

Laser Resonance Ionization Technique for Studies of Unstable Nuclear Properties (Postprint)

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Abstract

The fundamental properties of unstable atomic nuclei reflect their internal structure and effective interactions, and can be used to investigate the exotic structures of unstable nuclei. Laser nuclear spectroscopy, by measuring the hyperfine structure and isotope shift in atomic electron spectra, can extract multiple fundamental properties of atomic nuclei in a model-independent manner, and represents one of the powerful tools for studying the properties and structures of unstable nuclei. The multi-step laser resonance ionization method is one of the techniques for measuring the hyperfine structure and isotope shift of atoms or ions. Based on this, various resonance ionization spectroscopy experimental techniques have been developed internationally for studying the fundamental properties and structures of unstable nuclei at radioactive ion beam facilities. This paper first introduces the laser resonance ionization method and the various resonance ionization laser spectroscopy experimental techniques developed therefrom. Subsequently, it discusses in detail the collinear resonance ionization spectroscopy technique that has emerged in the past decade. This technique enables simultaneous high-resolution and high-sensitivity measurements of hyperfine structure spectra, and is playing an important role in studies of the properties and structures of unstable nuclei across a large mass range of the chart of nuclides. Finally, it analyzes the current status and application prospects of resonance ionization laser spectroscopy techniques for domestic radioactive ion beam facilities.

Full Text

Laser Resonance Ionization Techniques for Studying the Properties of Unstable Nuclei

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Abstract

The fundamental properties of unstable nuclei are highly related to nuclear structure and effective nucleon-nucleon interactions, and can be used to study various exotic structures of unstable nuclei. Laser spectroscopy is a powerful tool for studying nuclear properties and structure by probing the hyperfine structure and isotope shift of corresponding atoms or ions, from which nuclear properties can be extracted in a nuclear model-independent manner. Multi-step laser resonance ionization spectroscopy (RIS) can be used to measure atomic or ionic hyperfine structure. Based on this approach, various experimental techniques have been developed at radioactive ion beam (RIB) facilities worldwide to study the properties and structure of unstable nuclei. This paper first introduces the RIS method and related experimental techniques. Subsequently, the recently-developed collinear resonance ionization spectroscopy technique, which enables high-resolution and high-sensitivity measurement of hyperfine structure spectra and plays an important role in studying the properties and structure of unstable nuclei across large mass regions of the nuclear chart, is discussed in detail. Finally, the development status of RIS and its application prospects at domestic RIB facilities are analyzed.

Keywords: unstable nuclei, hyperfine structure, laser resonance ionization, laser spectroscopy technique

Unstable nuclei are mostly located in unexplored regions of the nuclear chart and exhibit rich structural phenomena such as halo structures, shell evolution, and shape coexistence. Related measurements continuously challenge existing nuclear structure models and theoretical frameworks. Therefore, research on unstable nuclei represents one of the frontier hotspots in international nuclear physics basic research [1-4]. With upgrades of existing RIB facilities worldwide and development of next-generation RIB facilities—such as ISOLDE (Isotope Separator On-Line DEvice) at CERN [5], RIBF (Radioactive Isotope Beam Factory) at RIKEN [6], FRIB (Facility for Rare Isotope Beams) at Michigan State University [7], FAIR (Facility for Antiproton and Ion Research) at GSI [8], and HIAF (High Intensity heavy-ion Accelerator Facility) at the Institute of Modern Physics in China [9]—more neutron-rich and proton-rich nuclides far from the β -stability line will be produced, providing important conditions for further studying the properties and exotic structures of unstable nuclei through various experimental means.

Currently, multiple experimental methods and techniques are used to study unstable nuclei, typically including nuclear reactions, nuclear decay, and fundamental property measurements, which are widely applied at international RIB facilities [1]. Different experimental techniques for studying unstable nuclei also

depend on the method of RIB production. RIB facilities mainly produce unstable nuclear beams through two approaches: the Isotope Separator On-Line (ISOL) method and Projectile Fragmentation (PF) [2]. The ISOL method uses light particle beams (such as p, D, etc.) to bombard thick targets, inducing nuclear reactions. Reaction products are stopped in the target, then diffused out and ionized in an ion source, extracted and electrostatically accelerated to form unstable nuclear beams of tens of keV. The PF method uses high-energy heavy ion beams (tens or hundreds of MeV) to bombard thin targets, inducing projectile fragmentation reactions to produce high-energy radioactive nuclear beams.

Laser spectroscopy is a powerful tool for measuring fundamental properties of unstable atomic nuclei [10-12], primarily for high-precision measurements of low-energy radioactive beams. Therefore, it is mostly applied at ISOL-type RIB facilities [13-14]. By measuring the hyperfine structure spectra of atoms, ions, or molecules of unstable nuclei, laser spectroscopy can precisely extract nuclear properties such as spin, magnetic moment, electric quadrupole moment, and charge radius in a nuclear model-independent manner, and has played an important role in studying the properties and exotic structures of unstable nuclei [2,11]. Laser spectroscopy mainly measures hyperfine structure spectra of atoms, ions, or molecules through two methods: laser-induced fluorescence and laser resonance ionization. Based on these two methods, various laser spectroscopy techniques have been developed internationally, notably collinear laser spectroscopy based on fluorescence detection and in-source laser spectroscopy based on laser resonance ionization. Collinear laser spectroscopy obtains hyperfine structure spectra of ions or atoms by resonantly exciting them using the Doppler tuning method with collinear laser and radioactive beams, then detecting de-excitation fluorescence [2,14]. This method effectively suppresses Doppler broadening and enables high-resolution measurement of hyperfine structure spectra, making it applicable for studying unstable nuclei across all mass regions of the nuclear chart [2]. In-source laser spectroscopy using laser resonance ionization offers higher detection sensitivity, but its resolution is limited by Doppler or collisional broadening, making it difficult to measure hyperfine structure spectra of light-mass atoms with small hyperfine splitting. It is currently mainly used for studying nuclear properties in heavy-mass regions [2]. Laser resonance ionization technology can be applied not only to frontier basic research on properties and structures of unstable nuclei, but also has various application potentials such as laser ion sources [15] and medical isotope production [16]. In recent years, the newly developed collinear resonance ionization spectroscopy method, combining collinear techniques, has achieved breakthrough progress by simultaneously realizing high-resolution and high-sensitivity measurements of unstable nuclei, playing an important role in studying properties and structures of unstable nuclei across large mass ranges of the nuclear chart [17-18]. Recently, this method was also successfully applied for the first time to spectroscopic measurements of radioactive molecules containing unstable nuclei, providing new approaches and opportunities for research in nuclear structure,

nuclear astrophysics, and fundamental symmetries [19-20].

