

# Resonance ionization scheme development for europium

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January 23, 2017

**Abstract** Odd-parity autoionizing states of europium have been investigated by resonance ionization spectroscopy via two-step, two-resonance excitations. The aim of this work was to establish ionization schemes specifically suited for europium ion beam production using the ISOLDE Resonance Ionization Laser Ion Source (RILIS). 13 new RILIS-compatible ionization schemes are proposed. The scheme development was the first application of the Photo Ionization Spectroscopy Apparatus (PISA) which has recently been integrated into the RILIS setup.

**Keywords** Resonance laser ionization · Laser ion source · RILIS · Europium · Ionization scheme development · Autoionizing states

## 1 Introduction

The Resonance Ionization Laser Ion Source (RILIS) [1,2,3] at the ISOLDE radioactive beam facility at CERN [4] is the most intensively used ion source, providing > 75 % of the ion beams in 2016. The RILIS is based on the principle of stepwise laser resonance ionization which is an element (and in some cases isotope or isomer) selective process. Europium belongs to the lanthanides which have ionization potentials (IP) of relatively low values in the range 5.4 - 6.3 eV [5]. Europium, with an IP of 5.67 eV [6], has therefore been produced at ISOLDE with a standard surface ion source [7] in the past. The effective ionization potential of europium lies at  $\sim 5.60$  eV<sup>1</sup>, which is slightly higher than the effective ionization potential of e.g. samarium ( $\sim 5.55$  eV). For samarium earlier measurements during on-line beam production at ISOLDE have shown, that an enhancement from laser to surface ionization of a factor of 6 can be reached (target at 2000°C, ion source at 1600°C). Another

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<sup>1</sup> the effective ionization potentials have been calculated for a temperature of 2000°C, according to  $IP_{\text{effective}} = IP - kT \ln \left( \frac{Q_i}{Q_a} \right)$ , where  $k$  is the Boltzmann constant,  $T$  is the temperature and  $Q_{i,a}$  are the partition functions of the ion and atom respectively (see [8,9])

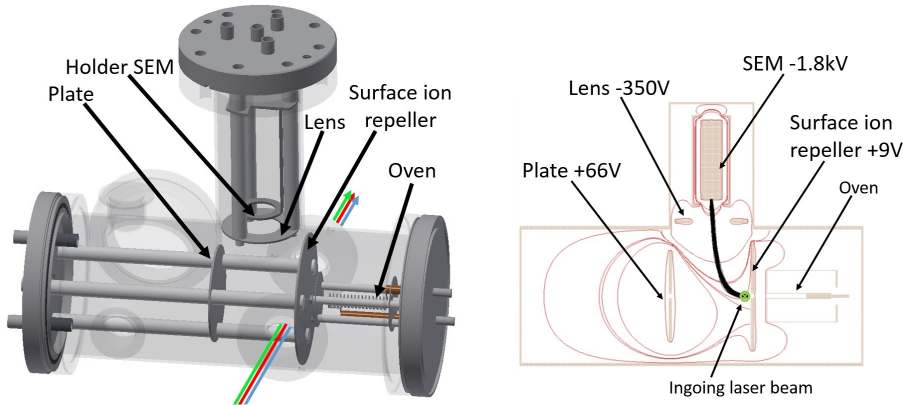
rare earth element for which resonance laser ionization has been used in the past for on-line applications is neodymium with an effective ionization potential of  $\sim 5.43$  eV. Depending on the target and ion source temperatures, laser to surface ionization enhancement factors of 3 (target at 1800°C, ion source at 2130°C) up to 20 have been observed (target at 1800°C, ion source at 1930°C). It is therefore reasonable to expect that a significant enhancement factor in the ion rate of europium should be seen when applying an efficient laser ionization scheme in a standard ISOLDE hot-cavity environment. In addition the isobar purity can be further enhanced if the RILIS is used in conjunction with surface ion suppression methods, e.g. reduced ion source temperatures, fast beam gating [10, 11], low work function cavities [12] or the Laser Ion Source Trap (LIST) [13].

Presently several resonance laser ionization schemes for europium exist, making use of either three step schemes [14] or two step schemes [15]. In both cases dye lasers were used, where the two step schemes have made use of UV pumped dye lasers, emitting light in the blue to green spectral range. None of the existing schemes are specifically well suited to convenient operation of the current RILIS laser setup (see section 2), because they either require a longer setup time (three-step excitation) or operation with UV pumped dyes. This work is the first application of a newly commissioned off-line atomic beam unit, the so-called Photo Ionization Spectroscopy Apparatus (PISA) [16, 17]. This device was used in combination with the ISOLDE RILIS laser system in order to develop convenient and effective RILIS ionization schemes for europium.

