采用激光烧蚀离子源的高分辨率共线激光光谱装置的调试

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Abstract

Collinear laser spectroscopy is known as one of the powerful tools for the study of nuclear spins, electromagnetic moments and charge radii of the exotic nuclei. Aiming at studying these nuclear properties of unstable nuclei at the Beijing Radioactive Ion-beam Facility (BRIF) and the future High Intensity Heavy-ion Accelerator Facility (HIAF), we have firstly developed a collinear laser spectroscopy apparatus integrated with an offline laser ablation ion source and a laser system. The overall performances of this state-of-the-art technique and device have been commissioned by using the bunched stable ion beam. High-resolution optical spectra of ^{40,42,44,48}Ca isotopes were successfully measured for the $4s\ ^2S_{1/2}$ $ rightarrow$ $4p\ ^2P_{3/2}$ (D2) ionic transition and the extracted isotope shifts relative to the ^{40}Ca show an excellent agreement with the literature values. This system is now ready to be applied at the ratioactive ion beam facility, such as BRIF, and has paved the way for further development of the higher-sensitivity collinear resonant ionization spectroscopy.

Full Text

Commissioning of a High-Resolution Collinear Laser Spectroscopy Apparatus Using a Laser Ablation Ion Source

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Collinear laser spectroscopy is recognized as one of the most powerful tools for investigating nuclear spins, electromagnetic moments, and charge radii of exotic nuclei. With the goal of studying these nuclear properties of unstable nuclei at the Beijing Radioactive Ion-beam Facility (BRIF) and the future High Intensity Heavy-ion Accelerator Facility (HIAF), we have developed a collinear laser spectroscopy apparatus integrated with an offline laser ablation ion source and a laser system. The overall performance of this state-of-the-art technique and device has been commissioned using bunched stable ion beams. High-resolution optical spectra of 40,42,44,48Ca isotopes were successfully measured for the 4s $^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ (D_2) ionic transition, and the extracted isotope shifts relative to 40Ca show excellent agreement with literature values. This system is now ready for application at radioactive ion beam facilities such as BRIF and has paved the way for further development of higher-sensitivity collinear resonant ionization spectroscopy.

Keywords: Nuclear properties, Collinear laser spectroscopy, Laser-ablation ion source, Photon detection, Isotope shift

Introduction

Understanding the evolution of nuclear structure in short-lived exotic nuclei toward the proton and neutron driplines represents one of the central themes in contemporary nuclear physics research, driving continuous developments in both experimental techniques and theoretical approaches [1, 2]. The static properties of ground and isomeric states of unstable nuclei are indispensable for studying exotic nuclear structure [3–5] and provide stringent tests of various nuclear models [6, 7]. Collinear laser spectroscopy (CLS) has proven to be a powerful tool for accessing multiple nuclear properties of ground and isomeric states of exotic nuclei [8, 9]. This is achieved by probing the hyperfine structure (hfs) and isotope shift resulting from the interaction between the atomic nucleus and surrounding electrons, which allows precise extraction of nuclear spins (I), magnetic dipole moments ( ), electric quadrupole moments (Qs), and changes in mean-square charge radii ($\delta\ r^2$) of an isotopic chain in a nuclear model-independent manner.

Motivated by the observation of unexpected nuclear phenomena in short-lived exotic nuclei, existing radioactive ion beam (RIB) facilities are being upgraded and next-generation facilities are being developed to produce more exotic radioactive beams. Meanwhile, considerable efforts have been made to continuously enhance the experimental sensitivity and precision of the CLS method [10, 11], facilitating ongoing studies of exotic nuclei. To date, this experimental technique has been established at various RIB facilities worldwide, including ISOLDE/CERN [12], IGISOL/JYFL [13], ISAC/TRIUMF [14], NSCL/MSU [15], and ALTO [16], yielding major contributions to nuclear structure studies and providing important benchmarks for the development of state-of-the-art nuclear theory [7, 17–19].