This paper focuses on laser resonance ionization methods and their applications. It begins with the principles of laser resonance ionization, then introduces in detail various resonance ionization laser spectroscopy techniques developed internationally, such as hot-cavity in-source resonance ionization spectroscopy, in-gas-cell and in-gas-jet resonance ionization spectroscopy, radiation-detected resonance ionization spectroscopy, and collinear resonance ionization spectroscopy. Finally, combined with the current development status of laser spectroscopy technology in China, the paper discusses application prospects at existing and next-generation RIB facilities.

Basic Principles of Laser Resonance Ionization

The basic principle of laser resonance ionization is illustrated in Figure 1 [Figure 1: see original paper], which shows the use of multi-step lasers to resonantly excite and ionize extranuclear electrons step by step. Figure 1 also presents three common schemes of laser resonance ionization: non-resonant ionization, resonant ionization, and field ionization [2]. In non-resonant ionization, the final step uses a high-power laser with fixed frequency to excite electrons above the ionization potential. Resonant ionization uses multi-step lasers with continuously tunable frequencies to resonantly excite electrons to autoionizing states. Field ionization uses multi-step lasers to resonantly excite atoms to Rydberg states, forming Rydberg atoms that are easily ionized by electrostatic fields or thermal effects. This is because Rydberg atoms have loosely bound valence electrons at large distances from the atomic core (with extremely low binding energies) that are easily ionized through collisions or external field perturbations. Experimentally, hyperfine structure spectra are obtained by directly detecting the ionized ions, or by detecting decay products of the ions after ionization, or by using high-sensitivity ion traps to detect ionized ions, and correlating these with the frequency of the resonantly excited laser (such as the frequency ν_1 of the first-step laser shown in Figure 1).

In practice, the most commonly used method is still non-resonant ionization, since suitable autoionizing states cannot be found for all elements of interest. However, because the final step of non-resonant ionization requires high-power lasers to achieve efficient ionization, the intense laser can directly ionize target atoms or impurity atoms already in excited states due to collisional excitation, creating large detection backgrounds that greatly affect the sensitivity of hyperfine structure spectrum measurements. Field ionization is considered one way to further improve experimental detection sensitivity. Figure 2 [Figure 2: see original paper] shows two commonly used field ionization modes in resonance ionization laser spectroscopy experiments. Figure 2(a) describes how a Rydberg atom beam of tens of keV is ionized into ions when entering a high electric field and experiencing a sudden increase in field strength (large field gradient) [21]. Figure 2(b) illustrates collisional ionization of atoms in Rydberg states in the hot-cavity in-source resonance ionization spectroscopy technique [22].

In-Source Resonance Ionization Laser Spectroscopy Techniques

The spectral resolution of Resonance Ionization Spectroscopy (RIS) is limited by the environment and conditions of the ion source (such as temperature, pressure, etc.), typically above GHz [28]. Currently, various in-source resonance ionization laser spectroscopy techniques have been developed internationally using different radionuclide reaction target sources, including hot-cavity in-source resonance ionization spectroscopy, in-gas-cell and in-gas-jet resonance ionization spectroscopy, and Radiation Detected Resonance Ionization Spectroscopy (RADRIS). Figure 3 [Figure 3: see original paper] qualitatively shows the detection sensitivity and resolution of several in-source resonance ionization spectroscopy techniques, demonstrating that resolutions are all above GHz and sensitivities can achieve yields below 10 pps. It is worth noting that such in-source laser spectroscopy often needs to be combined with high-sensitivity detection methods (such as decay particle detection or high-sensitivity ion trap detection) to achieve even higher sensitivity (below 1 pps) for hyperfine structure spectrum measurements. The following sections provide detailed examples of these in-source resonance ionization spectroscopy techniques.

2.1 Hot-Cavity In-Source Resonance Ionization Spectroscopy The hot-cavity in-source resonance ionization spectroscopy technique was first developed by Alkhazov et al. based on the laser ion source at the ISOL-type RIB facility IRIS, enabling measurement of hyperfine structure spectra of proton-rich $^{154,156}\text{Yb}$ isotopes and demonstrating that this method could measure nuclides with yields as low as 1–10 pps [23,29]. Currently, this hot-cavity in-source resonance ionization spectroscopy technique is widely used at ISOL-type RIB facilities. ISOL facilities produce nuclear reactions by bombarding thick targets with high-energy light ions (such as protons). Reaction products are typically stopped in the thick target; when heated above 2,000 K, the reaction products diffuse out of the target and into the ion source. Inside the ion source, atoms of the reaction products of interest are resonantly ionized by multi-step lasers. The resulting ions are extracted and accelerated, then mass-separated and detected (Figure 1 in reference [25]). By measuring the relationship between ion counts and laser frequency, the hyperfine structure spectrum of the target atomic nucleus can be obtained. At 2,000 K, the transit time of thermally moving atoms is on the order of 100 ns. Therefore, to improve the duty cycle between the ion beam and laser beam, the repetition rate of multi-step pulsed lasers used for resonance ionization is generally 10 kHz or higher [2]. The resolution of hyperfine structure spectra measured by in-source resonance ionization spectroscopy is affected by Doppler and pressure broadening, typically on the order of GHz, and is thus currently mainly used for studying heavy-mass nuclides with large hyperfine structure splitting [30].

