

# Laser cooling of anions for ultracold antihydrogen

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Nuclear Physics

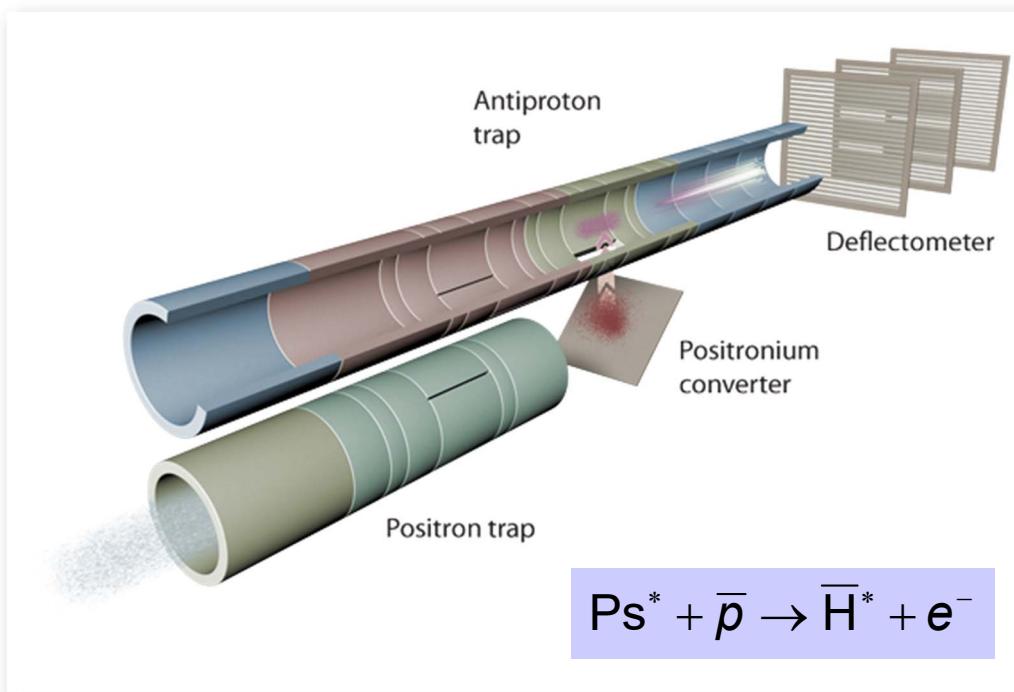


European Research Council

# AEGIS experiment

- AEGIS: Antimatter Experiment: Gravity, Interferometry, Spectroscopy
- Main goal: **Measurement of  $g$**  with 1% precision on antihydrogen
- Requirements / challenges:
  - Production of a **bunched cold beam of antihydrogen** (100 mK)
  - Measurement of vertical beam deflection (10 μm drop over 1 m)

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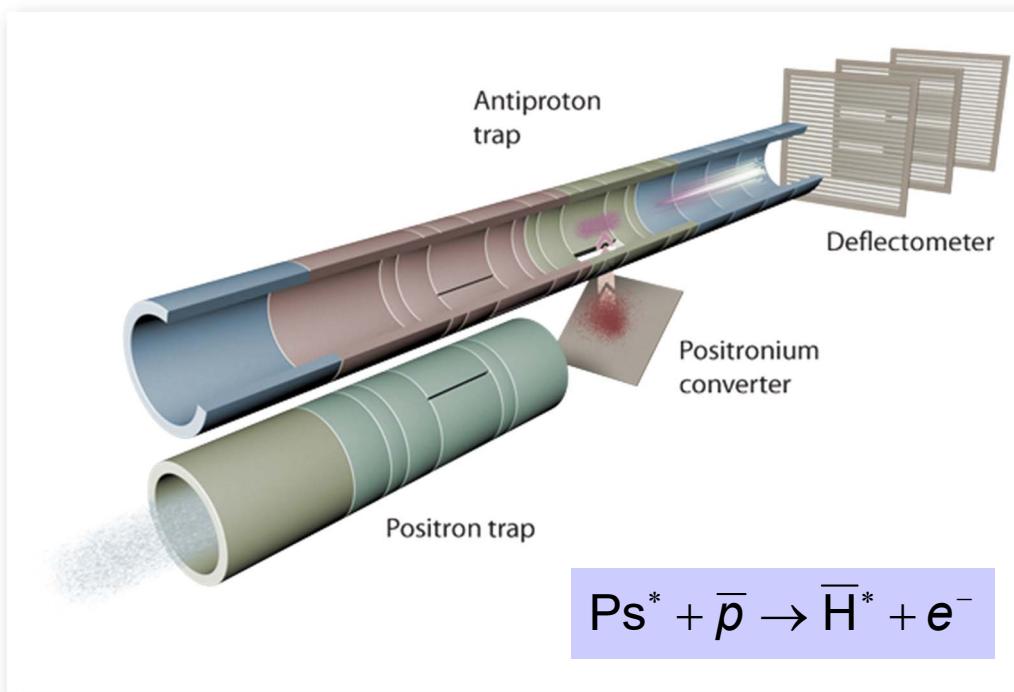


[A.K. et al. (AEGIS Collaboration),  
Nucl. Instrum. Methods B **266** (2008) 351]

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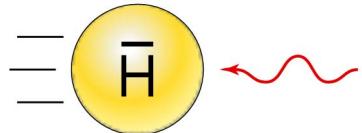
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# Towards ultracold antihydrogen

- Proposed techniques:

1

Direct laser cooling of  $\bar{H}$



- requires cw Lyman- $\alpha$  laser for lowest temperatures
- doesn't increase  $\bar{H}$  yield or trapped fraction

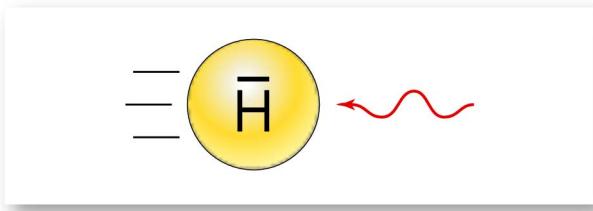
[P. H. Donnan *et al.* (ALPHA Collaboration),  
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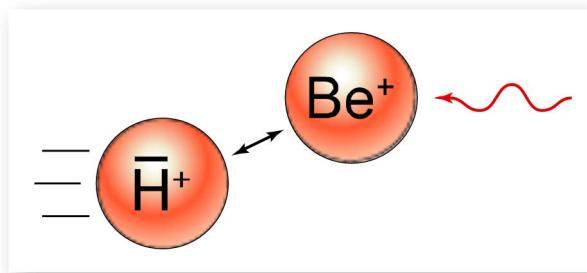


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Sympathetic cooling of  $\bar{H}^+$  by laser-cooled cations



- low yield of  $\bar{H}^+$
- complicated scheme

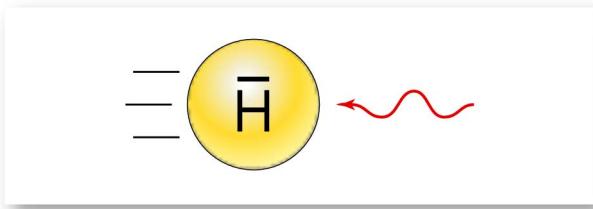
[L. Hilico *et al.* (GBAR Collaboration),  
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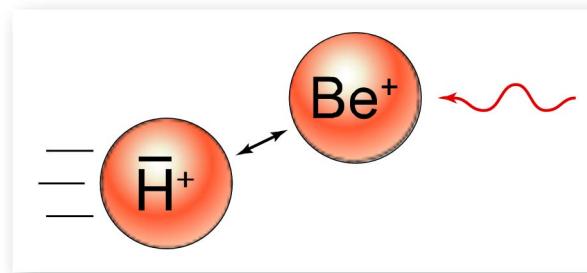


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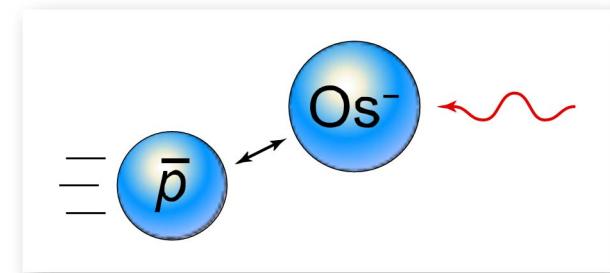


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3

Sympathetic cooling of  $\bar{p}$  by laser-cooled anions

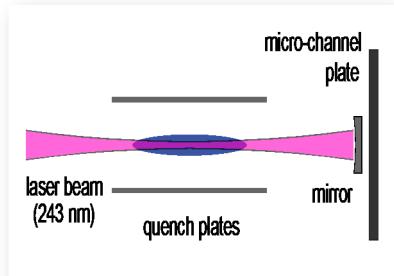


- laser cooling of anions has never been achieved

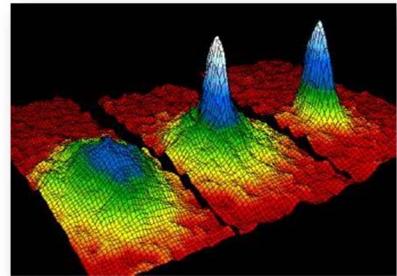
[A. K. & J. Walz,  
New J. Phys. **8** (2006) 1460269]

# Laser cooling of atoms and ions

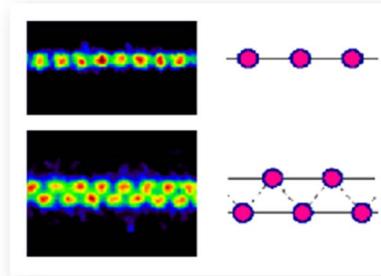
- Game-changing technique in many areas of AMO physics



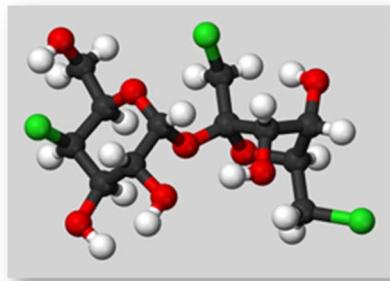
Atomic spectroscopy



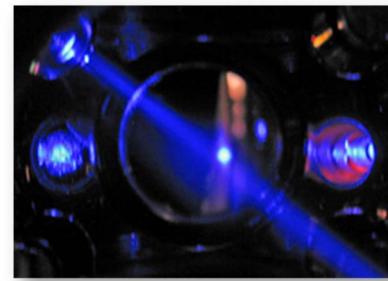
Bose–Einstein condensation



Coulomb crystals



Molecular ions



Atomic clocks

- Laser cooling is an established technology

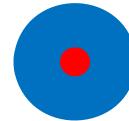
- Positive ions      1978 (Toschek/Wineland)
- Neutral atoms      1985 (Chu)
- Negative ions      ???

# Anion formation and binding

- Binding in atomic systems:

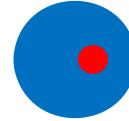
Coulomb potential

$$\propto r^{-1}$$



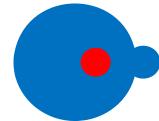
Polarization

$$\propto r^{-4}$$



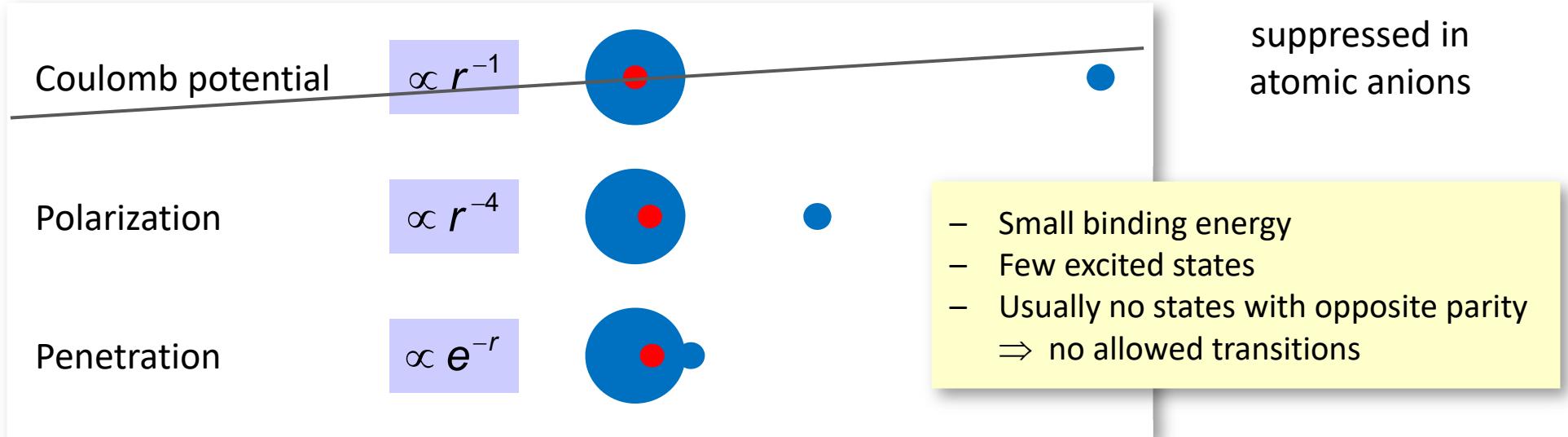
Penetration

$$\propto e^{-r}$$



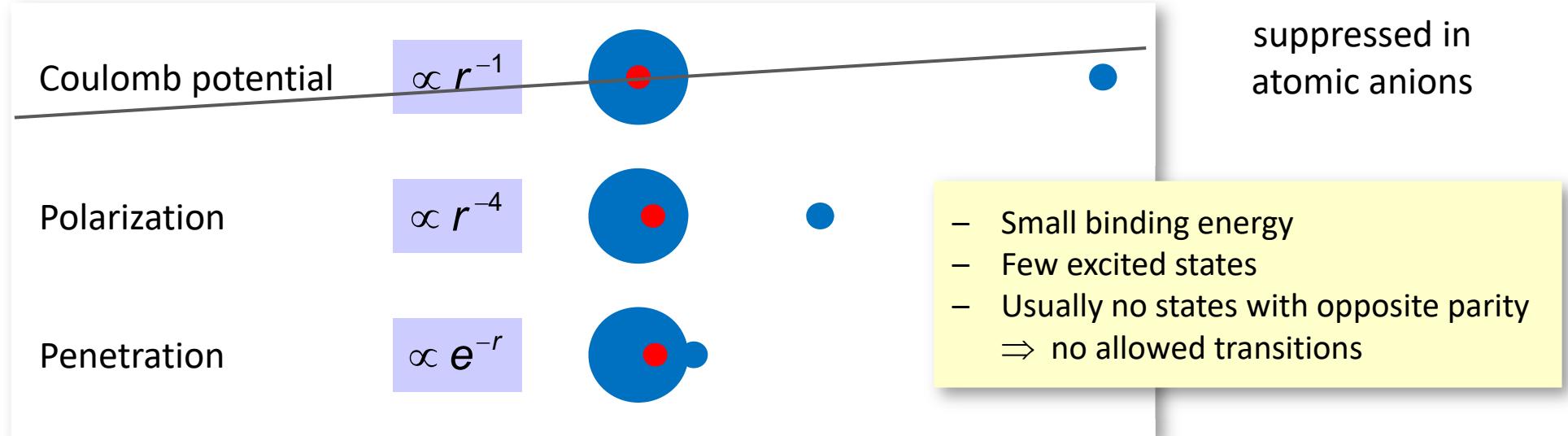
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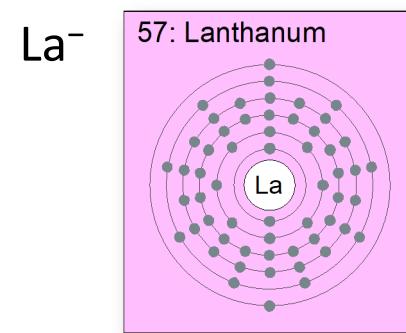
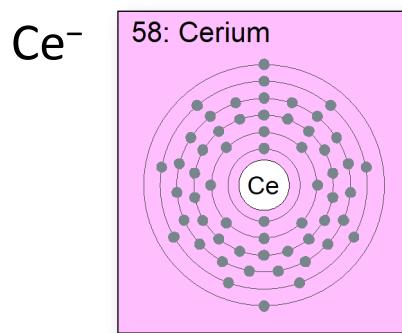
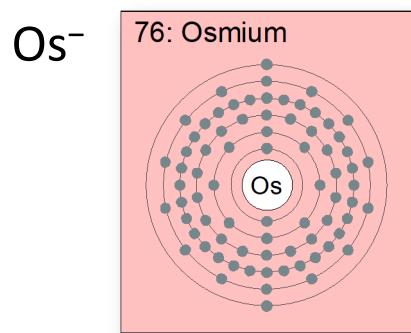


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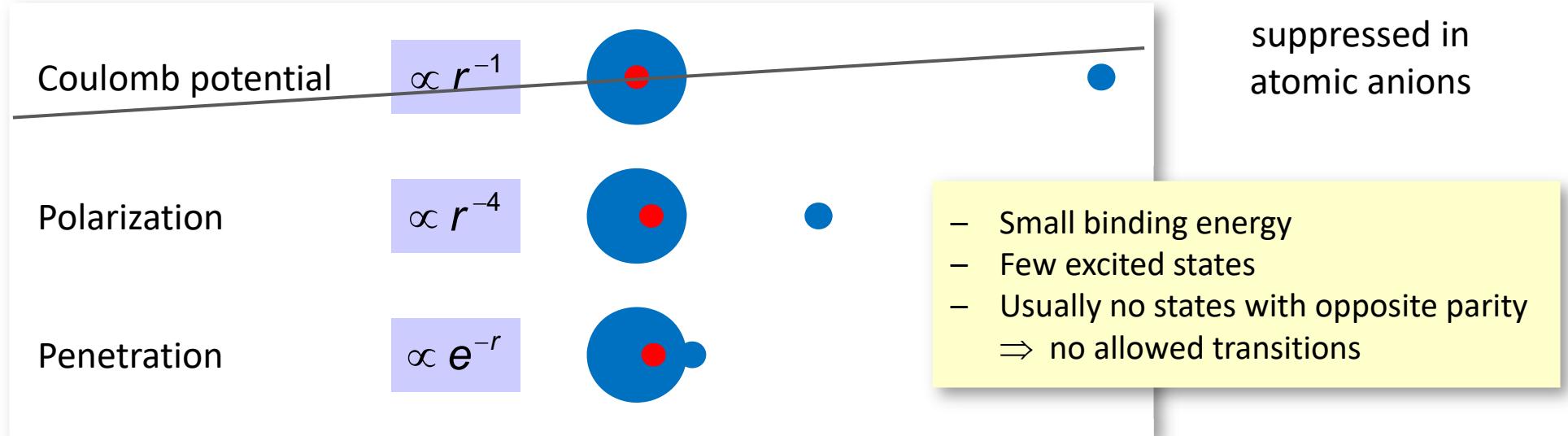


- Known atomic anions with *opposite-parity* excited states

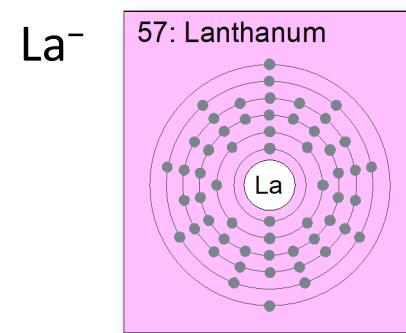
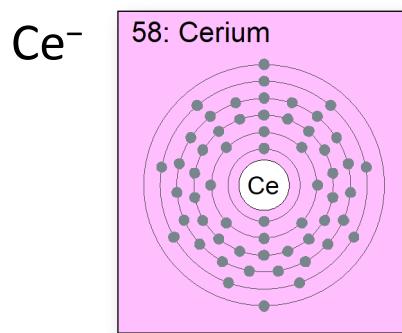
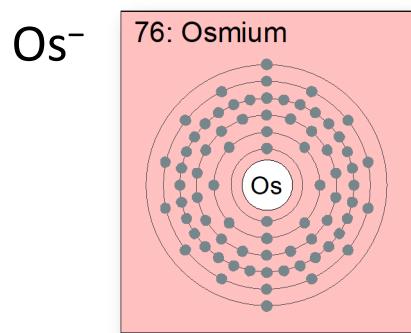


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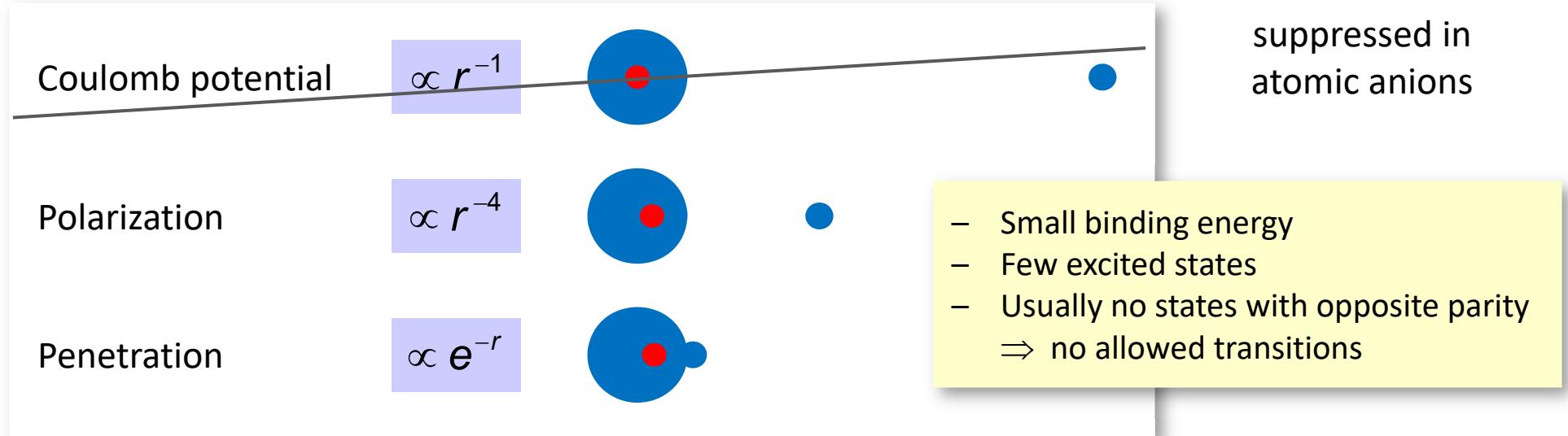
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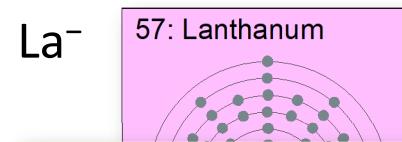
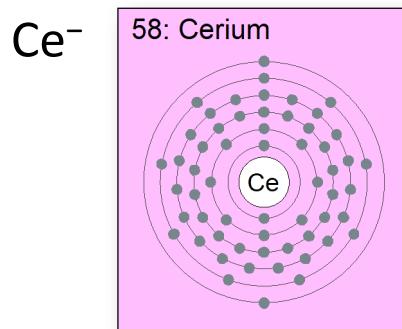
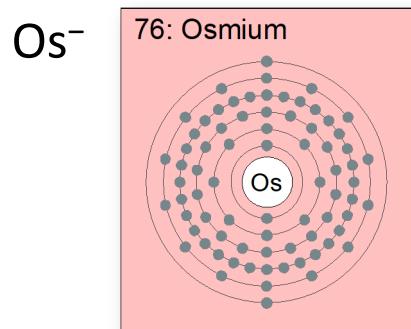
$$\Gamma \approx 50 \text{ Hz}$$

# Anion formation and binding

- Binding in atomic systems:



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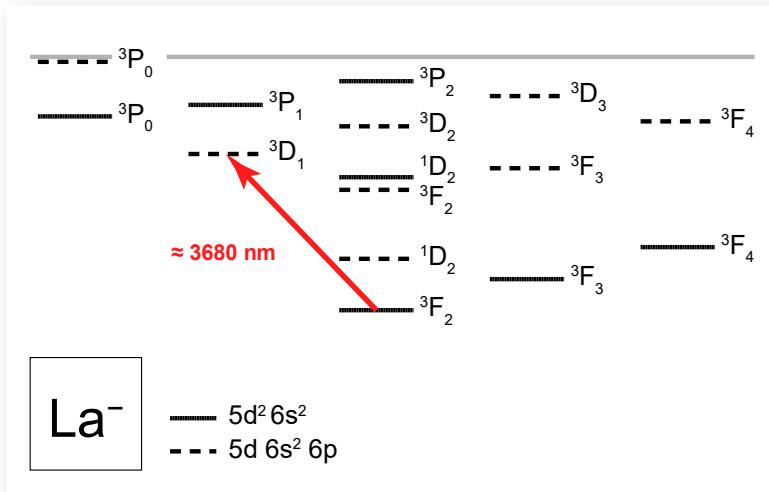


For a similar approach using molecular ions, see Julian Fesel's talk, Fri 10:20

$$\Gamma \approx 50 \text{ Hz}$$

# Laser cooling candidate La<sup>-</sup>

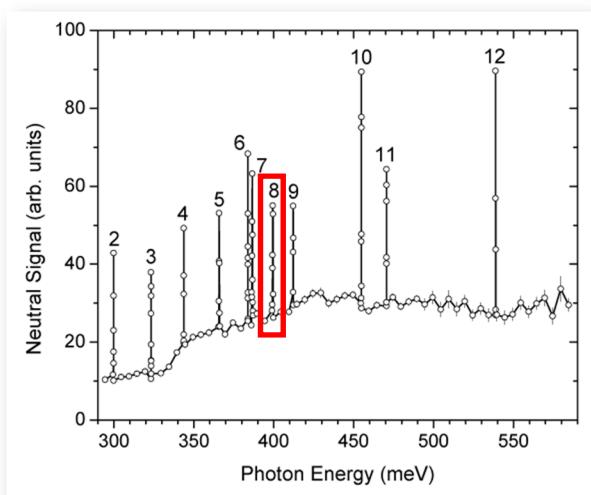
- 2010: Theoretical prediction of bound – bound transitions in La<sup>-</sup>:



Transition rate  $\Gamma \approx 4.5 \text{ kHz}$   
99.98% closed transition

[S. M. O'Malley & D. R. Beck,  
Phys. Rev. A **81** (2010) 032503]

