

First photodetachment studies on radioactive negative ions

Towards the electron affinity measurement of astatine

S.Rothe for the I-148 Collaboration



ENGINEERING
DEPARTMENT

The rarest element on Earth: Astatine

PERIODIC TABLE
Atomic Properties of the Elements

NIST
National Institute of Standards and Technology
U.S. Department of Commerce

Physics Laboratory Reference Data
physics.nist.gov www.nist.gov/srd

Group 1 IA 2 IIA 3 IIIB 4 IVB 5 VB 6 VIB 7 VIIB 8 VIII 9 IIB 10 IB 11 IIB 12 IIB 13 IIIA 14 IVA 15 VA 16 VIA 17 VIIA 18 VIIIA

1 H He
2 Li Be
3 Na Mg
4 K Ca
5 Rb Sr
6 Cs Ba
7 Fr Ra

11 Na 12 Mg
19 K 20 Ca
26 Fe 27 Co
33 As 34 Se
41 Nb 42 Mo
49 In 50 Sn
57 La 58 Ce
65 Dy 66 Ho
73 Ta 74 W
81 Th 82 Pa
89 Ac 90 Th
97 Bk 98 Cf
105 Uu

13 Al 14 Si 15 P 16 S 17 Cl 18 Ar
21 Sc 22 Ti 23 V 24 Cr 25 Mn 26 Fe 27 Co 28 Ni 29 Cu 30 Zn 31 Ga 32 Ge 33 As 34 Se 35 Br 36 Kr
37 Rb 38 Sr 39 Y 40 Zr 41 Nb 42 Mo 43 Tc 44 Ru 45 Rh 46 Pd 47 Ag 48 Cd 49 In 50 Sn 51 Sb 52 Te 53 I 54 Xe
55 Cs 56 Ba 57 La 58 Ce 59 Pr 60 Nd 61 Pm 62 Sm 63 Eu 64 Gd 65 Tb 66 Dy 67 Ho 68 Er 69 Tm 70 Yb 71 Lu
87 Fr 88 Ra 89 Ac 89 Ac 90 Th 91 Pa 92 U 93 Np 94 Pu 95 Am 96 Cm 97 Bk 98 Cf 99 Es 100 Fm 101 Md 102 No 103 Lr

85 At
Astatine (210)
[Hg]6p⁵

missing

Atomic Number, Symbol, Name, Atomic Weight, Ground-state Configuration, Ionization Energy (eV)

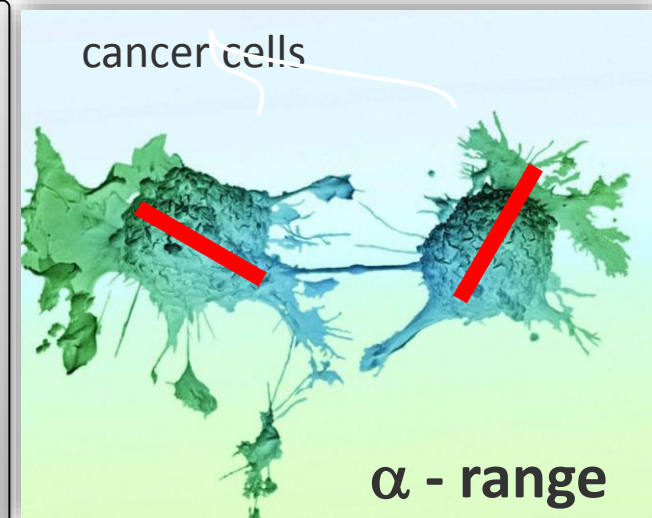
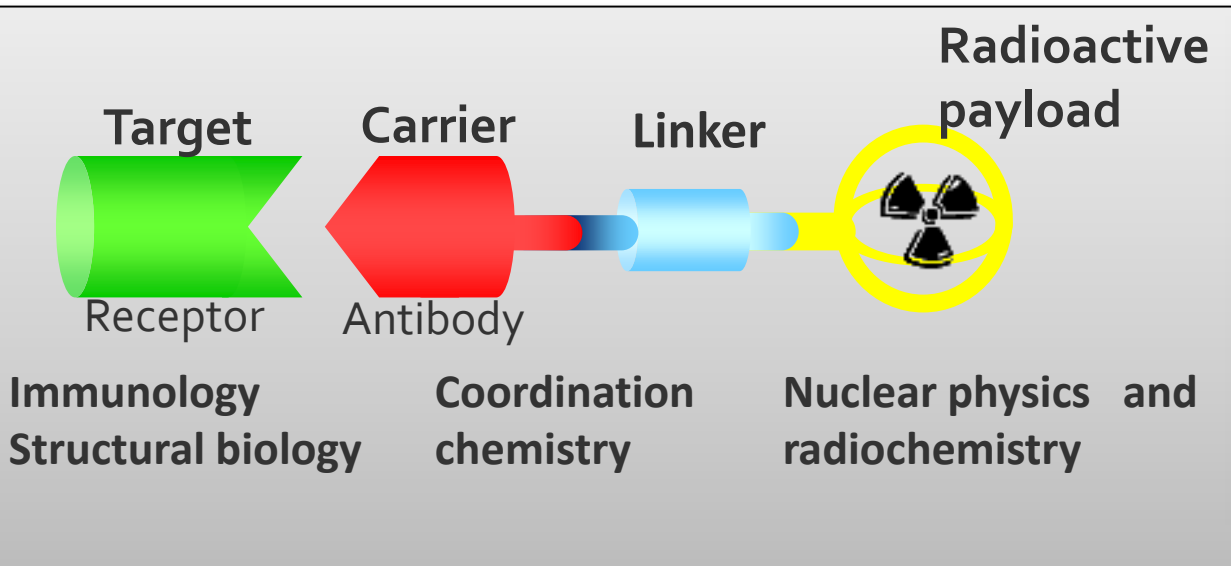
Ground-state Level, Lanthanides, Actinides

