Developments and Opportunities in Precision Laser Nuclear Spectroscopy for Unstable Nuclei Research

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Abstract

The basic properties of unstable nuclei constitute an important basis for investigating exotic nuclear structures. Precision laser nuclear spectroscopy technology, grounded in multi-disciplinary integration, enables the extraction of fundamental nuclear properties—including nuclear spin, magnetic moment, electric quadrupole moment, and charge radius—through measurements of hyperfine structure spectra of atoms, ions, or molecules, and currently plays a vital role in exotic structure studies of unstable nuclei across all mass regions of the nuclear chart. This article begins with the historical development of hyperfine structure spectroscopy research, expounding upon the fundamental principles and measurement methodologies of precision laser nuclear spectroscopy. Building upon this foundation, the article briefly illustrates the unique advantages of laser spectroscopic methods in nuclear structure research, exemplified by structural studies of unstable nuclei on the proton-rich side of the Pb (= 82) region. Additionally, this article presents the current status and recent advances in domestic collinear laser spectroscopy and collinear resonance ionization spectroscopy technologies. Finally, it outlines the promising prospects for laser nuclear spectroscopy applications at future domestic large-scale nuclear physics facilities, in areas ranging from unstable nuclear properties and structures to fundamental symmetry studies based on radioactive molecular spectroscopy.

Full Text

Preamble

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Progress and Opportunities of Precision Laser Spectroscopy for the Study of Unstable Nuclei

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Abstract

The basic properties of unstable nuclei serve as crucial foundations for investigating exotic nuclear structures. Precision laser spectroscopy, as an interdisciplinary technique, enables the extraction of fundamental nuclear properties—including spin, magnetic moment, electric quadrupole moment, and charge radius—by measuring the hyperfine structure spectra of atoms, ions, or molecules. This approach currently plays a vital role in studying exotic structures across various mass regions of the nuclear chart. This paper begins with the historical development of hyperfine structure spectroscopy and elucidates the fundamental principles and measurement methods of precision laser spectroscopy. Using the structure studies of proton-rich unstable nuclei in the Pb (Z = 82) region as an example, we illustrate the unique advantages of laser spectroscopy in nuclear structure research. Furthermore, we review the recent development status and latest progress of collinear laser spectroscopy and collinear resonance ionization spectroscopy in China. Finally, we prospect the broad application prospects of laser spectroscopy at future domestic nuclear physics large-scale scientific facilities for studying unstable nuclear properties and structures, as well as for fundamental symmetry research based on radioactive molecular spectroscopy.

Keywords: hyperfine structure; laser spectroscopy; unstable nuclei; fluorescence detection; ion detection

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The atomic nucleus is a quantum complex many-body system governed by strong, weak, and electromagnetic interactions. With the development of radioactive beam facilities and isotope separation techniques, unstable nuclei far from the β-stability line have become accessible for exploration. Experiments have gradually revealed that these unstable nuclei exhibit novel exotic features and structures distinct from stable nuclei, continuously challenging existing nuclear structure models and promoting their innovative development. Since the discovery of halo nuclei by I. Tanihata et al. in the 1980s [**?**], numerous experimental and theoretical investigations have emerged, leading to concepts such as cluster structures, new magic numbers, shell evolution, and shape coexistence, making radioactive nuclear beam physics one of the frontier fields in contemporary nuclear physics research.

The basic properties of nuclei—including mass, spin, electromagnetic moments, and radii—are not only effective tools for studying nuclear structure but also sensitive probes for investigating nucleon-nucleon interactions, playing important roles in testing and developing nuclear theoretical models [**?**]. Various detection methods have been developed experimentally to measure these fundamental nuclear properties. For example, storage rings, Penning traps, and multi-reflection time-of-flight spectrometers can precisely measure nuclear masses [**?**]; electron scattering and muonic atom methods can determine charge radii; transfer reactions and decay experiments can provide nuclear spins [**?**]; and nuclear magnetic resonance and nuclear quadrupole resonance methods can measure magnetic and electric quadrupole moments [**?**]. Precision laser spectroscopy, as an interdisciplinary technique, simultaneously measures nuclear spin, magnetic moment, electric quadrupole moment, and charge radius, offering unique advantages particularly for unstable nuclei, especially extremely neutron- or proton-rich nuclei.