Two operational RIB facilities are available in China: HIRFL (PF-type) at IMP, Lanzhou [20] and BRIF (ISOL-type) at CIAE, Beijing [21], which have played significant roles in nuclear physics research [20–22]. To gain access to more rare isotopes, two next-generation RIB facilities—the High Intensity Heavy Ion Accelerator Facility (HIAF) [23] and the Beijing Isotope-Separation-On-Line neutron-rich beam facility (BISOL) [24]—are under construction and planned, respectively, offering new opportunities for nuclear physics studies in the near future. However, the well-established CLS technique has not yet been implemented at these domestic RIB facilities. Therefore, to fully exploit the short-lived isotopes available at these facilities, we have, as a first stage, developed a CLS device combined with an offline laser ablation ion source to fully master the laser spectroscopy technique for nuclear property measurements. This integrated system also paves the way for further development of high-sensitivity collinear resonance ionization spectroscopy, enabling exploration of more exotic cases.

Here, we present the details of the newly developed CLS apparatus, including the laser ablation ion source with up to 30 keV high-voltage platform, the beamline with photon detection system, the laser system, and the data acquisition system. Thanks to the ion bunches (with approximately 10 µs temporal length) provided by the laser ablation ion source, the first commissioning experiment was successfully performed to probe the 4s $^2S_1/_2 \rightarrow$ 4p $^2P_3/_2$ (D_2) ionic transition of calcium. High-resolution optical spectra of four stable 40,42,44,48Ca isotopes were measured, achieving a typical linewidth of 55 MHz, comparable to well-established standard CLS setups worldwide [6, 15, 25]. Isotope shifts (δ^{40},) of 42,44,48Ca isotopes relative to the 40Ca reference isotope were extracted, showing excellent agreement with literature values [6, 26, 27].

II. Collinear Laser Spectroscopy System

Figure 1 presents a detailed sketch of the CLS apparatus, laser ablation ion source, and laser system. The entire setup is constructed according to the ConFlat (CF) standard for high vacuum conditions, currently achieving 10^{-8} mbar, permitting subsequent upgrade to a high-sensitivity collinear resonance ionization spectroscopy setup. With the current system, the hfs spectra of stable ion beams produced by the ion source can be measured with high resolution. In brief, the bunched ion beam extracted from the laser ablation target (solid material) is accelerated to up to 30 keV. After a 90° electrostatic deflection, the ion bunch is delivered into the CLS beamline, where it is anti-collinearly overlapped with a continuous-wave (cw) laser beam. The velocity of the ions can be tuned by applying a scanning voltage to an electrode in the interaction region. The fluorescence photons emitted from the laser-excited ions are collected and recorded by the photon detection system and data acquisition system as a function of the tuning voltage. When probing a transition from neutral atoms, a charge exchange cell (CEC, see Fig. 1) is required, and the scanning voltage is applied to an electrode upstream of the CEC. This component will not be discussed further

here, as we focus primarily on the Ca ion beam measurement. The functional details of the individual components are introduced in the following sections.

A. Laser Ablation Ion Source

The laser ablation ion source is developed to produce stable ion beams of a wide range of elements with energies up to 30 keV [25, 28]. Using a pulsed-laser ablation process, bunched ion beams with a typical temporal length of about 10 µs, high ion intensity, and low energy spread can be generated. These features of the ion beam are suitable for optimization and commissioning of the CLS system and future resonance ionization spectroscopy, for developing laser excitation and ionization schemes [28], and for measuring atomic hfs parameters of stable isotopes [29].

The inner structure of the ion source is shown schematically in Fig. 1 (left bottom inset). A 532-nm Nd:YAG laser (Litron TRLi 250-100), operated at 100 Hz repetition rate with a pulse width of 8 ns, is employed for ablation of the solid target. The target holder, floated at the acceleration (platform) potential (U_0: up to 30 keV), is tilted at 45° with respect to the laser beam and ion beam. The pulsed laser beam is focused onto the target material with a diameter of approximately 1 mm. The ions generated at the ablation area are extracted and refocused with a multiple-step extraction system, namely the first extraction electrode (Ext.1), the first einzel lens (L1), and the second extraction electrode (Ext.2). A negative potential (up to 6 keV) relative to the platform potential U_0 can be applied independently to these electrodes.