## 2 Experimental method

The current laser setup of the RILIS (see [2]) consists on the one hand of three tunable, commercial dye lasers (2 x CREDO from Sirah Lasertechnik GmbH and 1 x MSS from DMK Laser Microsystems), pumped by a Nd:YAG (532 nm and 355 nm) laser from Edgewave GmbH. On the other hand there are three solid state Titanium:Sapphire (Ti:Sa) lasers, based on the design by Mainz university (2 x Z-cavities [18, 19] and 1 x tunable grating Ti:Sa [20]) available which are pumped by two Photonics Industries DM-60 frequency doubled Nd:YAG lasers. Additionally a Blaze laser from Coherent (frequency doubled Nd:YVO<sub>4</sub>, 532 nm) is available and most commonly used for RILIS ionization schemes which employ a non-resonant last step for ionization. All lasers operate at a repetition rate of 10 kHz. The laser beams pass through telescopes on the laser table (as the last focusing elements) and are usually directed into the ion source of one of the two ISOLDE target stations (see e.g. [2]). For this work the laser beams were instead sent past the GPS launch mirrors (see Figure 1) and directed into the PISA. Spatial overlap at the interaction region was achieved by overlapping the laser beams before and after the laser windows of the PISA. The diameter of the laser beams at the laser-atom interaction region was 3 – 4 mm.





**Fig. 2** A drawing of the PISA is depicted on the left next to a simulation of the electric field potentials and the resulting ion beam trajectory (drawings from [22], adjusted).

### 3 Results

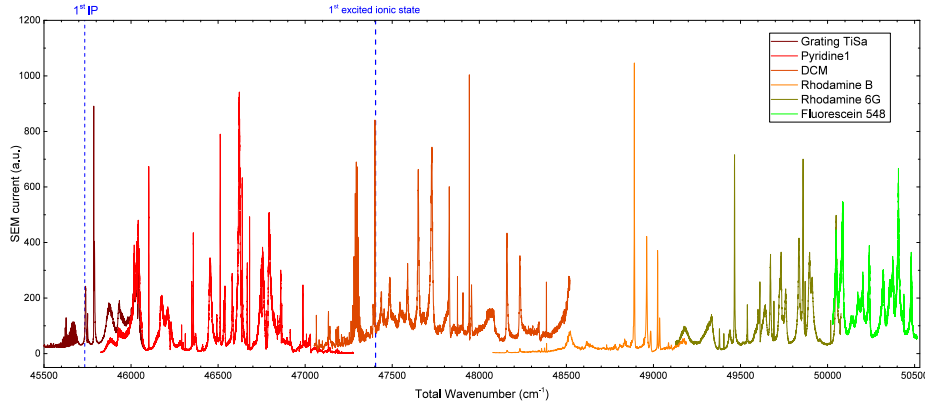
The first resonant step was chosen from literature. It is a transition from the ground state  $4f^7 6s^2 a^8 S_{7/2}^o$  to the  $4f^6 5d 6s^2 8 P_{9/2}$  excited state at  $32130.25 \text{ cm}^{-1}$  [25]. Several frequency scans were performed in order to find a suitable, resonant transition to an autoionizing state. The scans in a wavelength range of 544 to 730 nm were performed with different laser dyes (ethanol solvent) which are listed in Table 1.

**Table 1** Dyes used for second step scans of europium

Dye name	Wavelength range [nm]
Pyridin 1	660 – 730
DCM	610 – 670
Rhodamine B	586 – 627
Rhodamine 6G	557 – 588
Fluorescein 548	544 – 559

Additionally the grating Ti:Sa was scanned in a wavelength range of about 714 to 918 nm. This gave the possibility of scanning above and below the  $\text{IP}=45734.74 \text{ cm}^{-1}$  [6], revealing some additional transitions to autoionizing states (AIS) and Rydberg levels. The analysis of the Rydberg levels leading to the ionization potential will be the scope of a future publication.

A complete outline of the scans is given in Figure 3, where the ion currents for each scan have been scaled in order to gain a better overview. The transition energies  $v_{\text{AIS}}$  of the AIS are given in Table 2 along with the relative intensities for each scanned region and the total energy ( $E_{\text{AIS}} = E_1 + v_{\text{AIS}}$ ). The individual errors of the transitions result from the fitting errors and the error of the wavemeter which was assumed to be a systematic error



**Fig. 3** Overview of the second (final) step scans performed in the PISA.

of  $\Delta E_{\text{wm}} = 0.03 \text{ cm}^{-1}$ . Each region was scanned in two directions, so that each resonance was measured two times. The final value for the transitions  $\nu_{\text{AIS}}$  are therefore a result of the weighted mean value of at least two measurements (up to four in the case of resonances observed in the overlapping regions of the scanning ranges). For determining the relative intensities, the lowest peak in each of the scanned wavelength regions was chosen as reference, so that the lowest intensity is 1 and the numbers grow for higher intensities. It should be noted that due to time restrictions and the multitude of observed AIS there were no attempts to verify the saturation of the different transitions. Therefore a comparison of the relative intensities of the different scanning ranges has not been attempted. Some of the observed AIS have already been reported in the paper by S. Bhattachrya et al. [26] and have been marked with an \*. Additionally peaks that have been observed in more than one scan are presented in italic characters.