Laser spectroscopy extracts nuclear properties by measuring the hyperfine structure and isotope shifts of atoms, ions, or molecules [**?**]. This nuclear experimental technique emerged in the 1970s [**?**] and has been widely applied at radioactive beam facilities worldwide with the deepening of unstable nuclear research. At the turn of this century, the sensitivity of laser spectroscopy experiments was significantly enhanced by combining with radiofrequency quadrupole (RFQ) beam cooling and bunching technology [**?**], enabling high-precision measurements of nuclides with beam intensities as low as 10^{3-4} pps (particles per second). It has now become a standard terminal configuration at major international radioactive beam facilities, playing crucial roles in studying basic properties of unstable nuclei across all mass regions of the nuclear chart [**?**]. For instance, the rich deformation and shape coexistence phenomena in the heavy-mass Pb region were first discovered through laser spectroscopy and remain a hot topic in exotic nuclear structure research today [**?**]. Beyond fundamental research on nuclear properties and structures, laser spectroscopy also holds broad application prospects in interdisciplinary fields such as biomedicine [**?**, **?**].

Previously, no experience existed in using laser spectroscopy to study nuclear properties and structures at domestic radioactive beam facilities. In recent years,

the Peking University team has collaborated with several domestic institutions to develop fluorescence-detection-based collinear laser spectroscopy equipment, successfully applying it at the Beijing Radioactive Ion-beam Facility (BRIF) [**?**] and conducting China' s first online laser spectroscopy experiment on unstable nuclei [**?**]. Recently, important progress has also been made in the construction and offline testing of ion-detection-based collinear resonance ionization spectroscopy equipment [**?**]. This paper elaborates on the technical principles of laser spectroscopy methods, particularly collinear laser spectroscopy, starting from hyperfine structure. Using the exotic deformation of proton-rich nuclei in the heavy-mass Pb (Z = 82) region as an example, we illustrate the role and advantages of laser spectroscopy in exotic nuclear structure studies. We also introduce the development of domestic collinear laser spectroscopy equipment, including China' s first online laser spectroscopy experiment on unstable nuclei and recent progress in collinear resonance ionization spectroscopy instrumentation.

2.1 Discovery and Historical Study of Hyperfine Structure

Although physicists such as Michelson, Fabry, and Perot first observed the hyperfine structure of spectral lines in the late 19th century [**?**], a physical explanation for this phenomenon was lacking at the time. Regarding this splitting or broadening of fine structure spectral lines, some believed these splittings were isotope-related or that additional quantum numbers should be introduced for electrons. In 1924, Pauli first proposed the nuclear spin hypothesis to explain hyperfine structure phenomena, suggesting that the angular momentum of the nucleus itself could couple with extranuclear electron angular momentum to produce total angular momentum [**?**]. He also noted that analysis of spectral lines could enhance understanding of nuclear structure.

In 1927, Back and Goudsmit analyzed the hyperfine structure spectra of Bi ($Z = 83, A = 209$) in detail, concluding that the nuclear angular momentum of ^{209}Bi was $9/2\hbar$ [**?**]. After further studying the Paschen-Back effect, they confirmed Pauli' s view on nuclear spin and proposed that nuclei possess magnetic moments. In 1930, Fermi quantitatively calculated the contribution of magnetic dipole hyperfine interaction to hyperfine structure spectra [**?**]. The discovery of the neutron and measurement of proton magnetic moment in 1932 changed understanding of nuclear composition and structure. In 1935, Schüler and Schmidt found significant deviations between experimentally observed hyperfine structure splitting distances and theoretical calculations of magnetic dipole hyperfine interaction [**?**]. Based on this, Casimir first proposed the contribution of electric quadrupole hyperfine interaction to hyperfine structure spectra [**?**, **?**].

Today, the theoretical interpretation of hyperfine structure is well understood: nuclear electromagnetic properties affect extranuclear electron motion, causing energy levels of atoms, ions, and molecules to further shift and split based on fine-structure levels. Precisely because these shifts and splits originate from electromagnetic interactions between the nucleus and extranuclear electrons,

precise measurement of hyperfine structure spectra provides a unique pathway to study nuclear electromagnetic multipole moments.

