Novel radioactive ion-beam production techniques
Report on new schemes and novel laser-ion-source techniques

Subtask 1.1 (KU-Leuven): New LIS techniques (ion manipulation with DC electrical fields in gas cells, new resonance ionisation zones in RILIS) and novel methods for the collection of fusion-reaction products with half-lives beyond 100 ms.

New LIS techniques

1.1.1 In-gas-jel laser spectroscopy

The high efficiency and high selectivity of the in-gas laser ionization and spectroscopy (IGLIS) technique allows performing in-gas-cell resonance ionization spectroscopy studies of exotic atoms. Experiments conducted so far at the Leuven isotope separator on line (LISOL) facility using this technique have provided important ground- and isomeric-state properties, such as mean-charge radii, magnetic moments or spins, of neutron deficient Cu and Ag isotopes. A typical spectral resolution of the obtained experimental traces in these experiments was found to be of 5-8 GHz. These studies were recently extended to the heavy-mass region by resolving the hyperfine structure of the neutron-deficient actinium isotopes $^{212\text{-}215}$Ac.

However, a profound knowledge of the underlying nuclear structure in very exotic nuclides requires a technique that provides high efficiency in combination with high spectral resolution. A recent experimental campaign at LISOL was devoted to laser spectroscopy studies that made it possible to reconcile these requirements by employing the IGLIS technique in the supersonic gas jet produced by a de Laval nozzle installed at the buffer gas cell exit. Carrying out laser ionization in the low-temperature and low-density supersonic gas jet allows eliminating the pressure broadening thus improving significantly the spectral resolution.

An all-solid state narrow bandwidth, high repetition rate laser system based on the amplification of a CW diode laser in an injection-locked Ti:sa cavity was used to investigate the hyperfine structure of the 438 nm atomic transition in ^{214,215}Ac. The data obtained revealed a total spectral resolution of ~400 MHz. Thus, the isotope shifts as well as the hyperfine A- and B- parameters were extracted with a 25-fold higher precision than that obtained for these isotopes by in-gas-cell spectroscopy studies, and additionally a better spin assignment for the N=126 ^{215}Ac ($T_{1/2}$=0.17 s) nuclide was possible. Furthermore, the results showed that the total ionization efficiency in the gas jet is comparable to that measured in the gas cell (~ 0.5 %) and can potentially be improved up to one order of magnitude by increasing the duty cycle. This significant improvement of the overall efficiency might enable future laser spectroscopy studies of species with very low productions (~10^3 pps) or with half lives well below 100 ms.

