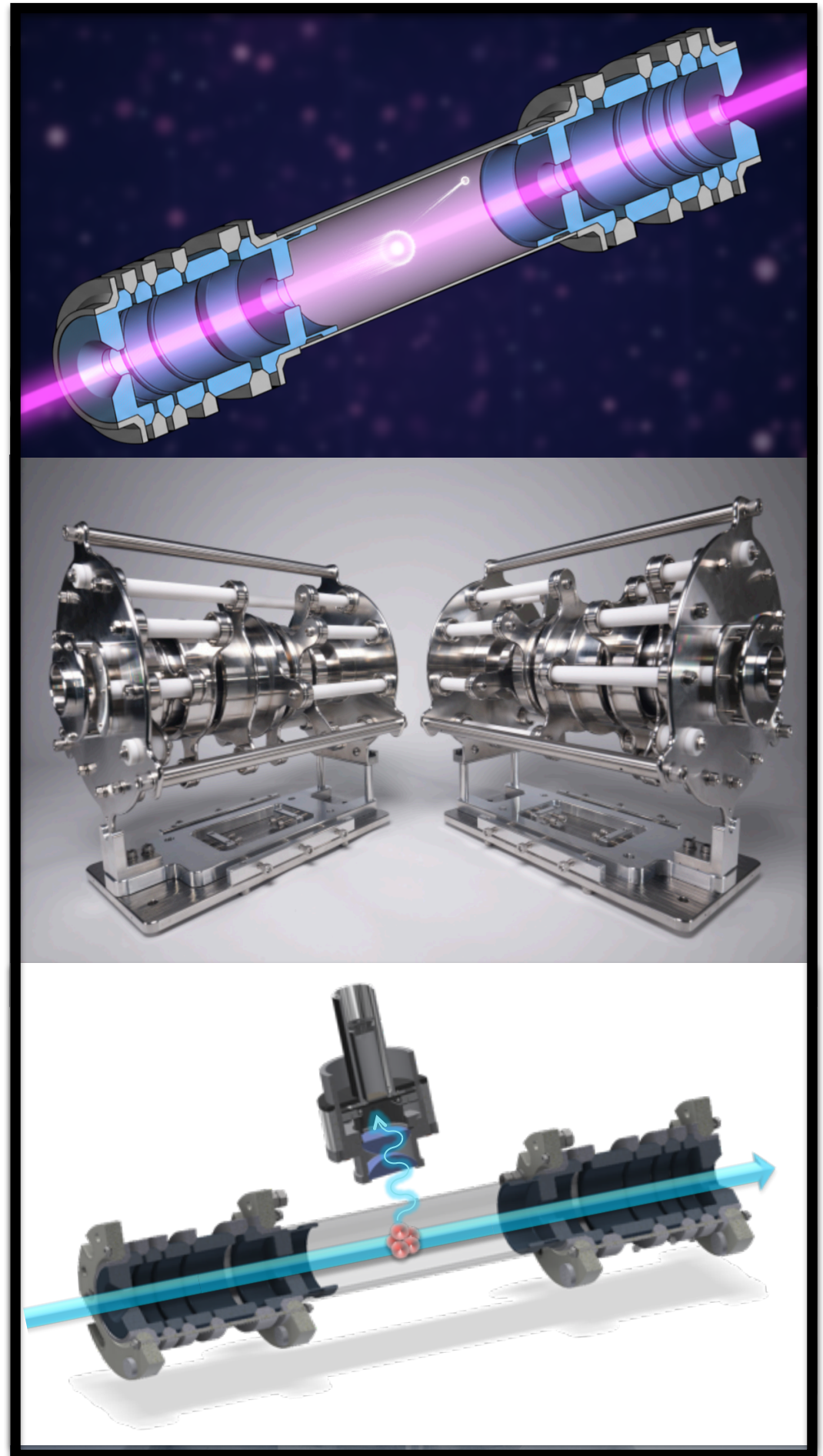


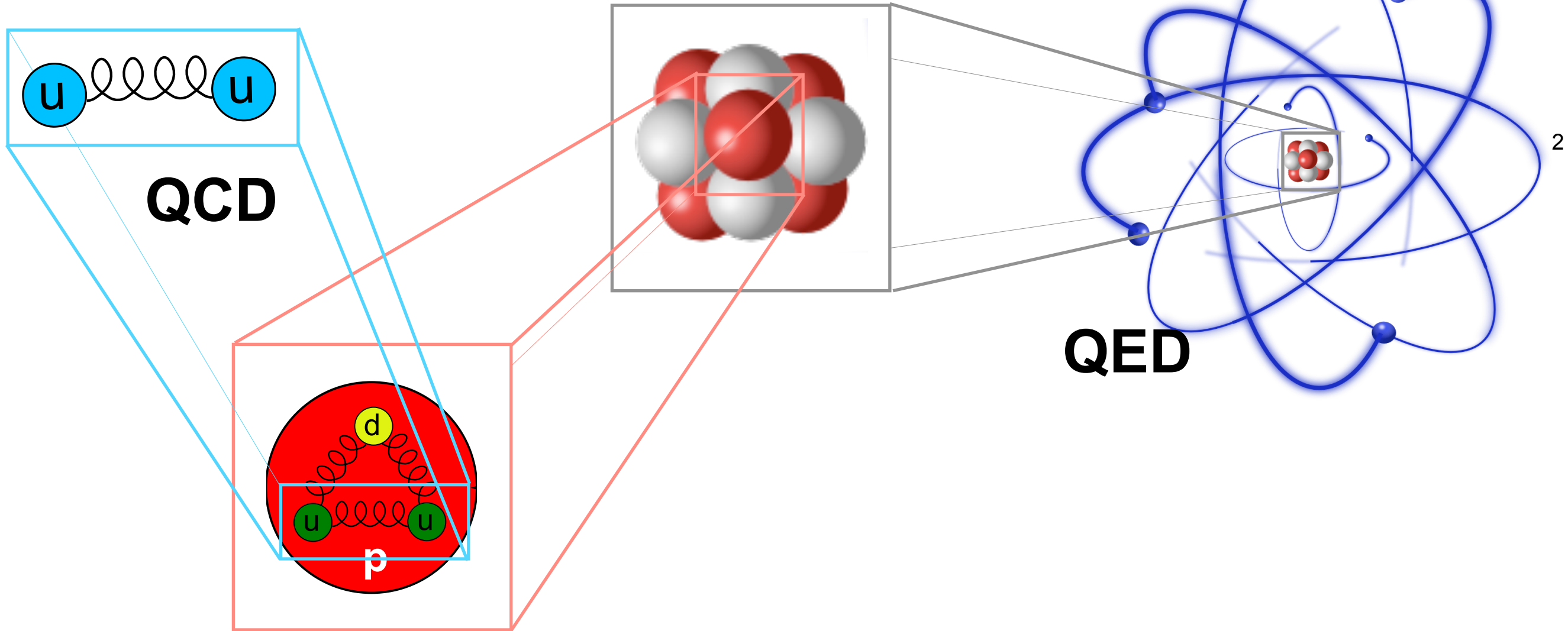
# Collinear Laser Spectroscopy for the Investigation of Short-lived Radionuclides