Early hot-cavity in-source resonance ionization spectroscopy primarily used Faraday cups or ion detectors to measure hyperfine structure spectra [25]. For measurements of increasingly lower-yield nuclides accompanied by large

amounts of isobaric impurities, in-source resonance ionization spectroscopy has been combined with decay detection that measures decay products of target ions, or with ion detection using high-sensitivity ion trap technology, greatly improving experimental sensitivity. For example, at CERN's ISOLDE facility, in-source resonance ionization spectroscopy combined with a Multi-Reflection Time-of-Flight Mass Spectrometer (MR-ToF MS) has enabled measurement of the charge radius of ^{177}Hg ($Z = 80$) with yields of only a few ions per minute [26]; at the IGISOL (Ion Guide Isotope Separator On-Line) facility at the University of Jyväskylä in Finland, in-source resonance ionization spectroscopy combined with position-sensitive Penning trap mass spectrometry PI-ICR (Phase-Imaging Ion-Cyclotron Resonance) has enabled measurement of the charge radius of ^{96}Ag ($Z = 47$) with yields of only 0.005 pps [27].

To improve the spectral resolution of hot-cavity in-source resonance ionization spectroscopy, new technologies are continuously being developed, such as the recently developed Perpendicularly Illuminated Laser Ion Source and Trap (PI-LIST) [31-32]. The LIST technology was initially developed to improve the purity of beams produced by hot-cavity ion sources by installing a positively charged repelling electrode at the extraction aperture of the hot cavity to suppress upstream positive ions, thereby addressing the poor selectivity caused by isobaric impurities [33]. This technology has been applied at CERN-ISOLDE, improving the suppression of surface-ionized isobaric impurities by more than 1,000 times [34]. The PI-LIST technology, developed based on LIST, suppresses Doppler broadening and improves spectral resolution by aligning the laser perpendicular to the ion source for interaction with atoms. This technology was first developed and operated on the offline RISIKO (Resonance Ionization Spectroscopy in KOLinear Geometry) device at the University of Mainz in Germany, achieving high-resolution (FWHM (Full Width at Half Maximum) ~ 100 MHz) measurements of hyperfine structure spectra of long-lived nuclides such as $^{97,99}\text{Tc}$ ($Z = 43$) and $^{143,147}\text{Pm}$ ($Z = 61$) [31-32]. However, the geometric cross-section for interaction between perpendicularly incident laser and atoms is significantly reduced, thereby decreasing resonance ionization efficiency.

2.2 In-Gas-Cell and In-Gas-Jet Resonance Ionization Spectroscopy

In the 1980s, the University of Leuven in Belgium proposed and developed the in-gas-cell resonance ionization spectroscopy technique [35-38] for use at the LISOL (Leuven Isotope Separator On-Line) RIB facility [39]. Using the measurement of ^{215}Ac ($Z = 89$) at Leuven as an example [40], a primary beam of ^{22}Ne or ^{20}Ne bombards a ^{197}Au target, inducing fusion-evaporation reactions. The resulting $^{215}\text{Ac}^+$ ions are neutralized in a gas cell filled with argon, and the ^{215}Ac atoms enter the ionization region with the gas flow, while unneutralized $^{215}\text{Ac}^+$ ions are removed by electrostatic deflection electrodes. The ^{215}Ac atoms are resonantly ionized by multi-step lasers, and after passing through an RF ion guide and mass separator, their decay α particles are detected to obtain the hyperfine structure spectrum of ^{215}Ac atoms. Although energy dispersion caused by collisions with buffer gas results in a spectral line width of 5.8 GHz for

the ^{215}Ac hyperfine structure spectrum, in-gas-cell resonance ionization spectroscopy achieved high-sensitivity measurement of ^{215}Ac with yields of only 8.3 ions/s. This technology has currently been developed and applied at multiple RIB facilities, such as IGISOL at the University of Jyväskylä [41] and KISS (KEK Isotope Separation System) at RIKEN-RIBF [42-43].

To further improve the spectral resolution of hyperfine structure spectra measured by in-gas-cell resonance ionization spectroscopy, the Leuven team has recently developed the in-gas-jet resonance ionization spectroscopy technique [44-45] (Figure 1 in reference [40]), greatly improving experimental spectral resolution. In this technique, target atoms are ejected from the gas cell at high speed through a de Laval nozzle in the resonance ionization region, forming a collimated supersonic gas jet that provides a low-temperature, low-pressure, quasi-collision-free environment, significantly reducing energy dispersion caused by collisions and temperature. Target atoms are resonantly ionized by perpendicularly incident lasers in this environment, enabling high-resolution measurement of hyperfine structure spectra of unstable nuclear atoms. The first online experiment using this technique achieved high-resolution measurement (FWHM: 394 MHz) of unstable ^{215}Ac nuclides, improving spectral resolution by nearly 15 times compared to in-gas-cell resonance ionization spectroscopy [40]. This high-resolution, high-sensitivity in-gas-jet resonance ionization spectroscopy technique will also be applied at the S3 Low-Energy Branch (S3-LEB) of GANIL-SPIRAL2 in France [46] and the Separator for Heavy Ion reaction Products (SHIP) terminal at GSI in Germany [47].

2.3 Radiation-Detected Resonance Ionization Spectroscopy Laser spectroscopy experiments on unstable nuclei of superheavy elements ($Z \geq 100$) can provide important information for testing atomic theory and studying nuclear structure in the superheavy region. The yields of these superheavy nuclides are often very low; for example, at the SHIP RIB facility at GSI in Germany, superheavy nuclides produced through fusion-evaporation reactions typically have yields of only a few pps [48]. In the 1990s, radiation-detected resonance ionization spectroscopy developed at GSI enabled measurement of the magnetic moment and charge radius of the heavy-mass unstable nucleus ^{208}Tl ($Z = 81$) [46]. In recent years, to conduct laser spectroscopy studies of superheavy nuclei, the experimental team at GSI has optimized and developed radiation-detected resonance ionization spectroscopy, conducting high-efficiency (1%) measurements of ^{155}Yb nuclides [48] and ultimately achieving studies of nobelium (No, $Z = 102$) isotopes [49].

