Precision Laser Spectroscopy Technique for Exotic Radioactive Beams at CERN-ISOLDE

Xiaofei Yang and the COLLAPS and CRIS collaboration
KU Leuven, Instituut voor Kern- en Stralingsfysica, B-3001 Leuven, Belgium
School of Physics and State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100871, China
E-mail: xiaofei.yang@pku.edu.cn

Abstract. The combination of high-resolution laser spectroscopy techniques and radioactive ion beams has provided major input for nuclear physics studies in different regions of the nuclear chart by accessing nuclear intrinsic properties, such as spins, magnetic and quadrupole moments and charge radii of exotic nuclei. As more and more exotic radioactive beams become available, laser spectroscopy and its relevant techniques start to be renascent with many advanced experimental setups being established and planned at radioactive-beam facilities world-wide. Meanwhile, a further exploration to expand the laser spectroscopy of radioactive beams into not only nuclear physics but also multiple interdisciplinary fields is being in progress. In this contribution, by using the relevant laser spectroscopy setups at ISOLDE as examples, the details of high precision laser spectroscopy experimental techniques applied to radioactive beams will be presented, together with a few highlights and achievements during the past years.

1. Introduction
It is well-known nowadays that the laser spectroscopy technique can be applied in various fields of science and technology. It was more than 30 years ago that this technique was firstly applied to nuclear physics studies [1, 2]. By probing the atomic hyperfine (HF) splitting and isotope shift (IS) using a high-precision laser spectroscopy method, multiple nuclear properties of ground/isomeric states of exotic nuclei far from stability, such as nuclear spins, nuclear magnetic and quadrupole moments and charge radii can be accessed simultaneously in a model independent way. With continuous development of radioactive ion (RI) beam facilities and production of bunched ions beam using the rf-quadrupole ion cooler and trap [3], the sensitivity of the laser spectroscopy technique has been significantly enhanced by several orders of magnitude in the last 10 years. The more and more exotic isotopes far from stability became accessible for laser spectroscopy, yielding many major inputs for nuclear structure study and providing important benchmark for the development of state-of-art nuclear theory [4, 5, 6, 7].

On another hand, considerable efforts have also been dedicated on the development of the laser spectroscopy technique with higher sensitivity and precision by a combination of many advantages offered by the conventional techniques [8, 9, 10] and by initiating many novel techniques [11, 12, 13]. These techniques have been established or are under construction or planned at different RI beam facilities across the globe and now can take advantages of RI beams produced from not only isotope separation on line (ISOL) but also in-flight project fragment (PF) facilities, as the The BEam COoler and LAser spectroscopy (BECOLA) [14] setup by using the RI beam at NSCL of MSU, US, and PALIS gas cell laser spectroscopy [15] developed to use
Figure 1. Example for HF structure of a given atomic transition from $^2S_{1/2}$ to $^2P_{3/2}$ with a nuclear spin of $I = 3/2$. It also presents the basic principle for (a) a collinear laser spectroscopy based on photo detection and (b) a typical collinear resonance ionisation spectroscopy by using ion detection.

The on-line laser spectroscopy method was firstly used for RI beams at ISOLDE-CERN. Until today, ISOLDE is still a well-known venue to produce most of the measurements of basic properties of RI beams by two complementary laser spectroscopy techniques, the conventional collinear laser spectroscopy technique (using the COLLAPS set-up) and the newly developed collinear resonance ionization spectroscopy (CRIS) by using the cooled and bunched ion beams. In this contribution, both COLLAPS and CRIS laser spectroscopy techniques currently active at ISOLDE will be introduced in detail as well as a few main inputs for nuclear structure studies in the last few years.

2. Precision lasers spectroscopy at ISOLDE

Figure 1 shows a HF splitting of atomic energy levels ($^2S_{1/2}$ and $^2P_{3/2}$) for a given atomic transition with a nuclear spin of $I = 3/2$, which is in a typical energy scale of $10^{-7}$eV or MHz and results in 6 allowed transition. This HF splitting is due to the interaction of the nucleus and electrons, and therefore provide directly several observables: nuclear spin ($I$), magnetic dipole ($A$) and electric quadrupole ($B$) HF parameters, and IS, as described in detail in Ref. [20] (Section 3). Therefore, by obtaining these parameters of a given isotope, fundamental properties (spins, moments and charge radii) of the nucleus can be extracted in a nuclear model independent way. Laser spectroscopy has acted as a powerful experimental technique for nuclear physics studies of RI beams over the past two decades. Certainly, several techniques can be applied for such measurement, for example in-source resonance ionization spectroscopy (RIS), collinear laser spectroscopy and spectroscopy of trapped atoms. The merit and demerit of different method has been evaluated in several review publication [16, 20], mainly regarding to three aspects: resolution, efficiency, accessible elements. Currently, the precision laser spectroscopy technique at ISOLDE is a compromise or balance between experimental efficiency and resolution by probing the ions or atoms with the low energy of 30-60 keV in a collinear geometry with the laser beam.
This technique has been employed at several RI beam facilities world-wide, and contributed the major part of the summarized nuclear chart for ground state properties measurements of RI beams (Figure 1 of Ref. [16]).