Stephan Malbrunot-Ettenauer  
TRIUMF, University of Toronto

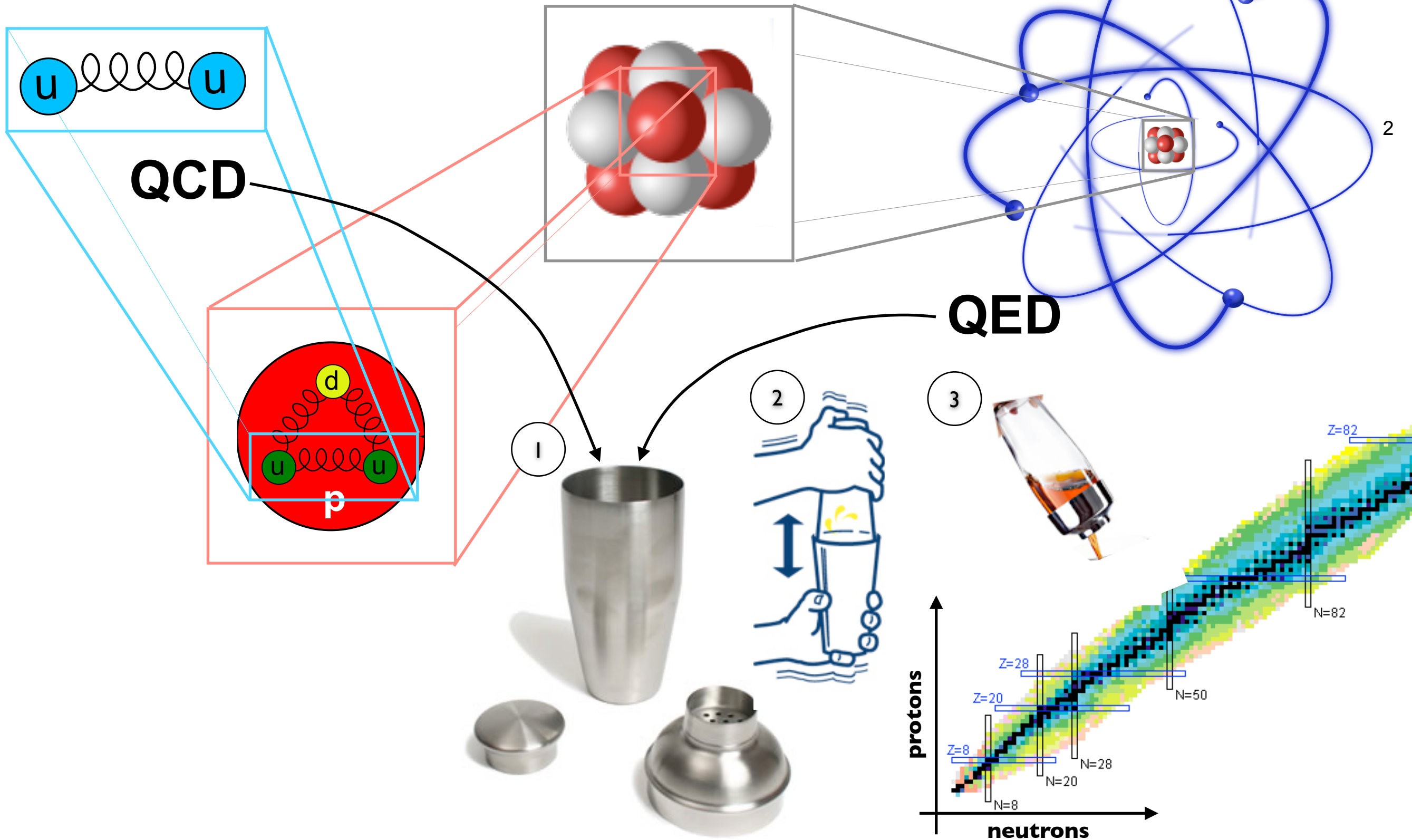
African Nuclear Physics Conference 2025



# particle - nuclear - atomic

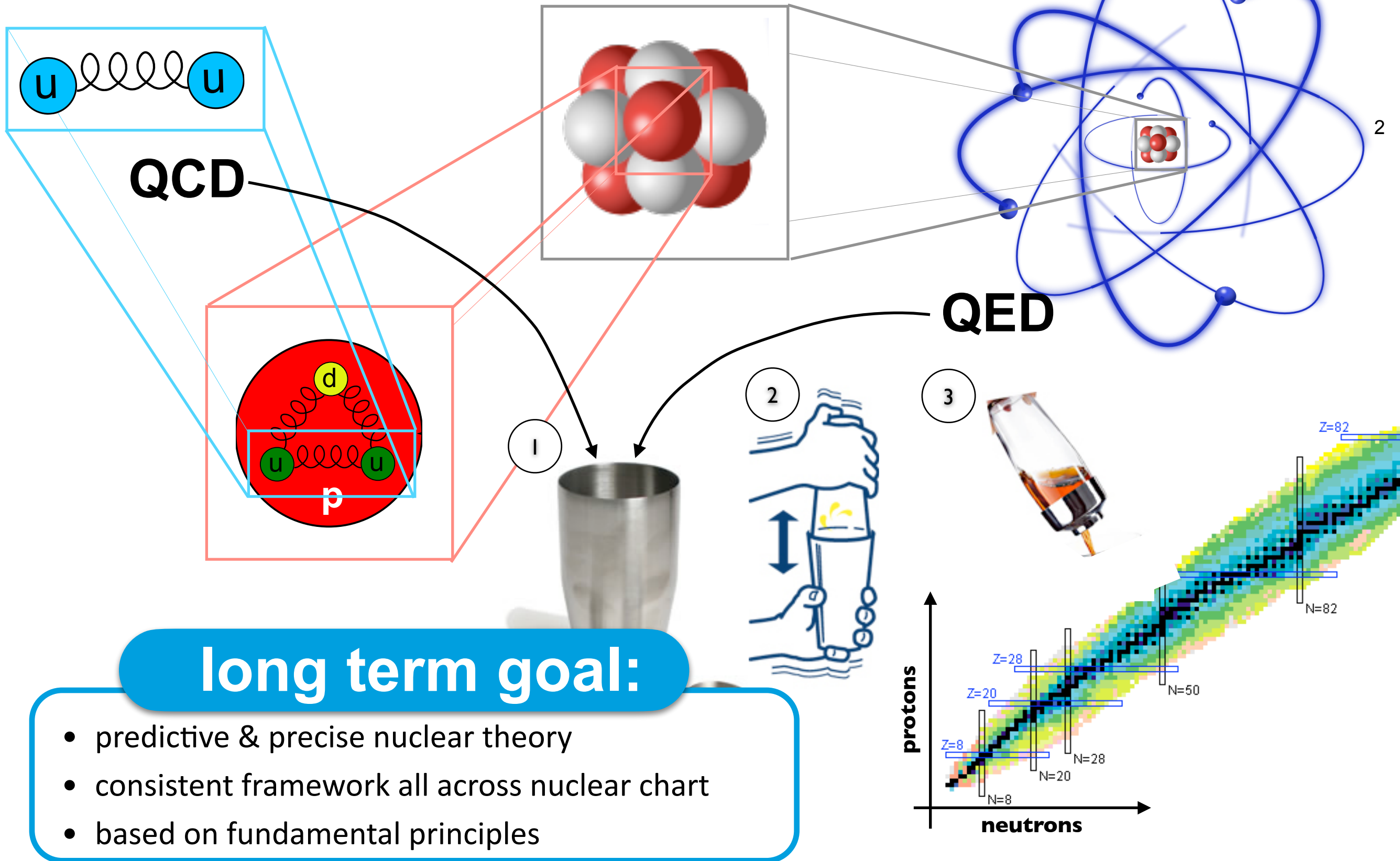


# particle - nuclear - atomic



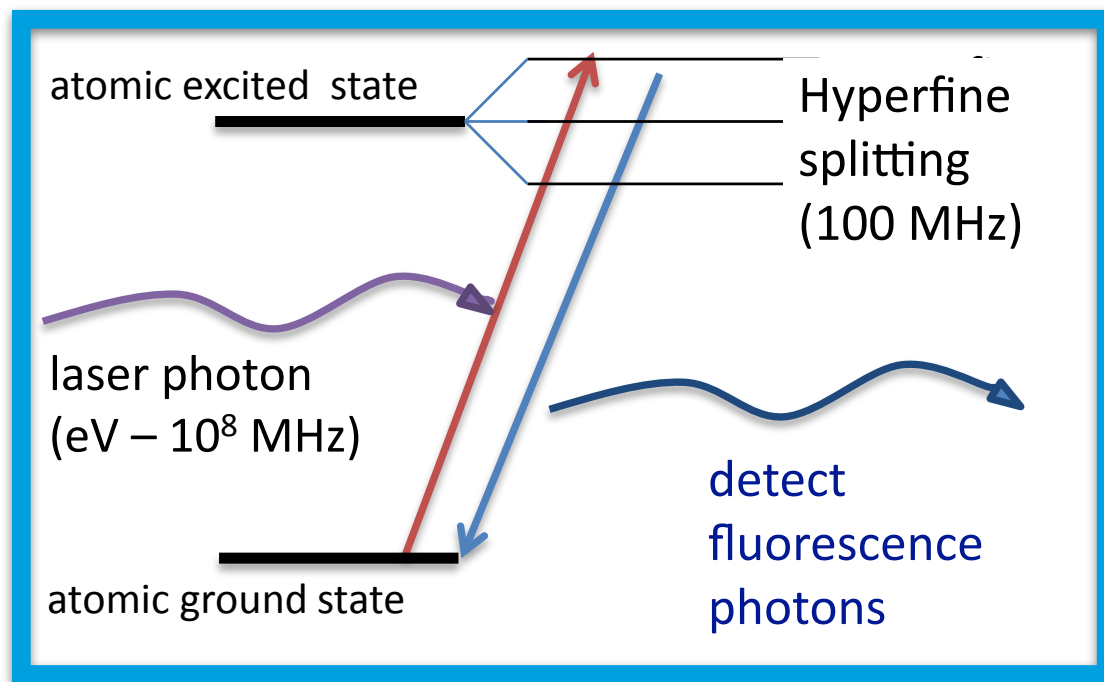


# particle - nuclear - atomic

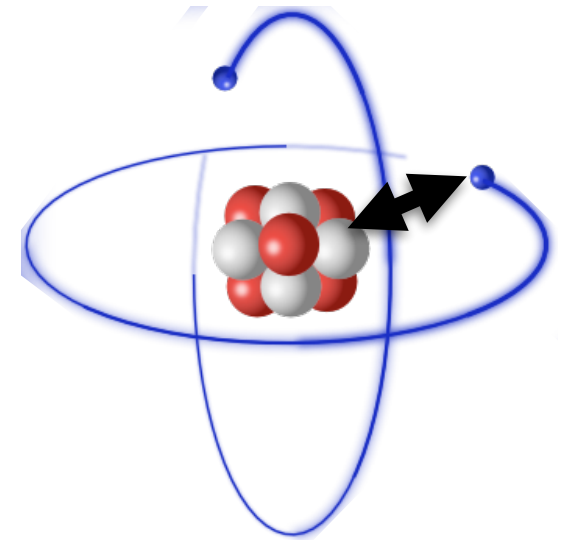




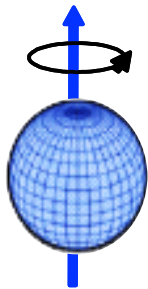
# laser spectroscopy of radionuclides



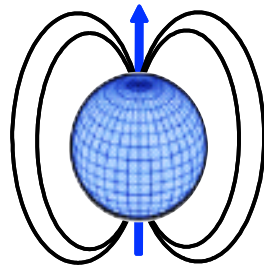
nucleus - electrons interaction  
 $\Rightarrow$  atomic hyperfine structure