- 2014: Experimental demonstration:



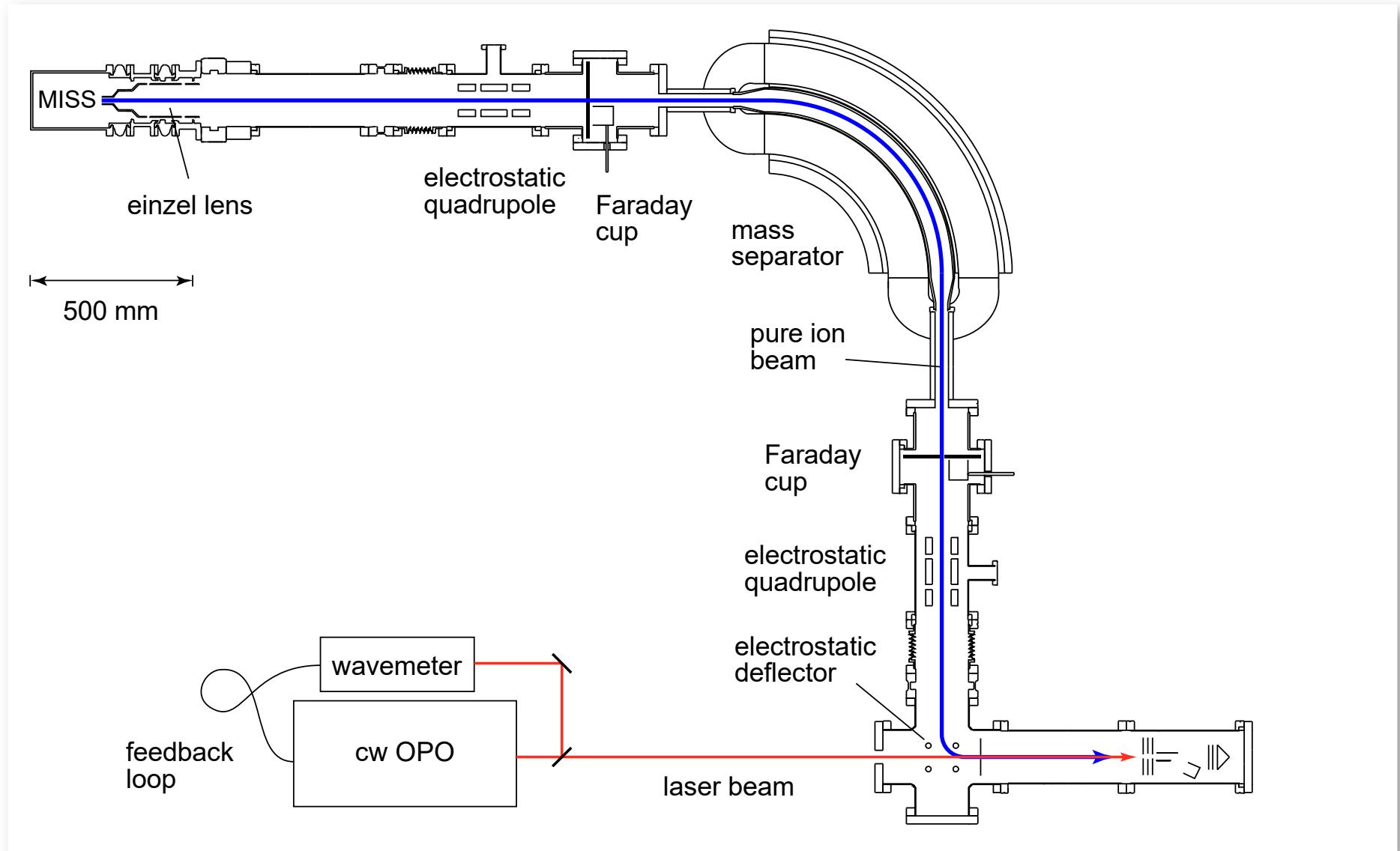
Laser spectroscopy with tunable  
OPO pumped by pulsed Nd:YAG

Peak 8  
 $E = 399.42(3) \text{ meV}$   
 $\lambda = 3104.4(2) \text{ nm}$

[C. W. Walter & N. D. Gibson,  
Phys. Rev. Lett. **113** (2014) 063001]