As shown in Figure 4 [Figure 4: see original paper], radioactive heavy nuclides (such as $^{252,254}\text{No}$) produced through fusion-evaporation reactions enter a buffer gas cell through an entrance window. In this buffer gas stopping cell, large numbers of reaction products remain in positive charge states. These positive ions accumulate on a tantalum filament and are neutralized to atoms. By heating the tantalum filament to 1,350 K, the adsorbed $^{252,254}\text{No}$ atoms evap-

orate from the filament. The evaporated atoms are resonantly ionized by lasers and rapidly guided by extraction electrodes into an α detector. By measuring the relationship between the decay α particle count of the target nuclide and the frequency of the resonantly excited laser, the hyperfine structure spectra of $^{252,254}\text{No}$ atoms can be obtained. Through this radiation-detected resonance ionization spectroscopy technique, the GSI team achieved, for the first time internationally, high-efficiency measurement of hyperfine structure spectra of superheavy $^{252,254}\text{No}$ nuclides (total efficiency of 3.3(1.0)% for ^{252}No and 6.4(1.0)% for ^{254}No). The yield of ^{254}No was only 4 pps, and the measured spectral broadening was about 4 GHz [49-50]. This represents the heaviest element studied by laser spectroscopy to date. This experiment also provided the first precise determination of the ionization potential of nobelium, (6.62621 ± 0.00005) eV [49,51]. These measurements provide important experimental data for future theoretical work on superheavy atoms and lay the foundation for laser spectroscopy measurements of even heavier elements [52].

Collinear Resonance Ionization Spectroscopy

As described above, in-source resonance ionization laser spectroscopy methods, due to their limited resolution, are mainly used for studying properties and structures of heavy-mass unstable nuclei. For numerous light-mass and medium-mass nuclides, high-resolution laser spectroscopy techniques are required. Typical high-resolution laser spectroscopy methods for unstable nuclei include atom/ion traps and collinear laser spectroscopy [2]. Atom/ion trap technology is limited by element type and can mostly only be applied to alkali metal and noble gas elements. Collinear laser spectroscopy uses collinear laser and high-speed (tens of keV) atomic or ion beams, greatly reducing Doppler broadening caused by energy dispersion. Specifically, ions with a certain energy dispersion δE accelerated to energy E have a velocity uncertainty of $\delta v = \delta E/mv$, resulting in Doppler broadening of: $\delta\nu = \nu_0 \delta v/c$, where ν_0 is the transition frequency and m is the ion mass. As can be seen, Doppler broadening δ decreases as the ion beam energy E increases. When ion energies are 30-60 keV, Doppler broadening caused by ion energy dispersion can be reduced to tens of MHz, comparable to the natural broadening of atomic transitions. Typical collinear laser spectroscopy based on this technique usually employs laser-induced fluorescence, so measurement sensitivity is limited by large background counts from laser-scattered photons and is commonly used for studying unstable nuclides with yields of 10^3 - 10^4 pps.

The collinear resonance ionization spectroscopy (CRIS) technique developed in recent years adopts resonance ionization based on collinear ion and laser beams, simultaneously achieving high-resolution and high-sensitivity measurements of unstable nuclei [17-18]. The CRIS concept was first proposed by Kudriavtsev et al. in 1982, initially envisioned for measuring ultra-low-abundance long-lived rare radioactive nuclides (such as ^{26}Al ($Z = 13$): $T_{1/2} = 7.4 \times 10^5$ years, relative natural abundance of 10^{-14}) [53]. In 1991, Schulz et al. [54] verified the feasibility of this method through online experiments at CERN-ISOLDE.

The experiment used two-step collinear lasers to excite atoms of nuclides such as $^{157,159,175}\text{Yb}$ to Rydberg states and measured hyperfine structure spectra through field ionization, thereby obtaining fundamental properties of these nuclides. This experiment used a continuous radioactive beam, achieving a total detection efficiency of only 0.001%, mainly due to the low duty cycle between the continuous beam and pulsed laser beam. To avoid duty cycle losses, after the development of beam bunching technology based on RadioFrequency Quadrupole cooler and buncher (RFQ) [55], Flanagan et al. [56] conducted offline verification experiments of the CRIS technique using pulsed stable ^{27}Al beams, achieving an experimental detection efficiency of about 3%. Subsequently, Flanagan et al. [58] successfully applied this CRIS technique at CERN-ISOLDE for studying properties and structures of unstable nuclei.

Figure 5 [Figure 5: see original paper] shows a schematic of the first CRIS experimental setup developed internationally at CERN-ISOLDE for studying unstable nuclei [58]. Radioactive beams of 30–60 keV produced by the ISOLDE facility are mass-separated, then cooled, bunched, and released in an RFQ [59] to achieve beam pulsing. After electrostatic deflection into the CRIS experimental terminal, the pulsed ion beam is first neutralized in a charge exchange cell filled with K or Na vapor. Unneutralized ions are removed by electrostatic deflection electrodes, and the neutralized pulsed atomic beam enters the interaction region through a differential pumping section. In this region, multi-step pulsed lasers overlap spatially and are synchronized temporally with the atomic beam to achieve resonant ionization of atoms. The interaction region is maintained at ultra-high vacuum (10^{-7} – 10^{-8} Pa) to minimize background from collisional ionization of atoms with stray gas. Ions resonantly ionized by lasers are electrostatically deflected into a detector for collection, ultimately obtaining hyperfine structure spectra of target atoms.