Table 2: Transitions  $\nu_{\text{AIS}}$  to autoionizing states observed from the laser-excited europium level  $4f^6 5d 6s^2 \ ^8P_{9/2}$ . The italic characters indicate that the AIS were found in two different spectra (relative intensities are given for both recorded spectra). The \* indicate that the levels have been observed in [26] and match the results from this work within their error margin.

Transition energy $\nu_{\text{AIS}}$ [ $\text{cm}^{-1}$ ]	Relative intensity	Total energy $E_{\text{AIS}}$ [ $\text{cm}^{-1}$ ]
13608.39±0.08	3	45738.64
13614.72±0.09	1	45744.97
13620.13±0.07	2	45750.38
13655.85±0.15	12	45786.10*
13659.31±0.11	5	45789.56*
<i>13739.46±0.35</i>	2, 1	45869.71
<i>13794.99±0.21</i>	1, 1	45925.24
<i>13887.04±0.17</i>	4, 6	46017.29
<i>13908.18±0.20</i>	4, 9	46038.43
13911.20±1.01	3	46041.45
13971.76±0.03	17	46102.01*

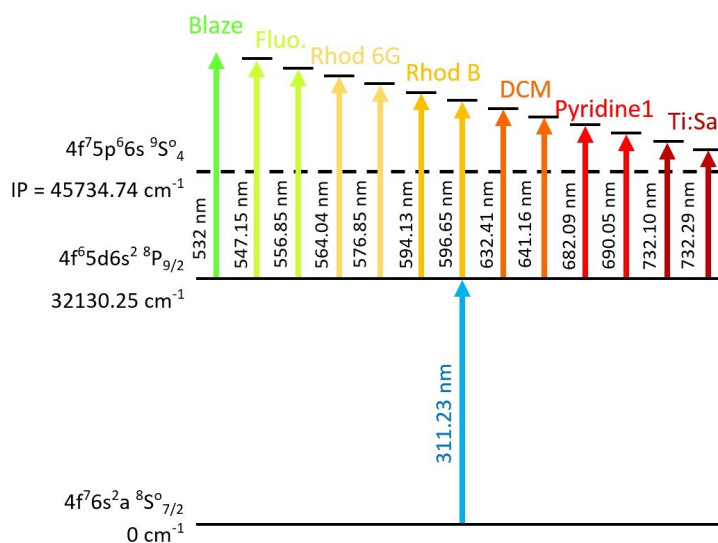
14040.58±0.20	4	46170.83
14089.89±0.27	3	46220.14
14097.50±0.17	2	46227.75
14160.47±0.04	2	46290.72
14168.62±0.02	1	46298.87
14181.27±0.03	1	46311.52
14218.37±0.02	6	46348.62*
14220.99±0.03	1	46351.24*
14227.41±0.02	9	46357.66*
14241.18±0.20	1	46371.43*
14322.95±0.08	7	46453.20*
14361.92±0.05	3	46492.17
14381.92±0.01	17	46512.17*
14402.38±0.10	4	46532.63
14408.95±0.04	5	46539.20*
14450.53±0.08	5	46580.78
14455.32±0.16	4	46585.57
14491.71±0.08	21	46621.96
14501.68±0.04	6	46631.93*
14508.44±0.03	14	46638.69
14537.23±0.03	8	46667.48*
14550.53±0.02	12	46680.78
14621.56±0.29	6	46751.81
14620.55±0.73	8	46750.80
14646.73±0.03	3	46776.98*
14660.41±0.07	12	46790.66*
14710.96±0.04	3	46841.21*
14730.15±0.04	7	46860.40
14734.94±0.03	4	46865.19*
14783.56±0.03	2	46913.81
14856.83±0.02	5	46987.08*
14877.13±0.04	1	47007.38
14900.71±0.09	1	47030.96
14934.40±0.04	1, 4	47064.65
14949.91±0.05	1, 1	47080.16
15003.22±0.04	1, 4	47133.47
14998.53±0.03	2	47128.78
15012.54±0.29	3	47142.79
15023.91±0.07	1	47154.16
15054.02±0.04	3	47184.27
15061.42±0.11	3	47191.67
15066.53±0.07	1	47196.78*
15123.40±0.05	2	47253.65
15133.38±0.04	3	47263.63*
15142.22±0.03	7	47272.47*
15150.43±0.04	9	47280.68*
15157.55±0.05	15	47287.80*
15163.31±0.05	18	47293.56
15167.83±0.07	20	47298.08