2.2 Fundamental Principles of Laser Spectroscopy for Nuclear Physics

The hyperfine interaction between the nucleus and extranuclear electrons includes electric multipole interactions. Under symmetry requirements, magnetic dipole and electric quadrupole hyperfine interactions dominate the hyperfine splitting, while higher-order electromagnetic interaction contributions are generally negligible. The energy shift of each newly split hyperfine level relative to the original fine-structure level can be expressed as:

$$\Delta E = a\frac{K(K+1) - I(I+1)J(J+1)}{2(2I-1)(2J-1)IJ} + b\frac{\frac{3}{2}K(K+1) - 2I(I+1)J(J+1)}{2(2I-1)(2J-1)IJ}$$

where the total angular momentum quantum number of extranuclear electrons **J** couples with nuclear spin **I** to give quantum number **F**, with $K = F(F+1) - J(J+1) - I(I+1)$. The first term in equation (1) represents the magnetic dipole hyperfine interaction contribution, and the second term represents the electric quadrupole hyperfine interaction contribution. The magnetic dipole hyperfine structure constant a satisfies $a = \frac{\mu_I B_e}{IJ}$, while the electric quadrupole hyperfine structure constant b satisfies $b = eQ_S\langle\frac{\partial^2 V}{\partial z^2}\rangle$. Here, B_e is the magnetic field strength produced by electron motion at the nucleus, and $\langle\frac{\partial^2 V}{\partial z^2}\rangle$ is the electric field gradient at the nucleus. Thus, the hyperfine structure energy shift ΔE is directly related to nuclear spin I, magnetic moment μ_I, and electric quadrupole moment Q_S. Figure 1 [Figure 1: see original paper] specifically shows the hyperfine structure levels of the $6s^2{}_{1/2}^{2S}$ and $6p^2{}_{3/2}^{2P}$ energy levels for ^{197}Au ($I = 3/2$) extranuclear electrons, along with corresponding transitions and hyperfine structure spectra.

After experimentally measuring the hyperfine structure spectrum, the nuclear spin I can generally be easily determined directly from the number of spectral lines, their spacing, and relative intensities. Fitting and analyzing the measured hyperfine structure spectrum can extract the hyperfine structure constants A and B, as well as the transition frequency between original fine-structure levels (the central frequency) ν. For different isotopes of the same element (same Z, different N), the magnetic field B_e and electric field gradient produced by extranuclear electrons at the nucleus position can be approximately treated as constants in first-order approximation ignoring hyperfine structure anomalies. Using precisely measured magnetic moments (μ_{ref}) and electric quadrupole moments (Q_{ref}) of reference nuclei (generally stable or long-lived nuclei) via nuclear magnetic resonance or nuclear quadrupole resonance methods, the magnetic moment (μ_X) and electric quadrupole moment (Q_X) of the target nucleus can be extracted through relative measurement:

$$\mu_X = \mu_{\text{ref}} \frac{A_X I_X}{A_{\text{ref}} I_{\text{ref}}}$$

$$Q_X = Q_{\text{ref}} \frac{B_X}{B_{\text{ref}}}$$

For different isotopes of the same element, the central frequency ν also changes due to differences in mass and charge radius. The difference between central frequencies of two isotopes A and A' is called the isotope shift:

$$\delta\nu_{AA'} = \nu_{A'} - \nu_A = K_{\text{MS}} \frac{M_A - M_{A'}}{M_A M_{A'}} + F\delta\langle r^2\rangle_{AA'}$$

As shown in equation (4), the isotope shift consists of two main contributions: the first term is the mass shift, primarily caused by nuclear motion around the center of mass; the second term is the field shift, arising from the effect of nuclear charge distribution on extranuclear electron energy levels, which is directly related to nuclear charge radius. K_{MS} and F are the mass shift factor and field shift factor, respectively, which can be approximated as constants for different isotopes of the same element and are generally obtained through atomic theory calculations or calibration using existing charge radius data. Therefore, after experimentally measuring the isotope shift $\delta\nu_{AA'}$, the mean-square charge radius difference $\delta\langle r^2\rangle_{AA'}$ between two isotopes can be calculated based on equation (4) [**?**].

