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ATOMIC SPECTROSCOPY STUDIES OF SHORT-LIVED ISOTOPES AND NUCLEAR ISOMER SEPARATION WITH THE ISOLDE RILIS

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Abstract

The Resonance Ionization Laser Ion Source (RILIS) at the ISOLDE on-line isotope separator is based on the selective excitation of atomic transitions by tunable laser radiation. Ion beams of isotopes of 20 elements have been produced using the RILIS setup. Together with the mass separator and a particle detection system it represents a tool for high-sensitive laser spectroscopy of short-lived isotopes. By applying narrow-bandwidth lasers for the RILIS one can study isotope shifts (IS) and hyperfine structure (HFS) of atomic optical transitions. Such measurements are capable of providing data on nuclear charge radii, spins and magnetic moments of exotic nuclides far from stability. Although the Doppler broadening of the optical absorption lines limits the resolution of the technique, the accuracy of the HFS measurements examined in experiments with stable Tl isotopes approaches a value of 100 MHz. Due to the hyperfine splitting of atomic lines the RILIS gives an opportunity to separate nuclear isomers. Isomer selectivity of the RILIS has been used in studies of short-lived Ag, Cu and Pb isotopes.

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Abstract

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Introduction

A large number of radioactive isotopes can be provided by isotope separator on- line (ISOL) facilities. The intensity and physical composition of the produced radioactive ion beams are strongly dependent on the type of source used to ionize the isotope of interest. Due to the outstanding selectivity of the laser resonance photo-ionization technique [1, 2] laser ion sources based on resonant excitation of atomic transitions by wavelength tunable lasers have been developed and implemented at different ISOL facilities [3, 4]. The main advantages of resonance ionization laser ion sources (RILIS) are the chemical selectivity of ionization and a convenient way of manipulating the composition of the ion beams by turning on and off laser beams.

Coupled with a mass separator the ion source does not need to select the isotope of interest. Moreover, in general an ion source is required to provide equal ionization efficiency for different isotopes of the same chemical element. Therefore, when running an ISOL facility with RILIS, special attention must be paid to the isotopic and hyperfine structure of atomic transitions used in the ionization scheme. On the other hand, the RILIS does provide an opportunity to study isotopic effects in optical transitions with a very high sensitivity. Experiments on the atomic spectroscopy of short-lived isotopes of Yb and Tm have been performed using RILIS at the IRIS (LNPI, Gatchina) facility [5 – 8]. Spectroscopic applications of the ISOLDE (CERN) RILIS have been demonstrated for Ag, Be and Cu isotopes [9 – 11]. In this paper the laser system of the ISOLDE RILIS will be presented and its application for atomic spectroscopy will be considered.

Laser system and RILIS ion beams

The resonance ionization laser ion source of the ISOLDE facility is a hot cavity type RILIS. Its principles of operation are presented in [12, 13]. In brief, the laser beams interact with the atomic vapor in a hot metal tube (typically 30 mm long, 3 mm inner diameter). Radioactive isotopes are produced in a heated ISOLDE target by proton-induced reactions and diffuse to the cavity of the ion source, which is attached to the target container by one of its ends. The laser beams enter into the ion source from the other end, passing through the opening of the extraction electrode. A DC heating keeps the ion source cavity temperature at about 2000 ° C. After leaving the source, ions are accelerated to 60 kV, separated in a magnet field and guided by electrostatic ion-optical elements to the experimental setup.

The key elements of the RILIS laser setup (Fig. 1) are copper vapor lasers (CVLs) with high pulse repetition rate. The system consisting of a laser oscillator and two parallel amplifiers is built using commercially produced, sealed-off, discharge-heated CVL tubes. The CVLs run at pulse repetition rate of 11 kHz, with a pulse duration of 18 ns. All three

tubes are triggered simultaneously. The oscillator with an unstable resonator produces a beam of 12 mm diameter with about 6 W of average power. The low divergence part of its beam is extracted with a double telescope system, split into two and sent to the amplifiers. At the output of the two amplifiers the beams are of 20 mm diameter, each with an average power of up to 45 W. The CVL radiation is composed of two spectral lines of 511 nm (55 – 60 %) and 578 nm (45 – 40 %). Two or three dye lasers and some amplifier cells can be transversely pumped by CVLs to produce wavelength-tunable beams in the range of 530 –850 nm. However, one CVL beams can also be used to excite transitions to the ionization continuum from a highly excited atomic state. Depending on the required ionization scheme, the CVL beams are divided into several beams using mirror beam splitters.

