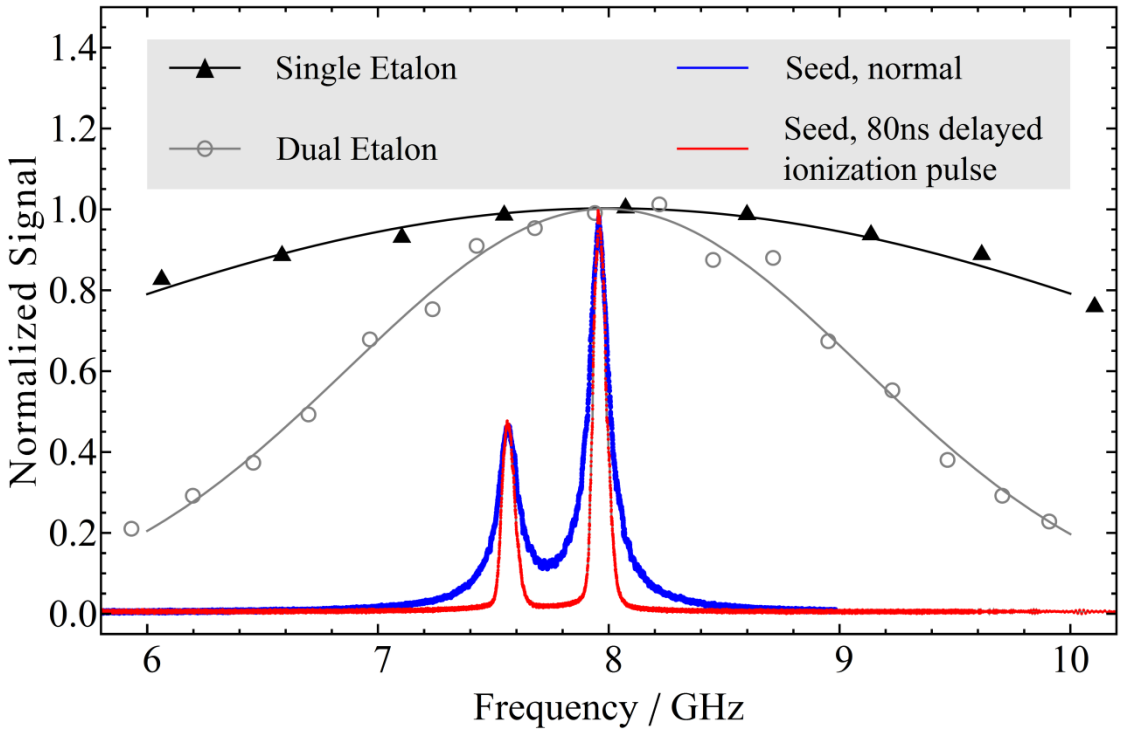

Fig. 4: Frequency scans of the $F_g=1 \rightarrow F_u=2$ hyperfine components of the 244-nm transition in ^{63,65}Cu. The improvements using the "seed" laser, the so-called injection-locking technique are demonstrated.

As highlighted in milestone MS106, the laser was used at Mainz to identify a more suitable transition for in-gas-cell spectroscopy of Ac at LISOL, using long-lived ^{227}Ac produced in the Mainz reactor. Later, in December 2014, the laser was successfully used to investigate the hyperfine structure of the 438-nm transition in ^{214,215}Ac. With a typical spectral resolution of ~400 MHz, isotope shifts as well as the hyperfine *A*- and *B*-

parameters were extracted with a 25-fold higher precision than that obtained using the same transition by in-gas-cell spectroscopy. Furthermore, the data afforded a better spin assignment for the N=126 ^{215}Ac ($t_{1/2}$=0.17s) nuclide. With a comparable efficiency to that measured in the gas cell (~0.5%) and a potential for an order of magnitude higher by increasing the duty cycle, gas-jet spectroscopy promises access to species with very low production rates (~10^3 pps) and/or with half-lives well below 100 ms.

[1]. Y. Kudryavtsev *et al*., Nucl. Instrum. and Meth. in Phys. Res. B **297** (2013) 7.
[2]. V. Sonnenschein, Ph.D. thesis, The University of Jyväskylä (2015).

Subtask 2.4 (Manchester): Optical manipulation of radioactive ions in gas-filled RF coolers leading to population of specific ionic states and nuclear polarizations.