2.3 Laser Spectroscopy Experimental Methods for Nuclear Property Measurement

There are two fundamental principles for measuring hyperfine structure spectra using precision laser spectroscopy methods: one is laser-induced fluorescence detection, and the other is ion detection after multi-step laser resonance ionization [**?**]. Based on these two detection approaches, various types of laser spectroscopy techniques have been developed internationally, each with respective advantages in measurement sensitivity and resolution.

Laser-induced fluorescence detection technology uses a continuous narrow-band tunable laser to resonantly excite extranuclear electrons from the ground state (or metastable state), obtaining hyperfine structure spectra by recording fluorescence photons emitted during de-excitation [**?**]. The typical experimental technique employing laser-induced fluorescence detection is collinear laser spectroscopy, which has played important roles in studying unstable nuclear structures and properties across all mass regions of the nuclear chart since its inception in the latter half of the 20th century. Currently, over 80% of the approximately 1000 nuclides studied using laser spectroscopy have been measured with collinear laser spectroscopy [**?**], primarily due to its high-resolution

advantage. Collinear laser spectroscopy employs collinear or anti-collinear laser and fast ion/atom beams for laser-induced fluorescence detection. In such experiments, the beam energy spread mainly depends on the energy dispersion δE from the acceleration voltage of the ion source or RF cooler-buncher, typically a few eV, remaining approximately constant with increasing acceleration voltage. Collinear laser spectroscopy experiments generally use beam energies of several tens of keV (e.g., 30-60 keV). After acceleration, the Doppler broadening caused by energy dispersion δE satisfies the relation:

$$\Gamma_D = \nu_0 \sqrt{\frac{2\delta E}{E}}$$

where ν_0 is the laser frequency for resonant excitation of ions or atoms. As the ion beam energy E increases, the Doppler broadening Γ_D caused by energy dispersion δE is suppressed. Figure 2 [Figure 2: see original paper] qualitatively illustrates the principle and effect of Doppler broadening suppression using collinear laser spectroscopy. As shown, when the beam is accelerated to several tens of keV, the Doppler broadening caused by beam energy dispersion is about several tens of MHz, comparable to the natural linewidth from excited-state lifetime (energy uncertainty). In light nuclear regions, hyperfine structure splitting is relatively small, generally on the order of several tens to hundreds of MHz. Therefore, only when the total spectral linewidth (Doppler broadening plus natural broadening) in laser spectroscopy experiments is sufficiently small can the resonance peaks of various hyperfine level transitions be better distinguished (as shown in the right panel of Figure 2), enabling comprehensive measurement of nuclear spin, magnetic moment, electric quadrupole moment, and charge radius. The excellent Doppler broadening suppression capability of collinear laser spectroscopy gives it high-resolution advantages, making it applicable for property measurements of unstable nuclei across all mass regions.

At the turn of this century, the development of pulsed beams based on RFQ cooling and bunching technology significantly enhanced the detection sensitivity of collinear laser spectroscopy, enabling high-precision measurements of unstable nuclides with beam intensities as low as 10^{3-4} pps while maintaining high resolution [**?**]. Consequently, this technology has been deployed at radioactive beam facilities worldwide in recent years, such as COLLAPS (COLlinear LAser SPectroscopy) at CERN' s ISOLDE (Isotope Separator On-Line DEvice) facility [**?**], BECOLA (BEam COoler and LAser spectroscopy) at the National Superconducting Cyclotron Laboratory of Michigan State University [**?**], IGISOL (Ion Guide Isotope Separator On-Line) at the University of Jyväskylä [**?**], and CFBS (Collinear Fast Beam laser Spectroscopy) at TRIUMF' s ISAC (Isotope Separation and ACceleration) facility [**?**]. These collinear laser spectroscopy experimental terminals have played important roles in studying properties and exotic structures of unstable nuclei. To investigate nuclides further from the β-stability line in the nuclear chart, new collinear laser spectroscopy experimental devices are continuously being developed with a focus on further improving

sensitivity, such as MIRACLS (Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy) based on multi-reflection time-of-flight spectrometers under development at CERN' s ISOLDE facility [**?**].