Period 1 to 7

53	²¹⁸ Po	54
	²¹⁰ Po	
	Iodine	
	126.90447	
	[Kr]4d ¹⁰ 5s ² 5p ⁵	
	10.4513	
85	²¹⁸ Po	86
	²¹⁰ Po	
	Astatine	
	(210)	
	[Hg]6p ⁵	
117	²⁹⁴ Uu	118
	Ununseptium	
	(294)	

- Most abundant isotope ²¹⁸At, (t_{1/2} = 1.5 s)
- I.Asimov: 1st mile of earth's crust : 70mg (~3.5 atoms/ kg)
- Artificial production: ²⁰⁹Bi(α, 2n)²¹⁰At, Carson et al. (1940)
- Physical and chemical properties hard to investigate

Astatine for targeted alpha therapy (TAT)



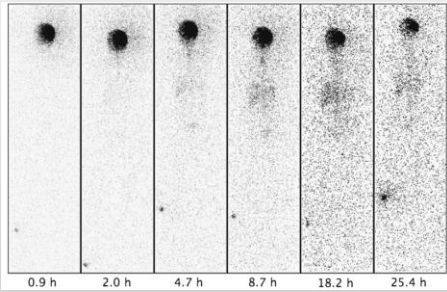
- Cancer cells have specific receptors
- Can be targeted by antibodies which carry radionuclides
- Alpha particles have ideal range to kill individual cancer cells
- Astatine is a good candidate for TAT (half life, α -energy)
- But At behaves sometimes like a metalloid sometimes like halogen - a **chemical hybrid**



A zorse (zebra-horse hybrid)

With material from *U.Köster, INPC 2013*

Astatine biochemistry studies



73 MBq ^{211}At -ch81C6
 γ ray image of full body

in vivo

M. Zalutsky et al., J. Nucl. Med. 49 (2008) 30.



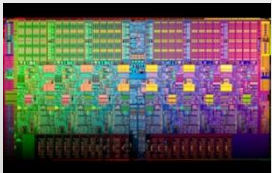
- Slow process due to absence of a long lived tracer

in vitro

S.H. Cunningham et al., Br. J. Cancer 77 (1998) 2061.

S.H.L. Frost et al., Cancer 116 (2010) 1101.

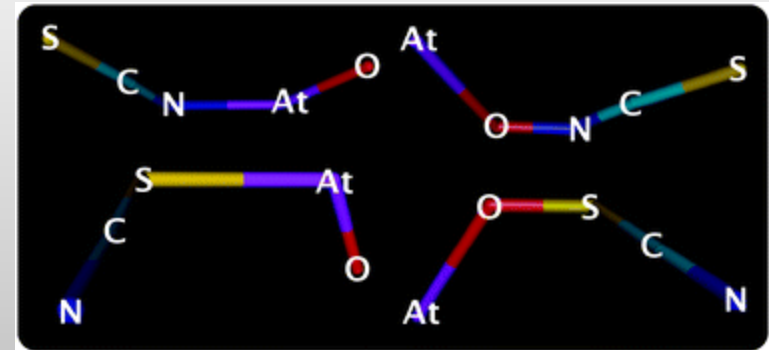
J. Champion et al., J. Chem. Phys. A114 (2010) 576.



in silico

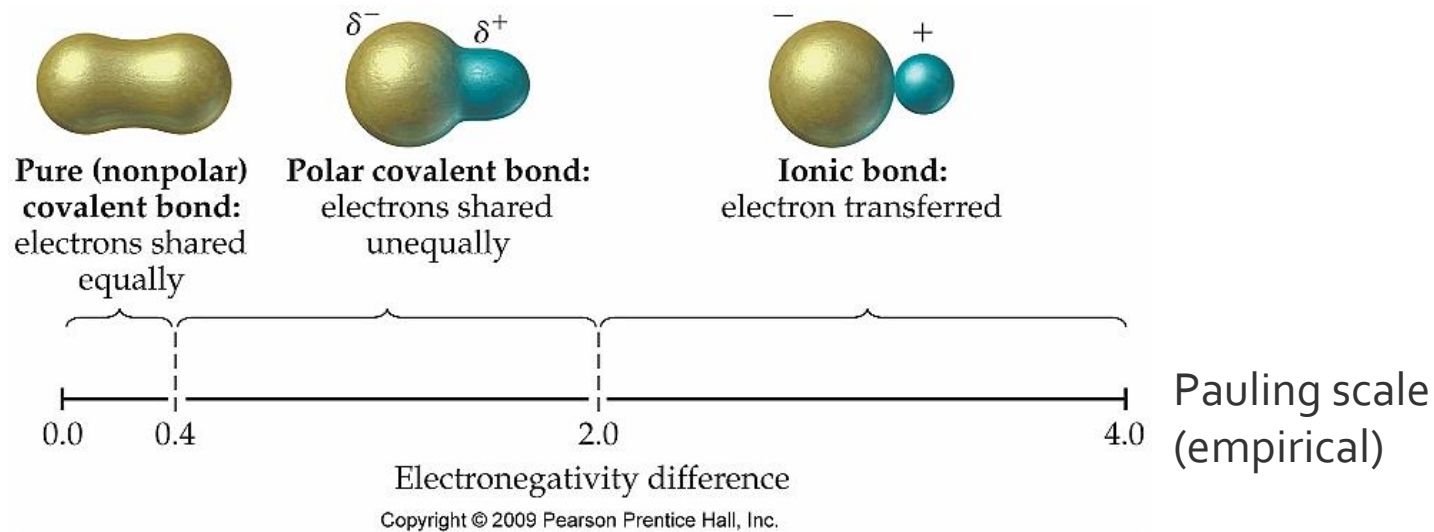
J. Champion et al., PCCP 13 (2011) 14984.

