Laser Resonance Ionisation for Isotope Separator Facilities



http://cds.cern.ch/record/2207615?ln=es



Overview

- Introduction to ISOLDE and the motivation for a Resonance Ionisation Laser Ion Source (RILIS)
- Atomic physics fundamentals relevant to achieving resonance ionisation
- Considerations for the implementation of a laser ion source
- The ISOLDE RILIS
- The RILIS as part of the array of ISOLDE Experiments





9	10	11	12	13	14	15 Pnictogens	16 Chalcogens	17 Halogens	
5 4	f f								Ate Sy Na We
. 11, 11, . 11, 11,				5 B Boron 3	6 C Carbon -4 4	7 N Nitrogen -3 3 5	8 O Oxygen -2	9 F Fluorine -1	10 N Ne
				13 Al Aluminium 3	14 Si Silicon -4 4	15 P Phosphorus -3 3 5	16 S Sulfur -2 2 4 6	17 Cl Chlorine -1 1 3 5 7	18 A Arg
27 Co Cobalt 2 3	28 Ni Nickel 2	29 Cu Copper 2	30 Zn ^{Zinc} 2	31 Ga Gallium 3	32 Ge Germanium -4 2 4	33 As Arsenic -3 3 5	34 Se Selenium -2 2 4 6	35 Br Bromine -1 1 3 5	36 K ry 2
45 Rh Rhodium 3	46 Pd Palladium 2 4	47 Ag Silver 1	48 Cd Cadmium 2	49 In Indium 3	50 Sn ^{Tin} -4 2 4	51 Sb Antimony -3 3 5	52 Te Tellurium -2 2 4 6	53 lodine -1 1 3 5 7	54 Xe 2 4
77 Ir Iridium 3 4	78 Pt Platinum 2 4	79 Au Gold 3	80 Hg Mercury 1 2	81 TI Thallium 1 3	82 Pb Lead 2 4	83 Bi Bismuth 3	84 Po Polonium -2 2 4	85 At Astatine -1 1	86 R I Ra 2
109 Mt Meitnerium	110 DS Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 MC Moscovium	116 LV Livermorium	117 Ts Tennessine	113 O 0g

Oxidation states are the number of electrons added to or removed from an element when it forms a chemical compound.

62 Sm Samarium 3	63 Eu Europium 2 3	64 Gd Gadolinium 3	65 Tb ^{Terbium} 3	66 Dy Dysprosium 3	67 HO Holmium 3	68 Er Erbium 3	69 Tm ^{Thulium} 3	70 Yb Ytterbium 3	71 Lu 3
94	95	96	97	98	99	100	101	102	10
Pu	Am	Cm	Bk	Cf	ES	Fm	Md	No	Li
Plutonium	Americium	^{Curium}	Berkelium	Californium	Einsteinium	Fermium	Mendelevium	Nobelium	Lav
4	3	3	3	3	3	3	3	2	3









Protons



Neutrons





"Isotope Separator On-Line" radioisotope production, selection and

transport to an experiment in one machine

















Focus on Exotic Beams at ISOLDE: A Laboratory Portrait

iopscience.iop.org/journal/0954-3899, /page/ISOI DE%20laboratory











Create the isotope





Create the isotope Release the isotope from the target

Н				(0		60	00									He
Li	Ве											В	С	Ν	0	F	Ne
Na	Mg				К	(Kelvi	n)		AI	Si	Р	S	CI	Ar			
κ	Са	Sc	Ti	v	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Υ	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	Ι	Xe
Cs	Ва	Lu	Hf	Та	w	Re	Os	lr.	Pt	Au	Hg	ті	Pb	Bi	Ро	At	Rn
Fr	Ra	Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	FI	Мс	Lv	Ts	Og



Boiling point www.webelements.com

u	Gd	Tb	Dy	Но	Er	Tm	Yb
m	Cm	Bk	Cf	Es	Fm	Md	No

© Wark Winter

•Create the isotope

 Release the isotope from the target Ionise the isotope

Н				(0		24	00									He
Li	Ве											В	С	Ν	0	F	Ne
Na	Mg				k	J mol	-	AI	Si	Р	S	CI	Ar				
к	Са	Sc	Ti	v	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	I	Xe
Cs	Ва	Lu	Hf	Та	w	Re	Os	lr	Pt	Au	Hg	TI	Pb	Bi	Ро	At	Rn
Fr	Ra	Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	FI	Mc	Lv	Ts	Og





