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# First on-line application of the high-resolution spectroscopy laser ion source PI-LIST at ISOLDE

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ABSTRACT

We report on the development, characterization, and first application of a specialized resonance ionization laser ion source for high-resolution spectroscopy applications below usual hot cavity Doppler broadening limitations, for on-line experiments at CERN-ISOLDE. The new PI-LIST ion source comprises perpendicular laser/atom beam interaction in a radio-frequency quadrupole unit directly downstream the hot atomizer cavity. A spectral linewidth of 200–300 MHz is demonstrated, with the potential to achieve below 100 MHz. Compared to standard in-source laser ionization, the efficiency reduction factor ranges from a few 100 to above 1000. The implementation of this ion source concept at thick target radioactive ion beam facilities will greatly enhance capabilities for nuclear structure investigations without demanding dedicated experimental beam line setups, and will facilitate the delivery of isomer-pure beams to experimental stations.

## 1. Introduction

The Resonance Ionization Laser Ion Source (RILIS) [1] has become a principal method exploited in the production of radioactive ion beams (RIBs) at world-wide leading Isotope Separation On-Line (ISOL) facilities, such as CERN-ISOLDE [2]. Radionuclides, created in a thick production target by particle bombardment, effuse into a hot cavity ion source, where they are exposed to laser beams as atomic vapor. The lasers' frequencies are tuned to electronic transitions in the atomic shell, subsequently resonantly promoting an electron into excited states, and eventually detaching it to form an ion. Due to the unique level structure of each chemical element, this method is inherently element-selective, and best-case ionization efficiency values exceeding 50% are reported [3–5].

Besides utilization for RIB production, the method is also employed for nuclear structure investigations via probing the isotope shift (IS) and the hyperfine structure (HFS) splittings of the involved atomic levels, arising from the interaction of the electronic shell with the nucleus [6]. While recent scientific results underline the efficiency of this approach down to an atom-at-a-time scale [7], an ultimate limit to experimental resolution is given by the Doppler broadening of spectroscopic lines in the hot vapor environment. Full-width-at-half-maximum (FWHM) linewidths typically are in the order of 1 to 10 GHz, depending on atomic mass, vapor temperature, and transition wavelength. In many cases, around one order of magnitude higher resolution is required. Dedicated laser spectroscopy setups at RIB facilities achieve this by, e.g., utilizing Doppler compression in accelerated beams [8], buffer gas pressure-driven supersonic jet formation [9], or trapping devices [10]. In this work, we report on the adaption and on-line implementation of an established hot cavity-type laser ion source, utilizing crossed laser/atom beam interaction geometry to reduce the range of addressed atomic velocity classes, thus achieving linewidths in the few 100 MHz regime.

The implementation of this technique is based on the high-purity Laser Ion Source and Trap (LIST) at ISOLDE, which is described in

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detail in [11,12]. It comprises spatial separation of the hot cavity from a dedicated laser ionization volume inside a radiofrequency quadrupole (RFQ) structure directly downstream. Here, similar limitations for the spectral resolution arise, as the lasers interact anti-collinearly with the atomic beam and thus probe the oncoming fraction of the Maxwell–Boltzmann velocity distribution. For the purpose of high-resolution spectroscopy at off-line facilities, a window was introduced at the side of the ion source vacuum vessel, to enable access for spectroscopy laser light in perpendicular geometry to the atom stream. In this way, only the lateral velocity components of the effusing atom cone contribute to the Doppler broadening. The effect is enhanced by keeping the subsequent, ionizing lasers in anti-collinear geometry, which thus only convert excited atoms on the central axis into ions, narrowing down the lateral angle acceptance even further. This application as the Perpendicularly Illuminated (PI-)LIST resembles the approach in prevalent atomic beam units and is detailed in [13,14]. At its development site at the RISIKO off-line mass separator [4] at Mainz University, it has become the standard tool for nuclear structure investigations, mainly focused on long-lived radioisotopes [15–17]. Altogether, it offers three modes of operation, that can be switched quickly during an experiment:

- **Ion guide mode** (all lasers collinear, repeller electrode on negative voltage to pre-extract and guide ions from hot cavity through RFQ): Highest efficiency, but no contamination suppression — resembling standard RILIS
- **LIST mode** (all lasers collinear, repeller electrode on positive voltage to suppress ions from hot cavity): High beam purity, but loss in efficiency
- **PI-LIST mode** (one laser perpendicular, repeller electrode on positive voltage): High-resolution spectroscopy

## 2. PI-LIST implementation at ISOLDE

For the on-line application of the PI-LIST, laser access via a window at the target vacuum chamber is not a practical option. The spectroscopy laser beam is instead transported through the vacuum chamber of the ion beam line, parallel to the ionization laser beams in "classic" anti-collinear fashion from their launch point 15–20 m away in the laser laboratory. Inside the RFQ structure and as close as possible to its entrance, where the atom density of the effusing cloud is the highest [18], the laser is perpendicularly deflected by a 45°-oriented mechanically polished stainless steel surface, and retro-reflected by an additional mirror surface to increase utilizable photon flux. The design with sketches of the atom stream and the laser beams is illustrated in Fig. 1. This additional operation mode does not affect the functionality of the unit as "standard" LIST using solely anti-collinear instead of perpendicular laser illumination.

A requirement for this mode of operation is optical line-of-sight for the off-center laser beam from its entrance into the vacuum vessel at the mass separator towards the mirrors. At ISOLDE, the tip of the ion extraction electrode had to be modified by adding four symmetrically placed orifices of 6 mm diameter (identical to central orifice) with a distance of 11 mm to the central axis. According cutouts in the LIST exit plate were added. The mirror surfaces themselves cover a square plane of 6 mm $x$ 6 mm (projected in laser beam direction) and are installed with a 0.5 mm gap behind the repelling electrode, between the RFQ rods. The distance of their inner edges to the central axis is 8 mm, as opposed to the 7.5 mm free field radius of the RFQ.

Prior to their introduction, consequences of these geometry changes on ion beam quality (shape, emittance, and intensity) were investigated with the particle trajectory and electric field simulation software SIMION 8.1 [19], and determined to show no significant impact [14]. In addition, a modified extraction electrode was used at RISIKO for over a year, including "conventional" standard RILIS operation, without deterioration in key characteristics like mass resolving power and overall efficiency. A back-to-back comparison of extraction electrodes with and

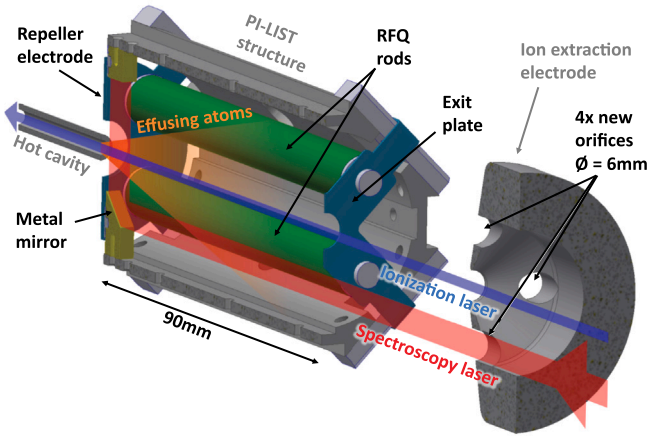


**Fig. 1.** Half-cut CAD model of the PI-LIST with internal polished steel surfaces as mirrors, and ion extraction electrode with added side bores for laser transport. Ionization solely occurs in the overlap volume of lasers and atoms, leading to greatly reduced Doppler broadening in direction of the spectroscopy laser.

without the additional orifices under otherwise identical conditions directly at ISOLDE showed neither a difference in the ion beam profile on various scanners, nor on tuning behavior. Thus, absence of influence from the changes on standard facility operation was confirmed and the new design permanently implemented.