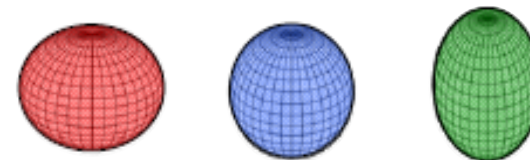
spin  $I$



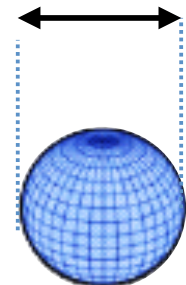
magnetic moment  $\mu$



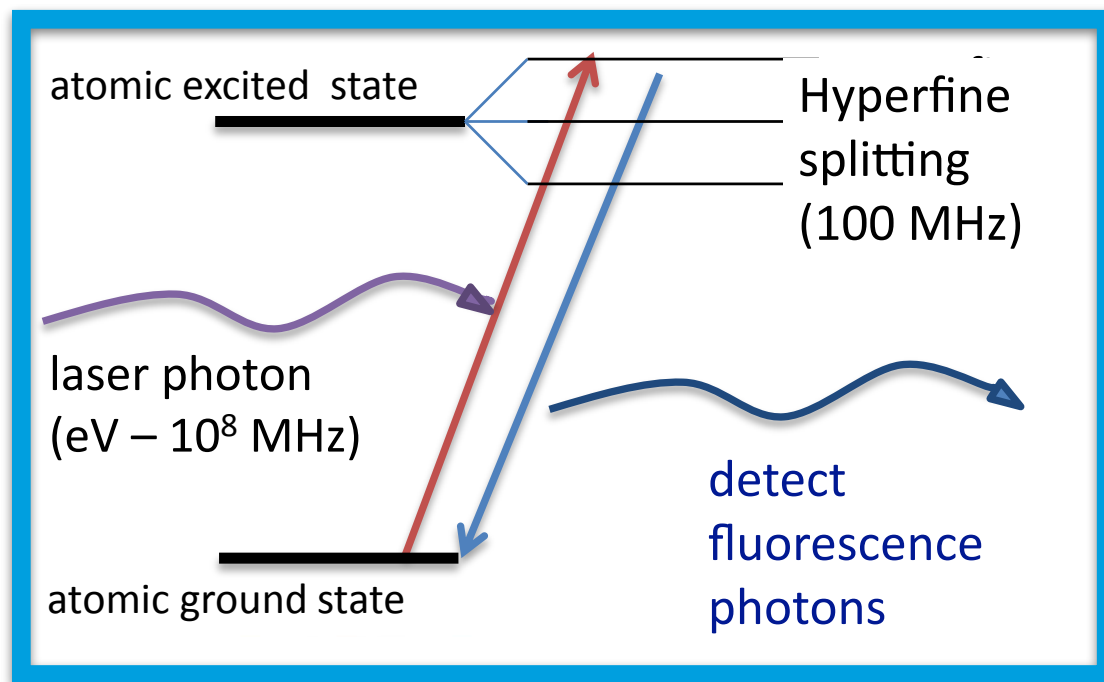
quadrupole moment  $Q_s$



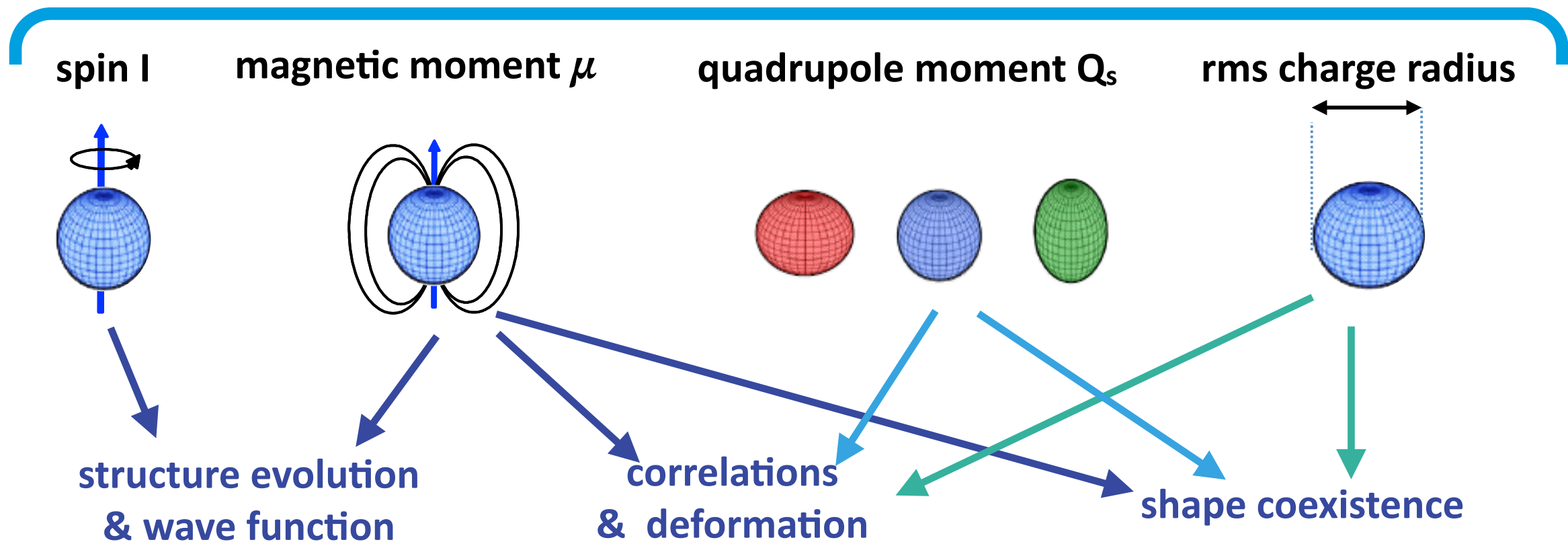
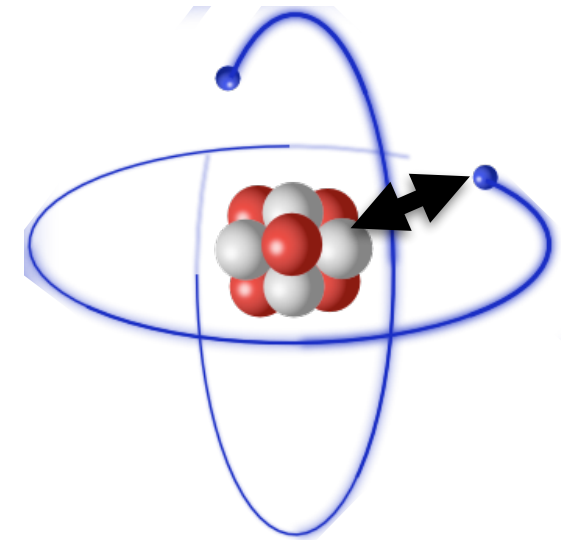
rms charge radius



# laser spectroscopy of radionuclides

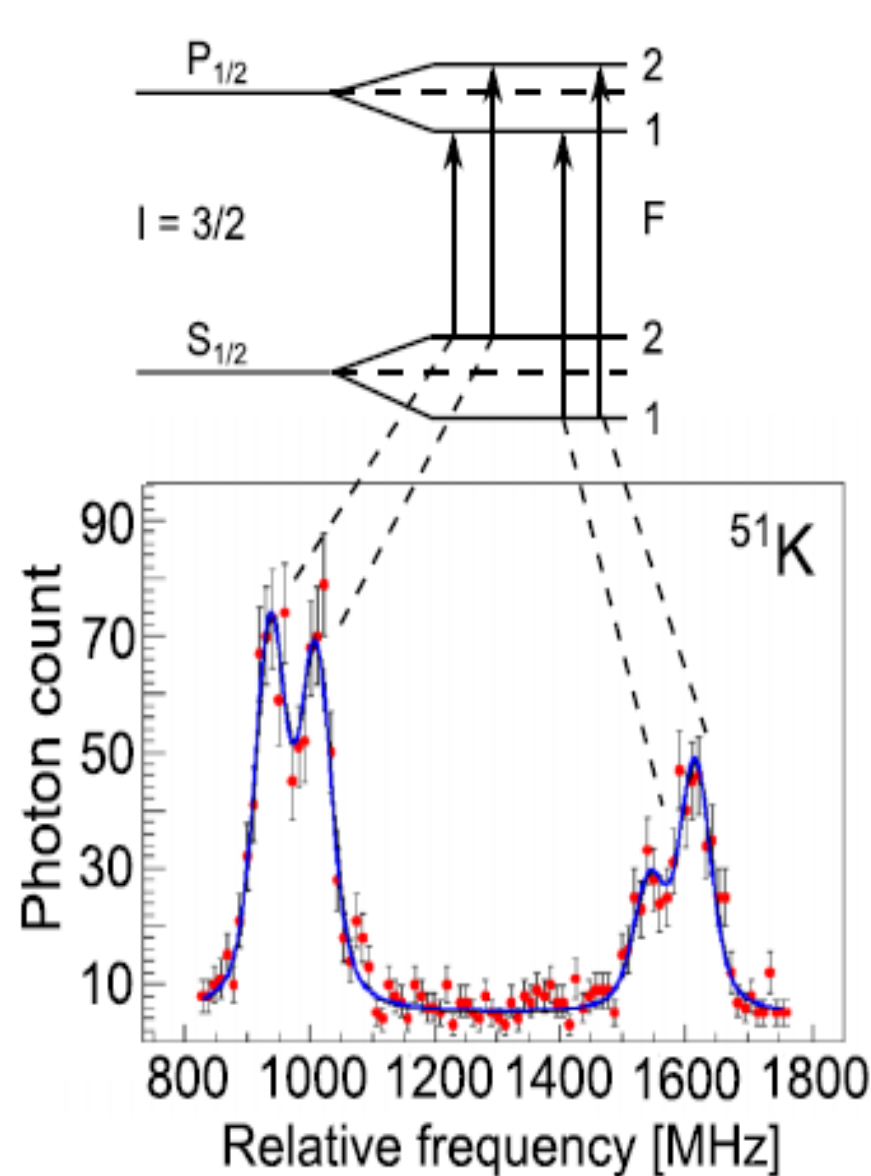


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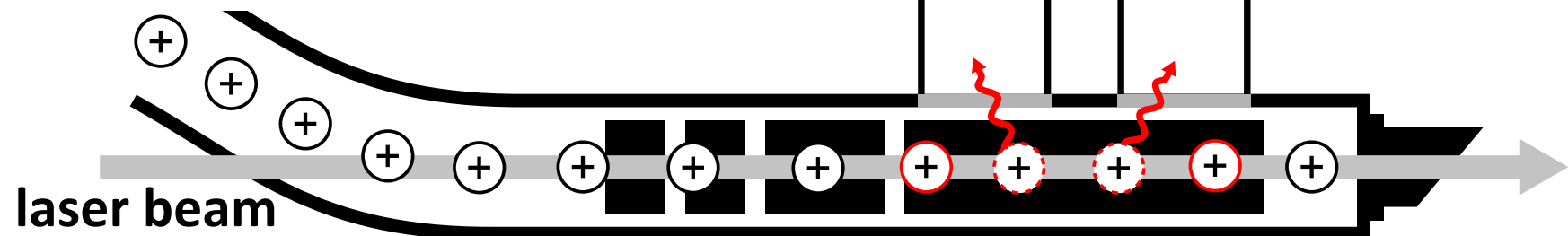
**benchmark for modern nuclear structure theory**