- 'In silico' calculations can predict behavior of compounds before synthesis
- **Needs experimental references:**
such as the IP and EA



With material from *U.Köster, INPC 2013*

Electronegativity



- Electron Affinity (EA)
= Binding energy of the negative ion
- Ionization Potential
= Binding energy of the positive ion
- IP + EA determine the chemical reactivity type of chemical bonds.

Mulliken electronegativity scale

$$X^M = \frac{IP + EA}{2}$$

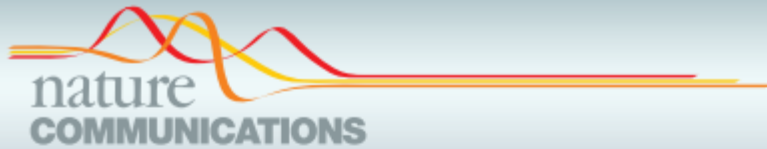
Can be connected to Pauling scale:

DOI: 10.1021/ct049942a

Contents

- 1 Measurement of the IP(At)
- 9 Measurement of the EA of radiogenic iodine
- 2 First results for astatine

The ionization potential of astatine



ARTICLE

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DOI: 10.1038/ncomms2819

OPEN

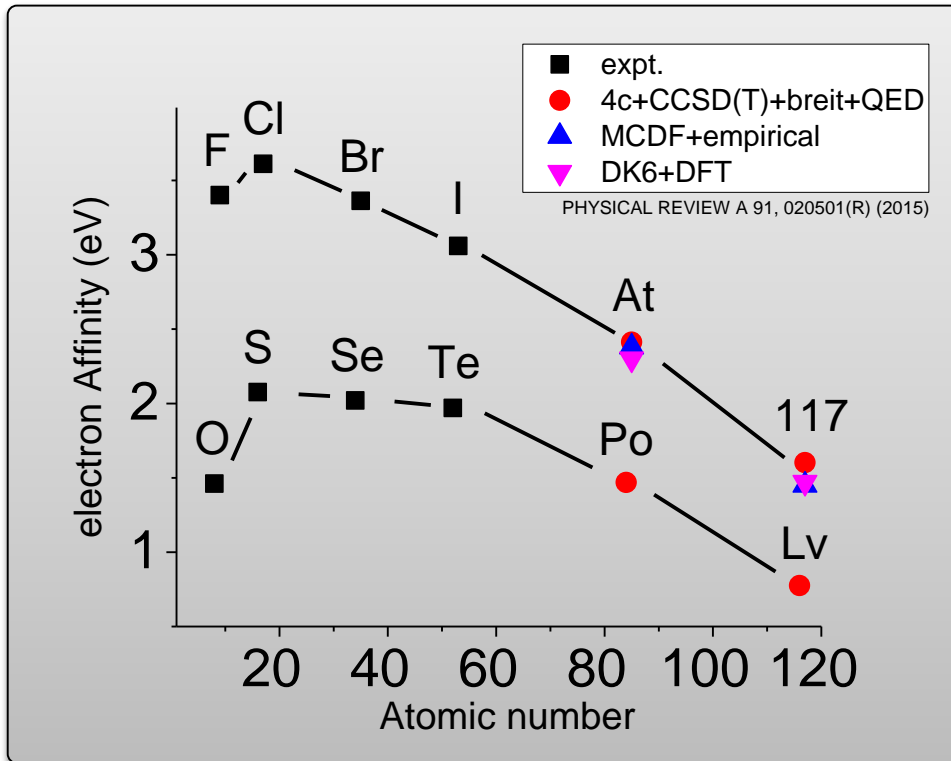
Measurement of the first ionization potential of astatine by laser ionization spectroscopy

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 Witte¹⁰, E. Eliav¹¹, D.V. Fedorov¹², V.N. Fedosseev¹, D.A. Fink^{1,13}, S. Fritzsche^{14,15,†}, L. Ghys^{10,16}, M. Huyse¹⁰, N. Imai^{1,17}, U. Kaldor¹¹, Yuri Kudryavtsev¹⁰, U. Köster¹⁸, J.F.W. Lane^{4,5}, J. Lassen¹⁹, V. Liberati^{4,5}, K.M. Lynch^{1,20}, B.A. Marsh¹, K. Nishio⁶, D. Pauwels¹⁶, V. Pershina¹⁴, L. Popescu¹⁶, T.J. Procter²⁰, D. Radulov¹⁰, S. Raeder^{2,19}, M.M. Rajabali¹⁰, 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

$$\text{IP (At)} = 9.31751(8) \text{ eV}$$

		10.4513	
2	85	² P _{3/2} ^o	8
	At Astatine (210) [Hg]6p ⁵ 9.31751		
	117		1

The electron affinity of astatine



Letter of Intent INTC-I-148

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Preparation of negative ion beams for the determination of the electron affinity of polonium and astatine by laser photodetachment

25.09.2013

S. Rothe¹, M. Bissell², T. Day Goodacre^{1,3}, V. Fedosseev¹, K. Flanagan³, N. Galland⁴, T. Giles¹, A. Gottberg¹, D. Hanstorp⁵, U. Köster⁶, T. Kron⁷, Yu. Kudryavtsev², B. Marsh¹, G. Montavon⁸, G. Neyens², S. Raeder², E. Renault⁴, R. Rossel^{1,9}, M. Stachura¹, T. Stora¹, K. Wendt⁷

Proposal INTC-P-462

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

(Following ISOLDE Letter of Intent I-148)