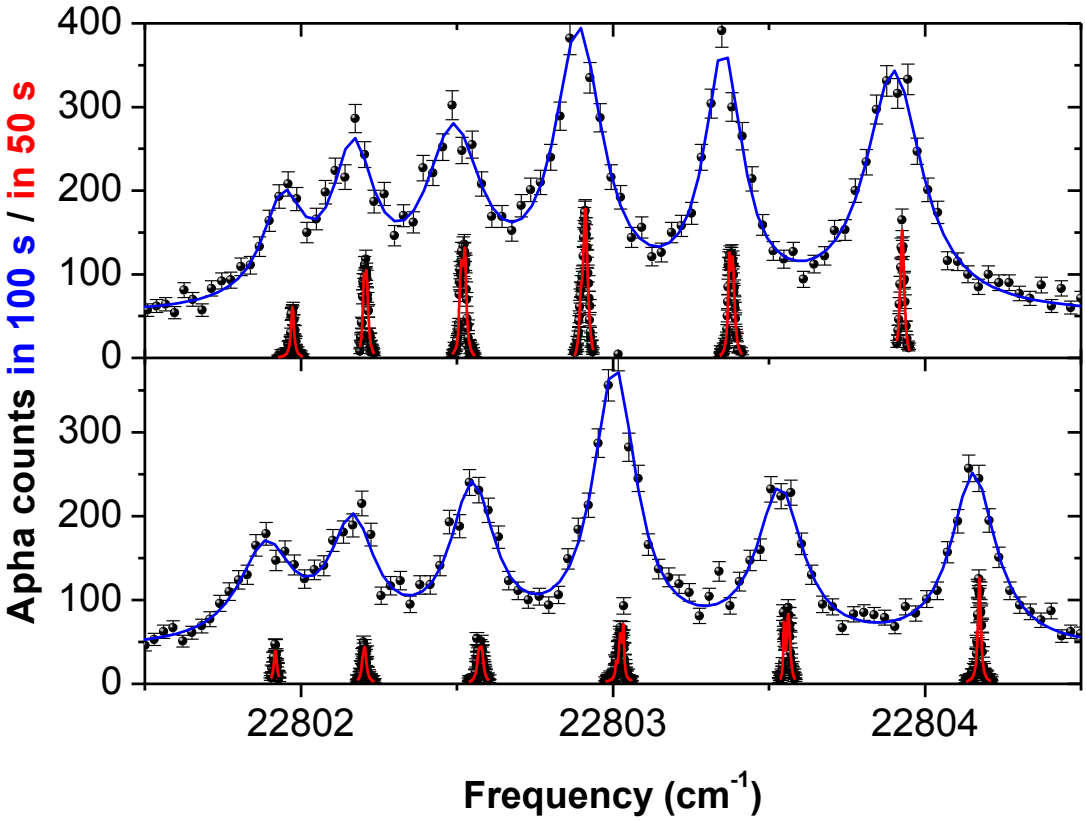

Figure 1. Experimental traces of the hyperfine structure in the $^{2}D_{3/2} \rightarrow {}^{4}P_{5/2}$ transition at 438 nm for the actinium isotopes ^{214,215}Ac using in-gas-cell (blue curve) and in-gas-jet (red curve).

Further characterization and optimization of this technique will be investigated at the off-line IGLIS laboratory, being commissioned at KU Leuven. Here, the physical and technical limits of the in-gas-jet technique will be explored. This will ensure the best performance in spectral resolution and ionization efficiency for the future IGLIS setup that will be linked to the Superconducting Separator Spectrometer (S3) at the new radioactive ion beam facility SPIRAL2 (GANIL).

1.1.2 Laser Ion Source and Trap (LIST)

With the purpose of suppression of surface ionized isobaric contaminations in radioactive ion beams produced with RILIS a Laser Ion Source and Trap (LIST) has been developed by Mainz University. It has been coupled to the ISOLDE target and ion source unit, providing a new mode of operation of the RILIS. The first on-line operation of the LIST took place in 2011 for the production of Mg isotopes. In particular, the isotopes ^{22}Mg and ^{27}Mg were produced with a suppression factor of more than 1000 for the isobaric contaminants ^{22}Na and ^{27}Al. Following this test, several modifications of the LIST were implemented to improve its efficiency, selectivity and reliability. During the five days of operation in September 2012, laser ionization of Mg and Po was demonstrated alongside the suppression of various surface-ionized elements (Na, K, Fr). The hyperfine-structure and isotope shifts of ^{217}Po and ^{219}Po could be measured for the

first time by in-source laser spectroscopy due to the suppression of surface-ionized francium by the LIST.

Ion manipulation with DC electrical fields in gas cells

1.1.3 Dual Chamber gas cell

The introduction of a new gas cell design in which the stopping volume of the nuclear reaction products, including the primary beam path, are separated from the laser ionization volume, is a key development for the production and spectroscopy studies of pure radioactive beams at the Leuven Isotope Separator Online (LISOL) facility. In such a "Dual Chamber" gas cell the direct ionization near the exit hole through hard X-rays is blocked, which enables the use of electrical potentials in a pair of collector electrodes within the gas cell. The use of these collector plates leads to a strong suppression of the isobaric contaminants surviving the neutralization and therefore to an increase of the selectivity, i.e. of the ratio between the signal with lasers on and lasers off. In the first experiments using this approach, a laser selectivity of 2200 was achieved for rhodium isotopes produced in fusion evaporation reactions , opening up new possibilities for in-source laser spectroscopy studies in, e.g., the N = Z and the actinide regions of the chart of nuclides.

A recent experimental campaign at LISOL has been devoted to in-gas-cell laser spectroscopy studies of neutron deficient actinium isotopes. During this campaign a very high production of these isotopes (~ 50 pps) in the absence of laser radiations observed, which could be significantly reduced (to ~ 0.03 pps) by applying a 40V potential in the collector plates. The suppression of this off-resonance count-rate resulted in an ion selectivity of ~ 300 and allowed us to determine isotope shifts, magnetic moments, and tentative spin values for the $^{212-215}$Ac isotopes.