# Collinear Laser Spectroscopy (CLS)



$\sim 30$  keV to eliminate Doppler broadening  $\delta\nu \propto \frac{\delta E}{\sqrt{E}}$

ion beam



*K. Blaum, et al., Phys. Scr. T152, 014017 (2013)*

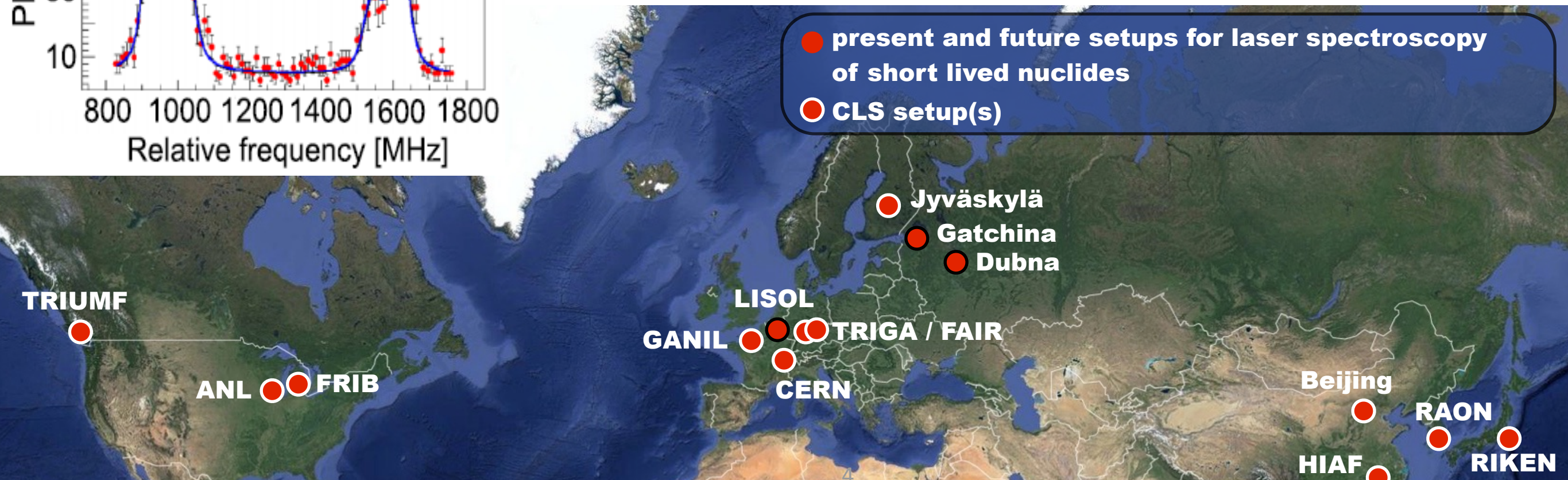
*P. Campbell et al., Prog. Part. and Nucl. Phys. 86, 127-180 (2016)*

*R. Neugart et al., J. Phys. G: Nucl. Part. Phys. 44, 064002 (2017)*

*X.F. Yang et al., Prog. in Part. and Nucl. Phys. 129, 104005 (2023)*

● present and future setups for laser spectroscopy of short lived nuclides

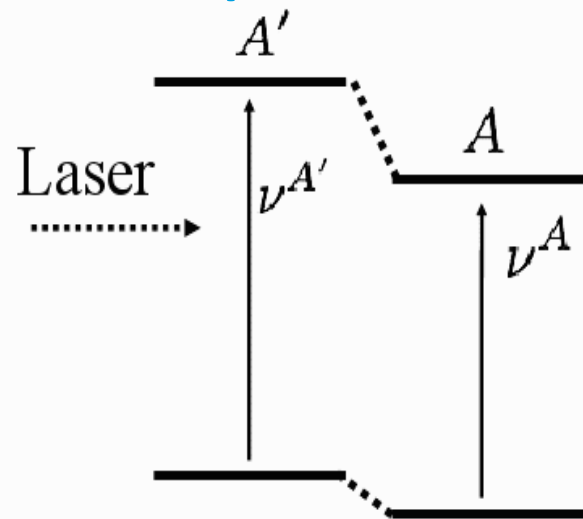
● CLS setup(s)





# CLS & nuclear charge radii

optical isotope shift



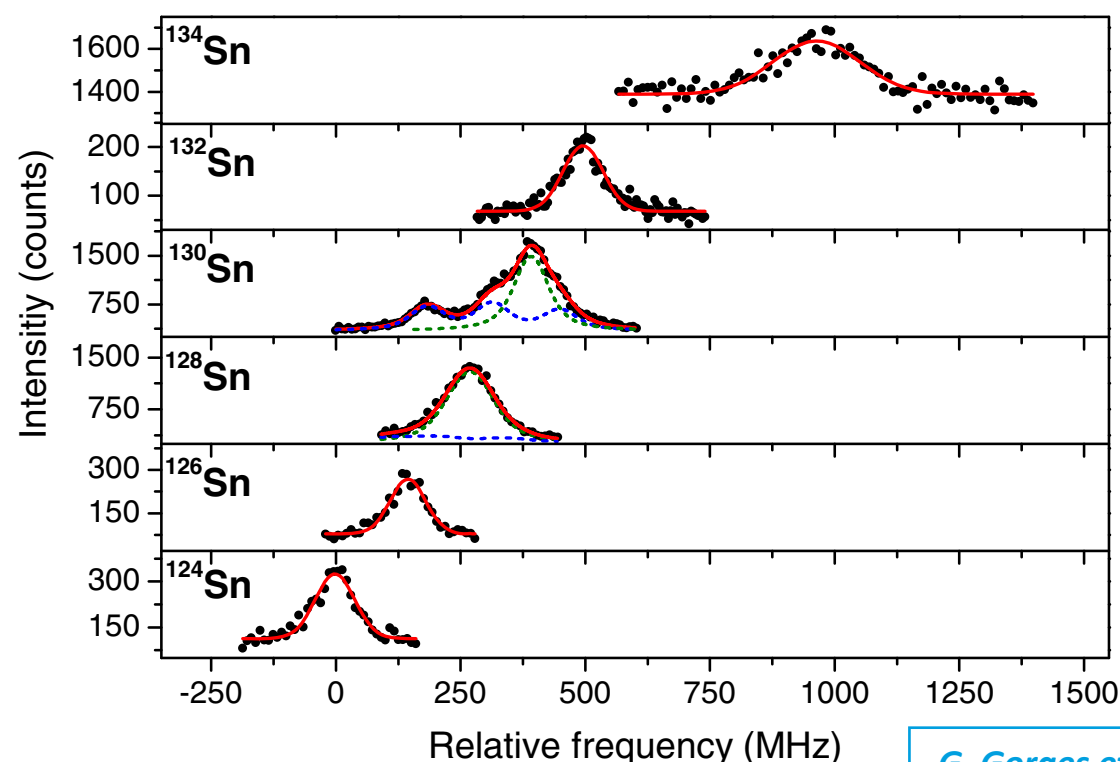
$$\delta\nu^{A,A'} = M \frac{A' - A}{A \cdot A'} + F \delta\langle r^2 \rangle^{A,A'}$$