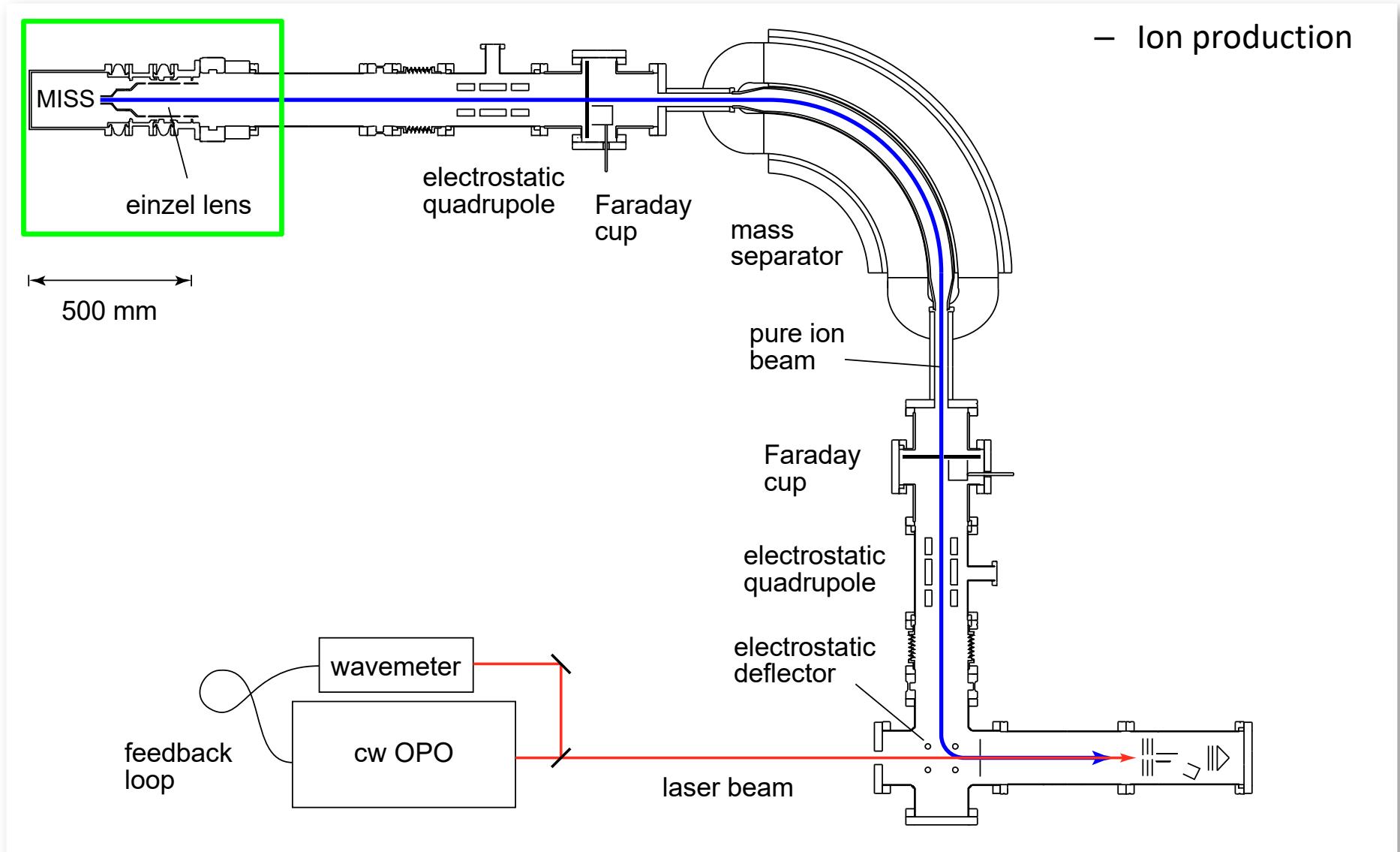
# Laser spectroscopy setup

- Overall schematic



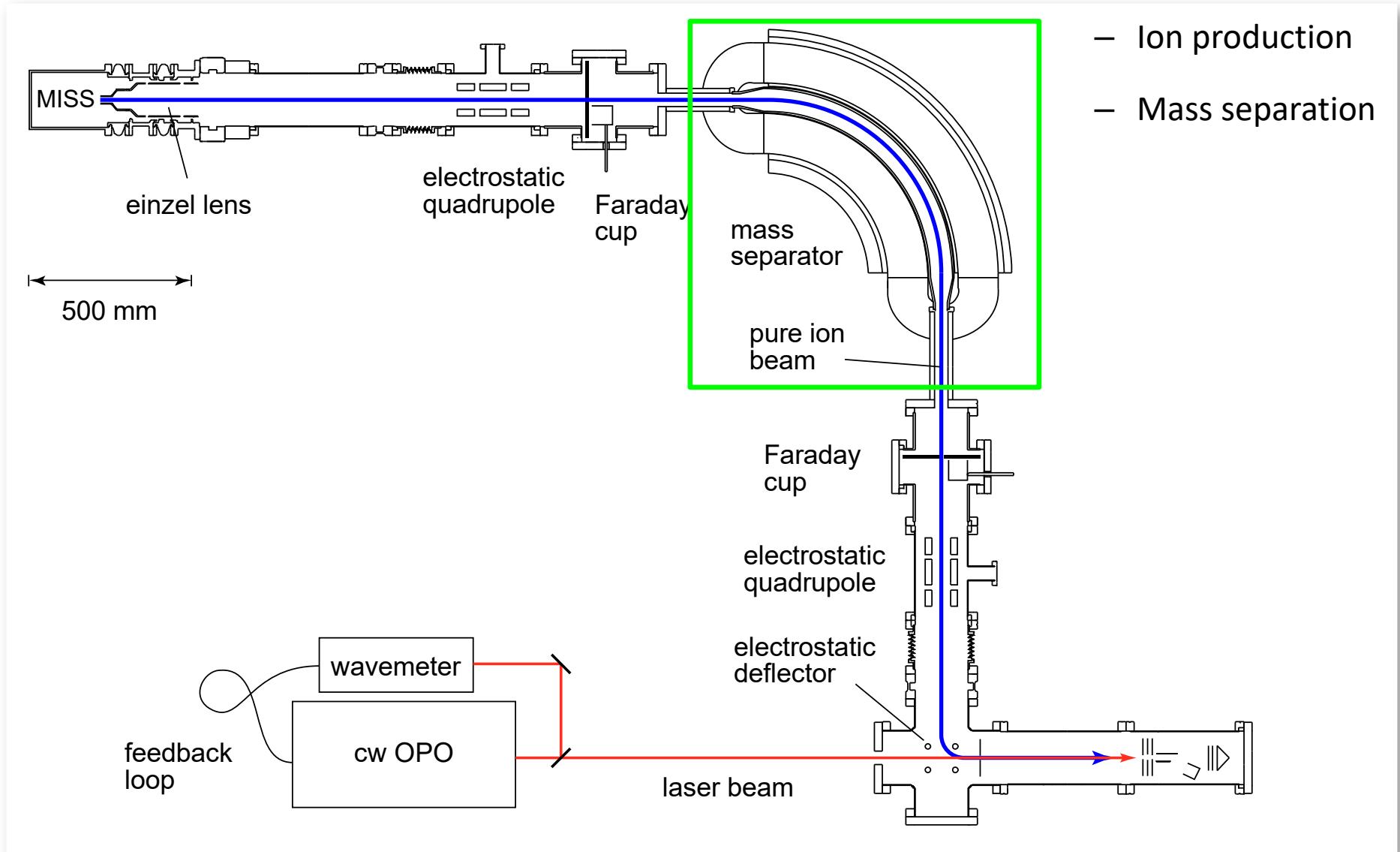
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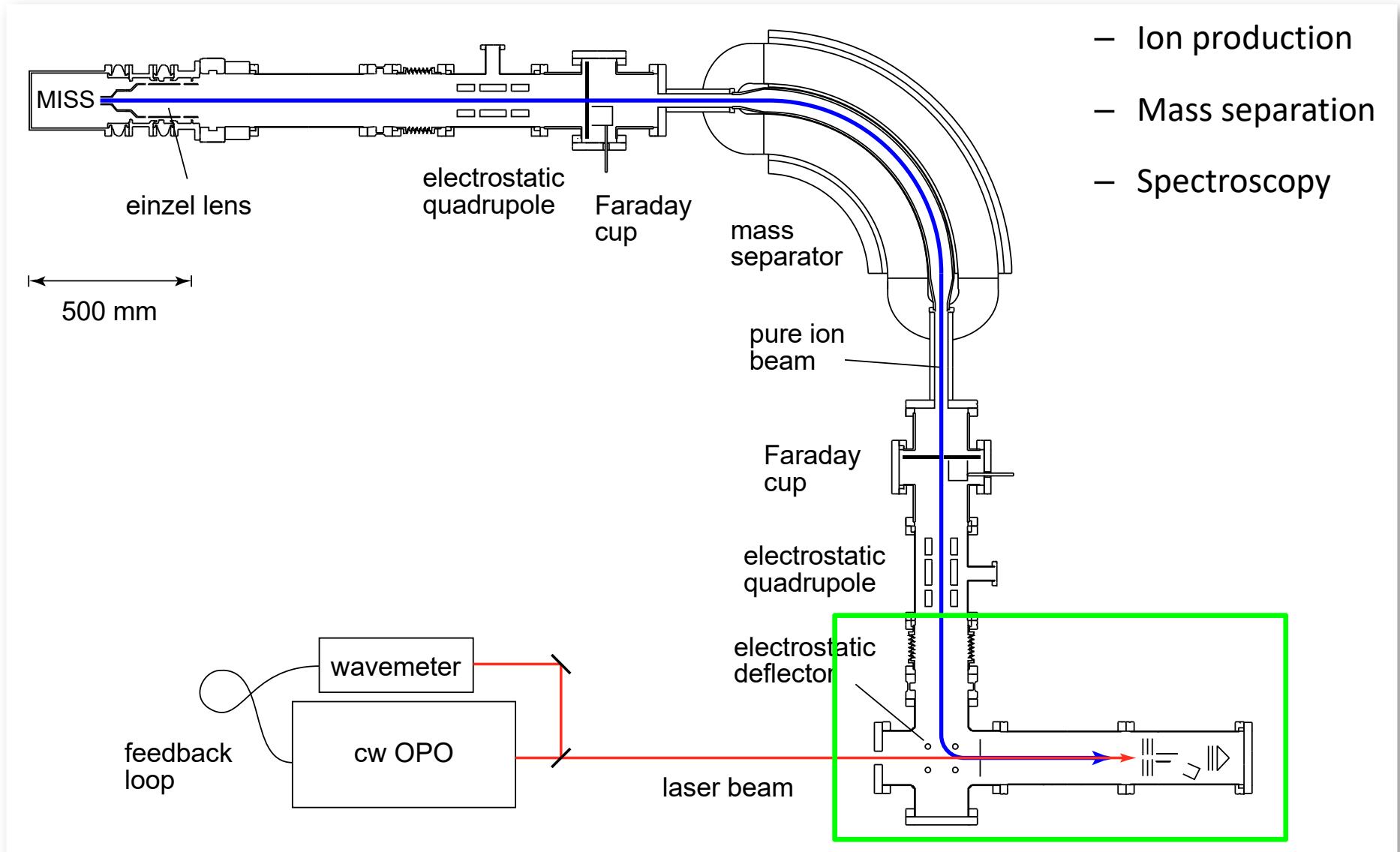
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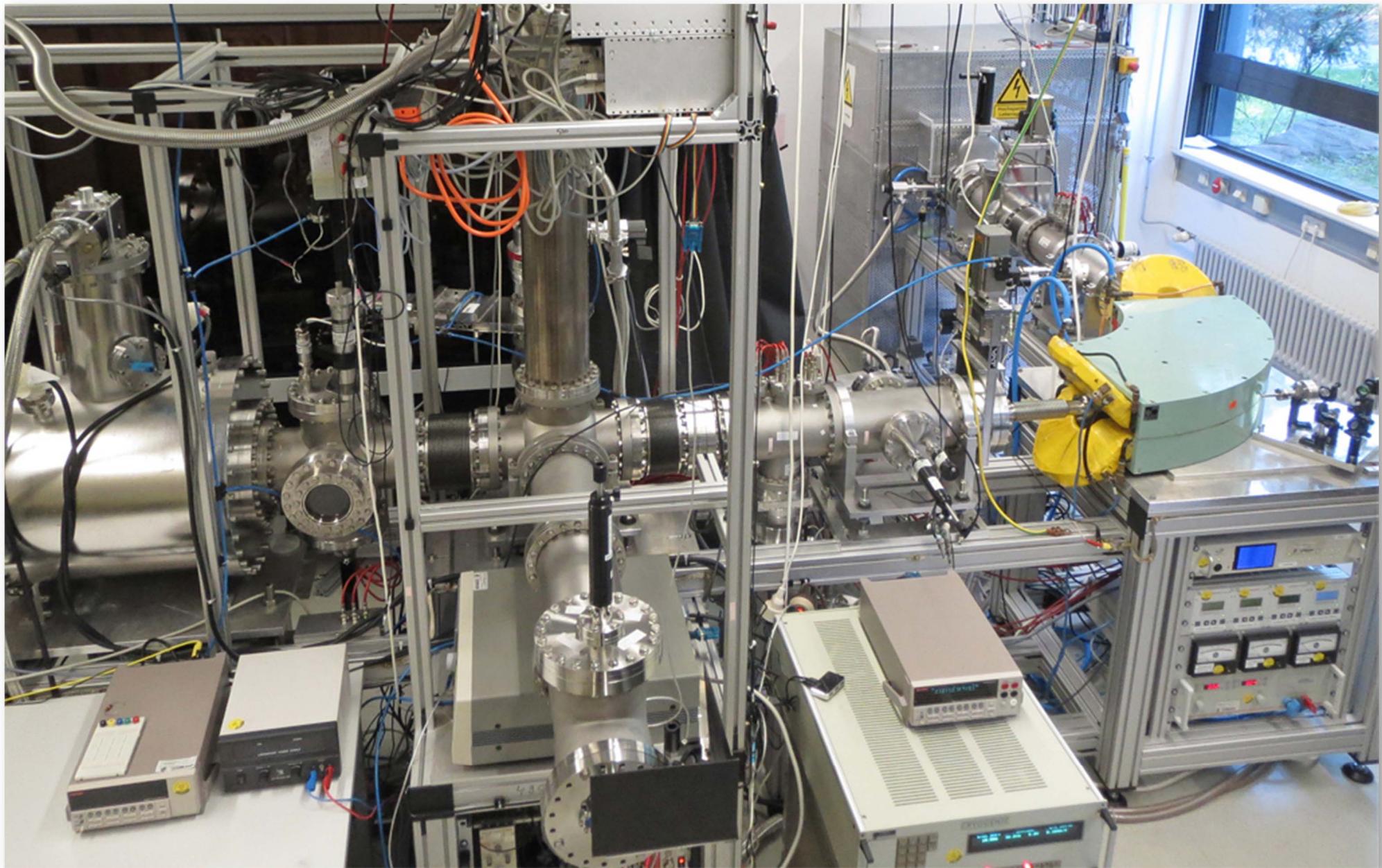


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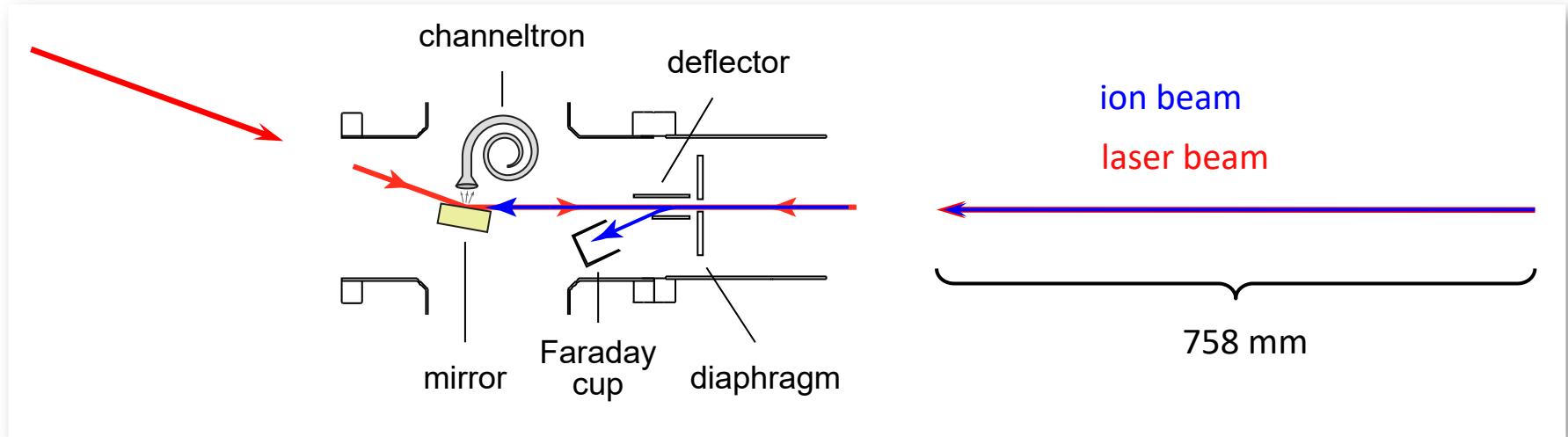


# Laser spectroscopy / trap setup

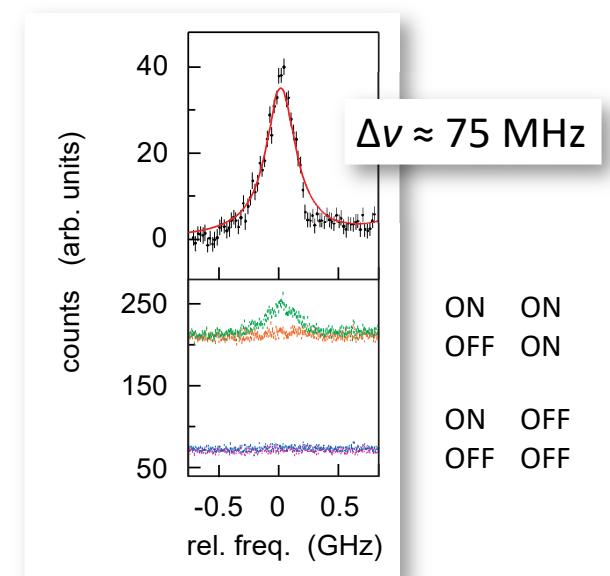


# Collinear laser spectroscopy of La<sup>-</sup>

- Measurement principle

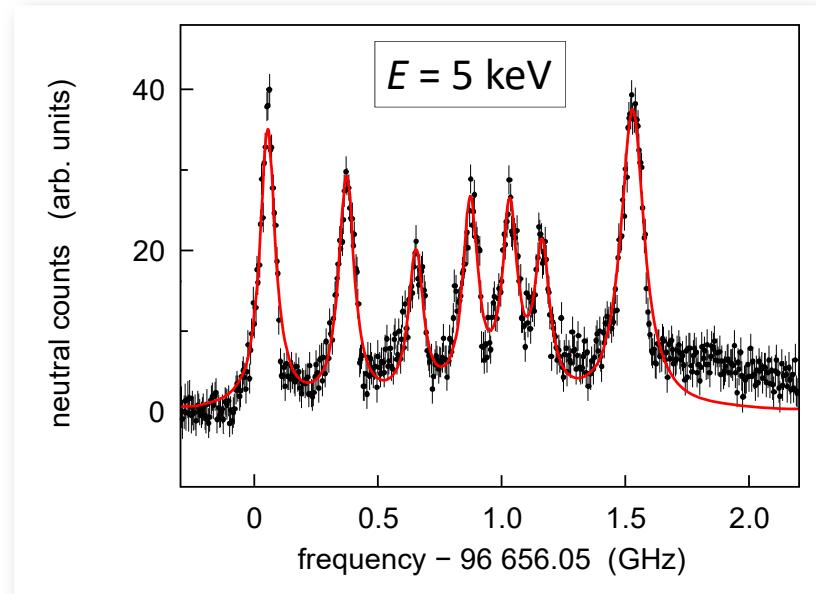


- Ion beam and laser beam are superimposed
- When laser in resonance, ions are excited, neutralized by photodetachment
- Neutral atoms are detected on forward channeltron
- 2nd laser anticollinear (out of resonance) to avoid power broadening

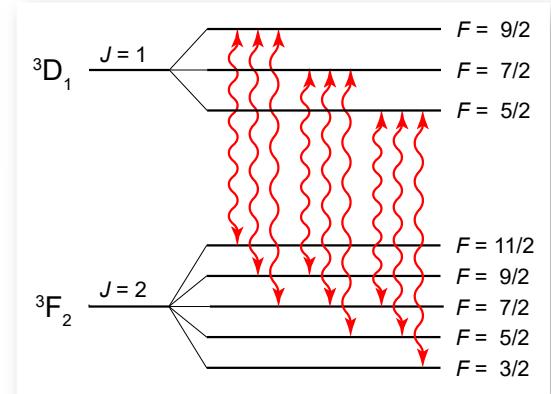


# Hyperfine structure of La<sup>-</sup>

- Resonance with resolved HFS (7 out of 9 peaks)



La139  
7/2<sup>+</sup>  
99.9



[E. Jordan *et al.*,  
Phys. Rev. Lett. **115** (2015) 113001]