Tuning of the dye laser wavelength is provided by precise rotation of the diffraction grating in the dye laser resonator. By using different gratings the laser line widths can be changed in a range of 9 – 30 GHz. Inserting a Fabry-Perot etalon in the dye laser resonator produces a narrow-band generation with a line width of 1.2 GHz. A synchronous rotation of the grating and the etalon is used to tune the wavelength of the narrow-band laser. Such rotation is controlled by a microprocessor unit. The wavelength or frequency of the dye laser beams is measured by using the lambdameter LM-007 of ATOS GmbH, which is specified for an accuracy of up to 90 MHz.

The tuning range of dye lasers can be extended to the ultraviolet region by doubling and tripling the light frequencies. With non-linear crystals BBO (beta-bariumborate) wavelengths from 214 to 414 nm have been generated at this setup.

Laser beam routing to ion sources of the General Purpose Separator (GPS) or the High Resolution Separator (HRS) is provided by 90° bending quartz prisms. The optical path lengths between the laser setup and ion sources are 18 m and 23 m for GPS and HRS, respectively. The space alignment of the laser beam is monitored at the reference points inside the laser hut by using low-power reflected beams produced by uncoated flat quartz plates, which are installed about half way to the ion sources.

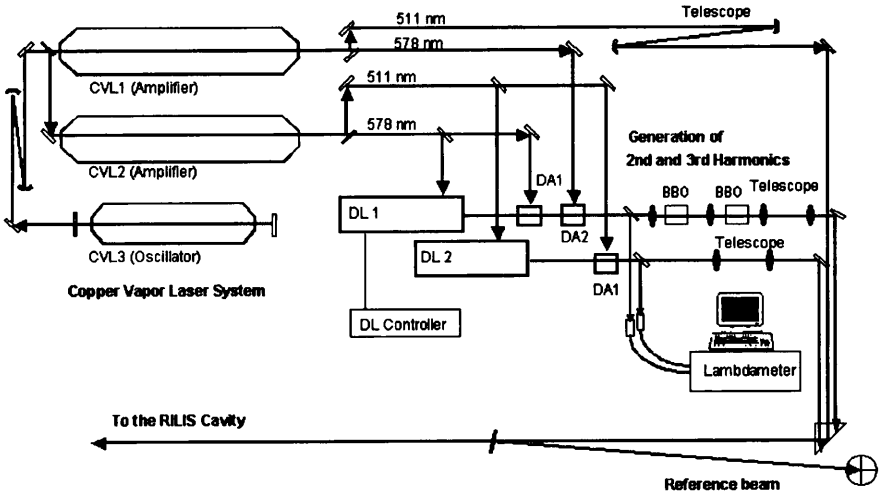

Figure 1. The simplified scheme of the RILIS laser set-up. CVL1, CVL3, CVL3 – copper vapor lasers; DL1, DL2 – dye lasers; DA1, DA2 – dye amplifiers; BBO – β-BaB_2O_4 nonlinear crystals.

Ion beams of isotopes of 20 elements have been produced at ISOLDE by using the RILIS setup. Excitation schemes used at the ISOLDE RILIS are presented in Table 1. They were defined in most cases from data available in literature. During searches for efficient excitation schemes, new highly excited atomic states of Mn [14] and Y [15] have been found by the resonance ionization spectroscopy with the RILIS. For a final choice of the optimal ionization scheme, off-line measurements of the ionization efficiency were needed. In many cases the absolute value of the efficiency could be measured directly by evaporating a small sample of the stable isotope of interest and integrating the produced ion current. In on-line experiments the efficiency can be obtained from yields of radioactive isotopes with known production cross-section. The measured values of the RILIS ionization efficiency are in the range of 0.5 – 30 % and depend on the atomic excitation scheme used, and on the laser power available. The power of the laser beams could not be measured directly in the ion source. Instead, the power of the reflected beams was measured at the reference points with a diaphragm of 3 mm diameter. Then knowing reflectance/transmittance ratios the laser power injected into the ion source cavity could be defined. Typical power values of injected beams are listed in Table 1.

Table 1. Excitation schemes used at the ISOLDE RILIS: IP – ionization potential; λ_1, λ_2, λ_3 - wavelengths of the first, second and third step excitation transition; P_1, P_2, P_3, – power values of laser beams injected into the RILIS cavity; η_{ion} – ionization efficiency.