1	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb
	Am	Cm	Bk	Cf	Es	Fm	Md	No

Ionization energy: 1st www.webelements.com

© Nark V

- •Create the isotope
- Release the isotope from the target
- Ionise the isotope
- Separate the isotope from contaminants



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- Release the isotope from the target
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Li	Ве											в	С	Ν	0	F	N
Na	Mg				K	(Kelvi	in)	-				AI	Si	Р	S	СІ	1
к	Са	Sc	Ti	v	Cr	Mn	Fe	Со	Ni	Cu	Zn	Ga	Ge	As	Se	Br	ł
Rb	Sr	Y	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	I	>
Cs	Ва	Lu	Hf	Та	w	Re	Os	Ir	Pt	Au	Hg	ті	Pb	Bi	Ро	At	F
Fr	Ra	Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	FI	Мс	Lv	Ts	C

		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb	
		Ac	Th	Ра	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	
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Li	Ве											в	С	Ν	0	F
Na	Mg					kJ mo) -'					AI	Si	Р	S	СІ
к	Ca	Sc	Ti	v	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	e As	s Se	Br
Rb	Sr	Y	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sr	n Sb	Те	I
Cs	Ва	Lu	Hf	Та	w	Re	Os	lr	Pt	Au	Hg	т	Pb	Bi	Ро	At
Fr	Ra	Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	FI	Mo	: Lv	Ts

L	₋a	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Тb	Dy	Но	Er	Tm	Yb
4	Ac	Th	Ра	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No



Ionization energy: 1st



Horizontal axis (mm)









ISOLDE beams: available for experiments





> **1000** isotopes available at ISOLDE >75 different elements In the form of an **ion beam** with individual isotope selection

Delivered ~immediately to an experiment for study!

https://isoyields2.web.cern.ch/Yield_Home.aspx































Different number of **protons**

Different electron configuration





[Ar] 3d10 4s2 4p1 [2, 8, 18, 3]

~109 ⁸⁵Rb

37: Rubidium

<<



[Kr] 5s1

[2, 8, 18, 8, 1]



Different number of **protons**

Different electron configuration





31: Gallium

[2, 8, 18, 3]



Different spectral properties

~109 ⁸⁵Rb

37: Rubidium

<<



[Kr] 5s1





90(



Stepwise Resonance Photo-ionization



An element-selective ionisation method!





History of the RILIS method

PROPOSAL of the Institute of Spectroscopy, Acad.Sci. USSR for experiments with ISOLDE-CERN Facility (V. S. Letokhov and V. I. Mishin) LASER PHOTOIONIZATION PULSED SOURCE OF

I. Purpose The development of a pulsed isobar-selective

effective source of ions at the mass-separator inlet on

the basis of the method of laser resonant atomic photoioni-

RADIOACTIVE ATOMS

1984







Early proposals: 1988

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN/ISOLDE IP 50

PROPOSAL TO THE ISOLDE COMMITTEE

DEVELOPMENT OF A LASER ION SOURCE

F. Ames, E. Arnold, H.J. Kluge, Y.A. Kudryavtsev, V.S. Letokhov, V.I. Mishin, E.W. Otten, H. Ravn, W. Ruster, S. Sundell and K. Wendt

University of Mainz, F.R.G., Institute of Spectroscopy, Troitzk, USSR and the ISOLDE Collaboration, CERN, Switzerland

> Spokesman: K. Wendt Contactman: E. Arnold

> > SUMMARY

Test experiments at Troitzk and Mainz have demonstrated the feasibility of step-wise multi-photon excitation and final ionisation by pulsed lasers as a selective and efficient tool for the production of isobarically pure ion beams. The development of a new type of ion source based on this concept is proposed. In combination with existing targets, this will open up the way to a further extension in respect to purity and availability for a number of elements at on-line mass separator facilities. The collaboration proposes to use the CERN-ISOLDE off-line separator for tests of appropriate target ion source configurations with respect to efficiency and purity. After succesful development the laser ion source shall be installed as an additional facility at the IS-3 separator.