15173.13±0.06	12	47303.38
15178.65±0.06	5	47308.90
15183.66±0.06	3	47313.91
15188.65±0.12	3	47318.90
15193.53±0.11	2	47323.78
15270.61±0.05	41	47400.86
15307.14±0.13	5	47437.39
15355.03±0.21	6	47485.28
15458.19±0.20	7	47588.44
15519.28±0.07	16	47649.53
15596.78±0.11	16	47727.03
15697.77±0.11	16	47828.02
15744.52±0.12	7	47874.77
15776.03±0.09	6	47906.28
15812.64±0.04	43	47942.89
15825.23±0.10	6	47955.48
16028.37±0.09	11, 1	48158.62
16102.39±0.11	9, 1	48232.64
16255.67±0.11	8, 3	48385.92
16227.49±0.19	2	48357.74
16244.84±0.07	3	48375.09
16388.49±0.86	6	48518.74
16760.15±0.05	87	48890.40
16831.37±0.09	35	48961.62
16855.36±0.31	7	48985.61
16893.81±0.05	75	49024.06
16905.45±0.11	11	49035.70
16955.19±0.29	3	49085.44
16973.25±0.18	3	49103.50
17249.07±0.07	1	49379.32
17274.30±0.04	1	49404.55
17307.63±0.19	1	49437.88
17335.61±0.06	10	49465.86
17409.26±0.05	2	49539.51
17470.06±0.15	2	49600.31
17481.72±0.05	4	49611.97
17512.20±0.33	3	49642.45
17541.69±0.06	5	49671.94
17561.73±0.05	3	49691.98
17594.20±0.93	3	49724.45
17604.26±0.19	5	49734.51
17630.67±0.14	3	49760.92
17705.35±0.11	5	49835.60
17729.28±0.05	8	49859.53
17741.18±0.06	4	49871.43
17764.80±0.18	4	49895.05
17783.24±0.32	3	49913.49
17918.52±0.05	5, 5	50048.77
17958.15±0.08	5	50088.40

18009.10±0.22	1	50139.35
18045.01±0.18	3	50175.26
18073.12±0.09	3	50203.37
18110.46±0.08	3	50240.71
18190.20±0.14	3	50320.45
18222.99±0.45	3	50353.24
18245.61±0.13	1	50375.86
18276.69±0.07	2	50406.94
18308.66±0.06	1	50438.91
18351.08±0.08	1	50481.33

#### 4 Conclusion

Extensive scans for new autoionizing states have been performed as the first application of the new atomic beam unit, PISA, of the ISOLDE RILIS laboratory. A total of 98 new AIS were found. We propose 13 new ionization schemes which are particularly well suited for convenient operation of the ISOLDE RILIS. A thorough evaluation of the relative performance of these ionization schemes should be conducted using a standard ISOLDE hot cavity laser ion source, as described in section 5. Once these final measurements will have been performed, the ISOLDE RILIS will be able to provide laser ionized europium, whenever the demand arises.



**Fig. 4** Proposed laser ionization schemes for europium (all except 'Blaze' are leading to AIS). The two most dominant peaks of each scanning range were chosen for the proposed schemes. Vacuum wavelengths are given.



## 5 Outlook

In order to finalize the scheme development, time with an ISOLDE target and ion source assembly (on-line or off-line) will be necessary. The following tests should be performed in order to identify the optimal scheme: saturation curves for the first step and second step transitions shown in Figure 4; relative efficiency measurements at the typical conditions of the hot cavity ion source. Additionally the schemes with a transition to an AIS as last step should be compared to the scheme with a non resonant last step at 532 nm. A comparison of the surface/laser ion ratios is a reliable means of evaluating the relative effectiveness of each ionization scheme. In the case of the use of the 40 W 532 nm laser, care should be taken to evaluate any misleading enhancement of the ion rate due to thermal effects or non-resonant ionization of molecular compounds. The former can be studied by time-delaying the first step laser pulse with respect to the 532 nm laser and performing all scheme comparisons with the 532 nm laser still on. The latter will require a more selective means of analyzing the ion beam constituents, such as the ISOLTRAP MR-ToF-MS [27].

## 6 Acknowledgments

The work was supported by the European Union's Seventh Framework Programme for Research, Technological Development and Demonstration under Grant Agreement 267194 (COFUND).

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