Element	IP eV	λ_1 nm	P_1 mW	λ_2 nm	P_2 mW	λ_3 nm	P_3 mW	η_{ion}, %	Separated isotop
$_4$Be	9.32	234.861	20	297.32	300	–	–	> 7	7, 9 – 12, 14
$_{12}$Mg	7.65	285.213	40	552.840	80	510.554 & 578.2	5000	10	23 – 34
$_{13}$Al	5.99	309.271 30	50 30	510.554 & 578	5000	–	–	> 20	26 – 34
$_{20}$Ca	6.11	272.164	100	510.554 & 578	5000	–	–	0.45	40, 42 - 44 (stab
$_{25}$Mn	7.44	279.827	50	628.270	1500	510.554	3000	19	48 – 69
$_{27}$Co	7.88	304.400	150	544.458	150	510.554 & 578.2	5000	> 4	59 (stable)
$_{28}$Ni	7.64	305.082	50	611.106	200	748.219	1000	> 6	56 – 70
$_{29}$Cu	7.73	327.396	50	287.894	250	–	–	> 7	57 – 78
$_{30}$Zn	9.39	213.857	20	636.234	150	510.554 & 578.2	5000	5	58 – 74
$_{31}$Ga	6.00	287.423	70	510.554	5000	–	–	21	61 – 85
$_{39}$Y	6.22	408.371	100	581.91	1900	581.91	1900		89 (stable)
$_{47}$Ag	7.58	328.068	70	546.550	100	510.554 & 578.2	5000	14	101 – 129
$_{48}$Cd	8.99	228.802	20	643.847	50	510.554		10	98 – 132
$_{49}$In	6.00	303.936	50	510.554 & 578	5000	–			100 – 135
$_{50}$Sn	7.34	300.914	50	811.399	100	823.49		~ 9	103 – 137
$_{65}$Tb	5.86	579.563	250	551.651	150	618.25			149
$_{70}$Yb	6.25	555.648	150	581.027	1000	581.027			155 – 178
$_{81}$Tl	6.11	276.787	70	510.554 & 578	5000	–		27	179 – 200
$_{82}$Pb	7.42	283.305	70	600.186	500	510.554 & 578.2		3	182 – 215
$_{83}$Bi	7.29	306.770	50	555.205	500	510.554 & 578.2		6	188 – 218

In the last column of Table 1 the mass numbers of produced radioactive ion beams are given. On-line yields obtained with the ISOLDE RILIS are published in [13, 14, 16 – 20]. Some ionization schemes have not yet been applied for on-line experiments, but have been tested using stable isotopes. With the goal of achieving the most narrow line width of the pulsed dye laser, the seeding of a single-mode cw laser beam into the cavity of the pulsed dye laser has been tested at the RILIS laser set-up.

High-resolution atomic spectroscopy measurements with RILIS

In the hot cavity RILIS, laser light interacts with atoms moving in different directions with thermal velocities. Although individual atoms have narrow spectral absorption line widths, the observable line width of atomic resonance is broadened due to the Doppler effect. The temperature of the cavity T and atomic mass number M define the Doppler line width according to

$$\Delta\nu_D = 7.16\cdot10^{-7}\nu_0\sqrt{T/M},$$

where ν_0 is the frequency of the atomic transition. At a temperature of 2300 K the Doppler broadening of atomic lines in the visible range is about 15 GHz for Be, 2 GHz for Cu, and 1.3 GHz for Tl. Thus, the resolution of the spectral measurements using RILIS dye lasers is limited by these values. Nevertheless, due to its high efficiency RILIS is very appropriate for atomic spectroscopy of rare isotopes, produced in very small quantities at ISOL facilities. The sensitivity of the spectral measurements can be enhanced by the selectivity of nuclear decay detection.

At ISOLDE the atomic hyperfine structure (HFS) of neutron-rich ^{68,70}Cu isotopes was studied by scanning the frequency of the narrow-band laser used for the first excitation step in RILIS. Variations of the radioisotope yields associated with the laser scan were monitored with a β–γ detector setup [10, 11]. The data analysis allowed determination of the

magnetic moments of five nuclear isomers with accuracies below 5%, while the uncertainties of optical HFS measurements were in the range of 0.12 – 0.5 GHz [11]. The experimental uncertainties were mainly due to the low number of measurements (about 5 per line profile) and due to fluctuations of the laser power during the frequency scan.