difference in ms  
charge radii

mass and field shift factors

- King-plot analysis of stable isotopes
- atomic theory

isotope shift  $\delta\nu^{A,A'}$

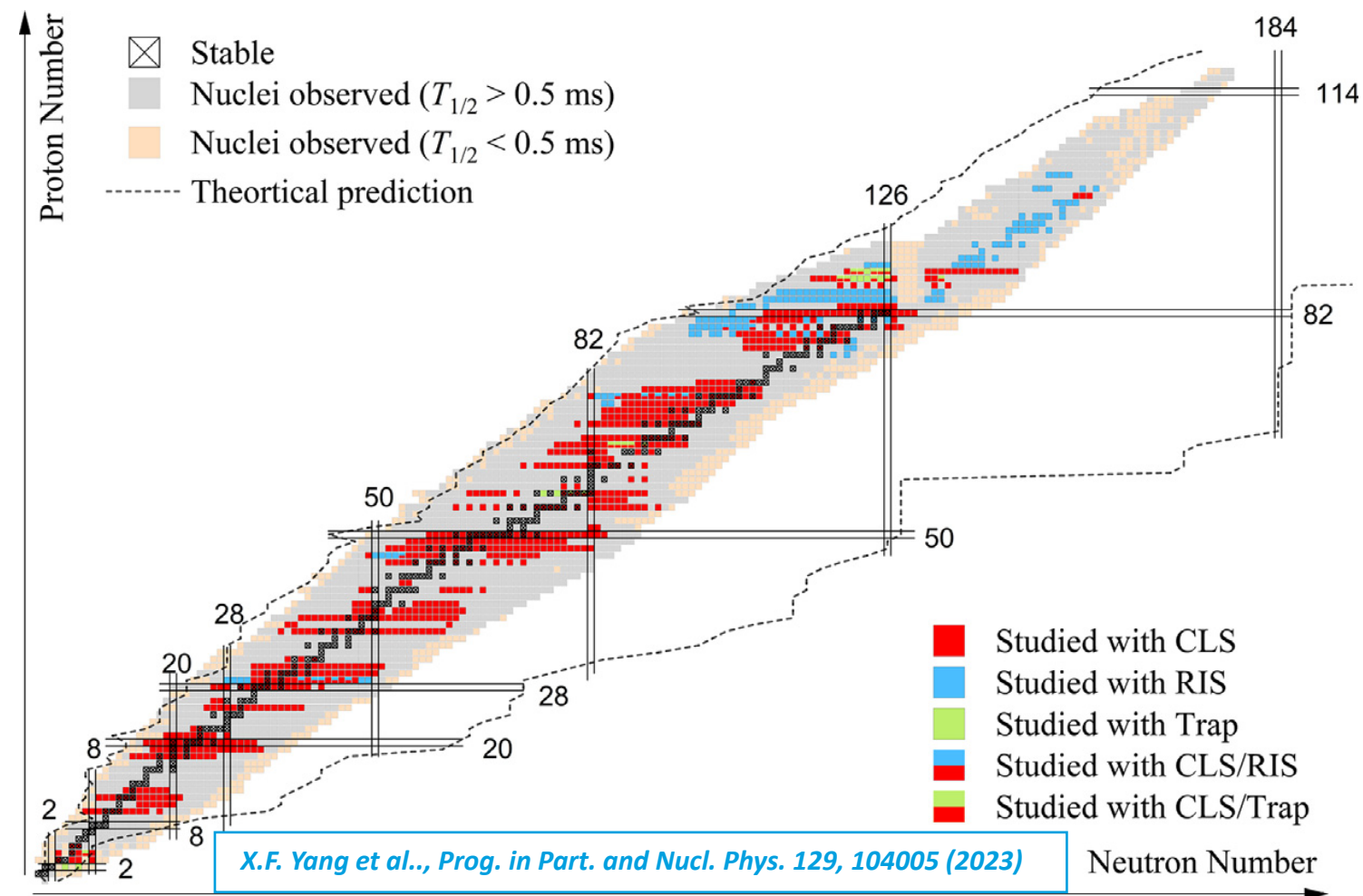


Absolute charge radii

$$R^{A'} = \sqrt{\langle r^2 \rangle^{A'}} = \sqrt{\delta\langle r^2 \rangle^{A,A'} + \langle r^2 \rangle^A},$$