1.1.4 New resonance ionisation zones in RILIS: VADLIS

R&D on new ionization zones has been performed at CERN. Laser resonance ionization was demonstrated inside the VADIS, ISOLDE's FEBIAD type plasma ion source. In VADIS electrons emitted by a hot cathode and accelerated to some 100 eV produce an impact ionization of species present in the VADIS cavity. By injection of laser beams into the cavity a resonance ionization mechanism was added. In off-line tests with gallium ions two modes of VADIS operation have been identified: an element selective RILIS-only mode; and a RILIS+VADIS mode for increased efficiency. These capabilities have since been verified on-line for the production of mercury beams as part of a feasibility study for a future in-source laser spectroscopy study of mercury isotopes. There are numerous immediate applications of these developments: it is now possible to couple a molten lead target with the laser ion source at ISOLDE; laser spectroscopy can be performed inside the VADIS cavity; and switching from VADIS mode to the element selective RILIS mode allows for signal identification.

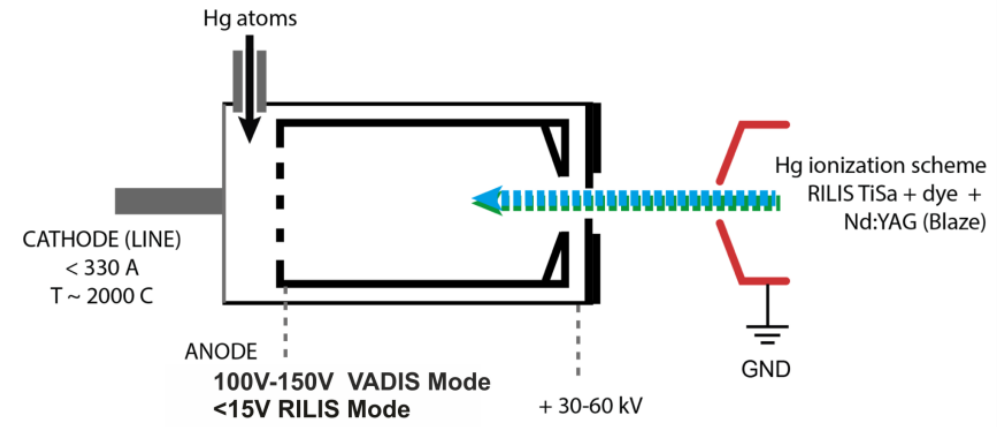

Figure 2. Schematic view of the laser ionization zone in VADIS.

Subtask 1.2 (Mainz): New LIS schemes (including atomic level searches in elements with unknown level schemes).

Development of New LIS Schemes

The majority of activities carried out for subtask 2 have already been reported in the milestone and intermediate report dated October 2013. Correspondingly, we refer here to this detailed presentation, which covered the successful development of LIS schemes as carried out at Mainz University for the elements Pd, Cu, Yb, Pu, and Ac. Further Information to be added comes from our international collaborations, where schemes for Nb, Ti and Te have been tested. The latter have not yet been fully characterized under on-line laser ion source conditions, so that no details will be given here.
Today most relevant developments concern line-width narrowing of the solid-state laser systems of Mainz University type by complementary passive or active approaches. This work is relevant for the use in high resolution on-line in-source spectroscopy at hot cavity facilities like ISOLDE (CERN, Geneve) or ISAC (TRIUMF, Vancouver) as well as for in-jet spectroscopy at gas cell units like IGISOL, (Jyväsklyä, Finland) and LISOL (Louvain la Neuve, Belgium) with the upcoming research opportunities at S3, (GANIL, Caen) PALIS (RIKEN, Japan) or TRIGA-SPEC (Mainz). Two competing experimental approaches for laser line width narrowing include passive wavelength selection by multiple etalon operation in a highly stabilized ring resonator laser cavity and on the other hand, injection locking of the high repetition rate pulsed lasers with single mode continuous wave laser light. First demonstrations of both techniques were carried out at LLN for hyperfine spectroscopy of Ac isotopes and are in preparation for studies on Pu at Jyväskylä. Laser line widths in the order of 20 MHz were demonstrated by generation of high resolution hfs spectra.
Classical development of excitation schemes in the last year of the PREMAS funding period concerned:

Holmium (Ho)

Apart of on-line nuclear physics studies, high interest in resonance ionization of the rare earth element holmium (Ho) is stemming from the novel upcoming ECHo project. Goal of this large collaboration is the measurement of the mass of the electron neutrino through a calorimetric spectrum of the electron capture of the radioisotope ^{163}Ho. Available sample material is highly contaminated with other radioactive and stable species, introduced by the ^{163}Ho production via high flux neutron irradiation. Resonance ionization thus is mandatory for initial isotope separation and purification in the sample.

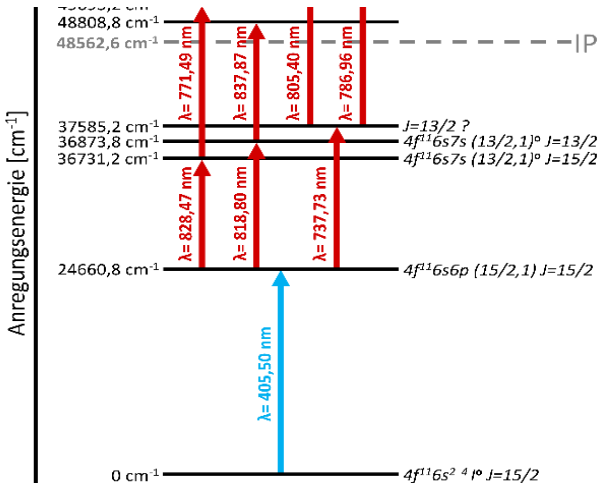

Figure 3. Three step excitation schemes in Ho.

An extensive search for the most efficient ionization scheme was carried out according to Fig. 3. Detailed measurements on the overall efficiency and its reproducibility was performed in the strongest excitation ladder with wavelengths of 405 nm, 818 nm and 837 nm. A remarkable high result of 25(5)% has been obtained reproducible, which is both pleasant for ECHo as well as for exotic isotope production. Based on this work presently on-going investigation concern level scheme development in the neighboring rare earth elements of Dysprosium, Terbium and Erbium as well as the all-radioactive element Pr, for which the ionization potential is one of the last missing atomic physics quantities.

Sodium (Na)

For the study of some specific elements, i.e. the alkaline element Na, tunable laser emission in the green to yellow spectral range of 500 to 680 nm is required. This is very easily achieved by conventional dye lasers but a particular challenge for solid state lasers, i.e. the titanium:sapphire lasers of Mainz University. Wavelengths in this spectral region were successfully obtained in a non-linear crystal by difference-frequency generation of the radiation of two titanium:sapphire lasers. From the latter, one was operating in the fundamental infrared range, and one in the frequency doubled blue range.

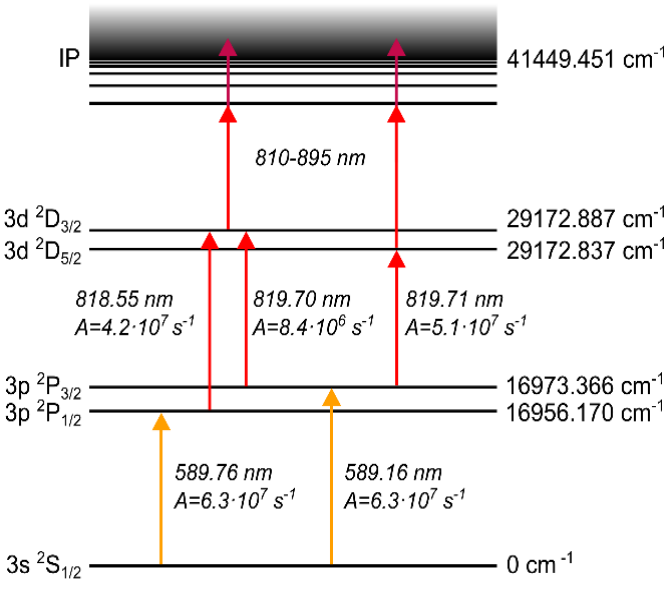

Figure 4. Three step excitation schemes in Na.