The extracted ions are then transported to the third extraction electrode (Ext.3) at ground potential and are simultaneously accelerated to a final energy of up to 30 keV. Before being directed into the CLS beamline by a pair of 90° bender plates, the accelerated ion beam trajectory can be further corrected by a group of horizontal and vertical steerers and by the second einzel lens (L2).

Figure 2 shows the schematic diagram of the high-voltage system applied for the laser ablation ion source, which can be operated at 0–30 keV. As mentioned above, the voltages applied to the electrodes (Ext.1, L1, and Ext.2) are floated on top of the acceleration potential U_0. Therefore, as shown in Fig. 2, a high-voltage (HV) platform and HV cage (indicated with dashed line) are built following safety guidelines. The acceleration potential U_0 is provided by a high-precision DC power supply (Heinzinger PNChp 40000-15pos) with a ripple of $<0.001\%$ and long-term stability of $<0.001\%$ over 8 hours. This U_0 potential is directly applied to the first CF 6-Way cross chamber (light red part in Fig. 2) of the ion source, which is fully isolated from the second 6-Way cross at ground potential (light green part in Fig. 2) by a CF-flanged vacuum ceramic break. A multichannel HV power supply module (iseg EHS 80-60n) installed in an HV crate (iseg ECH224), marked as power supply B, is used to control the negative potential of the three electrodes (Ext.1, L1, and Ext.2) inside the first 6-Way Cross. Power supply B is remotely controlled outside the HV cage via an optical

link (PEAK-System Technik, PCAN-LWL) for HV isolation and a USB adapter (PEAK-System Technik, PCAN-USB). An isolation transformer (50 kV) is used to isolate the high voltage (U_0) from ground potential and provide 220 VAC to power supply B. To determine the ion beam energy for hfs measurement, the acceleration potential U_0 is recorded in real time using a Keysight 34470A multimeter combined with a 1:1000 voltage divider (Ohm-labs, KV-30A).

Ion Beam Transport

The ion bunches are transported through the CLS beamline (approximately 4 meters long) using a series of electrostatic optical elements, including two sets of x-y steerers and two quadrupole triplet (QT1 and QT2) lenses, as shown in Fig. 1. The x-y steerer electrodes align the ion beam with the central axis of the beamline, while QT lenses control the beam spot. Ion beam diagnostic components, consisting of three Faraday cups (FC1, FC2-m, and FC3) with secondary-electron suppressors, one iris diaphragm, and one ion detector (ETP, 14924 MagneTOF Mini), monitor the beam position, intensity, and size. The iris diaphragm with a tunable diameter of 0–20 mm is placed in front of FC2-m (a multi-channel FC) to align the central axes of the ion and laser beams and can also serve as a simple annular FC to measure the ion beam current and size [30]. The FC ion beam current is recorded by a Keithley 6485 picoammeter. The potentials applied to the ion optics are provided by $±$6 kV HV modules combined with an HV crate (iseg ECH238). Using a program written in Python, the output voltage of the HV power supplies can be remotely controlled. The program is also used for visualizing the ion beam current, enabling remote optimization of ion beam transmission through the CLS beamline. The MagneTOF ion detector installed at the end of the beamline assesses the intensity and time-of-flight (TOF) of weak ion beams.

C. Photon Detection System

Figure 3 presents the schematic diagram of the photon detection system. The system consists of a 5-way-cross interaction chamber where the laser beam interacts with the atom/ion beam, and two identical detection units installed perpendicular to the CLS beamline. To suppress environmental photon background (i.e., scattered laser light), the interaction chamber is equipped with two dismountable arrays of tapered apertures, each having an inner diameter of 10 mm, as indicated in Fig. 3(a). The internal surface of the vacuum chamber is black-coated to further reduce background from laser scattering. A quartz window (VPZL-600 DU, Kurt J. Lester) with >90% transmission for a wide spectral range of 200–1200 nm isolates the detection units from the high-vacuum interaction region at 10^{-8} mbar. Note that an isolated electrode tube is mounted along the central axis of the interaction chamber, with two open rectangular holes in the direction of the detection unit for fluorescence photon transmission. To maintain a uniform field distribution inside the electrode tube (interaction region), the holes are covered with a metallic mesh with transmission efficiency

higher than 90%. By applying a scanning voltage to this electrode, the velocity of the ions (not applicable to neutral atoms) can be tuned to match the Doppler-shifted laser frequency for specific resonant excitation (see Sec. III for details).