reference radius of stable isotope

# (Some) Challenges & Opportunities

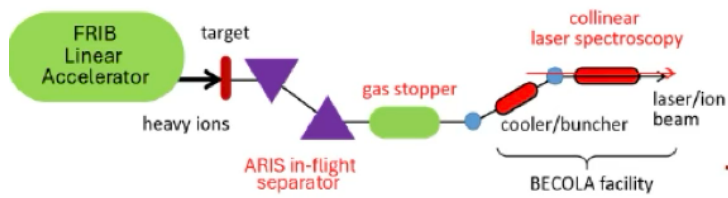


# (Some) Challenges & Opportunities

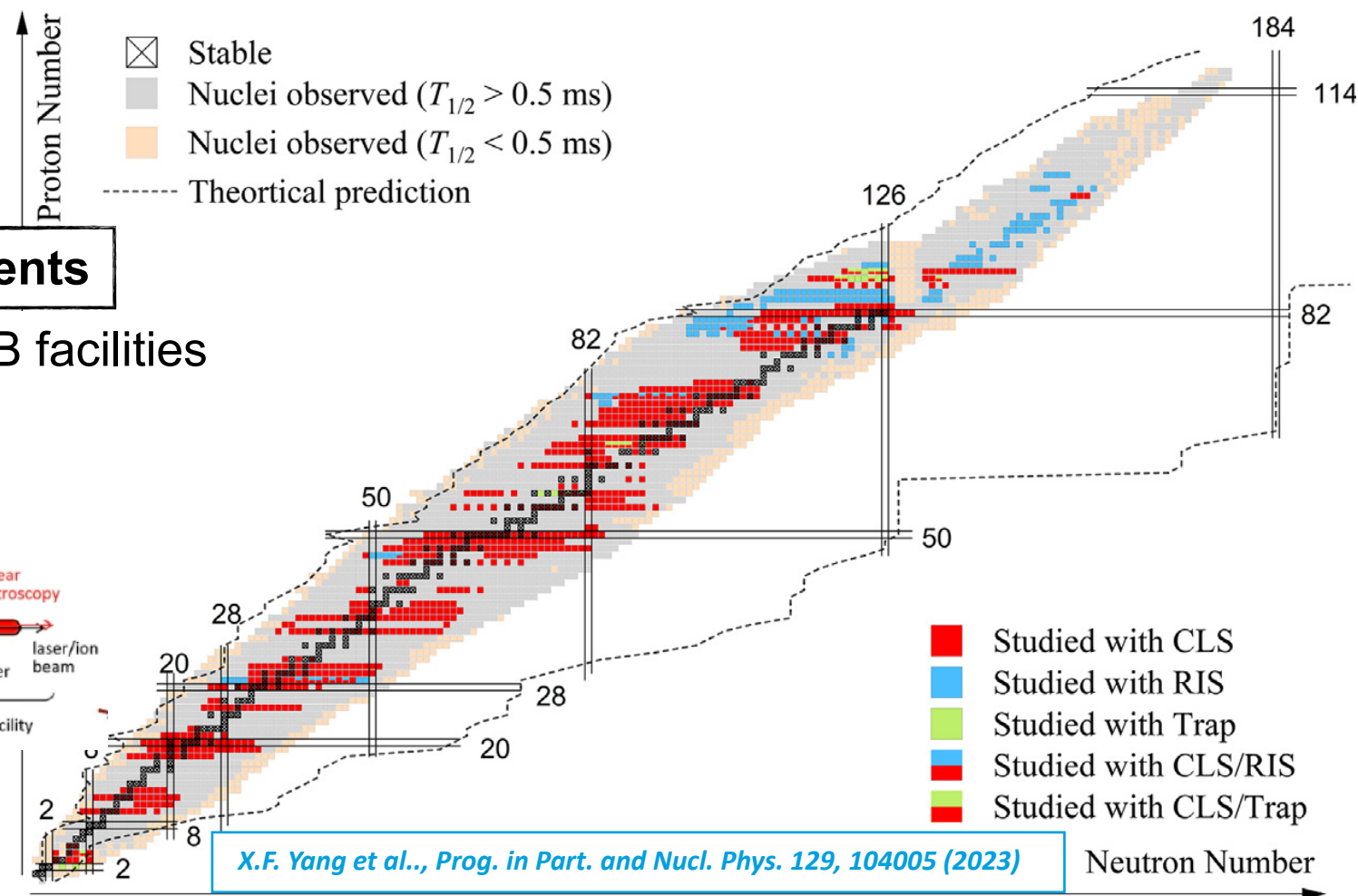
## Access to certain elements

variety in production & RIB facilities

- CLS @ IGISOL
- ATLANTIS @ CARIBU
- ~~BECOLA~~ @ FRIB



+ new CLS beamlines in construction



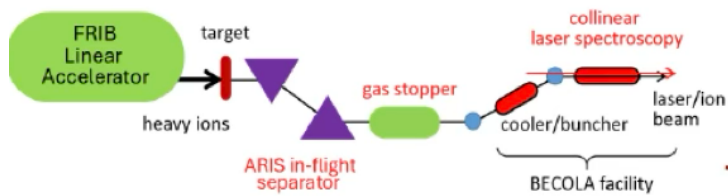


# (Some) Challenges & Opportunities

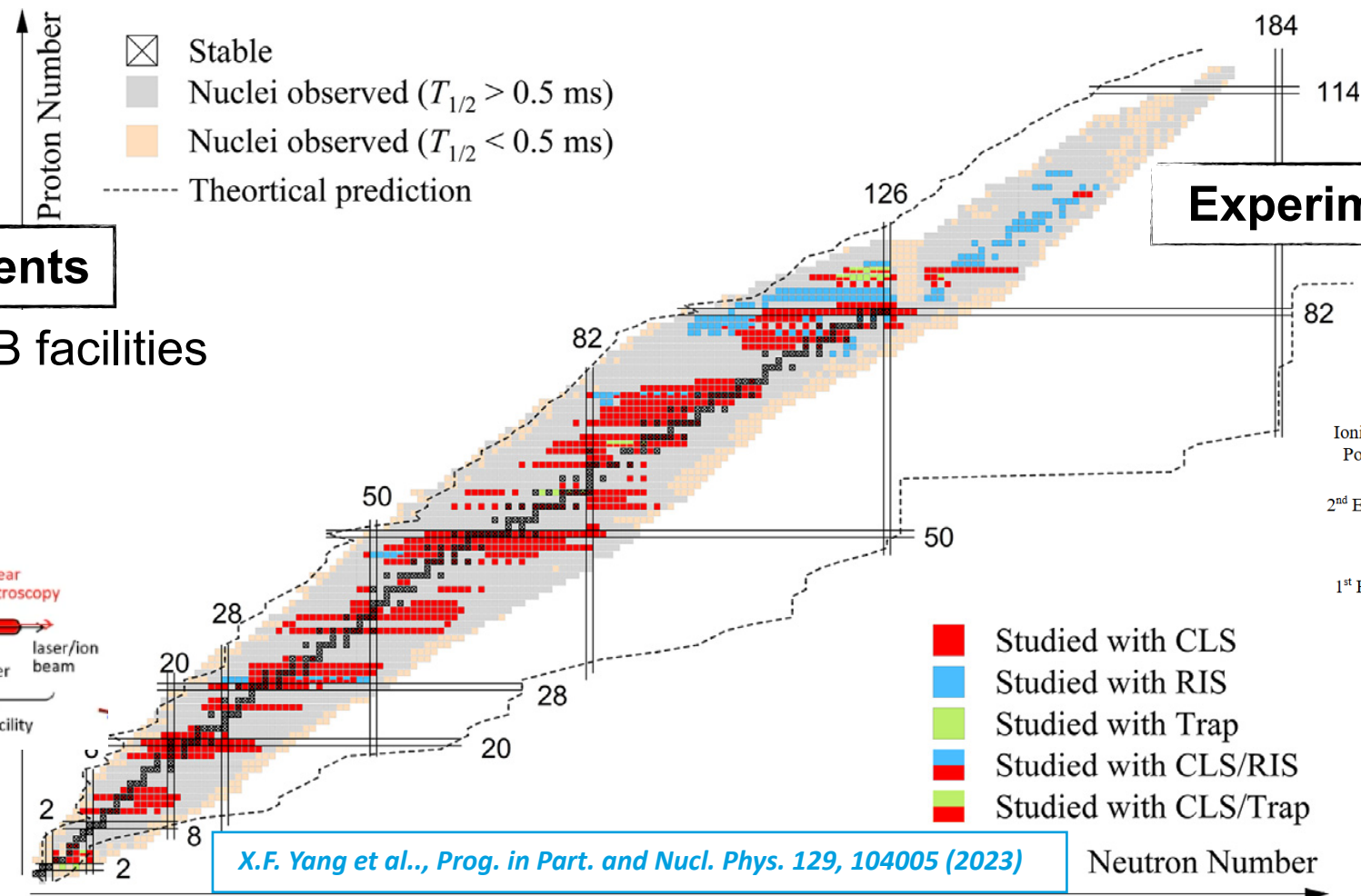
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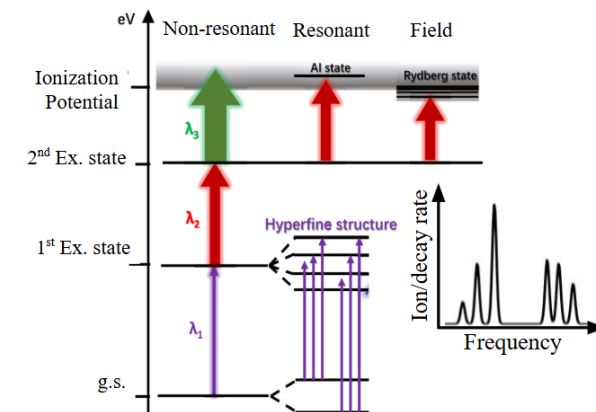
+ new CLS beamlines in construction



## Experimental sensitivity

new methods

- CRIS @ ISOLDE



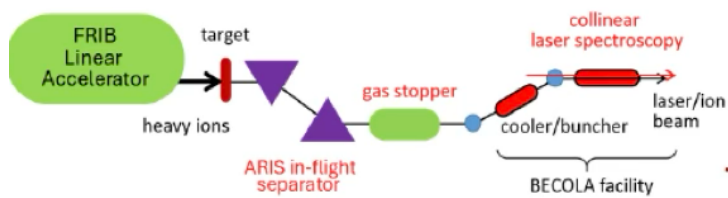
- MIRACLS@ISOLDE
- PRECIOSA @ MIT
- ROC @ ISOLDE
- ...

# (Some) Challenges & Opportunities

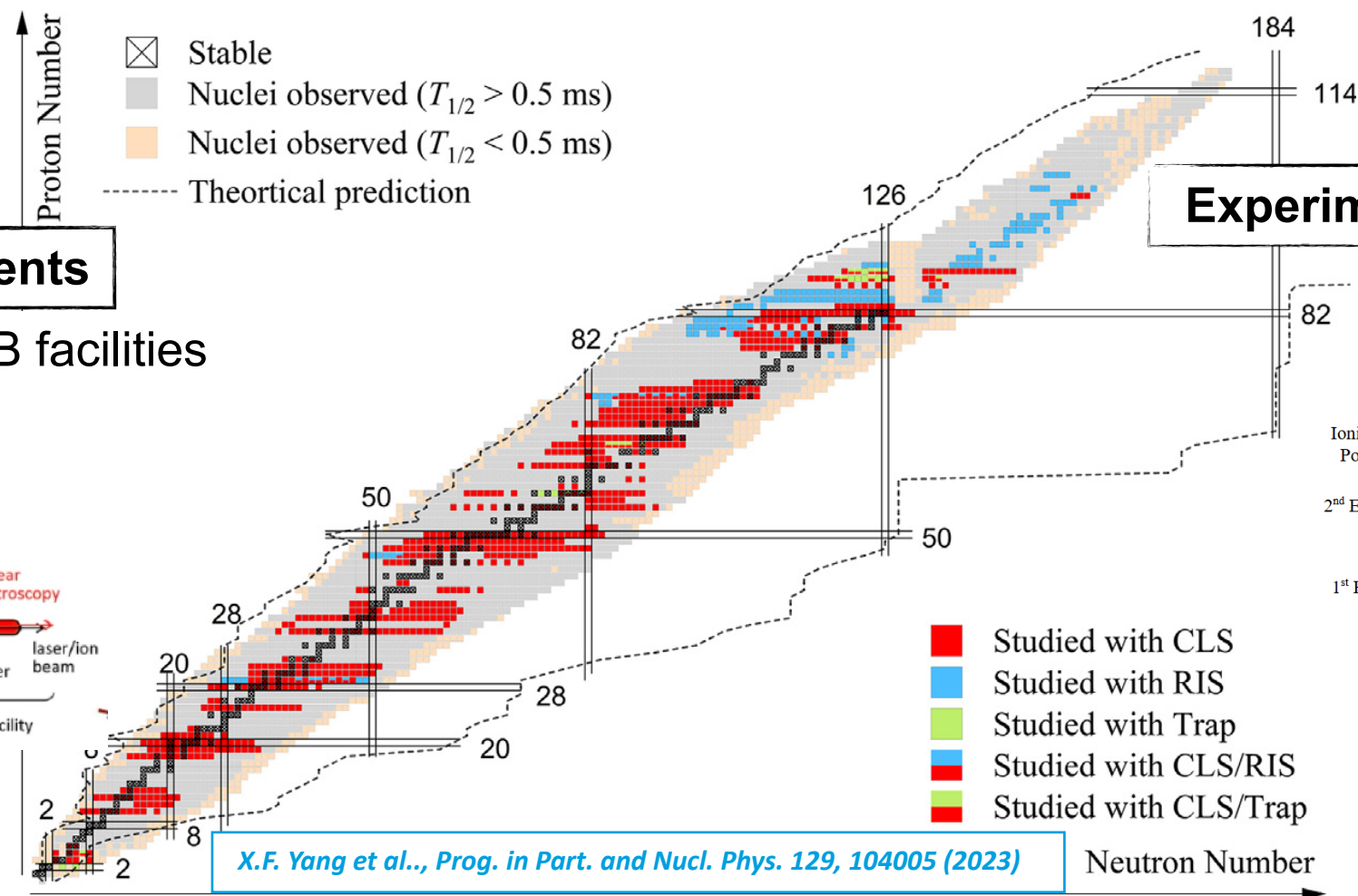
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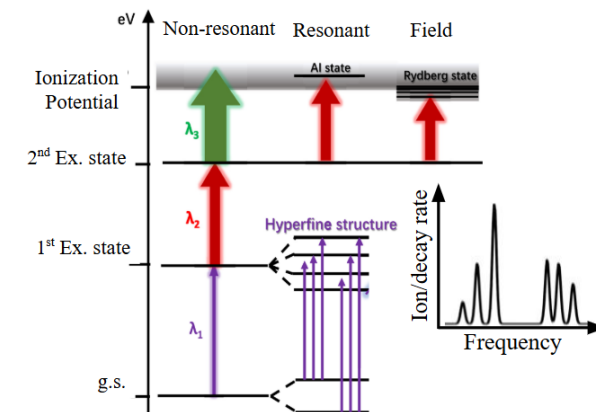
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## Experimental sensitivity

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- ...