The mirror surfaces were tested for their reflectivity at different laser wavelengths in the regime typically employed for laser spectroscopy. In their most simple form as mechanically polished stainless steel, a reflectivity of 52%, 55% and 25% was measured for 760 nm, 395 nm, and 302 nm, respectively. Preceding tests at Mainz University with a custom-built pulsed laser deposition device [20] showed that reflectivity in the range of 85% for the visible regime can be achieved with silver coating [14]. For narrow-bandwidth applications, where power broadening effects are to be avoided, the available laser power typically vastly exceeds the required level, and thus moderate losses like demonstrated here can be accepted. Laser beam shape after reflection was assessed with a beam profiling camera. While slight distortions in the intensity profile occur, the laser beam does not significantly broaden or diffuse [14], which would undermine the required solely perpendicular incidence to the effusing atoms.

## 3. Characterization and figures of merit

After successful technical implementation of the PI-LIST at ISOLDE, its first-time on-line application was nuclear structure investigations on neutron-rich actinium and polonium isotopes, continuing the work described in [9,21,22]. Performance parameters of the unit in its typical operation environment were assessed during this campaign. The narrow-bandwidth spectroscopy laser system comprised a continuous wave laser (Sirah Matisse) provided by the ISOLDE-CRIS setup, used as seed light for an injection-locked pulsed Ti:sapphire ring cavity [23] in a setup analogue to the work reported in [24]. A spectral bandwidth of $\approx$ 35 MHz was obtained after single pass second harmonic generation. For the subsequent ionization step, the standard RILIS Ti:sapphire laser system [25] with a bandwidth of a few GHz was used to ensure full spectral coverage of the upper state HFS for efficient ionization. The entire setup operates at a pulse repetition rate of 10 kHz.

### 3.1. Experimental linewidth

During commissioning phase, the experimental spectral resolution was investigated on the isolated $F = 3 \rightarrow 4$ HFS component in the $^{227}$Ac ground state transition (438.6 nm) of the scheme used in [21].

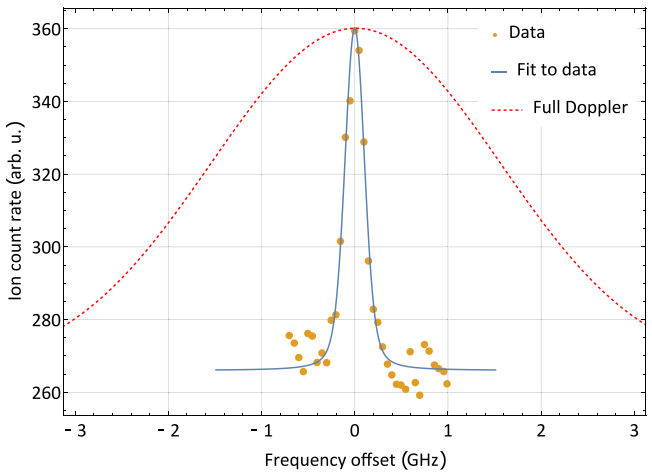


**Fig. 2.** Laser frequency scan of a $^{227}$Ac HFS singlet in PI-LIST mode. The resonance features an FWHM linewidth of slightly above 200 MHz, opposed to calculated 1.55 GHz from full Doppler broadening in a standard RILIS hot cavity.

The laser power of the first, spectroscopic transition step was reduced below saturation (< 1 mW before launch optics), and the second step laser pulse was delayed by 40 ns in respect to the first step to reduce broadening effects [26]. An ion bunch time-gating technique was applied for data acquisition (cf. Section 3.3). The resulting spectrum is shown in Fig. 2 and exhibits an FWHM linewidth of slightly above 200 MHz, significantly lower than the calculated 1.55 GHz full Doppler broadening in a 2000 °C vapor present in the hot cavity. Thus, the applicability of the ISOLDE version of the PI-LIST for high-resolution laser spectroscopy was ultimately confirmed.