Optical pumping (the use of light to manipulate atomic state or sub-state populations) features frequently in the spectroscopy of exotic species. In connection with PREMAS, optical pumping has been used to move populations to states more efficient or better suited to collinear laser spectroscopy. In the near future, optical manipulation will be attempted in the laser-interaction region of an electrostatic trap at JYFL, the device based on the ConeTrap pioneered by Schmidt *et al*. This dynamic multi-reflection trap has been shown to successfully contain cooled ions on millisecond time scales (without perturbation of the energy or energy spread of extracted ionic ensembles). In the trap, laser spectroscopy, at 800 eV total acceleration energy, can be performed with collinear interaction lengths corresponding to hundreds of meters.

A summary of this activity may be found in the attached milestone report MS107 – optical manipulation demonstrated on-line with several elements.

Subtask 2.5 (CSNSM): Efficient transport (variable energy, beam distribution, reduced temporal pulse structure) of the highest intensity of short-lived exotic species.

A summary of this activity may be found in the attached milestone report MS109 – design document on the schemes for element-selective transport and beam distribution.

MS102: New trap spectrometer characterized and experiment performed

Construction of a new compact spectrometer for precise beta spectrum shape measurements (miniBETA), including signal processing electronics and read-out software, has been completed (in collaboration with the Jagiellonian Univ. of Krakow). The spectrometer consists of two multi-wire drift chambers with hexagonal cells for tracking of beta particles, each combined with a 10 cm x 10 cm plastic scintillator used as trigger for the wire chamber, with the beta particle energy being determined from the curvature of its trajectory in a magnetic field. The main aims of the spectrometer are to obtain better understanding of effects in beta decay induced by the strong interaction, thereby improving the precision of beta-decay correlation measurements in search for exotic weak interaction components , and the direct search for such interactions via the contribution of the Fierz term to the beta spectrum. Commissioning and further optimizations of the system (including replacement of the two scintillators by double-sided silicon strip detectors) in view of first measurements are presently ongoing.

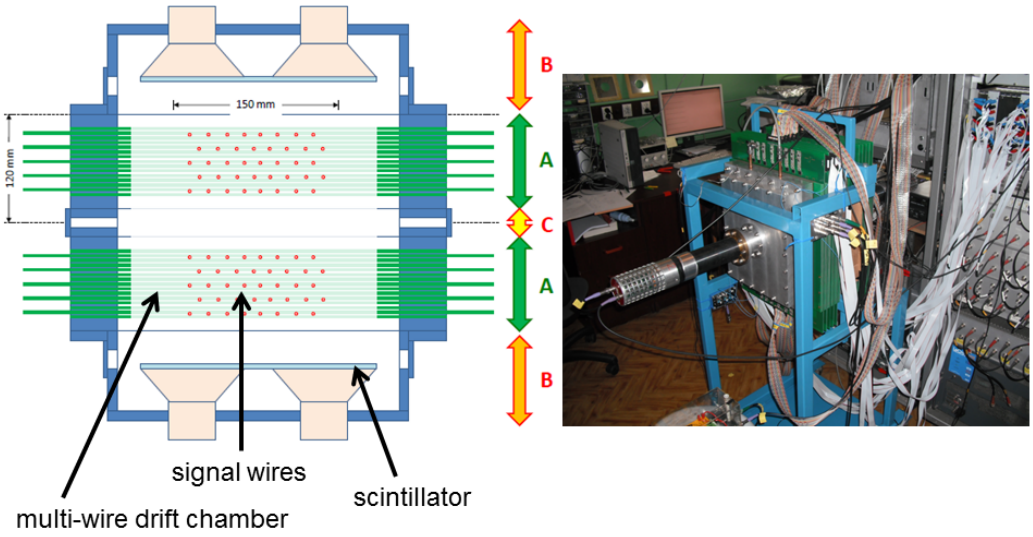

Fig. 1: Schematic of the new beta spectrometer, miniBETA, (left) and photograph (right).