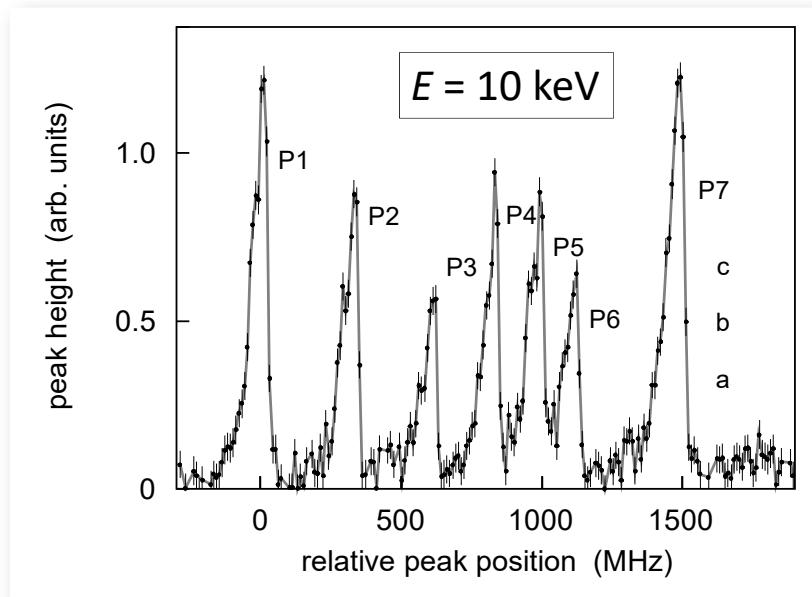
- 16 resonances recorded at beam energies  $\pm 5\ldots 10$  keV
- Center-of-mass frequency/wavelength determined from Doppler-corrected resonances:

$$\nu = 96.592,80(10) \text{ THz}$$
$$\lambda = 3.103,6729(32) \mu\text{m}$$

- Precision improved by factor  $\approx 75$ , deviation  $2\sigma$

# HFS coefficients

- Determination of A and B coefficients from fit



Hyperfine splitting:

$$\Delta E_{M1+E2} = \frac{A}{2} C + \frac{B}{2} D$$

where  $C, D$  are functions of quantum numbers  $I, J, F$

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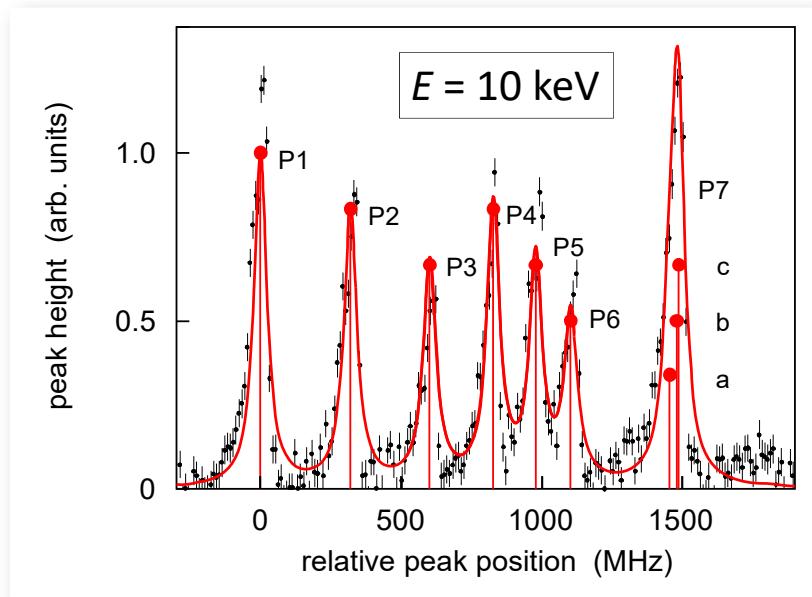
- Experimental HFS coefficients from fit to 9 best resonances

$$\begin{aligned} A_g &= 146.8(3) \text{ MHz} & B_g &= 36(9) \text{ MHz} \\ A_e &= 110.6(7) \text{ MHz} & B_e &= 5(1) \text{ MHz} \end{aligned}$$

⇒ Unambiguous assignment of peaks to HFS transitions  
(must fix relative heights for Peaks 7a–c to obtain their positions)

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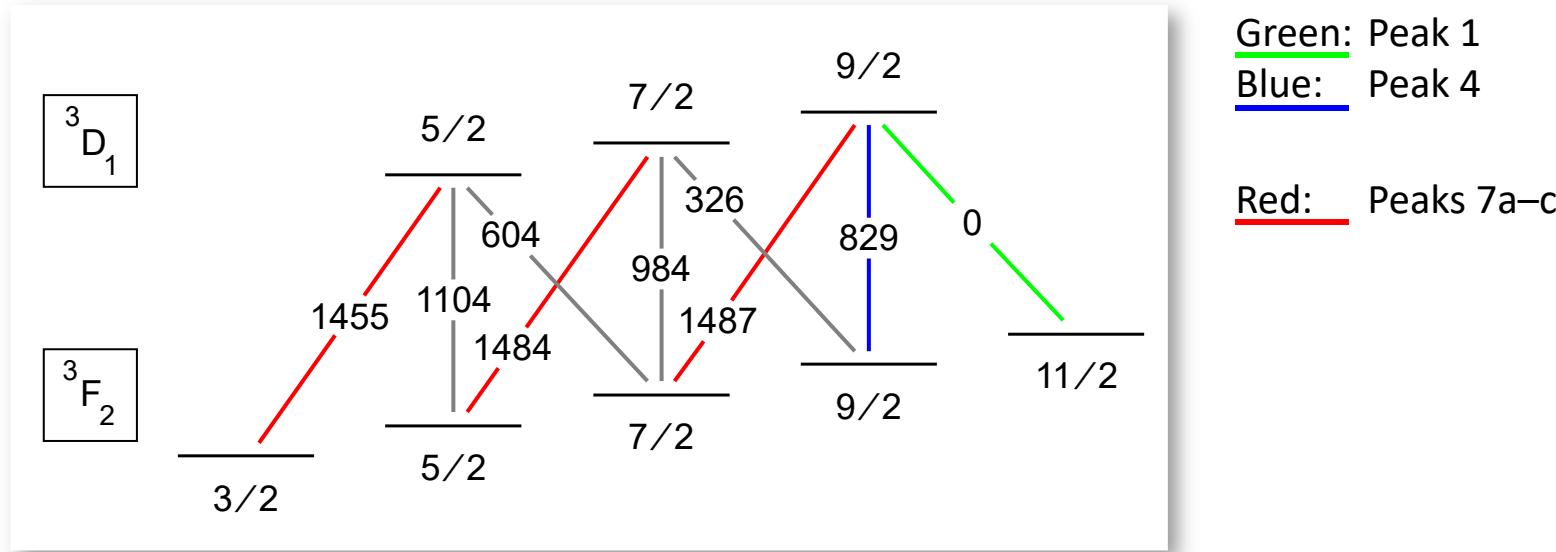
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# La<sup>-</sup> laser cooling scheme

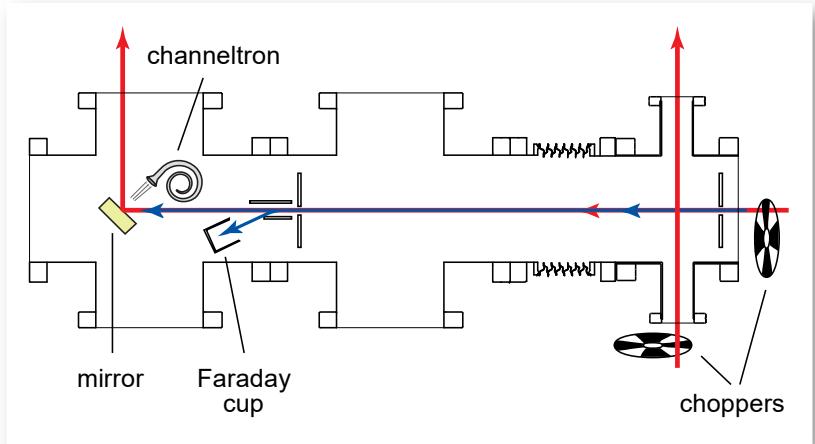
- HFS energy level diagram (no magnetic field)



- Cooling with  $F = 11/2 \rightarrow 9/2$  transition (Peak 1)  
 $\Delta F = 0, -1$  decays must be repumped
- Only three laser frequencies required (3rd laser > 40 MHz)  
→ Sideband generation by EOMs implemented
- Initially work in **Paul trap** to avoid additional Zeeman splitting

# Cross-section measurement

- Combined transverse/collinear spectroscopy:



$$\dot{N}_g = -\sigma_0 \varphi N_g + (\sigma_0 \varphi + \tau^{-1}) N_e$$

$$\dot{N}_e = \sigma_0 \varphi N_g - (\sigma_0 \varphi + \tau^{-1} + \sigma_d \varphi) N_e$$

$$\dot{N}_d = \sigma_d \varphi N_e$$

$\sigma_0, \sigma_d$  – excited / detached cross-section

$\tau$  – lifetime of excited state

$\varphi$  – photon flux

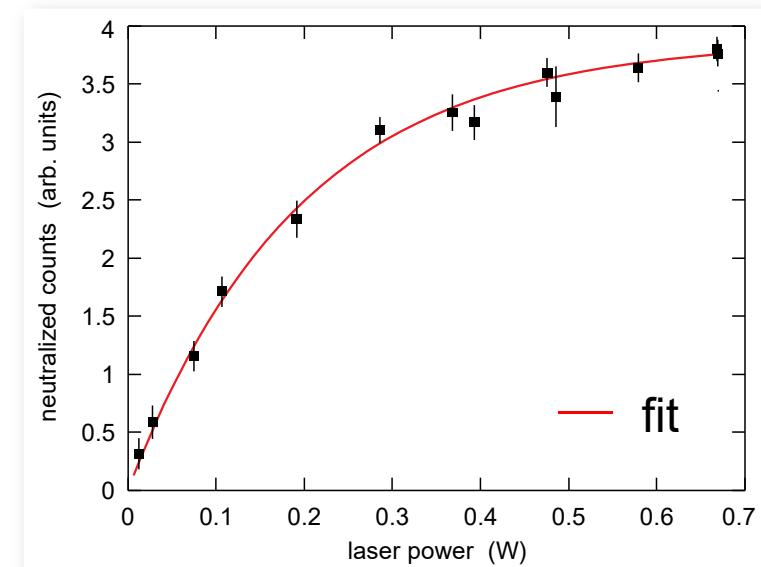
- rate equation for ground, excited state and detached populations
- analytical expressions for  $N_e$ ,  $N_d$  measured:  $N_d$
- resonant cross-section:

$$\sigma_0 = 1.0(1) \times 10^{-12} \text{ cm}^2$$

$$\Gamma \approx 9.7 \text{ kHz}$$

(factor 200 higher than in  $\text{Os}^-$ )

[G. Cerchiari *et al.*, accepted for publ. in Phys. Rev. Lett.]



# Decay rates and branching ratios

- Calculation of decay rates and branching ratios by hybrid CI+MBPT method

Upper level	Lower level	A (1/s)	Branching r.
$^3D_1^o$	$^3F_2^e$	45 400	0.999974
	$^1D_2^e$	1.18	0.000026
$^1D_2^e$	$^1D_2^o$	1.95	0.956
	$^3F_2^e$	0.09	0.044
$^1D_2^o$	$^3F_2^e$	168	0.791
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$^3D_2^o$	$^3F_2^e$	4 500	0.1059
	$^3F_3^e$	37 900	0.8924
	$^1D_2^e$	44.1	0.0010
	$^3P_1^e$	27.5	0.0006

⇒ Full information on cooling cycle

[G. Cerchiari *et al.*, accepted for publ. in Phys. Rev. Lett.]

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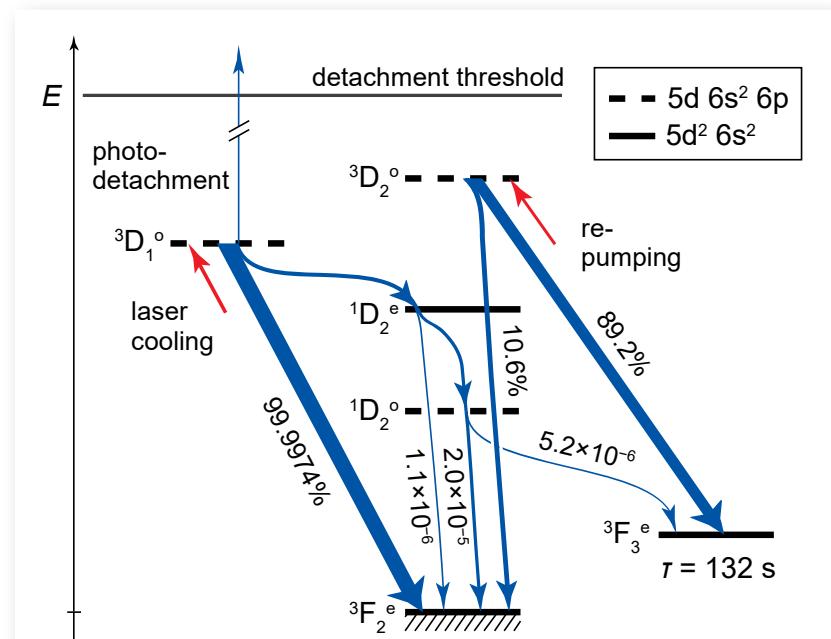
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$$\lambda = 3.104 \mu\text{m}$$

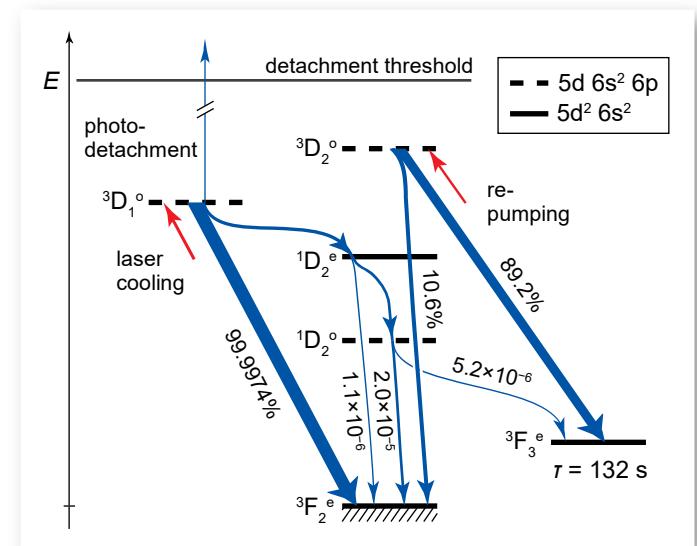
$$\lambda = 3.207 \mu\text{m}$$

⇒ Full information on cooling cycle

[G. Cerchiari *et al.*, accepted for publ. in Phys. Rev. Lett.]

