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# Towards in-jet resonance ionization spectroscopy: An injection-locked Titanium:Sapphire laser system for the PALIS-facility

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## ABSTRACT

This article presents a pulsed narrowband injection-locked Titanium:Sapphire laser aimed for high-resolution in-jet resonance ionization spectroscopy at the SLOWRI/PALIS at RIKEN. The laser has been integrated into the PALIS laser laboratory enabling it to be utilized with the existing broadband Titanium:Sapphire and dye lasers. The seed efficiency has been evaluated to be close to unity over the master laser wavelength range ∼ 753 to 791 nm, and the slope efficiency, namely the ratio of the pump power to the output power, was determined to be ∼ 30 % at 780 nm. A two-step ionization scheme with 386.4016 nm first step and 286.731 nm second step into an autoionizing state was developed for resonance ionization spectroscopy of $^{93}$Nb. Magnetic hyperfine coupling constants of $1866 \pm 8$ MHz and $1536 \pm 7$ MHz were measured for the ground and excited state, respectively, in a good agreement with the literature values. A Gaussian dominated Voigt linewidth of 434.5 ± 7.4 MHz was extracted from the hyperfine spectra measured for niobium. In addition, the resolution of the in-jet resonance ionization in PALIS is estimated through numerical methods.

## 1. Introduction

Hyperfine structures and isotope shifts in electronic transitions contain readily available model-free information [1] on the single-particle and bulk properties of exotic nuclei, namely the nuclear spin, magnetic dipole and electric quadrupole moments as well as changes in root-mean-square charge radii [2]. The part-per-million level perturbations of the atomic energy levels of an atom caused by its nucleus are readily probed by modern laser spectroscopic methods. Resonance ionization spectroscopy (RIS) [3] is a selective and efficient method built upon the unique excitation energies of atomic states of different elements for a step-wise excitation and subsequent ionization of an atom.

Through recent innovations in gas cell technology and laser techniques, RIS in-source has been demonstrated to be a powerful tool for probing rare nuclei with lifetimes as short as a few milliseconds and production rates often only a few isotopes of interest per second [4,5]. Typically, the laser ion sources at RIB facilities have linewidths of few gigahertz to maximize the ionization efficiency by covering the entire the Doppler ensemble in-source, and only in selected cases is the hyperfine splitting large enough to be resolved.

Recently, the implementation of RIS in a low-temperature supersonic gas jet [6,7] utilizing a narrowband first step excitation has gained considerable interest [8]. An optimal solution to combine high pulse powers required for efficient ionization with a narrow linewidth is the pulsed amplification of a narrow-band continuous wave (CW) laser. While for high-gain dye lasers a single pass amplification is sufficient, the lower gain Titanium:Sapphire gain medium requires a different approach. In a regenerative amplifier, the cavity length is locked to a multiple of the seed wavelength allowing Titanium:Sapphire-based lasers to reach a final output power of several kW (during the pulse) from the few mW of CW input.

A previous iteration of the laser presented in this article, was designed and tested offline at the University of Jyväskylä [9,10] on stable Cu isotopes, and later applied to high-resolution RIS of Ac, Pu and Th [11,9,12,13] at University of Mainz. Furthermore, the previous iteration has been successfully applied to online studies of $^{214,215}$Ac in the vicinity of the $N = 126$ neutron shell closure in LISOL-facility in Belgium [14] and of $^{73-78}$Cu in CRIS [15] experiment in CERN-ISOLDE [16]. In this work, we present a pulsed injection-locked Titanium:Sapphire laser operating at 10 kHz repetition rate aimed for future in-jet spectroscopy at the SLOWRI/PALIS at RIKEN, apply it for RIS of Nb in vacuum and numerically estimate the resolution of in-jet RIS in counter-propagating geometry.

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## 2. Experimental setup

The slow RI-beam facility (SLOWRI) [17] at the RI Beam Factory (RIBF) [18] accelerator complex of the RIKEN Nishina Center for Accelerator-Based Science is being developed towards precision mass [19] and atomic spectroscopy [20] of rare isotopes. Using RF-carpet ion guides, the facility will provide a broad selection of low-energy isotopic beams of all elements produced through projectile fragmentation and in-flight fission reactions in the BigRIPS. However, the use of the SLOWRI gas stopper requires primary machine time at the BigRIPS ; hence the available time for experiments will be restricted by the limited operation time. To address this issue a new gas stopper, namely PALIS—PArasitic RI-beam production by Laser Ion Source, was proposed [21,22].

With the PALIS approach, the 200 MeV/u in-flight reaction products are stopped in 25 cm long gas cell located the second focal plane chamber the BigRIPS separator. The stopped products are promptly neutralized by high purity atmospheric pressure argon buffer gas, transported towards an ⌀ 1 to 2 mm exit hole by the gas flow in 1–300 ms and subsequently selectively re-ionized using 10 kHz repetition rate pulsed lasers. Due to large atom density, the ionization mainly takes around the exit hole region both in gas cell and in a supersonic gas-jet within a SextuPole Ion-beam Guide (SPIG) [23]. The ions are assisted through the SPIG by the gas flow and into a quadrupole mass separator after which they detected by a channeltron detector located in a high-vacuum region. The extreme pressure difference from atmospheric conditions to $10^{-3}$ Pa is achieved by using a novel differential pumping setup described in Ref. [24].