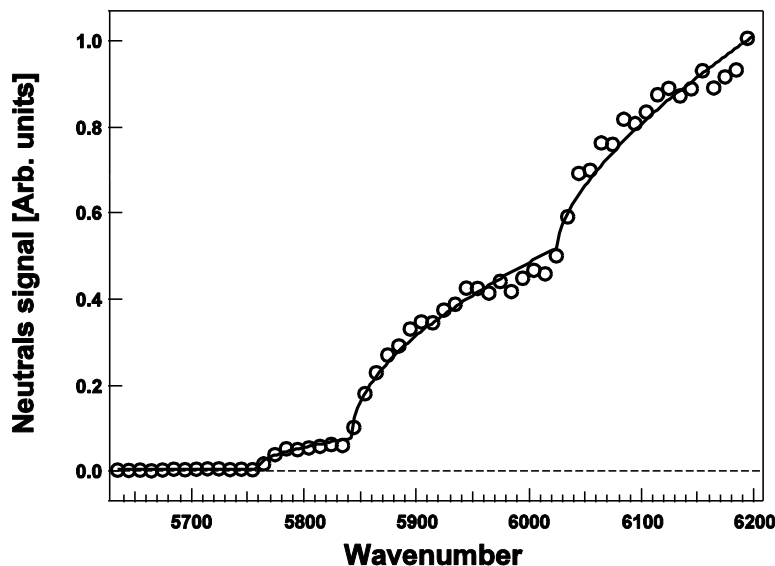
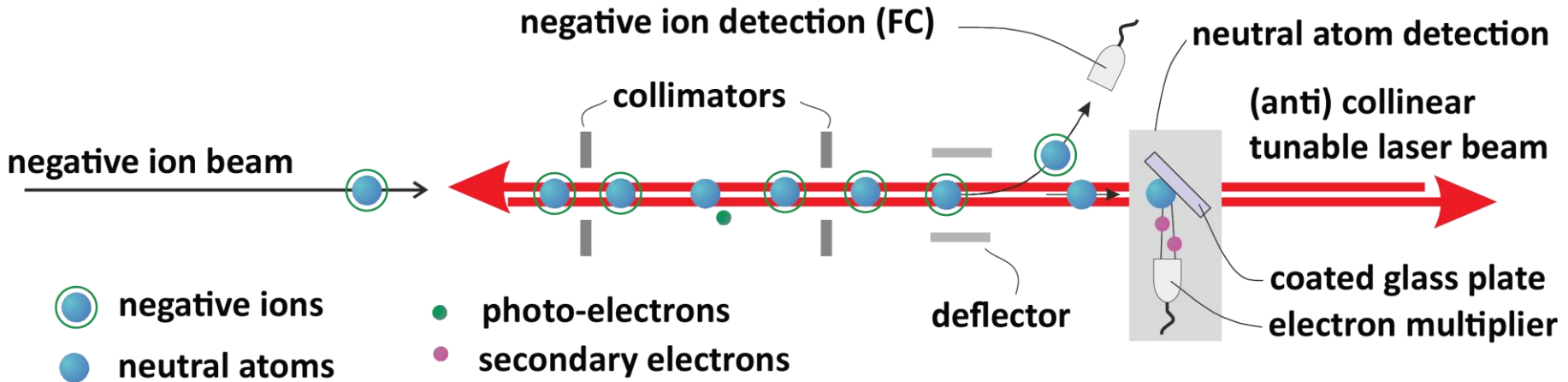
Determination of the electron affinity of astatine and polonium by laser photodetachment

January 12, 2016

S. Rothe^{1,2,3}, J. Champion⁴, K. Chrysalidis^{1,5}, T. Day Goodacre^{1,3}, V. Fedosseev¹, N. Galland⁶, D. Hanstorp², R. Heinke⁵, U. Köster⁷, T. Kron⁵, Y. Liu⁸, B. Marsh¹, G. Montavon⁴, E. Renault⁶, A. Ringwall-Moberg², R. Rossel⁵, C. Seiffert¹, J. Sundberg^{1,2}, J. Welander², and K. Wendt⁵

- No experimental value for EA(At) yet
- Scattering of all theoretical predictions and extrapolations ~1 eV