2.1. Collinear laser spectroscopy (COLLAPS)
COLLAPS is one of the collinear laser spectroscopic setups devoted to the study of radioactive isotopes, located at the isotope separator on-line facility ISOLDE at CERN. A detail overview about the past, present and future of COLLAPS setup can be found in a recent review publication [21]. As shown in Figure 1, this technique is based on the photon detection emitted from the laser resonance exited atoms or ions.

The different parts of the basic setup can be shortly described as (see FIG. 2 of Ref [22] for the schematic representation of the setup): The bunched ions beam delivered from ISOLDE is overlapped with the laser light from a cw Ti:sapphire or Dye laser (or together with a frequency doubling device), which is usually locked to a fixed frequency to match a Doppler shifted atomic/ionic transition and stabilized with an external stabilization system to avoid systematic errors and instability of the laser frequency. A charge exchange cell (CEC) filled with K or Na vapor (depends on the studied element) is used when an atomic transition from a certain atomic state was used for the HF structure measurement. This atomic state is populated when the RI beam passes thought the high-density vapor filled CEC. For the measurement process, instead of tuning the laser frequency, a variable voltage is applied to the light collection region (or CEC) to tune the velocity of the beam onto resonance with the laser beam. The photons emitted from the laser excited atoms or ions are recorded by four photomultiplier tubes (PMTs) as a function of the tunable voltage to obtain the HF spectra. Note that by using the bunched beam from ISOLDE (typical every ~ 100 ms, depends on the studied isotopes), the collection time for the PMTs can be gated with the bunch width of the beams (typical ~ 5 \mu s) in which the atom bunches passed through the light collection system. In this way, the background arising from the scattered laser light was reduced by a factors of \approx 10^4 (\approx 5 \mu s /100 ms ) and therefore the overall sensitivity of the experiment has been significantly improved, as have been demonstrated for many experiments in the last few years [4, 6, 23].

Another noteworthy point about COLLAPS is regarding to its versatility. Namely, the basic setup of COLLAPS can be modified or extended to a higher sensitivity measurement. For example, by a combination of a basic COLLAPS and ultra-sensitive collisional ionization detection [24], the exotic Kr isotopes were investigated, reaching a sensitivity of 10^3 atoms/s with continuous beam from ISOLDE; combing the optical pumping and \beta-NMR technique, COLLAPS has been used to studied exotic Mg isotopes [25], providing key input for the study of isotopes in the region of ‘island of inversion’, as has been reviewed in Ref. [26]; In-cooler optical pumping has been applied to Mn isotopes recently to populate a metastable state, enabling the measurement of the quadrupole moments of $^{53,63}$Mn and showing a clear onset of collectivity towards $N = 40$ [27]: a dedicated setup with ultra high sensitivity has been extended from the basic COLLAPS setup, aiming to measure the ground state properties of $^{53,54}$Ca with a production yield less than 10 ion/s [28].

2.2. Collinear resonance ionization spectroscopy
It is generally known that the production of exotic isotopes usually drops when go to the neutron-rich or proton-rich side, while the typical sensitivity for the COLLAPS technique is around $10^3$ ions/s. In-source spectroscopy using the efficient and selective resonant laser ionization method is another technique applied for rare isotope studies with very high efficiency [29]. Resonant laser ionization is also used in laser ion sources at many on-line facilities around the world [30]. However, due to pressure and Doppler broadening as well as the linewidth of commonly used pulsed lasers, the achieved resolution for the HF spectra from this technique is typically larger.
than 1GHz, which prevents a precision measurement of ground state properties of light and medium mass isotopes.