The novel idea behind the PALIS catcher is that it extends the available beam time at SLOWRI due its ability to run parasitically along primary experiments and utilize the otherwise discarded reaction products. Besides being a production device, PALIS has also been designed for in-jet RIS. The following sections describe a pulsed injection-locked Titanium:Sapphire laser system based on design presented in Ref's [25,26,10], aimed for high-resolution in-jet RIS. Furthermore, the laser is commissioned through RIS of $^{93}$Nb in vacuum.

Niobium was chosen as the test case due to experimental considerations, e.g. convenient transition frequencies considering the injection-locked Titanium:Sapphire, and for the possibility to apply the laser developments to future research projects. As niobium has only a single stable isotope, no mass separation is required and the experimental setup can be simplified considerably. In addition, the atomic structure for niobium is well documented (see Ref's [27–30]), thus enabling convenient evaluation of the accuracy of the setup.

Only limited optical spectroscopy has been performed on radioactive niobium isotopes [31] due to the refractory nature of the element which makes it difficult to produce with the standard thick target ISOL (Isotope Separation On-Line) method. However, high-resolution RIS capability at a gas-cell-based production facility such as PALIS would open a possibility for future on-line studies along the niobium isotopic chain. In addition, the isomer $^{93\mathrm{m}}$Nb($T_{1/2}$ = 16.13 a) has been proposed to be utilized in an integrated fast neutron dosimetry of nuclear reactor vessel and structures via resonance ionization mass spectrometry (RIMS) [28]. This technique aims to separate the ground state from the isomer $^{93m}$Nb, and it requires a narrowband laser to drive a transition with a large enough hyperfine splitting that can efficiently provide sufficient spectral selectivity [32]. Previous work utilizing Titanium:Sapphire lasers for this purpose can be found in Ref's [33,34].

### 2.1. PALIS laser system

The injection-locked Titanium:Sapphire laser has been designed to operate as a part of the PALIS-laser laboratory, described in detail in Ref. [23]. Two distinct laser systems (see Fig. 1 for a schematic layout) were utilized in this experiment. The first system is formed by two Sirah/Credo dye lasers pumped by a 10 kHz Edgewave Nd:YAG InnOSlab. The pump power can be split between the dye laser in various ways to produce up to 10 W average fundamental output power, which further converts to $\sim$1 W on the second harmonic with the linewidth in the GHz-region and a pulse width of $\sim$10 ns.

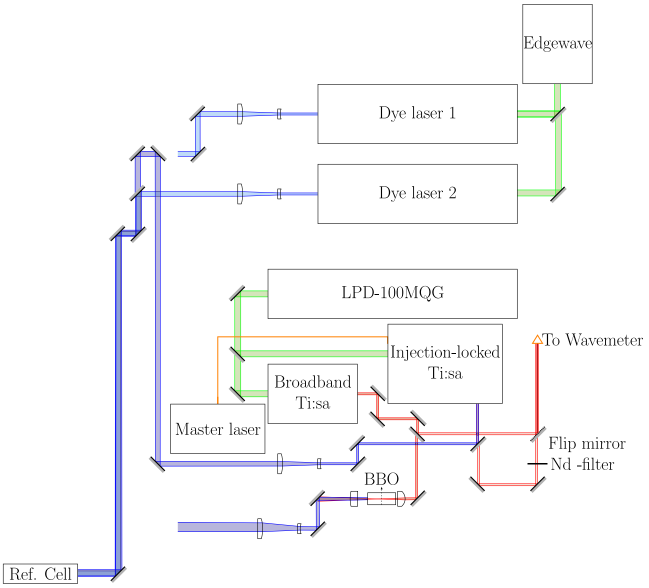

**Fig. 1.** A schematic optical table configuration at PALIS. The figure shows an overlap between the injection-locked Titanium:Sapphire and Dye laser 2 beams, but the systems allowed also dye laser 1 and the broadband Titanium:Sapphire to be utilized with the injection-locked Titanium:Sapphire.

The second laser system is a Titanium:Sapphire-based laser set-up pumped by a 10 kHz Nd:YAG (Lee Laser LDP-100MQG). The system consists of two lasers with one being a commercial broadband Titanium:Sapphire laser (Radiant Dyes) and the other laser is the in-house designed injection-locked Titanium:Sapphire laser. The broadband laser has a Z-shape cavity with an Etalon and a Birefringent filter resulting in a typical fundamental linewidth of 3–5 GHz, and an output power of ≤4 W with $\sim$35 ns pulse width.

Additionally the Titanium:Sapphire laser setup includes a single-pass harmonic generation setup capable of efficient generation of 2nd, 3rd and 4th harmonics. The setup also includes a precise beam shaping system realized with cylindrical lenses and beam expanders used compensate for beam astigmatism and divergence resulting from the harmonic generation, and to prepare the beam for the long transport to PALIS.

The pump lasers share a common master trigger, from which the signal is divided between the two pump lasers. The trigger for the Edgewave is passed through an Ortec 416A gate and delay generator to allow temporal synchronization of the output pulses from the dye lasers and the Titanium:Sapphire lasers. The two Titanium:Sapphire laser can further be synchronized by detuning either the laser cavities or the pump beam alignment.

When utilized for resonance ionization at the PALIS, the laser beams are expanded by a ∼ factor of 10, combined on the optical table and transported with more than 50% efficiency over a 70 m long optical path to the PALIS gas cell realized with broadband mirrors on a motorized mirror mounts [23].

### 2.2. Injection-locked Titanium: Sapphire laser

The lasers introduced in the preceding section were designed to operate in the GHz linewidth region to efficiently drive transitions in a high-pressure gas-cell environment. The injection-locked Titanium:Sapphire laser presented in this section, however, aims for resonance ionization