# Discussion

- 1D Doppler laser cooling:
  - Doppler temperature  $T_D = 0.17 \mu\text{K}$
  - cooling from 100 K to  $T_D$  requires scattering of  $\approx 8.4 \times 10^4$  photons  $\Rightarrow$  cooling time 3.7 s
  - after cooling, 40% of ions in metastable ‘dark’ state  ${}^3\text{F}_3^e$ , 10% photodetached
  - if necessary,  ${}^3\text{F}_3^e$  state can be repumped

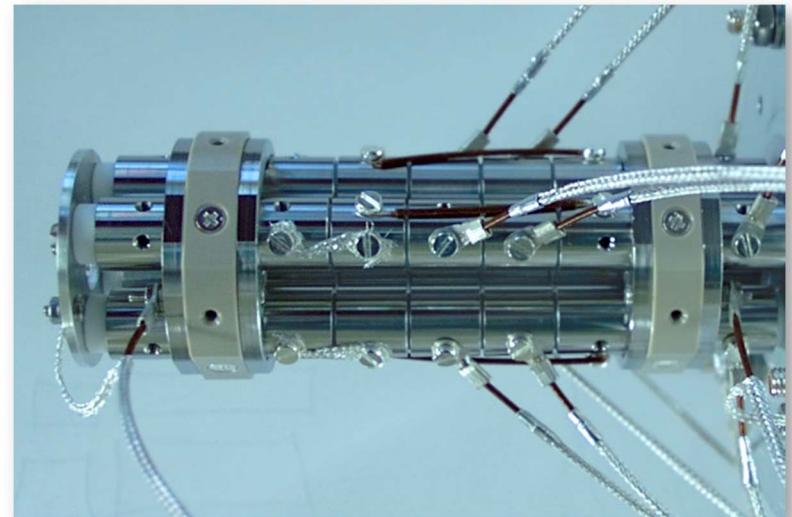


- Strategy towards demonstration of laser cooling:
  - must reach starting temperature of  $\approx 100 \text{ K}$
  - in Penning trap, easily achieved by electron cooling
  - in Paul trap, need new cooling mechanism
    - buffer gas cooling
    - “laser-assisted evaporative cooling”

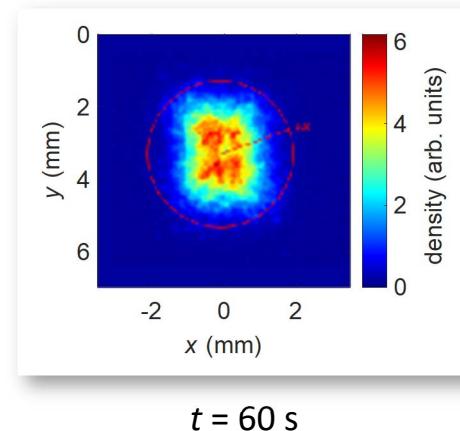
[A. Crubellier, J. Phys. B 23 (1990) 3585]

# Trapping and cooling anions

- Paul trap implemented downstream of spectroscopy section
  - linear RF quadrupole, 5 axial segments
  - radial and axial (oblique) laser access
  - MCP with phosphor screen as diagnostics for axial and transverse phase space

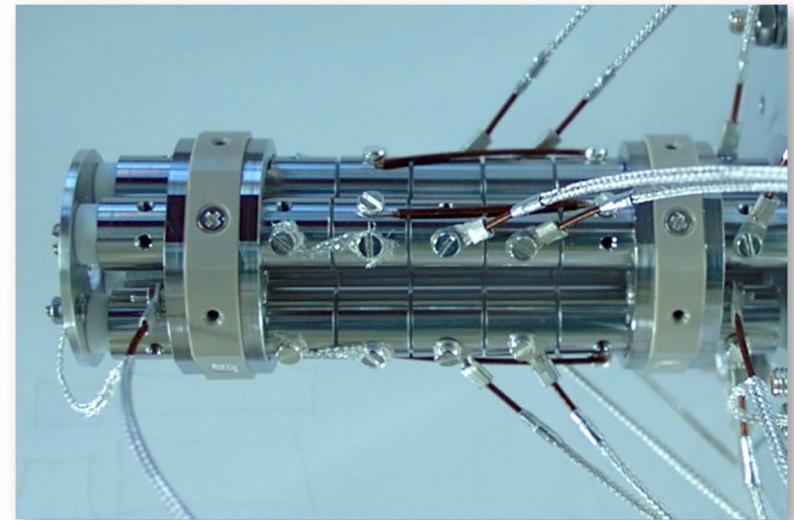


- Trap commissioned and characterized with  $\text{Au}^-/\text{O}^-$  ions:
  - max. number of ions loaded  $\approx 10^5 \dots 10^7$
  - storage time constant  $> \text{min}$
  - laser-assisted evaporative cooling of  $\text{O}^-$ 
    - geometrically selective neutralization with green laser (200 mW @ 532 nm)
    - velocity selection observed



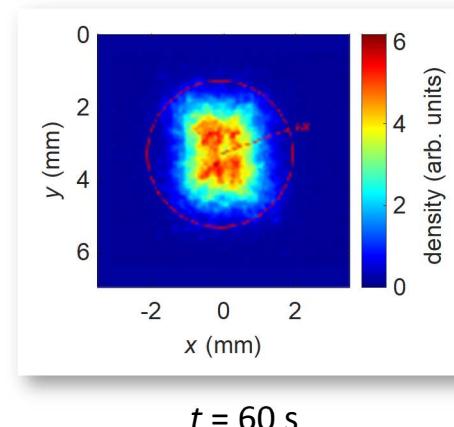
# Trapping and cooling anions

- Paul trap implemented downstream of spectroscopy section
  - linear RF quadrupole, 5 axial segments
  - radial and axial (oblique) laser access
  - MCP with phosphor screen as diagnostics for axial and transverse phase space

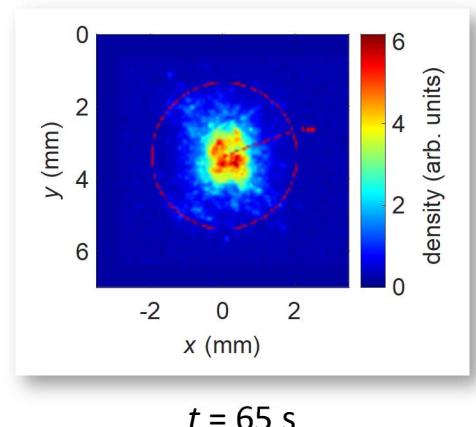


- Trap commissioned and characterized with  $\text{Au}^-/\text{O}^-$  ions:

- max. number of ions loaded  $\approx 10^5 \dots 10^7$
- storage time constant  $> \text{min}$
- laser-assisted evaporative cooling of  $\text{O}^-$ 
  - geometrically selective neutralization with green laser (200 mW @ 532 nm)
  - velocity selection observed



$t = 60 \text{ s}$



$t = 65 \text{ s}$

preliminary

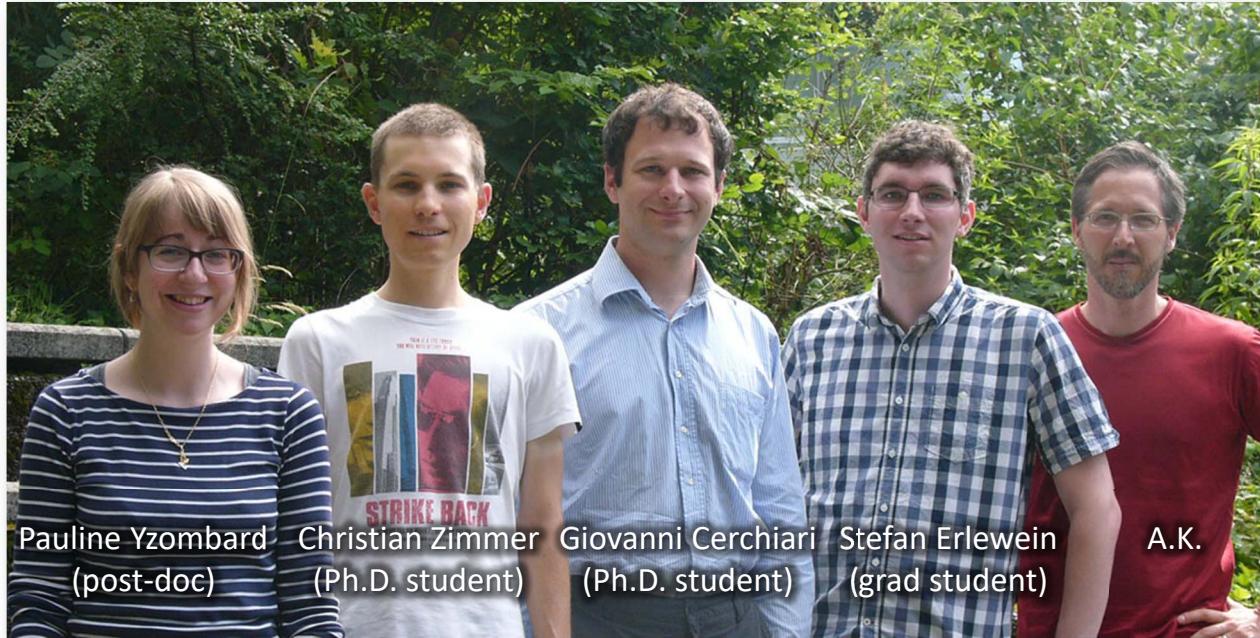
# Conclusions

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- Laser cooling of anions is a promising route to ultracold ensembles of negative ions
- Only few atomic anions exhibit suitable bound–bound transitions
- We have been investigating  $\text{Os}^-$  and  $\text{La}^-$  by optical spectroscopy to check their suitability for laser cooling
- $\text{La}^-$  is a promising candidate – attempt to laser-cool anions in a Paul trap ongoing

# Acknowledgments

- Team:



- Collaboration: Dag Hanstorp (Gothenburg U)  
Wesley Walter, Daniel Gibson (Denison U Ohio)  
Stephan Fritzsche (Helmholtz Institute and U Jena)  
Marianna Safronova (U of Delaware)

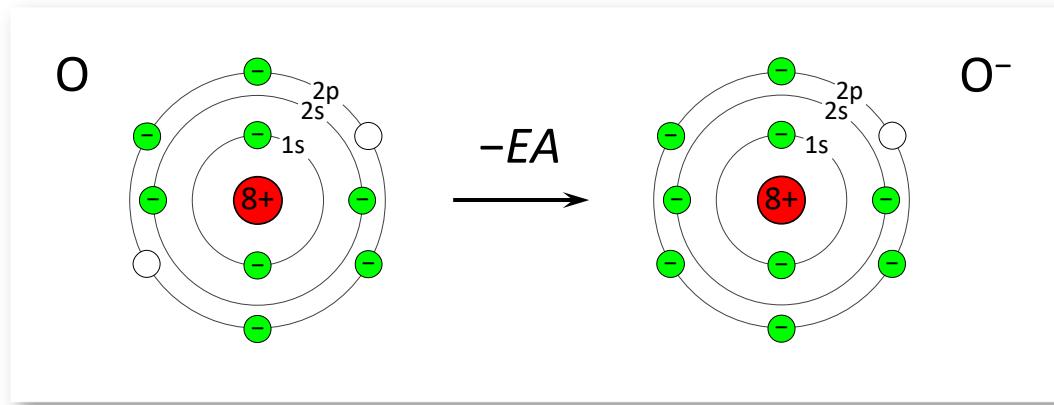
- Funding:



# Extra slides

# Anion formation and structure

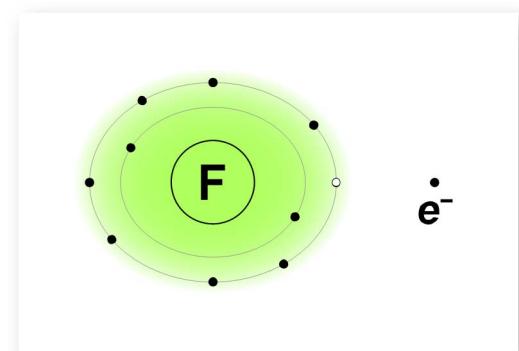
- Atomic anions form by “partial same-shell shielding”
  - Valence electrons only partially shield nuclear charge