As shown in Fig. 3, each detection unit is composed of two aspheric lenses (forming a telescope) and a photomultiplier tube (PMT). These lenses, with a diameter of 100 mm (N-BK7), have a transmittivity of 90% over a wide wavelength range of 350–1000 nm and guide the laser-induced fluorescence photons onto the sensitive area of the PMT. A PMT (R943-02, Hamamatsu) assembled with a socket (E2762-506) is employed to record the light. This PMT features an ultra-low dark count rate (typically 20 s^{-1} at –20°C) and wide spectral response (160–930 nm), but with a moderate sensitive area of 10 × 10 mm^2. The quantum efficiency of this PMT at 394 nm is 20%. Using two detection units, a total geometrical efficiency for fluorescence photons is simulated to be 15% [32]. The distance between the lens and PMT is adjustable by changing the thickness of the spacer (e.g., 27.3 mm for a wavelength of 394 nm) to ensure optimal geometrical efficiency for various wavelengths associated with specific ionic/atomic transitions.

To achieve a low dark count rate (dozens of counts per second), the R943-02 PMT must be maintained at a temperature of –20 to –30°C. To cool the GaAs(Cs) photocathode, the PMT head is covered by a spiral copper tube (copper heat exchanger in Fig. 3) through which ethanol is circulated by a refrigerant circulator (ECO RE 630S, LAUDA). To prevent frost/ice formation on the PMT glass window resulting from the low-temperature environment, dry nitrogen gas is circulated through the system for several minutes before cooling. The aforementioned spacer, made of PVC, also provides thermal isolation between the PMT and lens, effectively preventing lens damage due to thermal expansion and contraction. Based on these measures, the achieved typical background rate is about 1 kHz for a 1.2-mW laser beam (6 mm diameter). The entire detection unit (including the PMT) has proven robust and stable after frequent assembly and disassembly during commissioning.

D. Laser System

The laser system used for this CLS setup is shown in the top right inset of Fig. 1. The continuous-wave (cw) titanium-sapphire (Ti:Sa) laser (Matisse 2TS, Sirah Lasertechnik) is pumped by a 20 W 532 nm laser (Millennia, Spectra-Physics), which can also be converted into a dye system via an Exchange Kit (TIDYECW, Sirah Lasertechnik). This system provides a laser beam with a wavelength range of 650–1020 nm, which can be further frequency-doubled with the Wavetrain 2 (Sirah Lasertechnik), generating second-harmonic light covering the wavelength range of 325–500 nm. The Matisse cavity can be actively stabilized by the equipped reference cell, achieving a narrow linewidth of <50 kHz.

A high-precision wavelength meter (HighFinesse WS8-2) measures the real-time

frequency of the fundamental cw light. The wavelength meter can also perform long-term stabilization of the Matisse cavity through a built-in digital interface. To address drift of the wavelength meter (about several MHz per day) caused by temperature and pressure fluctuations, a commercial saturation absorption spectroscopy unit is introduced. This unit consists of a tunable diode laser (DLPRO780, TOPTICA Photonics AG), a temperature-controlled vapor cell filled with K, Rb, and Cs (COSY), and control and locking electronics. The frequency of the diode laser is locked to one of the hyperfine components of the available alkali atoms (e.g., ^{87}Rb). The frequency-locked diode laser is then used to calibrate the wavelength meter when it functions for long-term stabilization of the Matisse [33], or to correct the drift of the wavelength meter by recording both frequency-scanned Matisse laser and frequency-locked diode laser [34].