History of RILIS at ISOLDE







CVL lasers: v_{rep} =11.000 Hz Oscillator + 2 amplifiers 2-3 dye lasers with amplifiers, nonlinear crystals BBO:

V. Mishin

Institute of Spectroscopy of the Russian Academy of Sciences

Valentin Fedosseev (Former section leader of SY-STI-LP), now a Contributing Retiree @ CERN

Meet ISOLDE: Fresh faces bring fresh ideas

ears of ISOLDE physics, the third article in our Meet ISOLDE series looks at how people shape the facility and the importance of low-energy beams

https://home.cern/news/series/meet-isolde/isolde-50years-cutting-edge-science-benefitting-society







Laser ion sources worldwide



>600 isotopes have been ionized with RILIS

Inaccessible:

Non-metals (high IP or 1st excited state) < 210 nm is difficult to generate <205 nm absorption in transmissive optics



http://dx.doi.org/10.1063/1.4858015 B. A. Marsh, Resonance ionization laser ion sources for on-line isotope separators

Now for some atomic physics fundamentals..

Some terminology

Energy, eV E =



h - Planck's constant, 6.6261 x
10-34 J · s or 4.1357 x 10-15 eV · s

Some terminology

Energy, eV $E = \frac{h \cdot c}{\lambda}$

Wavelength, nm A 500 nm photon is ~2.5 eV

h - Planck's constant, **6.6261 x 10-34 J** · **s or 4.1357 x 10-15 eV** · **s**
Some terminology

$E = \frac{h \cdot c}{\mathbf{\lambda}}$ Energy, eV

Wavelength, nm



Wavenumber, cm⁻¹

Number or wavelengths per unit distance (cm) Proportional to energy (and to frequency).... Very convenient 1 cm⁻¹ x 30 (speed of light in in cm $500 \text{ nm} = 20000 \text{ cm}^{-1}$ per nanosecond) = 30 GHz

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Linewidth, MHz, GHz



Strong transition -> Short half-life O(ns) -> Broad linewidth

h - Planck's constant, **6.6261 x** 10-34 J · s or 4.1357 x 10-15 eV · s

A 500 nm photon is ~2.5 eV

 $\Delta \nu$ = 10s of MHz $\Delta t \ \Delta E \geq \frac{\hbar}{2}$



Some terminology

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Linewidth, MHz, GHz

Timescales, ns



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[2, 8, 18, 3]



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[2, 8, 18, 3]









Principal quantum number, n



Orbital angular momentum, L (s,p,d,f) = (0,1,2,3)

Related to the electron's motion in space, around the atom



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Spin angular momentum **S** Intrinsic motion (rotation) of the electron around its own axis 'Up' or 'down' +1/2 or -1/2





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Spin angular momentum **S** Intrinsic motion (rotation) of the electron around its own axis 'Up' or 'down' +1/2 or -1/2

Total angular momentum, **J**

 $J \equiv L + S$ to |L - S|





The Term Symbol, to describe the (excited) state of the atom



Total spin angular momentum



The Term Symbol, to describe the (excited) state of the atom



Total spin angular momentum

Ground state = 0 eV

 $2^{*}(1/2)+1P_{(1/2)+1}$ $^{2}P_{3/2}$



The Term Symbol, to describe the (excited) state of the atom



Total spin angular momentum

Ground state = 0 eV

 $2^{*}(1/2)+1P_{(1/2)+1}$ $^{2}P_{3/2}$



First excited state = 3.07 eV24788.530 cm-1 $2^{*}(1/2)+1 S_{(1/2)+0} S_{1/2}$





Energy

Ionization energy



Ground state (lowest energy electron ----configuration)



Energy

Ionization energy



Step 1 - Blue Laser light



Energy

Ionization energy





Energy



Step 1 - UV Laser light Step 2 - Red laser Step 3 - Green laser -> ionization!







Simpler, 2 step scheme, higher efficiency, reduced complexity



1	L-S = 1/2	826.19	cm-1			
J	L+S = 3/2	0	cm-1			

$$\frac{N_j}{N_1} = \frac{g_j}{g_1} e^{-(E_j - E_1)/k_B T}$$
55 % @ 220

Use a second laser @ 294 nm to also excite from 826.12 cm-1 thermally populated level!

Is it possible to saturate the transition with available laser?

Transition strength

Transition strength Is it possible to saturate the transition with available laser?

Wavelength (< 200 nm is difficult to generate and transport)

Is the wavelength convenient (or possible) to produce?

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Wavelength (< 200 nm is difficult to generate and transport)

Is the wavelength convenient (or possible) to produce?

Radiative decay paths Does the excited state decay to a level that will render the atom 'invisible'?