The experimental method based on multi-step laser resonance ionization first uses the first-step laser to resonantly excite target atoms, then employs one or more additional lasers to ionize the atoms, obtaining hyperfine structure spectra by detecting the ionized ions. Some elements have suitable autoionizing states, allowing multi-step laser resonance excitation to autoionizing states for higher ionization efficiency. Alternatively, atoms can be resonantly excited to Rydberg states via multi-step lasers and subsequently ionized by a strong electric field [**?**].

In-source laser spectroscopy is a typical representative of the multi-step laser resonance ionization method. ISOL-type radioactive beam facilities produce diverse reaction types and products in their target-ion sources. In-source laser spectroscopy employs multi-step laser resonance ionization of target nuclide atoms within the target-ion source and collects the extracted ions to measure hyperfine structure spectra. This technique offers extremely high sensitivity, providing significant advantages for measuring low-yield radioactive nuclides, and has achieved measurements of nuclides with beam intensities below 1 pps, playing important roles in exotic nuclear structure studies particularly in the heavy-mass Pb ($Z = 82$) region [**?**]. However, due to intense thermal motion in the reaction cavity of the target-ion source (generally >2000 K), Doppler broadening caused by thermal motion results in relatively poor resolution (generally > GHz) for hyperfine structure spectra measured with in-source laser spectroscopy, making it difficult to determine target nuclear spin and electric quadrupole moment.

In recent years, collinear resonance ionization spectroscopy (CRIS) based on collinear laser spectroscopy has rapidly developed [**?**]. This technique suppresses Doppler broadening using fast pulsed ion beams collinear with multi-step pulsed lasers, while simultaneously employing resonance ionization for ion detection to avoid the large background issues of fluorescence detection, thereby achieving both high resolution and high sensitivity. Currently, this technique has achieved precise measurements of nuclides with beam intensities as low as 10^{1-2} pps while maintaining high resolution (several tens of MHz) [**?**, **?**].

3. Studies of Unstable Nuclei Using Laser Spectroscopy

As described above, precision laser spectroscopy methods have played important roles in studying basic properties and exotic structures of unstable nuclei, discovering new exotic phenomena across all mass regions of the nuclear chart. For example, in light nuclear regions, ^{11}Li and ^{11}Be are typical representatives of neutron halo nuclei, with laser spectroscopy measurements showing charge radii significantly smaller than matter radii, confirming the existence of halo structures [**?**]. In the island of inversion region near $N = 20$, laser spectroscopy

of Na, Mg, and Al isotopes successfully explored the mechanisms of intruder states and deformation [**?**]. In medium-mass regions, laser spectroscopy measurements of Cu and Zn isotopes successfully verified theoretical images of shell evolution driven by tensor forces [**?**] and first discovered shape coexistence phenomena near ^{78}Ni [**?**]. Below, we use the exotic structures of proton-rich nuclei in the Pb ($Z = 82$) region as an example to illustrate the contributions of laser spectroscopy to exotic nuclear structure research.

Laser spectroscopy measurements of charge radii and other properties of neutron-deficient nuclei in the Pb region reveal rich deformation and shape coexistence phenomena. The hallmark representative is the shape staggering phenomenon observed in Hg ($Z = 80$) isotopes. The green points in Figure 3 [Figure 3: see original paper] show experimentally measured mean-square charge radius differences for the Hg isotope chain. At neutron numbers $N = 105, 103, 101$, the nuclear charge radii show abrupt changes compared to neighboring even-even nuclei, indicating large deformation. This shape staggering phenomenon disappears in more proton-rich regions. Experimentally, this charge radius mutation was observed in the 1970s and termed shape staggering [**?**]. Only in recent years has the formation mechanism of this shape staggering phenomenon been successfully explained by Monte Carlo Shell Model (MCSM) calculations. The results show that the proton $\pi 1h_{9/2}$ orbital above the $Z = 82$ closed shell is depressed, forming an intruder state. In Hg isotopes with $A = 181, 183, 185$, protons undergo cross-shell excitation to fill the $\pi 1h_{9/2}$ intruder state, and with increasing proton cross-shell excitation, neutrons also occupy the $\nu 1i_{13/2}$ orbital more frequently, leading to large quadrupole deformation and shape staggering phenomena. Recent laser spectroscopy measurements also indicate similar shape staggering phenomena in nearby ^{188}Bi ($Z = 83$) at neutron number $N = 105$ (blue points in Figure 3) [**?**], while neighboring Pb isotopes ($Z = 82$) remain near-spherical in their ground states (red circles in Figure 3) [**?**].