E. Electronics and Data Acquisition System

Two signals from the PMTs are amplified by a fast-timing amplifier (ORTEC FTA820A) and then discriminated by a constant fraction discriminator (CFD, CAEN model N605). After conversion to TTL logic signals, they are sent to different channels of a ChronoLogic TimeTagger4-2G time-to-digital converter (TDC) with 500 ps time resolution. Each event labeled by a timestamp can be tracked to obtain the TOF spectrum of the ion bunch. A master TTL signal with 100 Hz repetition rate (a period of 10 ms), labeled as $T_0 = 0$ µs and generated by a Quantum Composers 9528 (QC9528) digital-delay pulse generator, is used to externally trigger the 532-nm Nd:YAG laser. The laser pulse used for the ablation ion source arrives 490 µs later than T_0. Therefore, after accounting for the flight time of the ion bunch from the ion source to the detection region, the start time (trigger) and time window for the TDC are set to be $T_1 = T_0 + 498$ µs and $\Delta T = 100$ µs, respectively. This time window covers the period when the bunched beam traverses the photon detection region. A narrow time gate of 10 µs, corresponding to the width of the ion bunch, can be further applied to reduce background counts during offline analysis (more discussion in Sec. III).

The optical hfs spectrum of an isotope is measured by applying a scanned voltage (ΔU) to the electrode tube in the interaction detection region, while the laser frequency is fixed and stabilized with the wavelength meter. A scanned voltage between –1 keV and +1 keV can be provided by a DC amplifier (Kepco Model BOP 1000DM) with a gain of 100 and long-term stability of <0.01% over 8 hours, controlled by a USB device (USB-3106, Measurement Computing). Real-time measurement of the ΔU applied to the electrode tube and the starting potential U_0 of the ion beam are realized by a Keysight 34470A digital multimeter.

A program written in Python is used for the acquisition system. This program integrates functions for logging photon events from the TDC, controlling the scanning voltage via the USB device, recording frequencies of the Matisse and diode laser via the wavelength meter, reading the scanned voltage and starting potential of the ions through the multimeter, and displaying results via a

graphical interface.

III. Commissioning Test and Results

To validate the performance of the CLS system, we performed the first commissioning experiment on natural 40,42,44,48Ca isotopes by probing the 4s $^2S_1/_2$ $\rightarrow$ 4p $^2P_3/_2$ (D_2) ionic transition. The stable beams were produced by ablating a calcium target using the 532 nm laser (1 mm beam diameter on the target) with approximately 1.5 mJ/pulse power. In this test experiment, the extracted ion bunches were accelerated to 20 keV and delivered to the CLS beamline. The ion beam was anti-collinearly overlapped with the frequency-fixed cw laser beam (Fig. 1). The laser frequency was stabilized by the wavelength meter, which was calibrated by the diode laser locked to one hyperfine component of the ^{87}Rb atom. The used laser power was about 1.2 mW, and the laser spot diameter was 6 mm. The velocity of Ca ions was tuned in the interaction region by applying a scanning voltage (ΔU) to the electrode tube (Fig. 3). As a result, in the anti-collinear configuration, the Doppler-shifted laser frequency experienced by the calcium ion beam can be calculated in the rest frame as:

$$\nu = \nu_0 \times \frac{\sqrt{1-\beta^2}}{1-\beta}$$

where $_0$ is the fixed laser frequency, U is the total potential (U = U_0 + ΔU), and m is the mass of the Ca ion. By counting the emitted fluorescence photons from the resonantly excited ions as a function of the tuning voltage (ΔU), the optical spectra for Ca isotopes were obtained.

Figure 4 presents a typical two-dimensional spectrum for relative laser frequency and TOF. The projection of photon counts onto the x-axis and y-axis yields the hfs spectrum (Fig. 4(a)) and TOF spectrum (Fig. 4(b)), respectively. The typical temporal length of the ion bunch is about 10 µs, but with a visible tail on the higher-energy side (shorter TOF side), as shown in the TOF spectrum. This higher-energy tail is related to the field distribution within the plasma plume of the ablation process, as described in Refs. [25, 28]. A TOF correction method [28, 29] can be applied to compensate for the higher-energy component of the ion bunches, which leads to similar results as achieved by simply gating on the main TOF peak, as indicated by the dotted lines in the TOF spectrum. This low-probability tail is nearly invisible when a weak ion beam is adopted, e.g., ion beam current <1 pA as for 42,44,48Ca.