In 2012, the first online CRIS experiment achieved measurement of magnetic moments and charge radii of neutron-deficient $^{202,205,218\text{m},219,229,231}\text{Fr}$ ($Z = 87$) isotopes in the heavy-mass region, reaching a detection efficiency of 1% [58,60–61]. This experiment used pulsed Ti:sapphire lasers with 1 GHz linewidth for the first resonant excitation step, resulting in spectral resolution >1 GHz. In 2014, the CRIS team used Pockels cell-modulated narrowband continuous-wave lasers to generate narrowband periodic lasers for resonantly exciting francium, achieving high-resolution (FWHM: 20 MHz) hyperfine structure spectrum measurements of Fr atoms [62]. However, the pulsed laser power after Pockels cell modulation was relatively low, directly affecting laser resonance ionization efficiency. In 2016, the CRIS team used high-power narrowband pulsed lasers generated by seed amplification technology based on narrowband continuous-wave lasers to achieve, for the first time, high-resolution (FWHM: 75 MHz) and high-sensitivity measurement of neutron-rich ^{78}Cu ($Z = 29$) nuclides in the medium-mass region with yields of only 20 pps [17]. In 2018, for the short-lived (110 ms), low-yield (200 pps), high-stable-impurity (10^6 pps) light-mass nuclide ^{52}K ($Z = 19$), the CRIS team first employed β -decay-tagged collinear resonance ionization spectroscopy to measure the charge radius of the extremely

neutron-rich nucleus ^{52}K [18]. The β -decay-tagged collinear resonance ionization spectroscopy method exploits the short half-life characteristic of nuclides by detecting β -decay products instead of ions, avoiding interference from large numbers of stable impurity nuclei. In 2018, the CRIS team completed the first international spectroscopic measurement of short-lived radioactive molecules (such as RaF), providing new opportunities for fundamental symmetry studies at low energy scales [19-20]. In 2019, the CRIS team used field ionization technology to achieve high-resolution measurement of $^{113,115}\text{In}$ ($Z = 49$) nuclides, promising to further improve the sensitivity of the CRIS method [21].

After more than ten years of development, CRIS technology has matured and become an internationally leading high-resolution, high-sensitivity laser spectroscopy technique. Currently, this technique has been used to conduct experimental measurements of unstable nuclear properties across all mass regions of the nuclear chart, including light-mass K [18,63] and Sc ($Z = 21$) [64], medium-mass Cu [17] and Ga ($Z = 31$) [65], medium-heavy-mass In [66] and Sn ($Z = 50$) [67], and heavy-mass Ra ($Z = 88$) [68] and Fr [62], playing an important role in studying exotic structures of unstable nuclei. Currently, CRIS technology is being developed at Michigan State University's FRIB facility, the University of Jyväskylä's IGISOL facility, and China's BRIF (Beijing Radioactive Ion-beam Facility) for studying properties and structures of nuclei further from stability.

Summary and Outlook

This paper detailed the basic principles of laser resonance ionization methods and various online laser spectroscopy experimental facilities developed for studying properties and structures of unstable nuclei, including hot-cavity in-source resonance ionization spectroscopy, in-gas-cell and in-gas-jet resonance ionization spectroscopy techniques, radiation-detected resonance ionization spectroscopy, and collinear resonance ionization spectroscopy.

Currently, RIB facilities are being upgraded and next-generation RIB facilities are being developed internationally to produce nuclei even further from stability near the drip lines. Laser spectroscopy techniques are rapidly developing and being widely deployed at these RIB facilities. In China, existing BRIF [69] and the next-generation large-scale facility HIAF under construction [9] have not previously developed laser spectroscopy experimental platforms for studying properties and structures of unstable nuclei. Recently, domestically developed collinear laser spectroscopy equipment based on fluorescence detection has achieved important progress in offline and online experiments, reaching the advanced level of similar international facilities [14,70-72]. To study properties and structures of unstable nuclei with even lower yields at existing and future RIB facilities in China, domestic teams are also developing high-resolution, high-sensitivity collinear resonance ionization spectroscopy technology, which has already achieved important phased progress and is currently undergoing offline physics testing [73]. This technology will be combined with RFQ under development and applied at the BRIF facility and future HIAF facility for

studying fundamental properties and structures of unstable nuclei, as well as for spectroscopic studies of radioactive molecules.

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References

- [1] YE Yanlin, YANG Xiaofei, LIU Yang, et al. Radioactive ion beam physics related to HIAF[J]. *Scientia Sinica (Physica, Mechanica & Astronomica)*, 2020, 50(11): 112003. DOI: 10.1360/SSPMA-2020-0282.
- [2] BAI Shiwei, YANG Xiaofei. Study of nuclear structure by the measurement of the ground state properties of unstable nuclei[J]. *Nuclear Physics Review*, 2018, 35(4): 382-389. DOI: 10.11804/NuclPhysRev.35.04.382.
- [3] YANG X F, WANG S J, Wilkins S G, et al. Laser spectroscopy for the study of exotic nuclei[J]. *Progress in Particle and Nuclear Physics*, 2023, 129: 104005. DOI: 10.1016/j.pnpnp.2022.104005.
- [4] ZHOU L, WANG S M, FANG D Q, et al. Recent progress in two-proton radioactivity[J]. *Nuclear Science and Techniques*, 2022, 33(8): 105. DOI: 10.1007/s41365-022-01005-8.
- [5] BERGE M J G, JONSON B. ISOLDE past, present and future[J]. *Journal of Physics G: Nuclear and Particle Physics*, 2017, 44(4): 044011. DOI: 10.1088/1361-6471/aa5f03.
- [6] YANO Y, MOTOBAYASHI T. Radioactive isotope beam factory at RIKEN (RIBF)[J]. *Nuclear Physics News*, 2007, 17(4): 5-10. DOI: 10.1088/10506890701750368.
- [7] GLASMACHER T, SHERRILL B, NAZAREWICZ W, et al. Facility for rare isotope beams update for Nuclear Physics News[J]. *Nuclear Physics News*, 2017, 27(2): 28-33. DOI: 10.1317176.
- [8] ESCHKE J. International facility for antiproton and ion research (FAIR) at GSI, Darmstadt[J]. *Journal of Physics G: Nuclear and Particle Physics*, 2005, 31(6): S967-S973. DOI: 10.1088/0954-3899/31/6/041.
- [9] YANG Y, SUN L T, ZHAI Y H, et al. Heavy ion accelerator facility front end design and commissioning[J]. *Physical Review Accelerators and Beams*, 2019, 22(11): 110101. DOI: 10.1103/physrevaccelbeams.22.110101.
- [10] LIU Yongchao, BAI Shiwei, YANG Xiaofei. Development and prospect of precision laser spectroscopy techniques for nuclear physics study[J]. *Nuclear Physics Review*, 2019, 36(2): 161-169. DOI: 10.11804/NuclPhysRev.36.02.161.