In order to define the ultimate accuracy of the resonance ionization spectroscopy using the existing RILIS set-up we have performed HFS measurements for stable Tl isotopes. A sample of stable $^{203, 205}$Tl was loaded into the target container, ionized using RILIS and separated by the GPS mass separator. The produced ion current was measured by a Faraday cup at the GPS focal plane. The two-step single resonance ionization scheme was used for Tl atoms (see Table 1). The first step transition $6p\ ^2R^0_{1/2} \rightarrow 6d\ D_{3/2}$, $\lambda = 276.787$ nm was excited by a frequency-doubled light of the narrow-band dye laser. To avoid line broadening caused by the saturation, the power of the UV beam was attenuated down to 1 mW. The laser frequency was scanned linearly with simultaneous logging of the Tl ion current and lambda-meter readings. The time duration of a single scan was about 5 min. An example of the registered HFS spectrum is presented in Fig. 2.

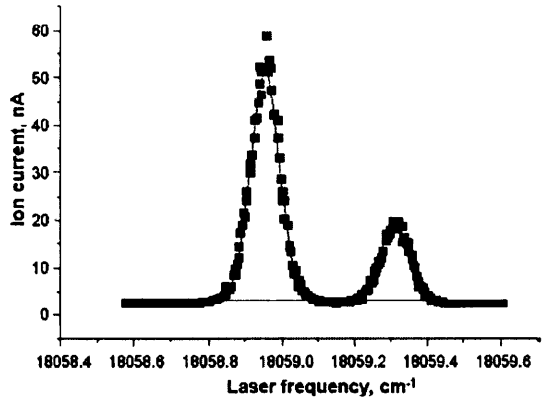

Figure 2. Example of the HFS spectrum of ^{205}Tl measured with RILIS by scanning the frequency of narrow band dye laser. The experimental results (squares) are displayed together with the result obtained from fitting two gaussian curves to the data (solid line).

Accurate measurements of the HFS of this transition are described in [21]. The observed structure is dominated by a large hyperfine splitting of the ground state, measured very precisely [22] to be 21105.447 (5) MHz for ^{203}Tl and 21310.835 (5) MHz for ^{205}Tl. The splitting of the upper d-state is much less, i.e. 85.0 (1.4) MHz and 85.8 (8) MHz, respectively [22]. In the conditions of our measurements the upper state splitting is unresolved and we thus restrict the following discussion to the ground state splitting. The registered spectra were fitted by two Gaussian profiles, which, after averaging over five consecutive scans yielded a peak separation of 10672 (86) MHz for ^{205}Tl. In the scale of the UV transition frequency this result represents the known value of the ground state hyperfine splitting with an error of 33 MHz.

Under these conditions the observed line width was equal to 2.4 GHz, where only 1.3 GHz were due to the Doppler broadening. Hence, a better resolution could be obtained with a more narrow laser line. With the goal of further narrowing the pulsed dye laser line width, the seeding of a continuous wave laser beam into the cavity of the pulsed dye laser was tested at the RILIS laser set-up. A cw beam from a commercial single-mode tunable dye laser (Coherent model 599) was delivered to the RILIS setup via 50 m long single-mode polarization preserving optical fiber. This beam with a power of about 10 mW was seeded into the resonator of the CVL-pumped dye laser. By tuning the laser resonator to the wavelength of the seeded beam a narrow-band pulsed laser beam was generated at its output. The line width of the laser, estimated by using interference patterns registered by the lambda-meter, was found to be less than 0.6 GHz. The line did become narrower, but a synchronous tuning of both dye laser resonators is needed to produce a frequency scan with such system.

Isomer separation with RILIS

Nuclear isomers with different values of magnetic moments and spins produce different hyperfine splitting of atomic lines. Therefore, with the RILIS an isomer selective ionization can be achieved by an appropriate tuning of the laser wavelengths. The feasibility of an isomer separation for given nuclides is defined by the HFS of the atomic transitions used for their ionization in RILIS. The line splitting should at least exceed the Doppler broadening. The separation of Ag and Cu nuclear isomers at ISOLDE by using the RILIS technique has been reported earlier [9, 10, 23]. Recently, by scanning the RILIS laser frequency over the HFS of the neutron-deficient ^{185}Pb isotope the existence of two nuclear isomers has been revealed. The isomers were identified on the basis of the atomic HFS registered by monitoring the α decay of ^{185}Pb at different α-particle energies. These measurements have supplied data for the determination of their half-lives, decay properties, magnetic moments and spins [24].

Outlook

Laser ion sources based on the resonance photo ionization technique of atoms can be used for a broad range of chemical elements. At ISOLDE the number of ion beams produced with RILIS continues to increase. A new series of experiments on short-lived isotopes of Sn, Pb, Bi by means of "in-source" resonance ionization spectroscopy is currently under preparation.

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