Using the full available laser power of 20 mW and the optimum temporal pulse overlap did not show a large influence on the observed linewidth. In a later experiment, using a newly manufactured PI-LIST unit, a resolution of 300 MHz was reached, again with little influence of these broadening effects. This suggests the governing factor to be the geometrical acceptance of lateral velocity classes. Previous off-line studies in Mainz yielded consistently lower linewidths of 100–150 MHz on holmium [14]. The best cases so far were 60 MHz on rubidium [24], albeit on extraordinary operation conditions of very low hot cavity temperatures, and 55 MHz on californium [17] with the lateral window variant. These comparisons indicate that improvements of the first on-line version of the PI-LIST presented here are possible, likely in the field of providing better lateral atom selection by providing, e.g., a better focused collinear laser beam (laser beam travel distance being a major difference to the Mainz setup). In turn, this comes with potential additional losses in efficiency.

### 3.2. Efficiency considerations

The efficiency of the PI-LIST was assessed in two ways. Relative values can be deduced by comparing the ion count rate in the various operation modes (cf. Section 1) as compiled in Table 1. Previous investigations reported a factor 20 loss between ion guide and suppression mode of the standard LIST [11], while the equivalence of the former to a standard RILIS ion source strongly depends on total ion load in the system. More recently measured radioactive ion yields at ISOLDE suggest that already under standard operation conditions, the LIST efficiency in ion guide mode can be a factor 2–5 lower. Switching from collinear LIST to PI-LIST resulted in additional loss factors up to 3, while in some occasions almost no loss was observed. This variation of the loss factors underlines the high importance of achieving good geometrical overlap of effusing atoms and both collinear and perpendicular laser beams. Overall, under standard working conditions, these losses

**Table 1**
Exemplary actinium ion production rate comparison in different PI-LIST modes. Absolute efficiency as deduced from the loss factors is estimated assuming 10% with standard RILIS [27]. See text for details.

| Operation mode | Mode loss factor | Combined loss factor | Est. total efficiency (%) |
|---|---|---|---|
| Standard RILIS | | | 10 |
| LIST ion guide | 3 | 3 | 3.3 |
| LIST high purity | 33 | 100 | 0.1 |
| PI-LIST | 2 | 200 | 0.05 |
| PI-LIST opt. | 10 | 2000 | 0.005 |

sum up to an overall factor of 100–500 efficiency reduction compared to a standard RILIS hot cavity ion source. These numbers are consistent with the results reported in [13] for the Mainz PI-LIST version with laser access through the lateral window. Additional losses can occur when further reduction of laser power or pulse delay techniques are applied for achieving the best possible resolution, especially on short-lived excited atomic states. Depending on the experimental requirements, compromises in linewidth versus efficiency can be applied.

An absolute efficiency assessment was conducted with the Mainz PI-LIST version with lateral window access during a HFS measurement of long-lived $^{225}$Ac on a sample of $3.7 \times 10^9$ atoms [14]. The ion count rate on the highest peak ($F = 0 \rightarrow 1$ of the 418 nm ground state transition) was interpolated between multiple points in time where it was scanned during the overall measurement duration of 70 min, until sample exhaustion. The resulting integrated ion number amounted to $\approx 4 \times 10^4$, thus yielding a total efficiency of $\approx$ 0.001%. As this experiment was conducted on best possible resolution of around 100 MHz, a factor 10 loss in count rate was accepted during optimization in PI mode. In contrast, best efficiency settings in PI mode yielded a linewidth of 400 MHz. Comparing these numbers to a best-case standard RILIS efficiency for actinium of 10% [27] (albeit with the scheme from [21] with potentially different total efficiency), a total loss factor in the order of 1000 can be derived for going to highest efficiency PI-LIST mode. This description naturally is defined for efficiency on a singly-standing HFS component, so highly dependent on the actual atomic structure.

These loss factors and efficiency values, specifically derived for actinium under the given operation conditions, cannot directly be adopted to different elements. The process depends on absolute efficiency of the employed laser scheme, the surface ionization fraction, and potential ion load limitations in both the hot cavity and RFQ structure.