MS103: Collinear RIS demonstrated with a stable beam (month 24)
MS104: First on-line results using collinear-RIS to study rare Fr isotopes (month 36)

Herewith we summarize both Milestones MS103 and MS104 together as the first results for the new method of Collinear Resonance Ionization Spectroscopy (CRIS) have been obtained on-line and supersede any previous stable isotope work. The CRIS beam line at ISOLDE, CERN, is an experiment which combines the Doppler-free resolution of collinear laser spectroscopy with the high detection efficiency of resonance ionization, performed within an ultra-low background. In a commissioning experimental campaign during 2012 the francium isotope chain was studied. Hyperfine structure of the isotopes $^{202-207,211,219-221,229,231}$Fr and of isomers in ^{202,204,206,218}Fr was extracted. An experimental efficiency of 1% has currently been demonstrated (using ^{202}Fr, see figure below) and a non-resonant equivalent efficiency was determined to be <0.001%. For additional selectivity, radioactive ion beams containing several isomeric states may be sent to a decay spectroscopy station. In the latest developments, laser-assisted nuclear decays spectroscopy (LANDS) was performed on ^{204g,m1,m2}Fr.

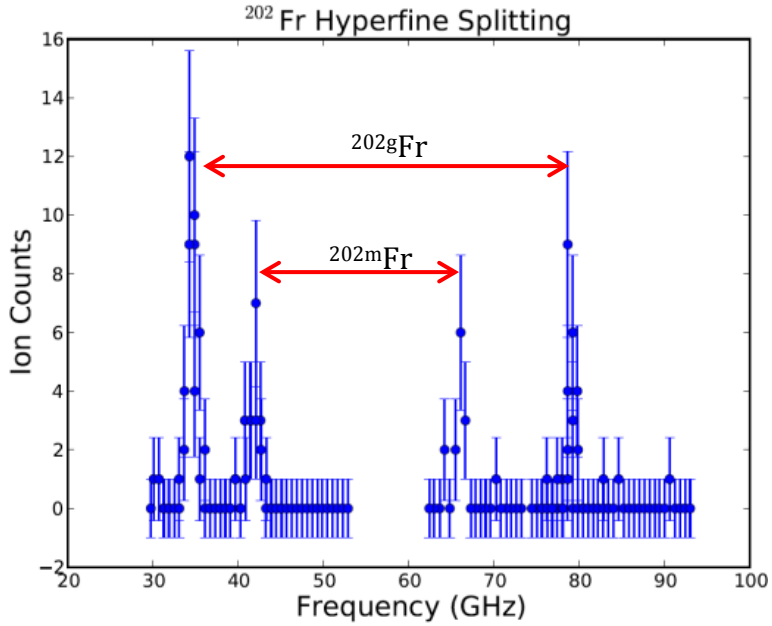

Figure: A single scan across the hyperfine structure splitting of the $7S_{½}$ state of ^{202}Fr showing the isomer and ground state.

MS105: Demonstration of on-line RIS in a gas cell and gas-jet environment (month 24)

In a large collaborative effort including KU Leuven, Mainz, JYFL, GANIL and institutes outside of the PREMAS participants, milestone MS105 was finally met on time by month 24 of the ENSAR project as planned. The well-established dye laser system (medium repetition rate, 200 Hz) operating at LISOL, Louvain la Neuve, was directly compared with a state-of-the-art Ti:sapphire laser system (high repetition rate, 10 kHz) in both off-line and on-line measurements at the LISOL facility. A systematic study of the ion signal dependence on laser pulse energy and repetition rate was performed in offline tests using stable copper and cobalt isotopes. On-line, the CYCLONE110 cyclotron delivered a primary beam of $^{3}He^{+}$ (25 MeV, 1μA) to LISOL that was used in combination with a Ni target to produce ^{59}Cu ($T_{½}$ 82s). The two laser systems performed comparably using in-gas-cell laser ionization however only the high repetition rate laser system resulted in in-gas-jet laser ionization (see Figure). In order to alleviate differences in efficiency between the two ionization volumes, on-going work to better collimate the expanding gas jets at JYFL and LISOL shows great promise.

In a second major achievement, in-gas-cell laser ionization spectroscopy has been performed at LISOL to extract magnetic moments and changes in mean-square charge radii in neutron-deficient silver isotopes in the vicinity of ^{100}Sn. This work has recently been published in Physics Letters B and is summarized in Milestone MS94 of this report.