The Method: Collinear laser photodetachment

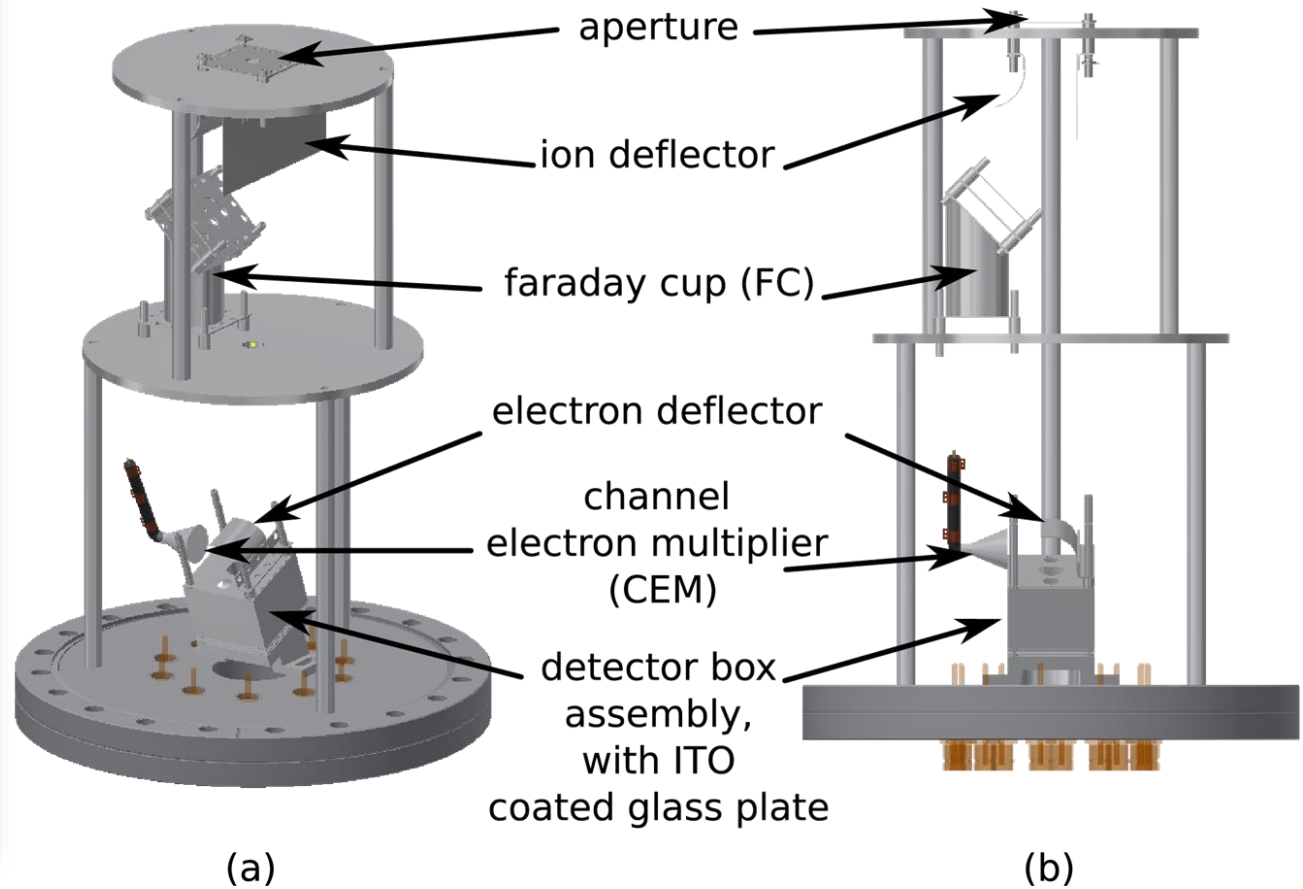
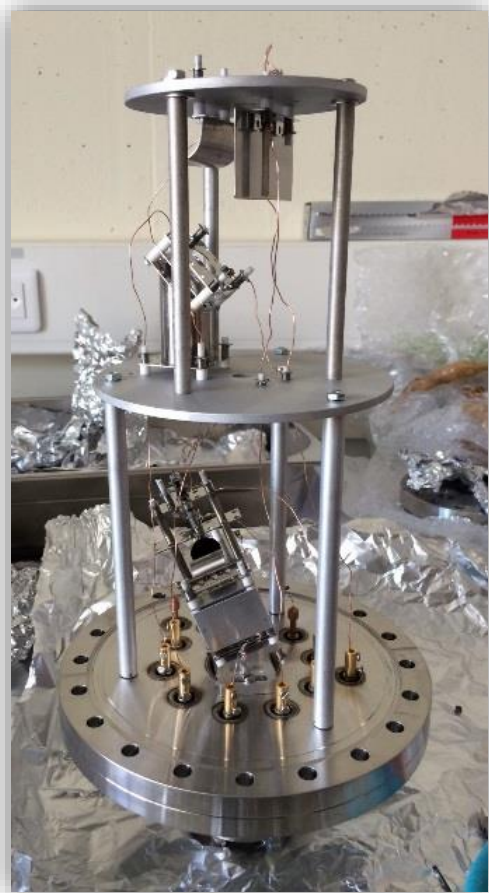


Wigner law:

$$\sigma = k (E - E_0)^{1+1/2}$$

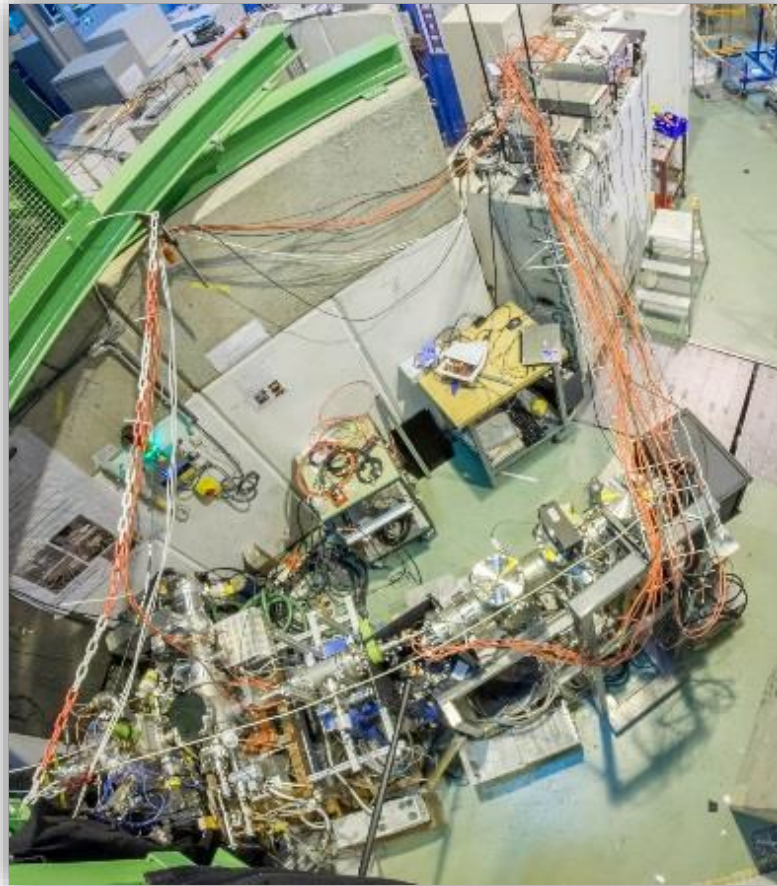
$l = 0$ for s-wave outgoing electron

The neutral atom detector



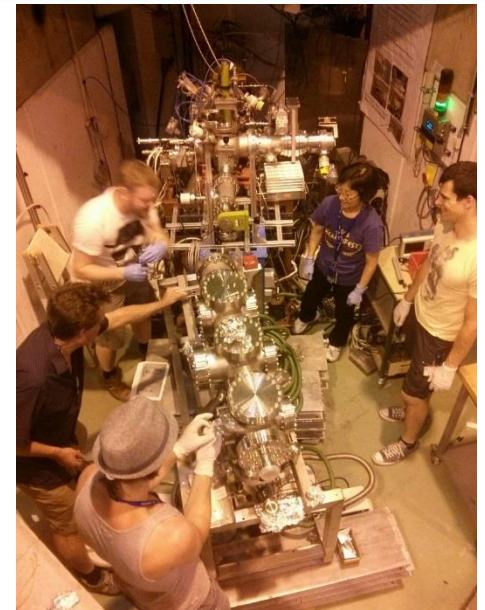
A new apparatus: GANDALPH 2015

Gothenburg ANion Detector for Affinity measurements by Laser PHotodetachment

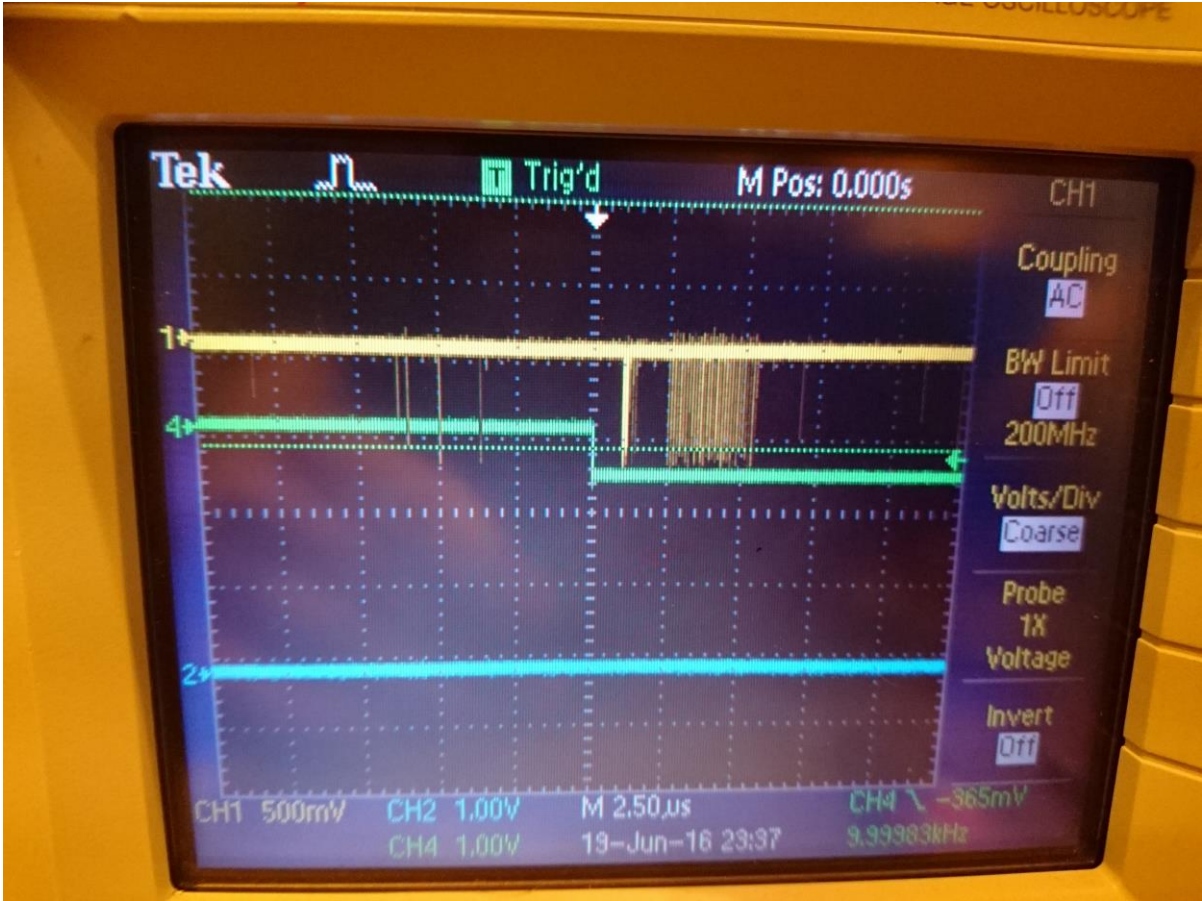


- Built in spring 2015 in Gothenburg
- Moved to ISOLDE, June 2015

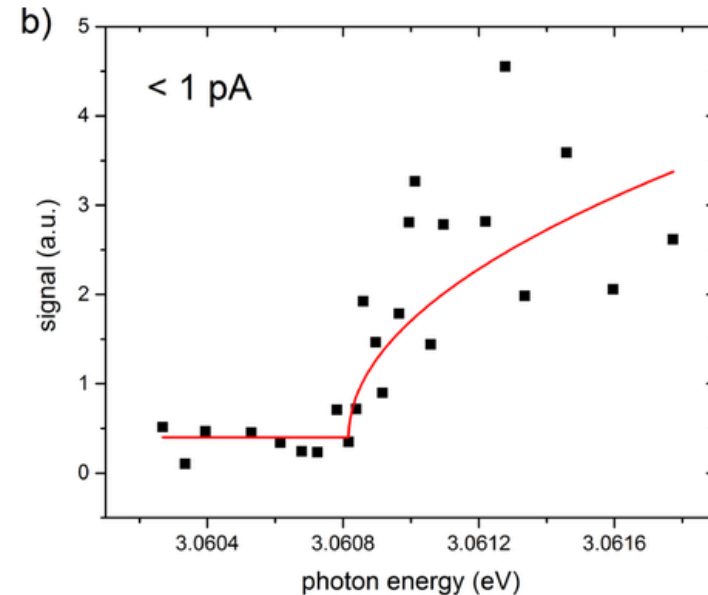
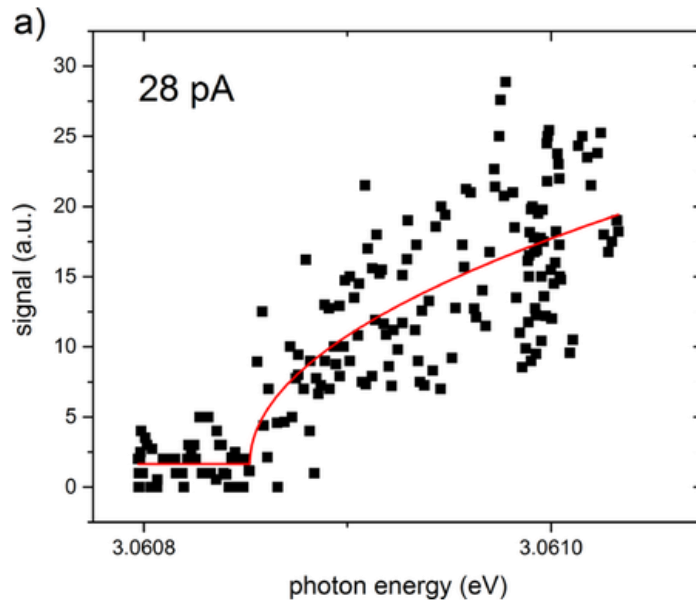
GANDALPH 2015