### 3.3. Ion bunch time structure

The ion bunch time structure originating from the pulsed laser ionization in the anti-collinear LIST has been studied in detail [12,14]. It features the majority of ions being ejected as a sharp bunch, having been created directly downstream of the repeller electrodes where atom density in the effusing cloud is highest, and being pushed out by their electric field. The following, slower and more washed out structure originates from ions created along the central volume of the LIST where no active ejection field is present. Fig. 3 shows a comparison of this structure with PI-LIST mode on the example of $^{48}$Ti: As in PI-LIST ionization solely happens close to the repeller electrodes, only the first, narrow feature remains in the time spectrum. The width of this bunch scales as the square-root of the ion mass [28]. For $^{227}$Ac, its total length is around 10 µs, compared to slightly below 5 µs for $^{48}$Ti.

This characteristic can be exploited for enhanced performance in two ways. On the one hand, a fast, laser pulse-synchronized ion beam gate can further enhance the ion beam purity by a factor of laser duty cycle (100 µs in our case) divided by gate width (around 10 µs) [28,29]. Further pulse duration compression can be achieved with higher repeller voltage settings. On the other hand, this gating technique also exactly suppresses ions that would be created by non-perpendicular

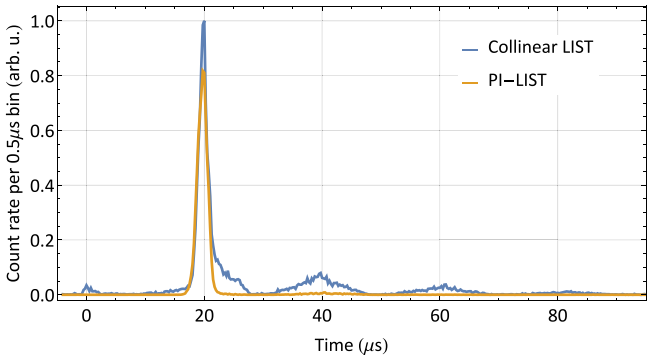


**Fig. 3.** Comparison of the time structures of $^{48}$Ti ion bunches created in LIST and PI-LIST mode. The latter only features the distinctly sharp bunch, as ions are solely created in the direct vicinity of the repeller electrode.

laser illumination, e.g. from potential diffuse reflections of the spectroscopy laser in the LIST corpus. These would lead to (asymmetric) broadening of the resulting spectra, and thus filtering them out improves experimental data quality and diminishes systematic uncertainty effects.

## 4. Conclusion and outlook

The first application of the PI-LIST as a specialized high-resolution laser spectroscopy ion source at ISOLDE was conducted successfully, and its performance demonstrated on a nuclear structure investigation experiment on actinium isotopes. The experimental linewidth was 200–300 MHz, while off-line studies show a potential to achieve even below 100 MHz under specific circumstances. Efficiency considerations yield a loss factor of at least a few 100 compared to standard RILIS operation, while further optimization towards narrower linewidth can cause additional losses. The distinctly sharp ion bunch time structure offers possibilities for further improvements in ion beam purity and experimental data quality.

The availability of this unit greatly enhances the capabilities at thick target on-line facilities to perform HFS investigation experiments directly in the ion source below the limits of hot cavity atomic vapor Doppler broadening, without the need of dedicated high-resolution spectroscopy beam line setups. Additionally, with improved spectral resolution, isomer-selective laser ionization becomes more widely applicable to provide ion beams to experiments in demand of high isomeric purity.

The limitations in efficiency mainly stem from geometrical and temporal losses due to the pulsed laser beam interaction with the atom cloud effusing from the hot cavity [18]. The possible means of improving the ionization efficiency include higher laser pulse repetition rate, revising the hot cavity tube geometry for more directed effusion, and further reducing the distance between hot cavity exit and volume in the LIST accessible for laser ionization and subsequent ion extraction.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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