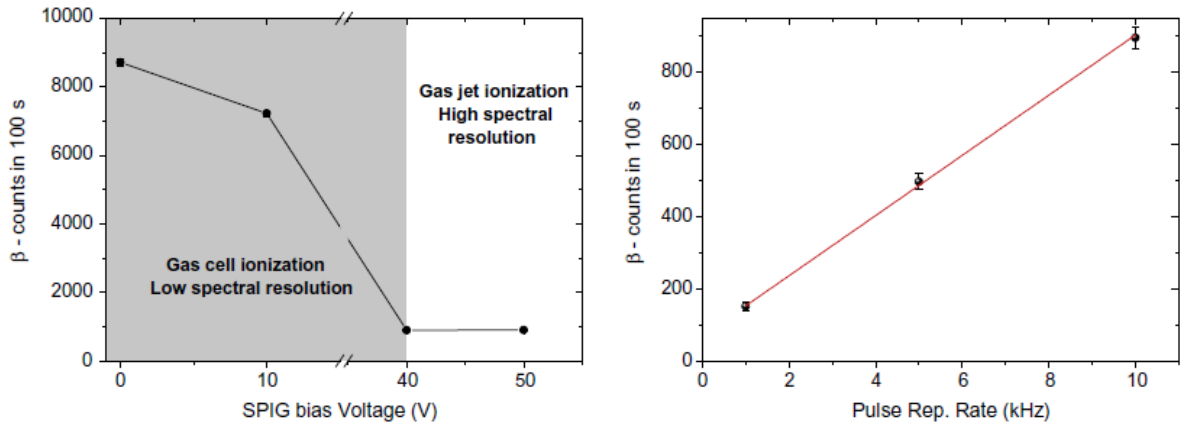

Figure: (Left) Ion signal as a function of bias voltage applied between the gas cell and SPIG rods. Only 40V is necessary to block ions form the gas cell and select only those ionized in the gas jet under optimal conditions for spectroscopy. (Right) Ion signal using in-gas jet ionization as a function of the Ti:sapphire repetition rate.

[1]. R. Ferrer, V.T. Sonnenschein et al., Nucl. Instrum. and Meth. In Phys. Res. B **291** (2012) 29

MS106: Demonstration of on-line RIS in a gas cell and gas-jet environment (month 48)

In our earlier milestone (MS105), we reported at month 24 of the ENSAR project on the collaborative effort which led to an on-line comparison between the LISOL medium-repetition rate dye laser system and the high-repetition rate laser system in use at JYFL, Mainz and GANIL for the laser ionization of ^{59}Cu in-gas-cell as well as in-gas-jet. Compared to in-gas-cell laser ionization, an improved resolution down to 2.6 GHz was achieved for in-gas-jet ionization due to the more favourable environment (low temperature and pressure) of the gas jet [1]. This experiment thus motivated developments associated with narrowing of the tunable laser linewidths for higher-resolution studies, in particular targeting the gas jet.

In a first step towards the very-heavy elements, a recent campaign of measurements at LISOL has focused on spectroscopy of Ac. Preliminary in-gas-cell broadband laser spectroscopy was performed on ^{212}Ac, revealing a linewidth of almost 30GHz for the excitation step [2]. Later, high-resolution RIS on long-lived ^{227}Ac (discussed in the second progress report) was performed in collaboration with JYFL and the University of Mainz to identify a more suitable transition for spectroscopy [3]. The most promising transition with the largest total splitting was subsequently used for isotope shift measurements of $^{212\text{-}215}$Ac at LISOL, the latter located at the N=126 shell closure [4].

In a very recent experiment, the same collaboration as reported in MS105 returned to LISOL to perform high-resolution in-gas-jet spectroscopy on ^{214,215}Ac. The JYFL contribution of the Ti:sapphire ring laser, injection-locked using the Mainz diode laser system, combined with the dual-chamber gas cell of Leuven with the GANIL-design de-Laval nozzle, resulted in a milestone for in-source spectroscopy and indeed the final experiment of LISOL. A spectral resolution as low as 300 MHz was achieved with a selectivity of ~200 and an efficiency of ~0.5% (the latter comparable to that measured in the gas cell). This resulted in isotope shifts as well as the hyperfine A- and B-parameters extracted from the data with a 25-fold higher precision than obtained via in-gas-cell spectroscopy. Both the earlier in-gas-cell measurements as well as the recent in-gas-jet measurements are currently under analysis and will be submitted for publication in 2015.

[1]. R. Ferrer, V.T. Sonnenschein *et al.*, Nucl. Instrum. and Meth. In Phys. Res. B **291** (2012) 29.
[2]. R. Ferrer *et al.*, Nucl. Instrum. and Meth. In Phys. Res. B **317** (2013) 570.
[3]. V. Sonnenschein, Ph.D. thesis, The University of Jyväskylä (2015).
[4]. C. Granados *et al.*, Phys. Lett. B. in preparation.