## Precision

needed for certain applications  
access to new observables

- 2-photon spectroscopy @  PLASEN  
Precision Laser Spectroscopy for Exotic Nuclei
- Laser cooling - MIRACLS @ ISOLDE  
- **STRIPE** @ Leuven

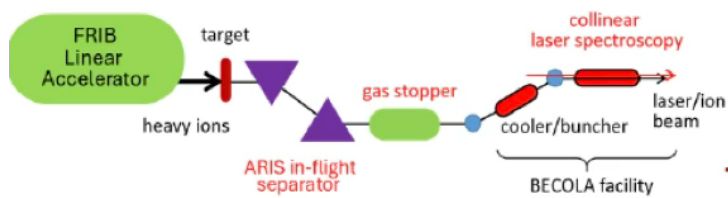
(apologies for any developments omitted due to time limitations)

# (Some) Challenges & Opportunities

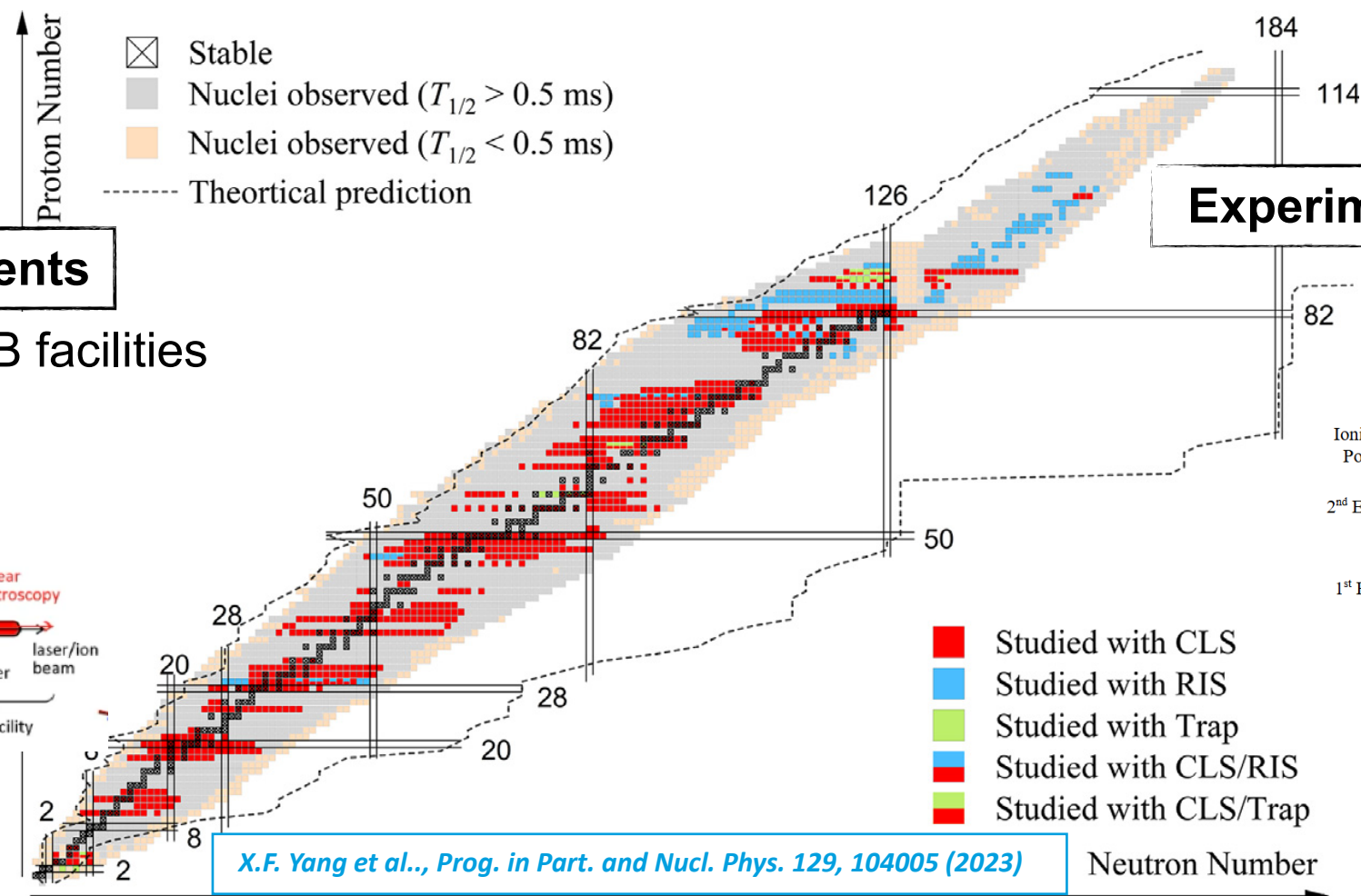
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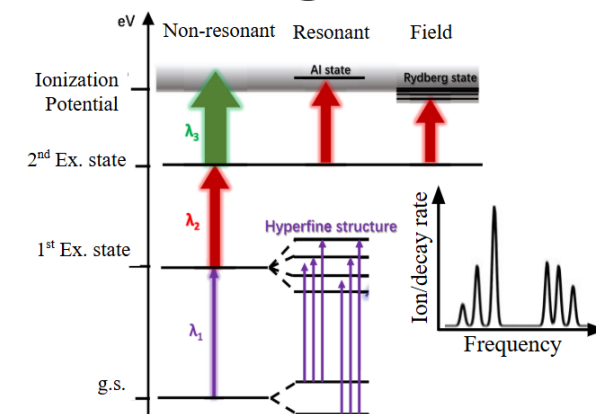
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## Experimental sensitivity

new methods

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- MIRACLS@ISOLDE
- PRECIOSA @ MIT
- ROC @ ISOLDE
- ...

## Mass and field shift factors



## Precision

needed for certain applications  
access to new observables

- 2-photon spectroscopy @ PLASSEN  
Precision Laser Spectroscopy for Exotic Nuclei
- Laser cooling - MIRACLS @ ISOLDE  
- **STRIPE** @ Leuven

## Reference radii

- revival of muonic X-ray spectroscopy

muX@PSI

e.g.  $^{40}\text{K}$  from



| 39K      | 40K                            | 41K     |
|----------|--------------------------------|---------|
| STABLE   | $1.25 \times 10^9$ y           | STABLE  |
| 93.2581% | 0.0117%                        | 6.7302% |
|          | $\beta^- = 89.28\%$            |         |
|          | $\epsilon + \beta^+ = 10.72\%$ |         |

(apologies for any developments omitted due to time limitations)

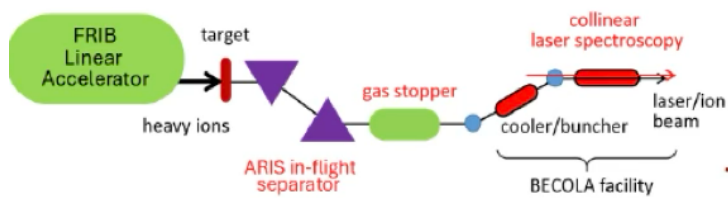


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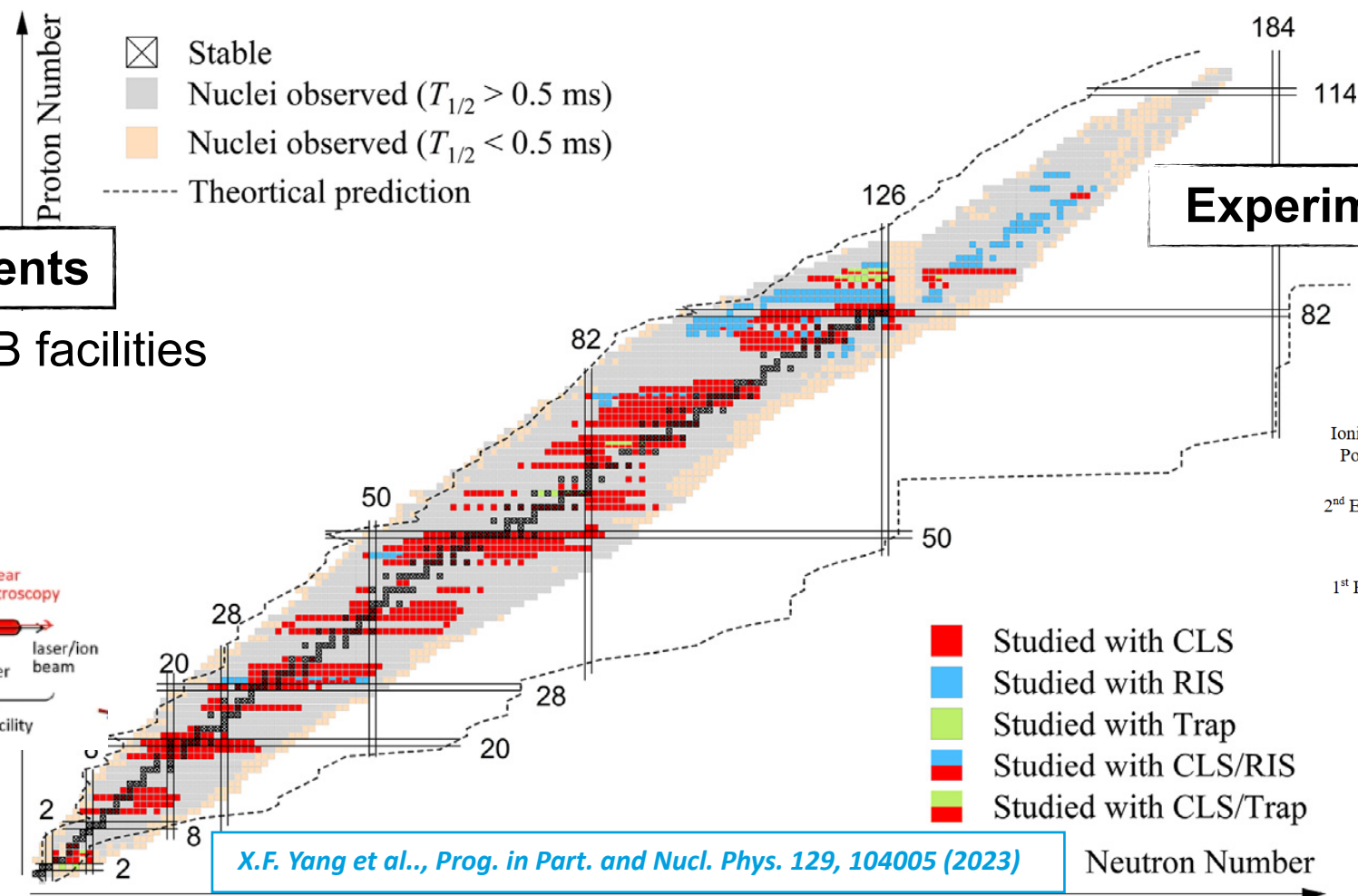
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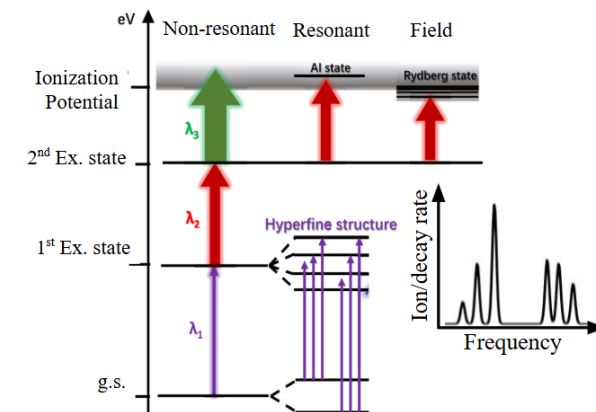
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## Experimental sensitivity

new methods

- CRIS @ ISOLDE



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- ...