Oxygen anion  
electron affinity:  
1.46 eV

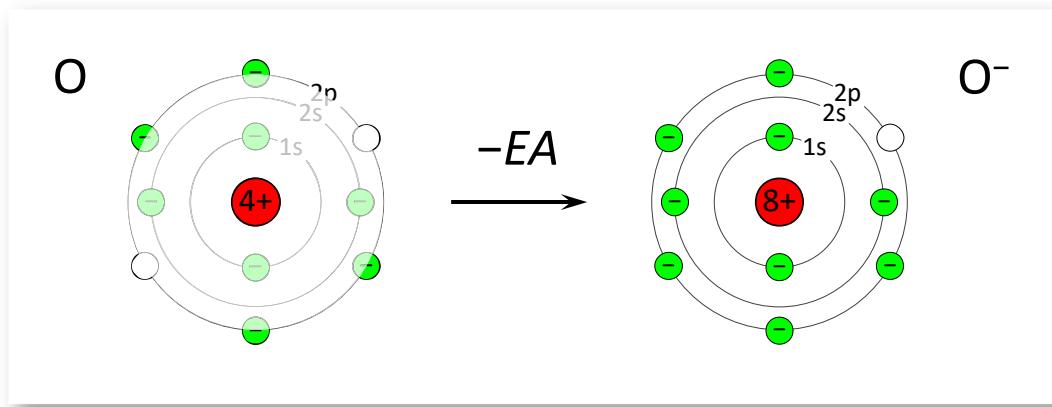
- Differences compared to neutral atoms and cations:
  - Effect of Coulomb potential ( $r^{-1}$ ) reduced
  - Main binding by (induced) short-ranged dipole potential ( $r^{-4}$ )
  - Contribution by correlation (quantum mechanical effect)

⇒ Few (or no) bound excited states



# Anion formation and structure

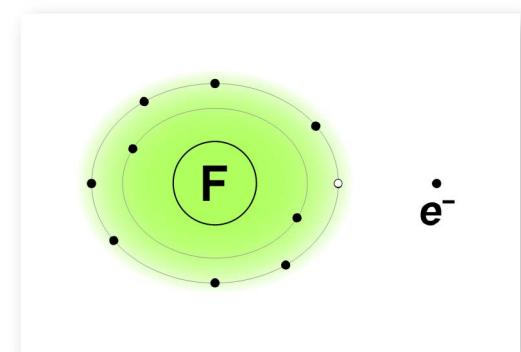
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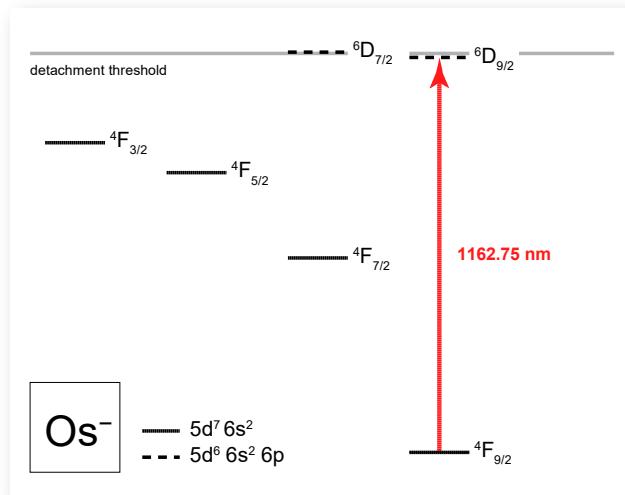
- Differences compared to neutral atoms and cations:
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# Laser cooling candidate $\text{Os}^-$

- Potential laser cooling transition:



Wavelength  $\lambda = 1162.7 \text{ nm}$   
Transition rate  $\Gamma = 1 \dots 10 \text{ kHz}$   
(initial measurement)

[P. L. Norquist & D. R. Beck, Phys. Rev. A **61** (1999/2000) 014501;  
R. C. Bilodeau & H. K. Haugen, Phys. Rev. Lett. **85** (2000) 534]

<b>Os183</b> 13.0 h 9/2+	<b>Os184</b> 0+ 0.02	<b>Os185</b> 93.6 d 1/2-	<b>Os186</b> 0+	<b>Os187</b> 1/2-	<b>Os188</b> 0+	<b>Os189</b> 3/2-	<b>Os190</b> 0+	<b>Os191</b> 15.4 d 9/2-	<b>Os192</b> 0+ 41.0	<b>Os193</b> 30.5 h 3/2-
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- Experimental results:

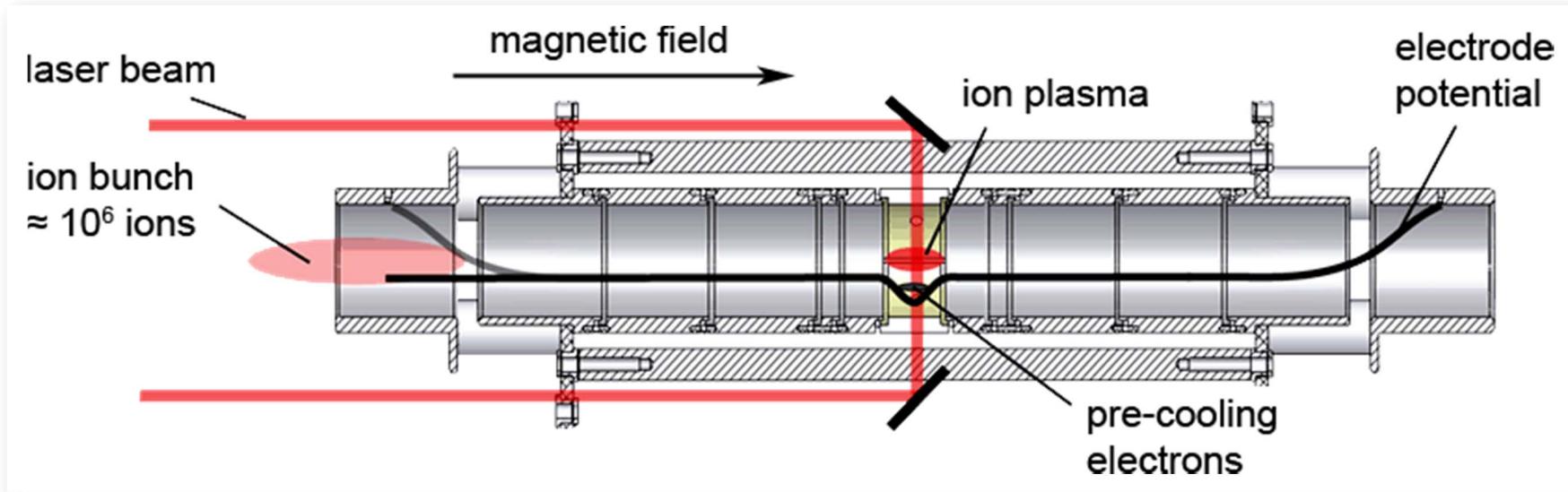
- Transition frequency confirmed
- Cross-section / rate of resonant transition and photodetachment  $\Rightarrow 50 \text{ Hz}$
- Hyperfine structure in  $^{187}\text{Os}^-$ ,  $^{189}\text{Os}^-$
- Zeeman splitting in  $^{192}\text{Os}^-$  in external magnetic field

} First-ever measurements  
in atomic anions

[U. Warring *et al.*, Phys. Rev. Lett. **102** (2009) 043001;  
A. Fischer *et al.*, Phys. Rev. Lett. **104** (2010) 073004;  
A.K. *et al.*, Phys. Rev. A **89** (2014) 043430]

# Indirect laser cooling of negative ions

- Principle:

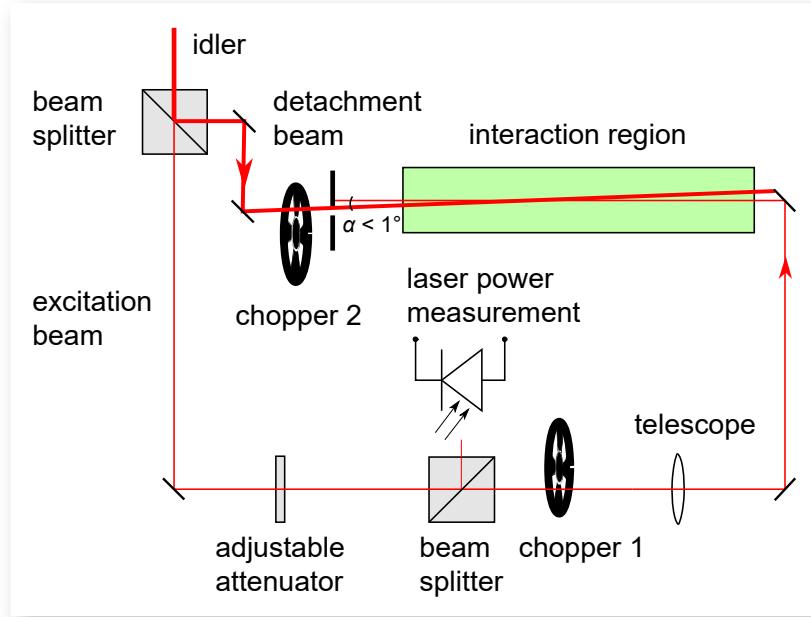


1. Capture anion bunch in Penning trap
2. Pre-cool anions with electrons
3. Laser-cool the anions
4. Simultaneously load other ions into trap
5. Sympathetically cool them with laser-cooled anions

[A. K. & J. Walz, New J. Phys. 8 (2006) 45]

# Spectroscopy setup for La<sup>-</sup>

- Laser beam setup



Laser powers:

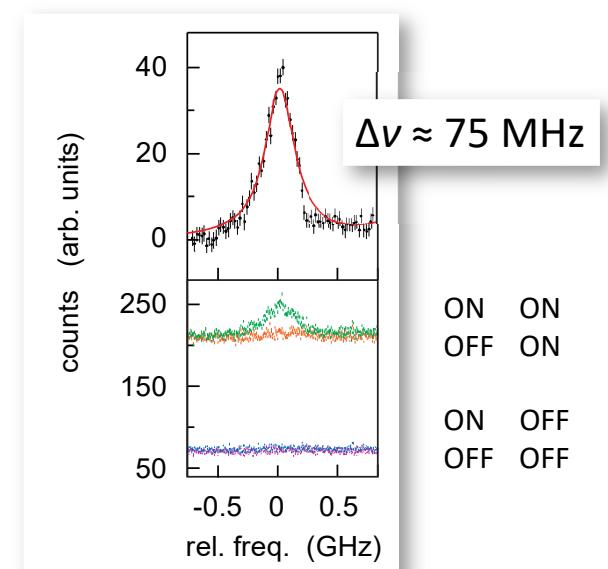
- Excitation: 2...60 mW
- Detachment:  $\approx 1$  W

Repetition frequencies:

- Chopper 1: 50 Hz
- Chopper 2: 5 Hz

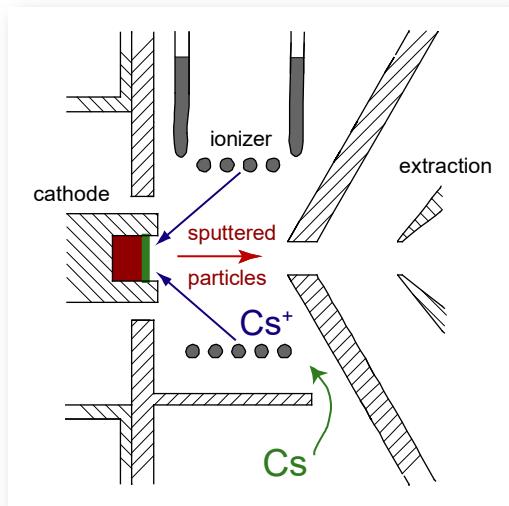
- Subtracted background signals

- OFF-ON: Ions arriving in interaction region in excited state
- ON-OFF: Two-photon detachment by excitation laser only
- OFF-OFF: Ions neutralized by other mechanisms  
(e.g. residual gas)