The signal



Results 2015: photodetachment of stable ^{127}I



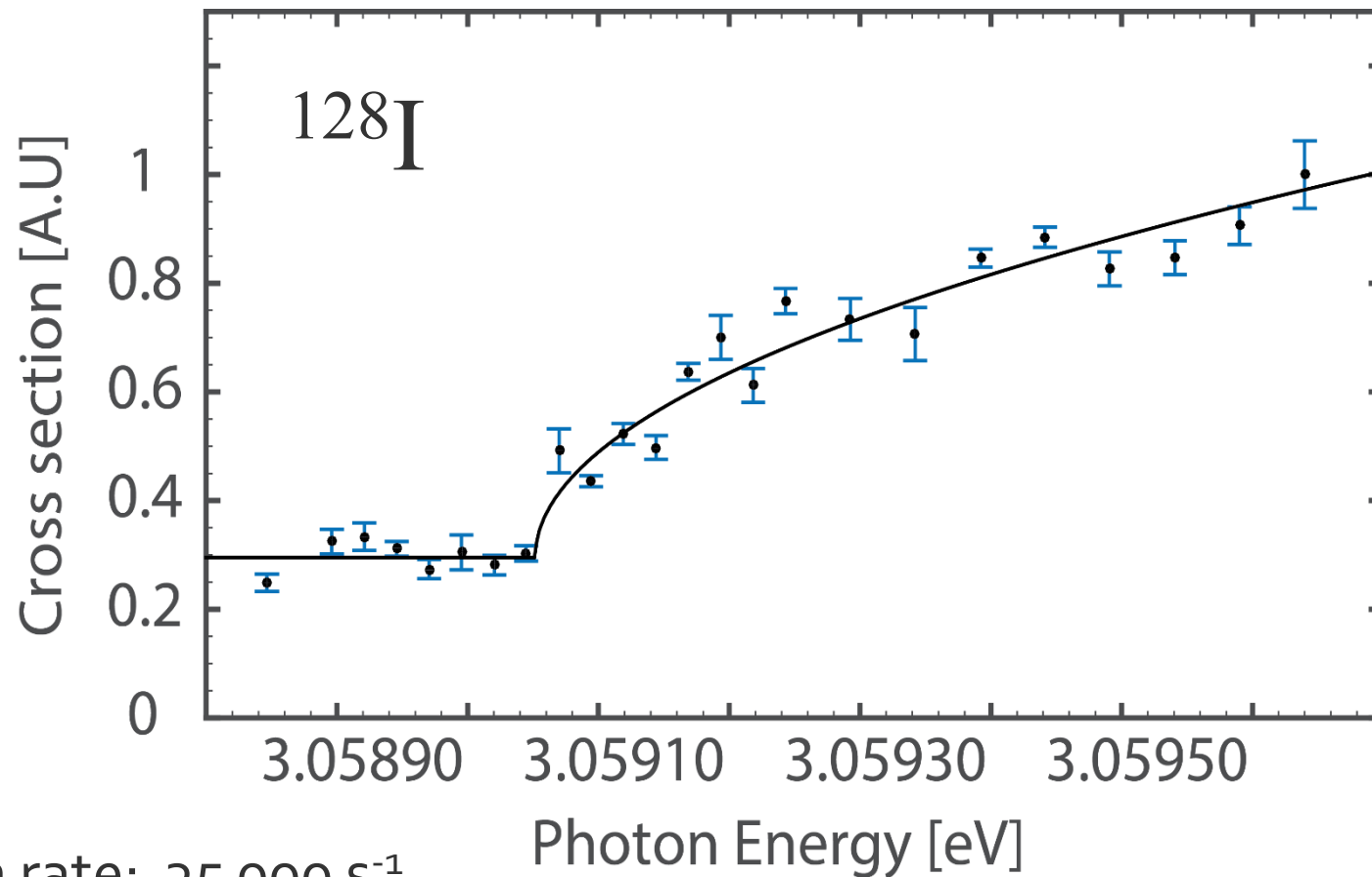
- First photodetachment signal of I^- at ISOLDE
- Threshold still measurable at very low ion rates (<1 pA, FC noise)
- No radiogenic iodine or astatine beams for LOI I-148

Upgraded GANDALPH 2016 vs 2015

	2015	2016
Baking	no	yes
Vacuum	2×10^{-8} mBar	$<10^{-10}$ mBar
DAQ	Lecroy	SRS Photon Counter
Transmission	8%	25%
Frame	Bricolage	ITEM profiles