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Wavelength (< 200 nm is difficult to generate and transport)

Is the wavelength convenient (or possible) to produce?

Radiative decay paths Does the excited state decay to a level that will render the atom 'invisible'?

Suitability / existence of subsequent transitions

We may compromise on the above considerations if it makes subsequent steps more convenient or efficient

Developing an ionisation scheme for astatine



Sebastian Rothe

Developing an ionisation scheme for astatine



Sebastian Rothe

a Initial exploration - confirm existence of 1st steps
 b Search for on-set of ionization (ionization energy)
 c Search for second steps that would allow the use of the high power green laser for ionization
 d Carefully compare best second steps

Developing an ionisation scheme for astatine



Sebastian Rothe

a	Initial exploration - confirm existence of 1st steps
b	Search for on-set of ionization (ionization energy)
С	Search for second steps that would allow the use of the high power green laser for ionization
d	Carefully compare best second steps
e	Careful scan across the ionization continuum - to precisely measure ionization energy and look for more efficient ionization steps



IP (At) = 9.31751(8) eV

ARTICLE

Received 21 Aug 2012 | Accepted 27 Mar 2013 | Published 14 May 2013

Measurement of the first ionization potential of astatine by laser ionization spectroscopy

S. Rothe^{1,2}, A.N. Andreyev^{3,4,5,6}, S. Antalic⁷, A. Borschevsky^{8,9}, L. Capponi^{4,5}, T.E. Cocolios¹, H. De V E. Eliav¹¹, D.V. Fedorov¹², V.N. Fedosseev¹, D.A. Fink^{1,13}, S. Fritzsche^{14,15,†}, L. Ghys^{10,16}, M. Huyse¹⁰, N U. Kaldor¹¹, Yuri Kudryavtsev¹⁰, U. Köster¹⁸, J.F.W. Lane^{4,5}, J. Lassen¹⁹, V. Liberati^{4,5}, K.M. Lynch^{1,20}, B. K. Nishio⁶, D. Pauwels¹⁶, V. Pershina¹⁴, L. Popescu¹⁶, T.J. Procter²⁰, D. Radulov¹⁰, S. Raeder^{2,19}, M.M. E. Rapisarda¹⁰, R.E. Rossel², K. Sandhu^{4,5}, M.D. Seliverstov^{1,4,5,12,10}, A.M. Sjödin¹, P. Van den Bergh¹⁰ P. Van Duppen¹⁰, M. Venhart²¹, Y. Wakabayashi⁶ & K.D.A. Wendt²



Ionization Energy, eV He (24 eV) Non metals (Not accessible due to high-lying 1st excited states >6 eV) Hg (10.4 eV) Metals

Cs (3.3 eV)

Wavelength range 210 - 950 nm







Pulse energy (ns pulse)

For a 10W laser with 10 ns pulses @ 10kHz

Peak power is ~100kW



Fluence φ condition φ $\sigma_{\text{lon}} \cdot \phi > 1$













Now how is this actually done practically..

ISOLDE operation in 2022....

		GPS schedule 2022																																		
	Ma	rch		Ap	oril				May		June					July					August				Septe	ember					Novemb					
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SU		IS691	IS685		(LOI 217)		15679 15703	3 IS668		LOI216		Colls		tests	IS684			Foll)	10 M eV/u		IS717		IS703 IS602	2	@ 8MeV/u		4.9MeV/u	110Sn@8MeV/u	IS668 + colls	IS717	7MeV/u	IS691	15679 15703			4
	RILIS : Dy	RILIS : Dy	RILIS : Cd	RILIS : Cd	RILIS: TI/TE	b	111Cd	8He/6He	1	RILIS: Ac	RILIS: Ac	RILIS: Ga			RILIS: Zn	RILIS: Zn		RILIS: Be	RILIS: Be	27Na	1.7GeV		199Hg	Stable to MB	RILIS: Mg	stable to MB/XT03	RILIS: Sn	RILIS: Sn	RILIS: Sn	1.7GeV	Kr beams	Xe beams	111Cd	RILIS: Sb	RILIS: Ni	Hg
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				RILIS : AI	RILIS : AI	RILIS : Te	RILIS : Te 49K RILIS : A				RILIS: Ag	RILIS: Ag RILIS: Po RILIS: Po					49K					RILIS: Cu				RILIS: Sn	RILIS: Sn						37K			