- [11] CAMPBELL P, MOORE I D, PEARSON M R. Laser spectroscopy for nuclear structure physics[J]. Progress in Particle and Nuclear Physics, 2016, 86: 127-180. DOI: 10.1016/j.pnpnp.2015.09.003.
- [12] YANG X F, COLLAPS and CRIS collaboration. Precision laser spectroscopy technique for exotic radioactive beams at CERN-ISOLDE[J]. Journal of Physics: Conference Series, 2018, 1024: 012031. DOI: 10.1088/1742-6596/1024/1/012031.
- [13] NEUGART R, BILLOWES J, BISSELL M L, et al. Collinear laser spectroscopy at ISOLDE: new methods and highlights[J]. Journal of Physics G: Nuclear and Particle Physics, 2017, 44(6): 064002. DOI: 10.1088/1361-6471/aa6642.
- [14] ZHANG Peng, LIU Yinshen, BAI Shiwei, et al. Progress on the development of the collinear laser spectroscopy setup for the study of unstable nuclei[J]. Chinese Science Bulletin, 2023, 68(9): 1054-1065. DOI: 10.1360/TB-2022-1116.
- [15] FEDOSSEEV V N, KUDRYAVTSEV Y, MISHIN V I. Resonance laser ionization of atoms for nuclear physics[J]. Physica Scripta, 2012, 85(5): 058104. DOI: 10.1088/0031-8949/85/05/058104.
- [16] LI R H, MOSTAMAND M, ROMANS J, et al. Recent RILIS developments at the TRIUMF offline laser ion source test stand[J]. Hyperfine Interactions, 2020, 241(1): 22. DOI: 10.1007/s10751-020-1694-4.
- [17] DE GROOTE R P, BILLOWES J, BINNERSLEY C L, et al. Dipole and quadrupole moments of $^{73-78}\text{Cu}$ as a test of the robustness of the $Z=28$ shell closure near ^{78}Ni [J]. Physical Review C, 2017, 96(4): 041302. DOI: 10.1103/physrevc.96.041302.
- [18] KOSZORÚS Á, YANG X F, JIANG W G, et al. Charge radii of exotic potassium isotopes challenge nuclear theory and the magic character of $N = 32$ [J]. Nature Physics, 2021, 17(4): 439-443. DOI: 10.1038/s41567-020-01136-5.
- [19] GARCIA RUIZ R F, BERGER R, BILLOWES J, et al. Spectroscopy of short-lived radioactive molecules[J]. Nature, 2020, 581(7809): 396-400. DOI: 10.1038/s41586-020-2299-4.
- [20] UDRESCU S M, BRINSON A J, RUIZ R F G, et al. Isotope shifts of radium monofluoride molecules[J]. Physical Review Letters, 2021, 127(3): 033001. DOI: 10.1103/physrevlett.127.033001.
- [21] NAUBEREIT P, MARÍN-SÁEZ J, SCHNEIDER F, et al. Resonance ionization spectroscopy of sodium Rydberg levels using difference frequency generation of high-repetition-rate pulsed Ti:sapphire lasers[J]. Physical Review A, 2016, 93(5): 052518. DOI: 10.1103/physreva.93.052518.
- [22] RAEDER S, ACKERMANN D, BACKE H, et al. Probing sizes and shapes of nobelium isotopes by laser spectroscopy[J]. Physical Review Letters, 2018, 120(23): 232503. DOI: 10.1103/physrevlett.120.232503.

- [23] ALKHAZOV G D, BATIST L K, BYKOV A A, et al. Application of a high efficiency selective laser ion source at the IRIS facility[J]. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 1991, 306(1-2): 400-402. DOI: 10.1016/0168-9002(91)90348-T.
- [24] MISHIN V I, FEDOSEYEV V N, KLUGE H J, et al. Chemically selective laser ion-source for the CERN-ISOLDE on-line mass separator facility[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 1993, 73(4): 550-560. DOI: 10.1016/0168-583X(93)95839-W.
- [25] MARSH B A, ANDEL B, ANDREYEV A N, et al. New developments of the in-source spectroscopy method at RILIS/ISOLDE[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2013, 317: 550-556. DOI: 10.1016/j.nimb.2013.07.070.
- [26] MARSH B A, DAY GOODACRE T, SELS S, et al. Characterization of the shape-staggering effect in mercury nuclei[J]. Nature Physics, 2018, 14(12): 1163-1167. DOI: 10.1038/s41567-018-0292-8.
- [27] REPONEN M, DE GROOTE R P, AL AYOUBI L, et al. Evidence of a sudden increase in the nuclear size of proton-rich silver-96[J]. Nature Communications, 2021, 12: 4596. DOI: 10.1038/s41467-021-24888-x.
- [28] FEDOSSEEV V N, KUDRYAVTSEV Y, MISHIN V I. Resonance laser ionization of atoms for nuclear physics[J]. Physica Scripta, 2012, 85(5): 058104. DOI: 10.1088/0031-8949/85/05/058104.
- [29] ALKHAZOV G D, BARZAKH A E, DENISOV V P, et al. A new highly efficient method of atomic spectroscopy for nuclides far from stability[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 1992, 69(4): 517-520. DOI: 10.1016/0168-583X(92)95309-F.
- [30] KRON T, BEERWERTH R, RAEDER S, et al. Hyperfine structure study of 97,98,99Tc in a new laser ion source for high-resolution laser spectroscopy[J]. Physical Review C, 2020, 102(3): 034307. DOI: 10.1103/physrevc.102.034307.
- [31] STUDER D, ULRICH J, BRACCINI S, et al. High-resolution laser resonance ionization spectroscopy of 143-147Pm[J]. The European Physical Journal A, 2020, 56(2): 69. DOI: 10.1140/epja/s10050-020-00061-8.
- [32] POHJALAINEN I, MOORE I D, KRON T, et al. In-gas-cell laser ionization studies of plutonium isotopes at IGISOL[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2016, 376: 233-239. DOI: 10.1016/j.nimb.2016.02.019.
- [33] BLAUM K, GEPPERT C, KLUGE H J, et al. A novel scheme for a highly selective laser ion source[J]. Nuclear Instruments and Methods in Physics Re-