MS107: optical manipulation demonstrated on-line with several elements (month 36)

Optical manipulation (in other words, optical pumping) of ion beams undergoing cooling and bunching within RF coolers is now an established tool for the collinear laser spectroscopy programme at JYFL. By pumping ions from the ground state, population may be moved to an excited, metastable state, from which high-resolution laser spectroscopy may be performed. Such metastable state spectroscopy may be preferable if the ground state transition is too weak, unfavourable for extracting nuclear parameters or beyond the wavelength reach of modern cw laser systems. On-line, spectroscopy has been successfully demonstrated on the refractory elements Nb and Y, and most recently Mn (see figure below). The last of these also demonstrated the ability to enhance and retain the population of specific hyperfine F-states. Beam time has also been awarded to develop in-cooler optical pumping at ISOLDE for continuation of Mn, and at TRIUMF for Ca and Y.

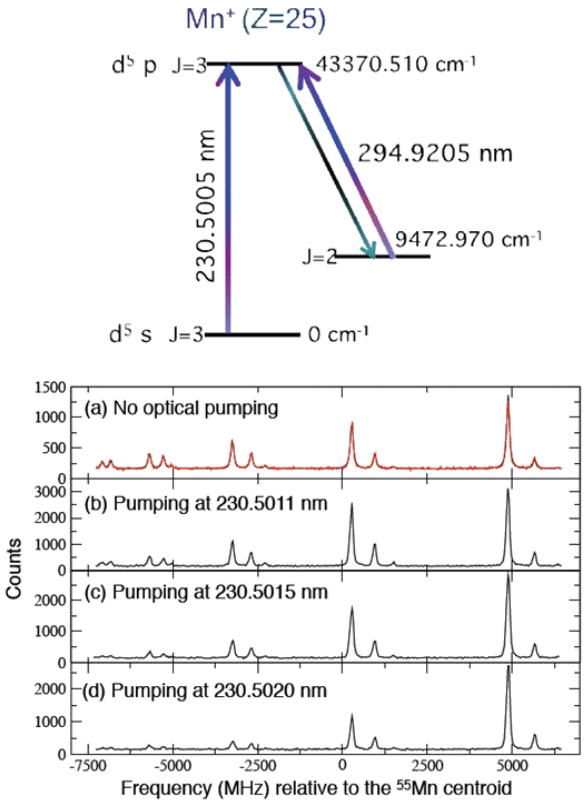

Figure: Metastable state collinear laser spectroscopy on Mn, following optical manipulation in the JYFL RF cooler.

MS109: design document on the schemes for element-selective transport and beam distribution (month 30)

The commissioning and subsequent successful operation of ISOLTRAP's Multi-Reflection Time-of-Flight (MR-TOF) device has ensured that the milestone associated with subtask 2.5 has been successfully accomplished. Briefly, the MR-TOF device has a number of operational modes and applications [1]. With a fast measurement cycle of typically 30 ms, very favorable compared to Penning trap cycles, lower half-life limitations on exotic nuclei can be tolerated. The device offers a high mass resolving power, sufficient for separation of most isobars, i.e. in excess of 10^5. By operating in a non-scanning mode, the MR-TOF can obtain the whole isobar spectrum from just a few injections, and as long as space charge limitations are not reached, contaminations may be used as mass calibrants. In addition to purification of ion beams, the device may be used as a high-precision mass spectrometer for nuclides that are either shorter lived or have lower yields than those accessible for Penning trap mass spectrometry.

In order to reduce the temporal pulse structure of the MR-TOF, important modifications had to be made to the RFQ buncher which acts as the ion source for the device. The time necessary to reach the maximum mass resolving power depends sensitively on the injected bunch width, therefore the RFQ trapping region was shortened and the extraction field strength increased. This resulted in an impressive reduction from 900 ns to ~60 ns.

By operating the MR-TOF-MS in combination with a Bradbury-Nielson gate, a means to quickly separate the ion of interest for delivery to ion manipulation devices downstream from contaminations has been achieved. The device has also been successfully operated in combination with the Resonant Ionization Laser Ion Source both for unambiguous ion identification as well as background-free hyperfine structure scans from low purity ion beams [2].

1. "ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer", R.N. Wolf *et al.*, Int. J. of Mass. Spec. **349-350** (2013) 123.
2. "New developments of the in-source spectroscopy method at RILIS/ISOLDE", B.A. Marsh *et al.*, Nucl. Instr. and Meth. in Phys. Res. B **317** (2013) 550.