In addition to shape staggering, shape coexistence is another exotic nuclear structure phenomenon in this region. Shape coexistence often occurs when protons (or neutrons) are at closed shells while neutrons (or protons) are at half-filled shells. In such cases, cross-shell excitation of particle-hole pairs is energetically enhanced, leading to deformed intruder states with low single-particle energy, i.e., isomeric states near the ground state. For example, laser spectroscopy measurements of charge radii for ground and isomeric states of ^{185}Hg ($N = 105$) show large differences, indicating shape coexistence in this nucleus.

Laser spectroscopy measurements of charge radii for Au ($Z = 79$) isotopes display completely different novel characteristics compared to neighboring Hg, Bi, and Pb isotopes, as shown by black points in Figure 3 [**?**]. In neutron-deficient Au isotopes, starting from $N = 108$, the ground-state charge radii show abrupt increases as neutron number decreases, exhibiting large deformation. This deformation maintains the same trend between $N = 101 - 107$, making this region known as the "island of deformation" in the Au isotope chain. This large deformation phenomenon disappears in ^{179}Au ($N = 100$), but both ground

and isomeric states of ^{178}Au ($N = 99$) show sudden charge radius increases, indicating obvious shape changes.

Currently, basic properties of nuclei in this region (particularly Au isotopes) have mainly been measured using in-source laser spectroscopy. While the high sensitivity of in-source laser spectroscopy is well-suited for measuring extremely low-yield unstable nuclei in heavy regions, its limited spectral resolution has left spin information undetermined for most nuclei and electric quadrupole moment information largely unknown. Therefore, to further investigate the formation mechanism of the Au isotope island of deformation, high-resolution collinear laser spectroscopy is needed to measure spins and electric quadrupole moments of neutron-deficient Au isotopes. Currently, a high-resolution collinear resonance ionization spectroscopy experimental program for measuring neutron-deficient Au isotopes at CERN' s ISOLDE radioactive beam facility has been approved [**?**]. In this experiment, we will utilize the high-sensitivity and high-resolution advantages of collinear resonance ionization spectroscopy to measure basic properties including spins and electric quadrupole moments of neutron-deficient Au isotopes.

4.1 Online Collinear Laser Spectroscopy Experiment Based on Fluorescence Detection

Although laser spectroscopy technology has been widely applied at radioactive beam facilities worldwide, no laser spectroscopy experimental terminal for unstable nuclear property studies existed previously at domestic radioactive beam facilities. In 2021, the Laser Spectroscopy and Nuclear Properties Group at Peking University collaborated with the China Institute of Atomic Energy to construct a fluorescence-detection-based collinear laser spectroscopy experimental terminal at the Beijing Radioactive Ion-beam Facility (BRIF) and successfully conducted China' s first online laser spectroscopy experiment on unstable nuclei [**?**].

The collinear laser spectroscopy experimental terminal at BRIF is shown in Figure 4 [Figure 4: see original paper]. Proton beams with energies of about 100 MeV produced by a cyclotron bombard a CaO target. Reaction products ^{38,39}K are ionized by a surface ion source, accelerated to 60 keV, and mass-separated. The ^{38,39}K ions enter the collinear laser spectroscopy beamline through an 80-degree electrostatic deflector, undergo beam optics shaping and Faraday cup diagnostics, then enter a charge exchange cell containing alkali metal vapor (K). In the charge exchange cell, ^{38,39}K ions undergo charge exchange reactions with K vapor and become neutralized. Meanwhile, voltage scanning electrodes upstream of the charge exchange cell can apply voltages within $±$2 kV range. Since the electrodes and charge exchange cell share the same potential, neutralized atoms maintain the same kinetic energy downstream of the beamline. Therefore, by changing the potential applied to the voltage scanning electrodes, the atom velocity can be varied, which changes the laser frequency experienced by the atoms through the Doppler effect while keeping the laser