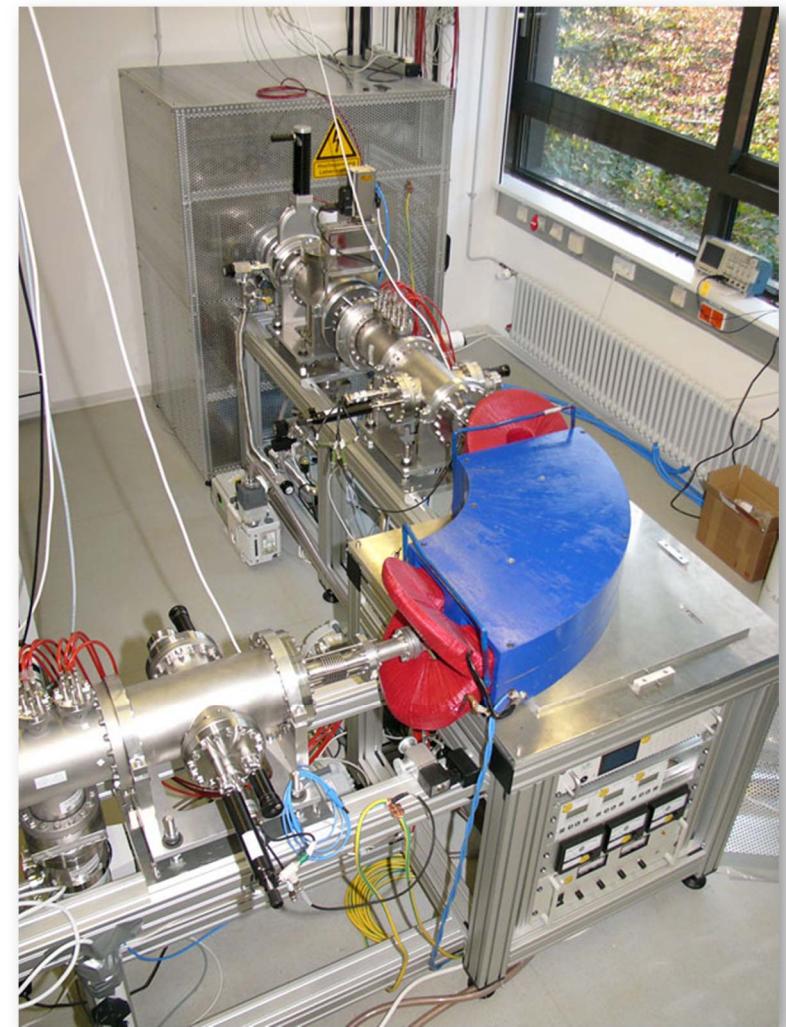
# Ion production

- Negative-ion source
  - Middleton-type ion source: Sputtering of  $\text{Cs}^+$  ions onto surface of desired target material



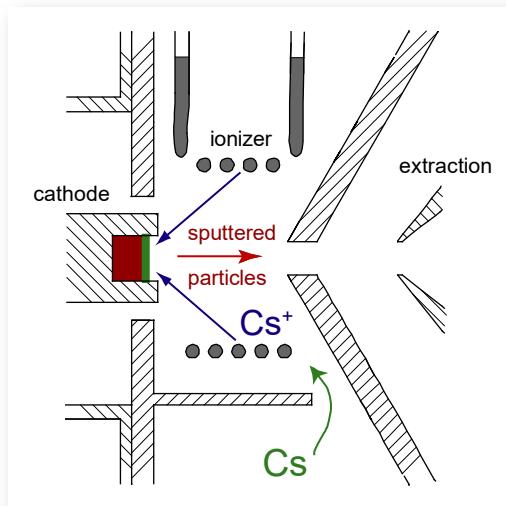
[R. Middleton,  
NIM 214 (1983) 139]

- Typical ion source performance:  
several 100 nA of  $^{192}\text{Os}^-$
- Mass resolving power:  $R \approx 180$   
 $\Rightarrow$   $^{192}\text{Os}^-$  can be separated from  
nearest isotope  $^{190}\text{Os}^-$



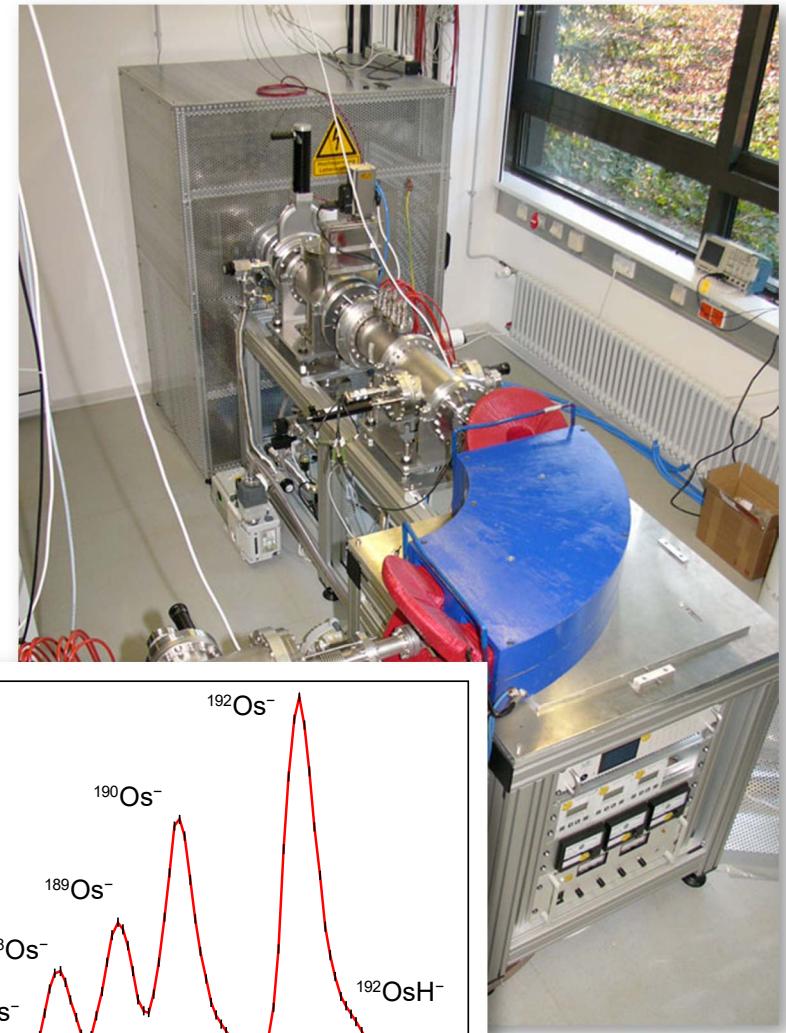
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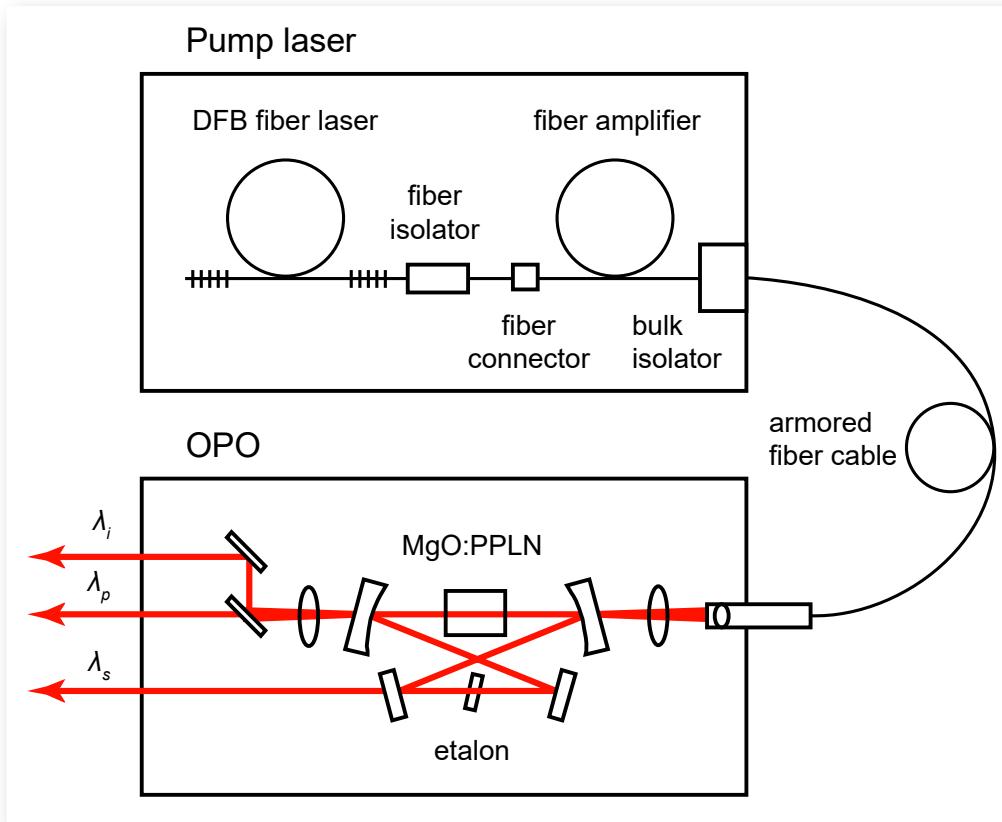
[R. Middleton,  
NIM 214 (1983) 139]

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nearest isotope  $^{190}\text{Os}^-$



# Laser system for La<sup>-</sup> spectroscopy

- cw-OPO laser seeded by DFB fiber laser:

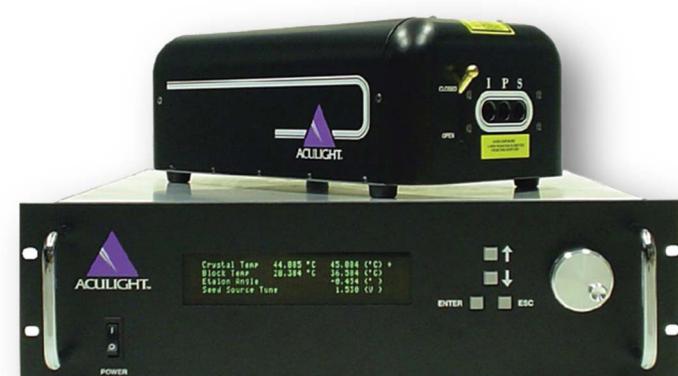


Aculight Argos SF10

$\lambda = 2.5 \dots 3.2 \mu\text{m}$

$P > 600 \text{ mW}$

$\Gamma < 1 \text{ MHz}$



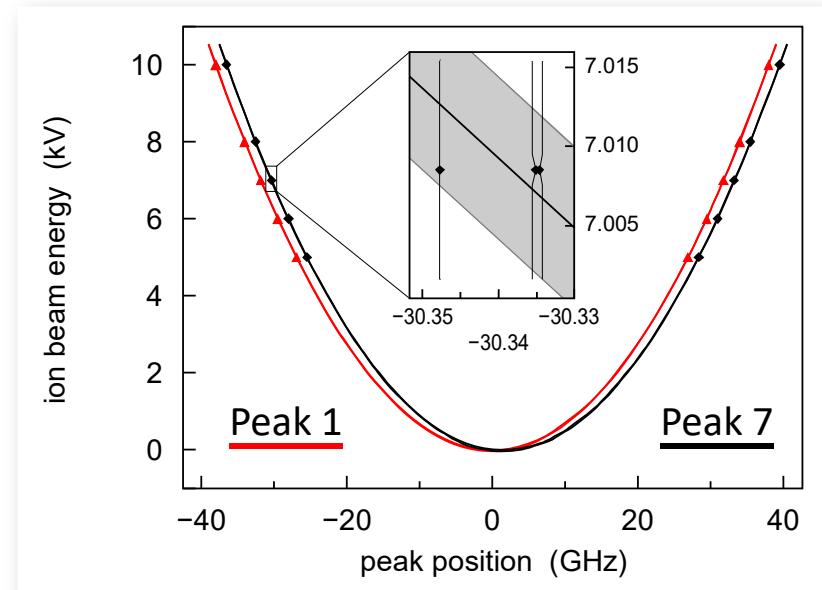
# Doppler correction of peak frequencies

- Doppler shift as a function of ion velocity/energy:

$$f = f_0 \sqrt{\frac{c + v(E)}{c - v(E)}}$$

- Frequencies for Peaks 1–7 from parabola:

Peak	Rel. intensity	Frequency (MHz)
1	1.00(11)	0.0(5.8)
2	0.77(8)	324.8(5.8)
3	0.51(6)	604.1(5.9)
4	0.81(8)	825.1(5.8)
5	0.74(8)	990.1(5.9)
6	0.53(7)	1116.2(6.1)
7	1.08(10)	1480.2(5.8)



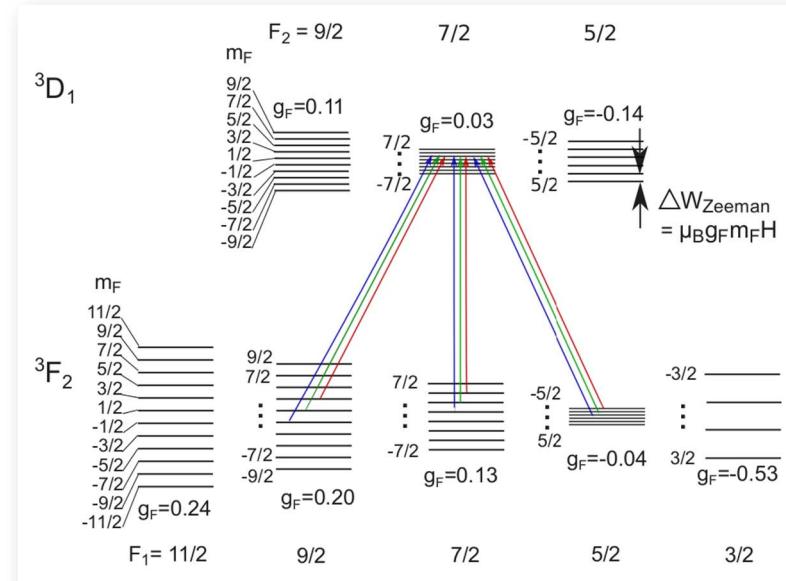
[E. Jordan *et al.*,  
Phys. Rev. Lett. **115** (2015) 113001]

[relative to Peak 1  
at 96.592,004(80) THz]

# La<sup>-</sup> in a magnetic field

- Zeeman splitting ( $B \ll 1$  T)

Transition splits 192-fold



- Paschen–Back splitting ( $B \gg 1$  T)

Transition splits 65-fold