Performance of this approach was demonstrated by two- and three-step resonance ionization spectroscopy of atomic Rydberg levels in the spectrum of atomic sodium involving as first excitation step the famous ground state doublet at 589 nm. The analysis of these data is still in progress but already now shows the successful extension of radiation from solid-state lasers into so far inaccessible spectral ranges

Table 1. New laser resonance ionizations schemes.

Element	Step 1, nm	Step 2, nm	Step 3, nm	Ionization efficiency	Place of development
Astatine (At)	216.9 224.4	795.4 793.2	532 532	> 4%	CERN, TRIUMF
Barium (Ba)	350.1	653.5		60% of surface efficiency	CERN
Barium ion (Ba^+)	456	223	532	1.2%	CERN
Calcium (Ca)	422.8	586.8	654		CERN
Chromium (Cr)	357.9	697.8	579		CERN
Germanium (Ge)	275.5	569.2	532	> 2%	CERN
Holmium (Ho)	405.5	623.3 828.5 818.8 737.7	532 771 837 805	25%	CERN MAINZ MAINZ MAINZ
Lithium (Li)	670.8	610.3	532	58% of surface efficiency	CERN
Mercury (Hg)	253.7	313.2	532	6 %	CERN
Sodium (Na)	589.8 589.16	818.6 819.7	810-895 810-895		MAINZ MAINZ
Actinium (Ac)	438.6 418.3	434.7 439.2			MAINZ MAINZ

A series of highly excited Rydberg levels have been studied by in-source laser resonance ionization spectroscopy of astatine and polonium at ISOLDE RILIS. Based on obtained data the atomic ionization potential (IP) of astatine has been measured for the first time and a new more precise value of polonium IP has been determined:

$$\text{IP (At I)} = 75150.8(7)\ \text{cm}^{-1}$$

$$\text{IP (Po I)} = 67896.310(13)(30)\ \text{cm}^{-1}$$

In combination with a surface ionization for the first time an ion beam of doubly charged Ba^{2+} ions has been produced by the laser resonance ionization at ISOLDE RILIS. Resonance ion-ionization selectively doubles the charge to mass ratio, shifting the

barium in the mass region, away from surface ionized 1^+ caesium and indium isobars, to a region where surface ionization is not considered to be efficient.

Subtask 1.3 (CERN): Isomer selectivity using the combination of lasers and ion traps.

A new way to perform the in-source laser spectroscopy was developed by combining the laser selectivity of RILIS with the high mass resolution of ISOLTRAP. In particular, the Multi-Reflection Time-of-Flight Mass Separator (MR-ToF MS) was used for photo-ion detection for the first time. This method is essential to obtain background-free laser spectra for many ion beams containing surface ionized isobars, particularly for α-inactive or long-lived isotopes.
On the other hand, the information on hyperfine structure obtained from in-source spectroscopy using the α-decay station has allowed to separately investigating masses of nuclear isomers. This newly developed method was applied for the study of astatine and gold isotopes at ISOLDE/CERN.
Efficient performance of such an experiment requires data exchange with several detector setups located in the experimental hall and the recording of all parameters relevant to the ion beam production and detection. A RILIS laser control software and data acquisition framework based on the Shared Variable Engine protocol has been developed and successfully implemented at the ISOLDE facility. This framework allows a flexible extension for exchange of any arbitrary set of data between RILIS and collaborative experimental setups.