RILIS overview

HC-RILIS





6 tuneable lasers

http://riliselements.web.cern.ch н 0 F Ν С 40 CI Ρ S ionisation K Ca V Zn Ga Br Cu Cr Co schemes Rb Nb Rh I Cd Cs Та W Re Os Pt Ir Hg At Fr Rf Db Sg Bh Hs Mt Rg Cn Uut Uuq Uup Ds Uuh Uus Uuo Bk Cf Es Fm Md Cm No Lr

Dye schemes tested

+ MEDICIS (MELISSA), Offline 2, LARIS

Ti:Sa schemes tested Dye and Ti:Sa schemes tested

532 nm

950 nm

532 nm

950 nm

Beam transport to targets



- Simple, robust, reliable
- Problem with surface-ionised isobars
- Ion capacity limit in the range of 10-100 nA



Grounded extraction electrode







Grounded extraction electrode



Surface ionization efficiency



** R. Kirchner. Progress in ion source development for on-line separators.

Grounded extraction electrode









Reinhard Heinke

Daniel Fink Sven Richter

http://dx.doi.org/10.1016/j.nimb.2013.06.039 http://www.ub.uni-heidelberg.de/archiv/16725



Versatile Arc Discharge and Laser Ion Source (VADLIS)



- Modified version of the standard FEBIAD source at ISOLDE
- Ability to turn on/off non selective electron impact ionization
- Molecular breakup with electrons or lasers
- Yisel Martinez Tom Day Goodacre





Adjustable extraction voltage and larger volume may improve ion capacity limit to 10 uA range







PhD Topic Ralitsa Mancheva, KU Leuven / CERN

RILIS as one of the ISOLDE experiments

Low energy nuclear physics (nuclear ground / isomer state properties)



















Measuring the charge radius systematics: isotope shifts





Measuring shape and nuclear moments: The Hyperfine Structure









1972

Sudden change in the nuclear charge distribution of very light mercury isotopes

J. Bonn ^{a, b}, G. Huber ^{a, b}, H.-J. Kluge ^{a, b}, L. Kugler ^{a, b}, E.W. Otten ^{a, b}



Physics Letters B 8, Issue 5, 6 March 1972, Pages 308-311





1972 Sudden change in the nuclear charge distribution of very light mercury isotopes

J. Bonn^{a, b}, G. Huber^{a, b}, H.-J. Kluge^{a, b}, L. Kugler^{a, b}, E.W. Otten^{a, b}



Incredible results for the time, but limited by low production rates and low sensitivity of the method

Physics Letters B 8, Issue 5, 6 March 1972, Pages 308-311



Z. Phys. A - Atomic Nuclei 325, 247-259 (1986)



Isotope Shift of ¹⁸²Hg and an Update of Nuclear Moments and Charge Radii in the Isotope Range ¹⁸¹Hg-²⁰⁶Hg

G. Ulm¹, S.K. Bhattacherjee², P. Dabkiewicz³, G. Huber, H.-J. Kluge¹, T. Kühl⁴, H. Lochmann⁵, E.-W. Otten, and K. Wendt Institut für Physik, Universität Mainz, Federal Republic of Germany

S.A. Ahmad⁶, W. Klempt, R. Neugart⁷, and the ISOLDE Collaboration CERN, Geneva, Switzerland









Ion source development





New ionization scheme for Hg (10 x efficiency improvement)













In-source spectroscopy 'niche'



Neutron Number

Sensitivity is unmatched!



In-source spectroscopy 'niche'



Neutron Number

Sensitivity is unmatched!



In-source spectroscopy 'niche'



Sensitivity is unmatched!



S

LASER IONISATION AND SPECTROSCOPY OF ACTINIDES







IISATION AND SPECTROSCOPY OF ACTINIDES









Reinhard Heinke



The single-mode diamond Raman laser



Courtesy D. Talan



[1] https://doi.org/10.1364/OL.44.003924 [2] https://doi.org/10.1364/OE.384630 568.15 568.16 568.17 568.18 Pump wavelength [nm]




LASER IONISATION AND SPECTROSCOPY OF ACTINIDES







Reinhard Heinke



LASER IONISATION AND SPECTROSCOPY OF ACTINIDES





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A significant experimental challenge ! e.g Fm, Md, No, Lr





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If you are interested in a PhD, Masters (technical studentship) or internship in the RILIS team.. Let me know!

bruce.marsh@cern.ch