The high-resolution hfs spectra of 40,42,44,48Ca, obtained by gating the TOF window as indicated in Fig. 4(b), are shown in the insets of Fig. 5, fitted using a Voigt profile (a convolution of Gaussian and Lorentzian). The full width at half maximum (FWHM) of the spectra from the Voigt fit is about 55 MHz, comparable to similar CLS measurements worldwide [6, 15, 25]. Considering the natural linewidth of about 25 MHz (main contribution to the Lorentzian

component Γ) for the 4s $^2S_1/_2 \rightarrow$ 4p $^2P_3/_2$ (D_2) ionic transition, the maximal Gaussian contribution (Γ_G) to the FWHM is about 40 MHz, mainly attributed to the energy spread of the ion beam. Assuming an energy spread δE for the total potential U of the ion beam, the resulting Doppler broadening of the spectral line is:

$$\delta\nu = \nu_0 \times \frac{2eU}{mc^2}$$

Thus, the 40 MHz Gaussian contribution (Γ_G) in linewidth corresponds to an energy spread of 2 eV for the ion beam, mainly attributed to fluctuations of the Heinzinger power supply (200 mV: 4 MHz), Kepco DC amplifier (20 mV), and field distribution at the ablation target position. From these high-resolution optical spectra (shown in the insets of Fig. 5), isotope shifts of 42,44,48Ca^+ relative to the reference isotope 40Ca^+ are extracted, showing good agreement with literature values [26, 27], as displayed in Fig. 5 and summarized in Table 1.

TABLE 1 . Isotope shifts (in MHz) of 42,44,48Ca isotopes relative to 40Ca measured for the 4s $^2S_1/_2 \rightarrow$ 4p $^2P_3/_2$ (D_2) ionic transition. These results are compared with literature values [26, 27]. Note that the isotope shifts from Ref. [27] are from a dedicated experimental setup for high-precision measurement.

Isotope	This work	Ref. [26]	Ref. [27]
42Ca	426.9(29)	426.4(15)(10)	426.04(15)
44Ca	848.8(37)	850.1(10)(19)	850.09(14)
48Ca	1705.7(39)	1710.6(35)(39)	1707.58(16)

IV. Summary and Prospects

In summary, a collinear laser spectroscopy apparatus integrated with a laser ablation ion source and a frequency-tunable laser system has been implemented at Peking University, aiming to study nuclear properties of unstable nuclei at domestic RIB facilities. The ion source was designed to provide bunched stable ion beams with energies up to 30 keV, commissioned with a 20 keV bunched stable calcium ion beam. The typical temporal width of the ion bunch was determined to be about 10 μs. Combined with an anti-collinear laser at 394 nm, high-resolution hfs spectra were measured for stable 40,42,44,48Ca isotopes, achieving a narrow linewidth of about 55 MHz (FWHM), comparable to similar CLS setups worldwide. The Gaussian component of the linewidth (FWHM) was determined to be about 40 MHz, corresponding to an energy spread of 2 eV for the stable calcium ion beam. Isotope shifts (δ^{40},) of 42,44,48Ca relative to the reference 40Ca^+, extracted from the obtained hfs spectra, are in excellent agreement with literature values, demonstrating the overall satisfactory performance of this CLS system.

Based on this successful implementation and operation, the entire system is now ready for application to unstable isotopes at RIB facilities such as BRIF at CIAE. This type of online experiment is scheduled and will be performed in the near term. In addition, further exploration and development of the CLS technique are planned for subsequent steps, e.g., application of the system for atomic hfs spectrum measurement using a charge exchange process and development toward resonant ionization spectroscopy measurement. A mass separator and a radio-frequency quadrupole cooler/buncher can also be incorporated into the system to provide ion bunches with improved time structure.

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