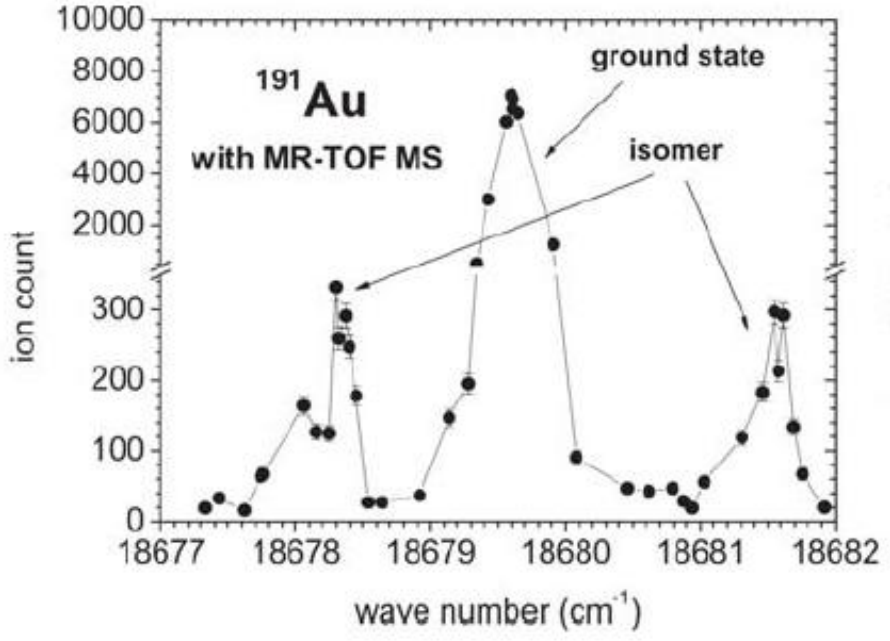

Figure 5. The ^{191}Au ToF- gated ion-count rate at the MR-ToF detector as a function of the laser frequency of first excitation step of the Au ionization scheme.

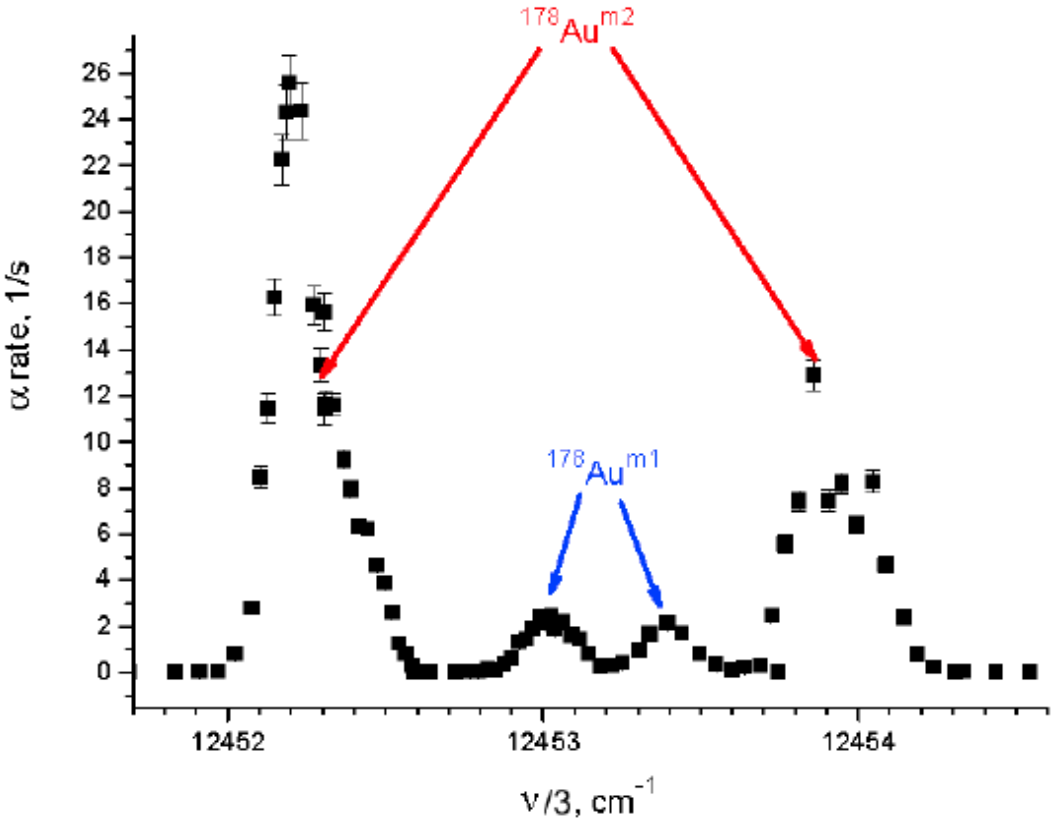

Figure 6. Hyperfine structure for ^{178}Au measured with the "Windmill" α-decay setup.