search Section B: Beam Interactions With Materials and Atoms, 2003, 204: 331-335. DOI: 10.1016/S0168-583X(02)01942-0.

[34] FINK D A, RICHTER S D, BLAUM K, et al. On-line implementation and first operation of the Laser Ion Source and Trap at ISOLDE/CERN[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2015, 344: 83-95. DOI: 10.1016/j.nimb.2014.12.007.

[35] VAN DUPPEN P, DENDOOVEN P, HUYSE M, et al. A laser ion source for on-line mass separation[J]. Hyperfine Interactions, 1992, 74(1): 193-204. DOI: 10.1007/BF02398629.

[36] QAMHIEH Z N, VANDEWEERT E, SILVERANS R E, et al. Resonance ionization in a gas cell: a feasibility study for a laser ion source[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 1992, 70(1-4): 131-136. DOI: 10.1016/0168-583X(92)95920-M.

[37] KUDRYAVTSEV Y, ANDRZEJEWSKI J, BIJNENS N, et al. Beams of short-lived nuclei produced by selective laser ionization in a gas cell[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 1996, 114(3-4): 350-365. DOI: 10.1016/0168-583X(96)00194-2.

[38] KUDRYAVTSEV Y, FACINA M, HUYSE M, et al. Beams of isotopes produced at LISOL by laser ionization after thermalization of energetic ions in a gas cell[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2003, 204: 336-342. DOI: 10.1016/S0168-583X(02)01946-8.

[39] VAN DUPPEN P. LISOL: Leuven Isotope Separator On-Line[J]. Nuclear Physics A, 1998, 626(1-2): 171c-180c. DOI: 10.1016/S0375-9474(97)00631-7.

[40] FERRER R, BARZAKH A, BASTIN B, et al. Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion[J]. Nature Communications, 2017, 8: 14520. DOI: 10.1038/ncomms14520.

[41] CHOI H, HIRAYAMA Y, CHOI S, et al. In-gas-cell laser ionization spectroscopy of 194,196Os isotopes by using a multireflection time-of-flight mass spectrograph[J]. Physical Review C, 2020, 102(3): 034309. DOI: 10.1103/physrevc.102.034309.

[42] MUKAI M, HIRAYAMA Y, WATANABE Y X, et al. In-gas-cell laser resonance ionization spectroscopy of 196,197,198Ir[J]. Physical Review C, 2020, 102(5): 054307. DOI: 10.1103/physrevc.102.054307.

[43] HIRAYAMA Y, MUKAI M, WATANABE Y X, et al. In-gas-jet laser ion source: resonance ionization spectroscopy of radioactive atoms in supersonic gas jets[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2013, 297: 7-22. DOI: 10.1016/j.nimb.2012.12.008.

- [44] ZADVORNAYA A, CREEMERS P, DOCKX K, et al. Characterization of supersonic gas jets for high-resolution laser ionization spectroscopy of heavy elements[J]. *Physical Review X*, 2018, 8(4): 041008. DOI: 10.1103/physrevx.8.041008.
- [45] ROMANS J, AJAYAKUMAR A, AUTHIER M, et al. First offline results from the S3 low-energy branch[J]. *Atoms*, 2022, 10(1): 21. DOI: 10.3390/atoms10010021.
- [46] LAUTH W, BACKE H, DAHLINGER M, et al. Resonance ionization spectroscopy in a buffer gas cell with radioactive decay detection, demonstrated using ^{208}Tl [J]. *Physical Review Letters*, 1992, 68(11): 1675–1678. DOI: 10.1103/physrevlett.68.1675.
- [47] BACKE H, LAUTH W, BLOCK M. Resonance ionization spectroscopy of nobelium[J]. *Nature*, 2016, 538(7626): 495–498. DOI: 10.1038/nature19345.
- [48] RAEDER S, BACKE H, BLOCK M, et al. Developments for resonance ionization laser spectroscopy of the heaviest elements at SHIP[J]. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms*, 2020, 463: 272–276. DOI: 10.1016/j.nimb.2019.05.016.
- [49] RAEDER S, BACKE H, BLOCK M, et al. Probing sizes and shapes of nobelium isotopes by laser spectroscopy[J]. *Physical Review Letters*, 2018, 120(23): 232503. DOI: 10.1103/physrevlett.120.232503.
- [50] BUDINČEVIĆ I, BILLOWES J, BISSELL M L, et al. Laser spectroscopy of francium isotopes at the borders of the region of reflection asymmetry[J]. *Physical Review C*, 2014, 90: 014317. DOI: 10.1103/physrevc.90.014317.
- [51] BUDINČEVIĆ I, BILLOWES J, BISSELL M L, et al. Measurement of the first ionization potential of nobelium[J]. *Physical Review Letters*, 2018, 120(26): 263003. DOI: 10.1103/physrevlett.120.263003.
- [52] BLOCK M, LAATIAOUI M, RAEDER S. Recent progress in laser spectroscopy of the actinides[J]. *Progress in Particle and Nuclear Physics*, 2021, 116: 103834. DOI: 10.1016/j.ppnp.2020.103834.
- [53] KUDRIAVTSEV Y A, LETOKHOV V S. Laser method of highly selective detection of rare radioactive isotopes through multistep photoionization of accelerated atoms[J]. *Applied Physics B*, 1982, 29(3): 219–221. DOI: 10.1007/BF00688671.
- [54] SCHULZ C, ARNOLD E, BORCHERS W, et al. Resonance ionization spectroscopy on a fast atomic ytterbium beam[J]. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 1991, 24(22): 4831–4844. DOI: 10.1088/0953-4075/24/22/020.
- [55] NIEMINEN A, HUIKARI J, JOKINEN A, et al. Beam cooler for low-energy radioactive ions[J]. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 2001,