2016 – first affinity measurement on radioactive ions



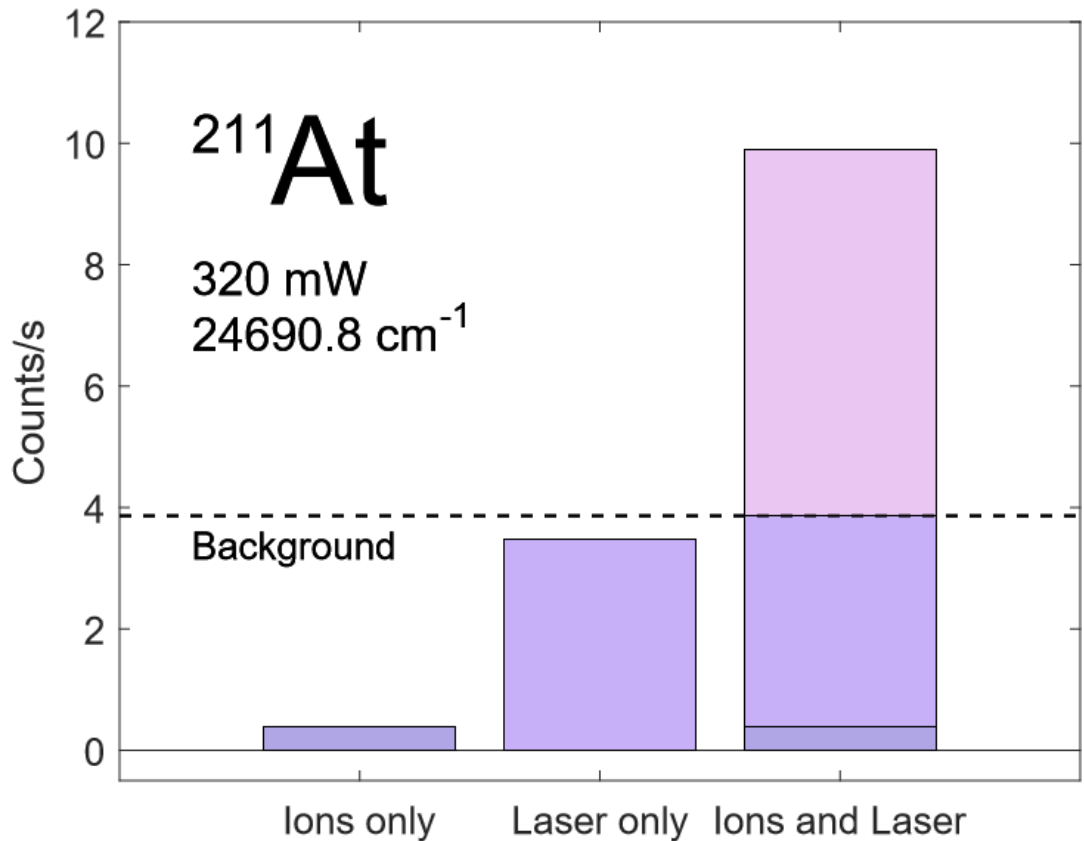
- Ion rate: $35\,000\text{ s}^{-1}$
- Measurement time: 39 min
- Extracted EA: $3.059\,052(38)\text{ eV}$

2016 - astatine yields

Mass	Signal (cps)	Background (cps)
204	2000	200
206	2500	170
208	1700	170
209	4000	150
210	4800	150
211	6200	170
212	about 800	170
213	170	170
217	170	170
218	170	170
219	170	170
204	2900	200

- 1.6uA p on target
- Signal = secondary electrons from ion beam on glass plate
- Increased background
- ²¹¹At most intense beam

First photodetachment signal of astatine



- Ion rate ($^{211}\text{At}^-$): $6\,200\text{ s}^{-1}$
- Measurement time: $\sim 1\text{ h}$
- Upper limit for EA(At): $< 3.06\text{ eV}$

Conclusion

- GANDALPH has been upgraded – better vacuum better ergonomics
- First electron affinity measurement on a radioactive ion performed
- First laser photodetachment of astatine negative ions achieved

Outlook: GANDALPH 201X

- Improved beam diagnostics
 - Dedicated ion detector for tuning
 - Segmented collimators
- Re-condition the ion pump
- Fix broken vacuum feed-troughs
- File a beam time request for P-462

Astatine EA team (2015)



I-148 Collaboration

Laser photodetachment of radioactive $^{128}\text{I}^-$

Sebastian Rothe^{1,2,3}, Julia Sundberg^{1,2}, Jakob Welander², Katerina Chrysalidis^{1,4}, Thomas Day Goodacre^{1,3}, Valentin Fedosseev¹, Oliver Forstner⁵, Reinhard Heinke⁴, Karl Johnston¹, Tobias Kron⁴, Ulli Köster⁶, Yuan Liu⁷, Bruce Marsh¹, Annie Ringvall-Moberg^{1,2}, Ralf Erik Rossel^{1,4}, Christoph Seiffert¹, Dominik Studer⁴, Klaus Wendt⁴, and Dag Hanstorp²

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³ School of Physics and Astronomy, The University of Manchester, Manchester, UK

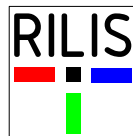
⁴ Institut für Physik, Johannes Gutenberg-Universität, Mainz, Germany

⁵ Friedrich Schiller Universität, Jena, Germany

⁶ Institut Laue-Langevin (ILL), Grenoble, France

⁷ Physics Division, Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee, USA

Submitted to J. Phys. G



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JOHANNES GUTENBERG
UNIVERSITÄT MAINZ

Thanks

- Isolde technical team for intense ion source tests and swapping ISOLDE to negative mode
- ISOLDE experimental setups (CRIS, COLLAPS, ISOLTRAP, SSP, ...) for helping out with equipment – especially for GANDALPH 1.0
- Vacuum group for assistance with baking of GANDALPH 2.0



/eof