CRIS is proposed to combine the high resolution achieved in collinear spectroscopy and the high sensitivity of resonance laser ionization spectroscopy. With a continuous effort for the development of high-resolution pulsed laser, CRIS has achieved a resolution of less than 100 MHz (in some cases even 20 MHz [10]) and sensitivity of 20 ions/s for Cu isotopes recently [31]. As detailed in Fig. 1 of Ref. [9], the delivery of the ISOLDE ion beam, the collinear geometry of the beam line, and the CEC system used in the CRIS setup are comparable to those in the COLLAPS setup. Specifically, instead of detecting the laser induced fluorescence, CRIS detects the laser resonantly re-ionized ions, which can be deflected from the remaining background atoms in the beam. Thus the sensitivity limitation of photon detection is overcome and also the background photon detection due to scattered laser light is avoided, resulting in a higher overall sensitivity [8]. This is realized by multiple-step laser excitation until the ionization potential of the studied element is reached. The process occurs in an interaction region with ultra-high vacuum (UHV: currently 10^{-9} mbar) to suppress the production of background ions from collisional ionization. This UHV region after the CEC is achieved by a differential pumping section [9]. The laser ionized ions are deflected onto an ion detector and recorded. Note that a high resolution laser system with a linewidth of at most a few 10 MHz is required for the first step excitation to reach high resolution HF spectra, and also this laser is required to be pulsed or chopped to eliminate the possible low sensitivity resulting from optical pumping processes for certain elements [10, 31]. Therefore, it is the combination of bunched ion beams, the resonance ionization process, ion detection, high-resolution pulsed laser system and the UHV to reduce non-resonant excitation that makes CRIS a high resolution and high sensitivity laser spectroscopy method for the measurement of ground state properties of rare isotopes with low production yields down to few 10 ions/s.

Furthermore, different long-lived states of a studied isotope can be selected by the multiple-step laser ionization procedure and send to a decay spectroscopy station for laser-assisted nuclear decay spectroscopy on a pure isomeric beam. This method has been successfully used for alpha-tagging of HF components and for the alpha decay of ground and isomeric states of neutron-deficient Fr isotopes [9].

2.3. Recent highlights from laser spectroscopy measurements at ISOLDE
As laser spectroscopy provides four nuclear parameters at the same time, it can offer different aspects of nuclear structure information. For example, the nuclear spins and magnetic moments can be used to study the shell evolution of the single particle states of nucleons [32]; the magnetic and quadrupole moments can be used to check the configuration of the wavefunction for a given state with the help of the shell model calculation [33, 27]; the quadrupole moments and mean square charge radii can probe the collectivity and deformation of the nucleus [27, 34]; charge radii itself can also be used to verify the halo structure observed in light nuclei [35]; a combination of spins, moments and charge radii of ground and isomeric states also signal the existence of shape coexistence in a given nucleus [7].

Recently, many efforts for laser spectroscopy measurements at ISOLDE have been applied to the isotopes near the double magic nuclei such as $^{52,54}$Ca, $^{78}$Ni, $^{100,132}$Sn, resulting in many fruitful optical spectra results, providing key inputs for nuclear structure studies. Around $^{52,54}$Ca, HF spectra of Ca isotopes have been measured between $N = 20$ and $N = 32$ at COLLAPS, yielding the nuclear spins, moments and charge radii up to $^{52}$Ca. The spins and moments of Ca isotopes show the single particle nature of the $^{41-49}$Ca, but with some mixing in the wave function of $^{51}$Ca due to excitations of neutrons across $N = 32$ [6]. The unexpected increase in the charge radii up to $N = 32$ challenges the earlier suggested magicity of neutron number 32 and tested the most advanced nuclear theory from both phenomenological and
microscopic interactions [36]. Neutron rich Cu isotopes up to \( N = 49 \) have been studied by the CRIS experiment, and achieved the highest efficiency until now for a high-resolution HF spectra measurement. With one proton added to the proton closed shell \( Z = 28 \), the magnetic and quadrupole moments of \(^{76-78}\text{Cu} \) are therefore very sensitive to the cross shell excitation of proton and neutron [31]. Assisted by theoretical calculations from state-of-the-art shell model, this measurement provides further evidence for the robustness of \( Z = 28 \) shell closure near \(^{78}\text{Ni} \) because of a subsequent decrease of proton and neutron excitations across \( Z = 28 \) and \( N = 50 \) when approach to \( N = 50 \). Experimental results in the Sn region from both laser spectroscopy techniques at ISOLDE are ongoing and under analysis, which will certainly provide important information on the nuclear structure around the doubly-magic isotopes \(^{100}_{50}\text{Sn} \) and \(^{132}_{50}\text{Sn} \). Further study of more exotic isotopes near these so called double magic nucleus are still on going.

3. Summary
Laser spectroscopy is a powerful tool for a systematic measurement of nuclear ground state properties of exotic isotopes. Here, we introduced two complementary precision laser spectroscopy techniques at ISOLDE. The measurements achieved from these setups have provided massive nuclear data on the fundamental properties (nuclear spins, moments and charge radii) of ground and long-lived isomeric states of exotic isotopes produced at ISOLDE, and contributed a lot to the nuclear structure studies in the different regions of nuclear chart.

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