469(2): 244-253. DOI: 10.1016/S0168-9002(00)00763-2.

[56] CAMPBELL P, NIEMINEN A, BILLOWES J, et al. First results from laser spectroscopy on bunched radioactive beams from the JYFL ion-beam cooler[J]. European Physical Journal A, 2002, 15: 45-48. DOI: 10.1140/epja/i2001-10223-y.

[57] FLANAGAN K T. Recent advances in laser spectroscopy at ISOLDE[J]. Acta Physica Polonica B, 2013, 44(3): 627-636. DOI: 10.5506/aphyspolb.44.627.

[58] COCOLOS T E, AL SURADI H H, BILLOWES J, et al. The collinear resonance ionization spectroscopy (CRIS) experimental setup at CERN-ISOLDE[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2013, 317: 565-569. DOI: 10.1016/j.nimb.2013.05.088.

[59] MANÉ E, BILLOWES J, BLAUM K, et al. An ion cooler-buncher for high-sensitivity collinear laser spectroscopy at ISOLDE[J]. The European Physical Journal A, 2009, 42(3): 503-507. DOI: 10.1140/epja/i2009-10828-0.

[60] FLANAGAN K T, LYNCH K M, BILLOWES J, et al. Collinear resonance ionization spectroscopy of neutron-deficient francium isotopes[J]. Physical Review Letters, 2013, 111(21): 212501. DOI: 10.1103/physrevlett.111.212501.

[61] DE GROOTE R P, BUDINČEVIĆ I, BILLOWES J, et al. Use of a continuous wave laser and pockels cell for sensitive high-resolution collinear resonance ionization spectroscopy[J]. Physical Review Letters, 2015, 115(13): 132501. DOI: 10.1103/physrevlett.115.132501.

[62] DE GROOTE R P, BUDINČEVIĆ I, BILLOWES J, et al. Laser spectroscopy of francium isotopes at the borders of the region of reflection asymmetry[J]. Physical Review C, 2014, 90: 014317. DOI: 10.1103/physrevc.90.014317.

[63] KOSZORÚS Á, YANG X F, BILLOWES J, et al. Precision measurements of the charge radii of potassium isotopes[J]. Physical Review C, 2019, 100(3): 034304. DOI: 10.1103/physrevc.100.034304.

[64] BAI S W, KOSZORÚS Á, HU B S, et al. Electromagnetic moments of scandium isotopes and $N = 28$ isotones in the distinctive $0f_{7/2}$ orbit[J]. Physics Letters B, 2022, 829: 137064. DOI: 10.1016/j.physletb.2022.137064.

[65] FAROOQ-SMITH G J, VERNON A R, BILLOWES J, et al. Probing the ^{31}Ga ground-state properties in the region near $Z=28$ with high-resolution laser spectroscopy[J]. Physical Review C, 2017, 96(4): 044324. DOI: 10.1103/physrevc.96.044324.

[66] VERNON A R, GARCIA RUIZ R F, MIYAGI T, et al. Nuclear moments of indium isotopes reveal abrupt change at magic number 82[J]. Nature, 2022, 607(7918): 260-265. DOI: 10.1038/s41586-022-04818-7.

[67] GUSTAFSSON F P, RICKETTS C M, REITSMA M L, et al. Tin resonance-ionization schemes for atomic- and nuclear-structure studies[J]. Physical Review

A, 2020, 102(5): 052812. DOI: 10.1103/physreva.102.052812.

[68] VERNON A R, GARCIA RUIZ R F, BINNERSLEY C L, et al. Laser spectroscopy studies of the nuclear structure of neutron-rich radium[J]. Physical Review C, 2018, 97(2): 024309. DOI: 10.1103/physrevc.97.024309.

[69] ZHANG T J, CUI B Q, LV Y L. BRIF: from the first proton beam to RIB production[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2020, 463: 123–127. DOI: 10.1016/j.nimb.2019.06.045.

[70] WANG S J, YANG X F, BAI S W, et al. Construction and commissioning of the collinear laser spectroscopy system at BRIF[J]. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2022, 1032: 166622. DOI: 10.1016/j.nima.2022.166622.

[71] BAI S W, YANG X F, WANG S J, et al. Commissioning of a high-resolution collinear laser spectroscopy apparatus with a laser ablation ion source[J]. Nuclear Science and Techniques, 2022, 33(1): 9. DOI: 10.1007/s41365-022-00870-7.

[72] ZHANG P, LIU Y S, BAI S W, et al. Development of a data acquisition system for collinear laser spectroscopy experiments[J]. Nuclear Science and Techniques, 2023, 34(3): 38. DOI: 10.1007/s41365-023-01197-0.

[73] ZHANG P, HU H R, YANG X F, et al. Progress in the development of a collinear resonance ionisation laser spectroscopy setup[J]. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions With Materials and Atoms, 2023, 541: 37–41. DOI: 10.1016/j.nimb.2023.04.020.

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