frequency constant, enabling hyperfine structure acquisition through voltage scanning. Unneutralized ions are removed by deflection electrodes downstream of the charge exchange cell. Ground-state ^{38,39}K atoms enter the fluorescence detection region and are resonantly excited by a narrow-band continuous laser. Fluorescence photons emitted from excited atoms are detected by four photomultiplier tubes, with signals recorded by electronics and sent to the online data acquisition system. By recording resonance signals versus scanning voltage, the hyperfine structure of ^{38,39}K can be obtained. The right panel of Figure 4 shows the online measured hyperfine structure spectrum of ^{38}K. Conical aperture arrays installed before and after the fluorescence detection region mainly reduce laser scattering photons entering the detection region to lower background. A MagneTOF counter installed after the fluorescence detection region can monitor beam conditions in real-time during experiments.

Fitting the ^{38}K hyperfine structure spectrum in Figure 4 yielded electric dipole hyperfine structure constants and isotope shifts, from which magnetic moments and charge radii of ^{38,39}K were deduced, showing good agreement with literature values. The detection efficiency is also comparable to the advanced level of similar international facilities, completely validating the performance of this fluorescence-detection-based collinear laser spectroscopy terminal. However, since the beam used in this experiment was continuous with relatively large energy dispersion, both measurement sensitivity and resolution were affected. Currently, we have begun developing a radiofrequency quadrupole (RFQ) cooler-buncher, which can convert injected continuous beams into pulsed beams and reduce beam energy dispersion through buffer gas, thereby greatly suppressing background noise from laser scattering photons and reducing Doppler broadening from energy dispersion, significantly improving the overall sensitivity and resolution of the laser spectroscopy terminal.

4.2 Offline Testing of Collinear Resonance Ionization Spectroscopy Based on Ion Detection

To further improve the sensitivity of high-resolution laser spectroscopy, our group has collaborated with domestic partners to develop collinear resonance ionization spectroscopy equipment based on the existing fluorescence-detection collinear laser spectroscopy. Recently, important progress has been made in the construction and offline testing of collinear resonance ionization spectroscopy [**?**].

The developed collinear resonance ionization spectroscopy apparatus is shown in Figure 5 [Figure 5: see original paper]. The entire beamline adopts CF vacuum standards, achieving a high vacuum environment below 10^{-8} mbar. High-power pulsed laser-produced 532 nm laser bombards a solid target in the ion source, generating singly-charged positive ion pulses at 100 Hz frequency. The pulsed ion beam is extracted from the ion source, accelerated to 20 keV, and enters the collinear resonance ionization spectroscopy apparatus through a 90-degree electrostatic deflector. The beam optics and charge exchange cell structure are

essentially the same as in Figure 4. The main differences are: (1) In resonance ionization spectroscopy, the more common spectrum scanning method is directly changing the resonant laser frequency rather than voltage, with the charge exchange cell at zero potential; (2) The conical aperture arrays before and after the fluorescence detection region now serve differential pumping purposes, enabling an ultra-high vacuum environment of 10^{-9}-10^{-10} mbar in the interaction region. This ultra-high vacuum greatly reduces ion detection background from collisional ionization of atoms with residual gas. After neutralization in the charge exchange cell, atoms in ground or metastable states enter the interaction region, are resonantly excited by the first-step frequency-tunable pulsed laser, then ionized by the second (and third) step laser. Ionized ions are deflected by 40-degree electrostatic deflection into the ion detection region and collected by a MagneTOF ion detector. By recording the relationship between collected ion signals and the first-step laser frequency, the hyperfine structure spectrum of extranuclear electrons can be obtained.

Currently, we have begun offline testing of collinear resonance ionization spectroscopy for stable ^{64,66,68}Zn isotopes. The first test experiment adopted the two-step resonance ionization scheme shown in the right panel of Figure 6 [Figure 6: see original paper]. The first-step laser with continuously tunable wavelength at 280.1 nm is produced by frequency-tripling pulsed Ti:Sa laser output, corresponding to Zn atom transitions from $3P_2$ to $3D_1$, $3D_2$, and $3D_3$. The second-step 532 nm laser is produced by frequency-doubling pulsed Nd:YAG laser output. Both laser beams enter the beamline through quartz windows after the 40-degree deflection chamber, anti-collinear with the stable Zn atomic beam. The preliminary resonance ionization spectra for ^{64,66,68}Zn atom $3P_2 \rightarrow 3D_{1,2,3}$ transitions obtained in this test experiment are shown in Figure 6, with further optimization testing ongoing.

The pulsed Ti:Sa laser used in this measurement has a linewidth of about 2 GHz, resulting in limited spectral resolution. Currently, our group is developing narrow-band ($ $20 MHz) pulsed lasers based on seeded amplification technology [**?**], aiming to achieve high-resolution measurements in subsequent collinear resonance ionization spectroscopy tests.

5. Summary and Future Outlook

Laser spectroscopy technology can model-independently extract basic nuclear properties including spin, magnetic moment, electric quadrupole moment, and charge radius by measuring hyperfine structure spectra of atoms, ions, or molecules. This paper briefly reviewed the research history of hyperfine structure spectroscopy and introduced the technical principles and corresponding experimental detection methods of laser spectroscopy. Using shape staggering, shape coexistence, and islands of deformation in the neutron-deficient Pb region as examples, we illustrated the role of laser spectroscopy in exotic nuclear structure research and introduced future experimental plans using collinear resonance ionization spectroscopy to study neutron-deficient Au isotopes. We

then described the development of collinear laser spectroscopy equipment at domestic radioactive beam facilities through collaboration between Peking University' s Laser Spectroscopy and Nuclear Properties Group and domestic partners, including China' s first online laser spectroscopy experiment on unstable nuclei. Finally, we briefly introduced the development status of high-sensitivity collinear resonance ionization spectroscopy equipment in China and preliminary offline test results for stable Zn isotopes.

ISOL-type radioactive beam facilities can produce and extract not only radioactive ions but also radioactive molecules. Recently, laser spectroscopy of radioactive molecules ($^{223-228}$RaF) was first realized using collinear resonance ionization spectroscopy [**?**]. Measurement of electric dipole moments in low-energy regions may be an important approach to finding CP violation effects that could explain the matter-antimatter asymmetry in the universe. Theoretical studies indicate that radioactive molecules containing octupole-deformed heavy nuclei could improve electric dipole moment measurement sensitivity by 3-7 orders of magnitude [**?**]. Therefore, radioactive molecular spectroscopy is considered a key candidate system for studying CP symmetry violation and has been included in new nuclear physics long-range plans in the US and Europe [**?**]. Furthermore, radioactive molecular spectroscopy provides new opportunities for studying nuclear electromagnetic multipole moments, nuclear structure, and nuclear astrophysics, and will promote further development of atomic and nuclear theories [**?**].

In recent years, domestic radioactive beam facilities have entered a period of rapid development, with expectations to produce unstable nuclides closer to the drip lines, continuously expanding the accessible nuclear chart. The BRIF team at the Beijing Tandem Accelerator National Laboratory is developing uranium target technology that has achieved important progress, enabling production of some medium-mass unstable nuclides and expected to provide more measurable nuclides in medium- and heavy-mass regions. We will further optimize and upgrade the collinear laser spectroscopy terminal at BRIF, expanding it into a more sensitive collinear resonance ionization spectroscopy terminal combined with an RFQ cooler-buncher to significantly improve measurement sensitivity and resolution. This collinear resonance ionization spectroscopy terminal is expected to be applied to basic property and structure studies of a broader range of unstable nuclei at BRIF. The High Intensity heavy-ion Accelerator Facility (HIAF) in Huizhou, Guangdong, is expected to be completed in the coming years [**?**]. The collinear resonance ionization spectrometer we are testing will also be combined with HIAF' s low-energy nuclear structure spectrometer terminal to conduct property and structure studies of heavy and superheavy nuclei, and to pioneer spectroscopic studies of radioactive molecules.

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Note: Figure translations are in progress. See original paper for figures.