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## Precision

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## Reference radii

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muX@PSI  
e.g.  $^{40}\text{K}$  from NRF  
National Research Foundation

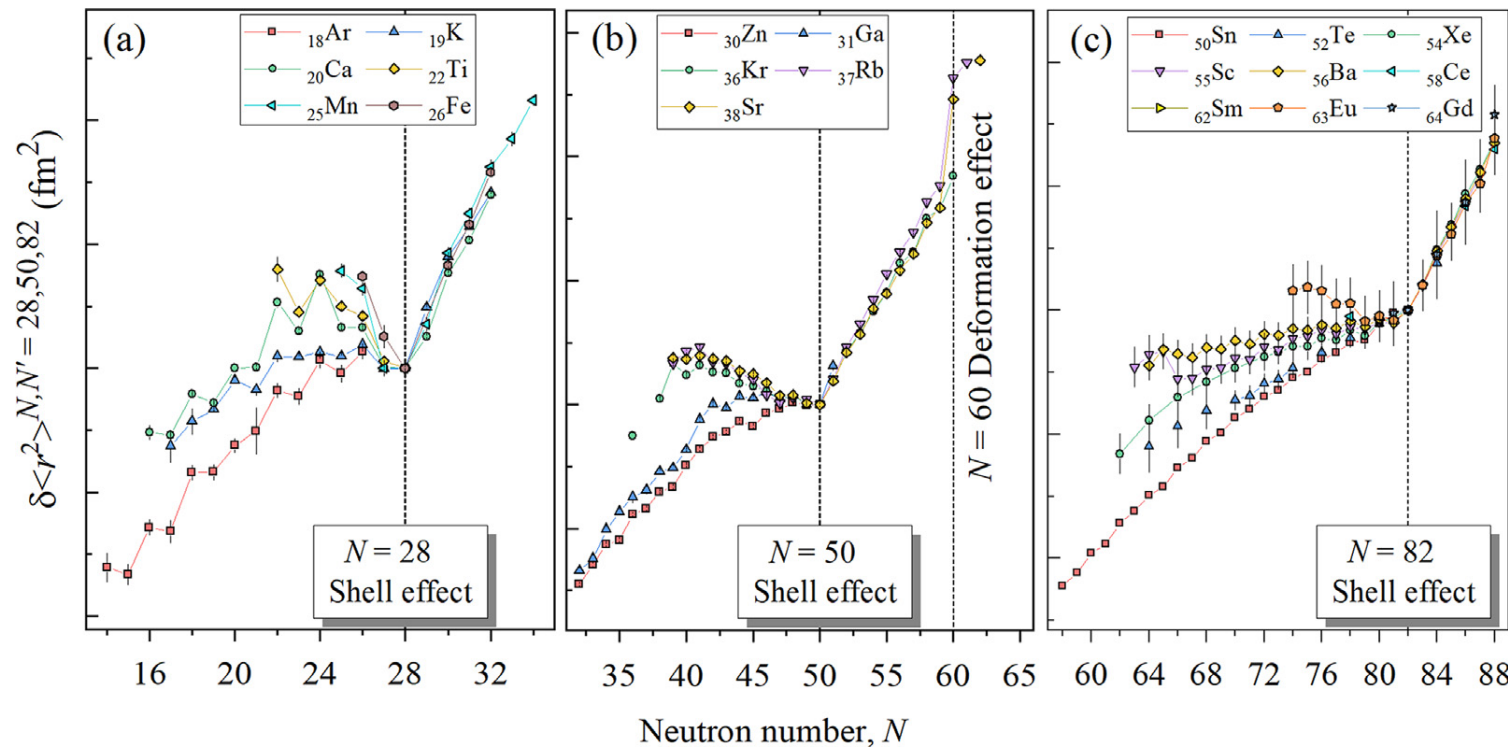
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## Contamination

- minimization at source
- MR-TOF after CRIS:  
REBEL@Leuven

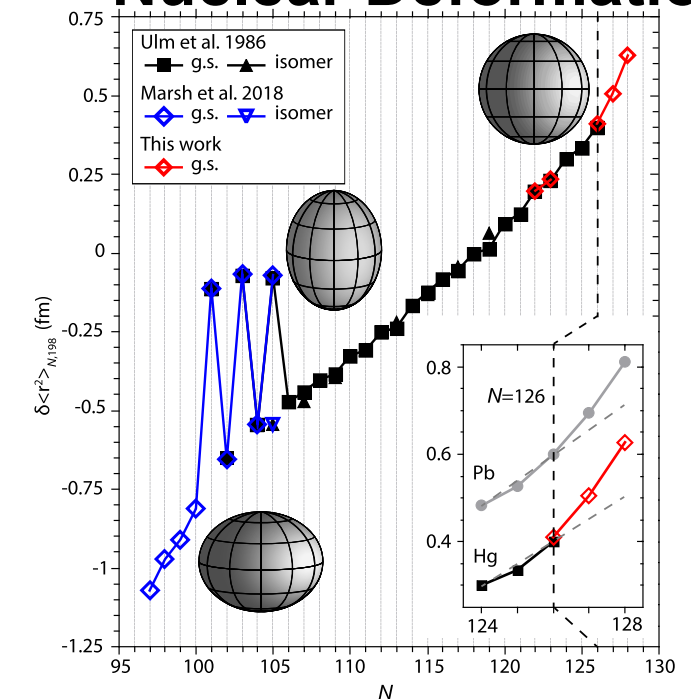
# Physics of Nuclear Charge Radii

## Nuclear Shell Structure



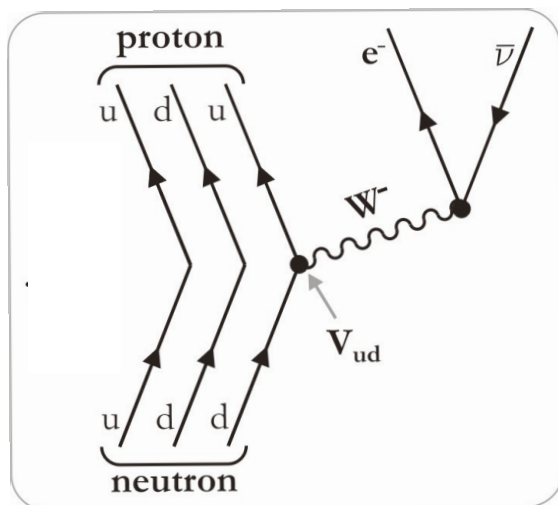
*X.F. Yang et al., Prog. in Part. and Nucl. Phys. 129, 104005 (2023)*

## Nuclear Deformation



*B. A. Marsh et al., Nature Physics, 14, 1163 (2018)*  
*T. Day Goodacre et al., PRL 126, 032502 (2021)*

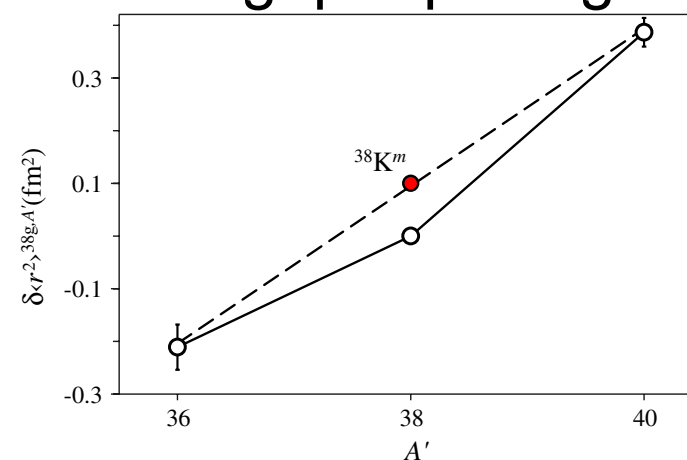
## Fundamental symmetries



*Mané et al., Phys. Rev. Lett. 107, 212502 (2011)*  
*Plattner et al., Phys. Rev. Lett. 131, 222502 (2023)*

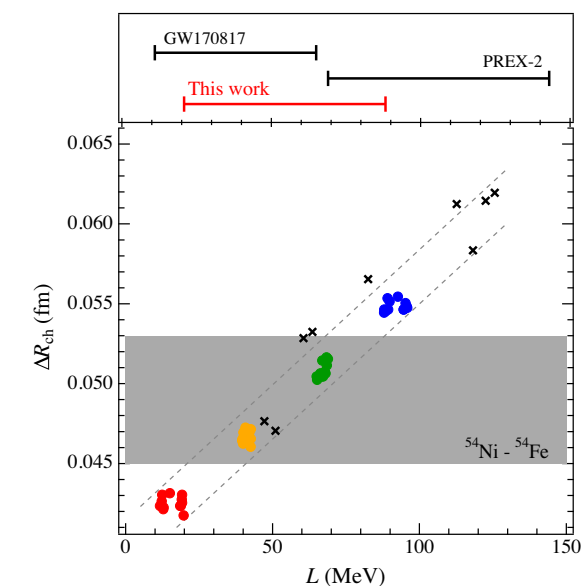
## Nuclear Pairing

e.g. p-n pairing



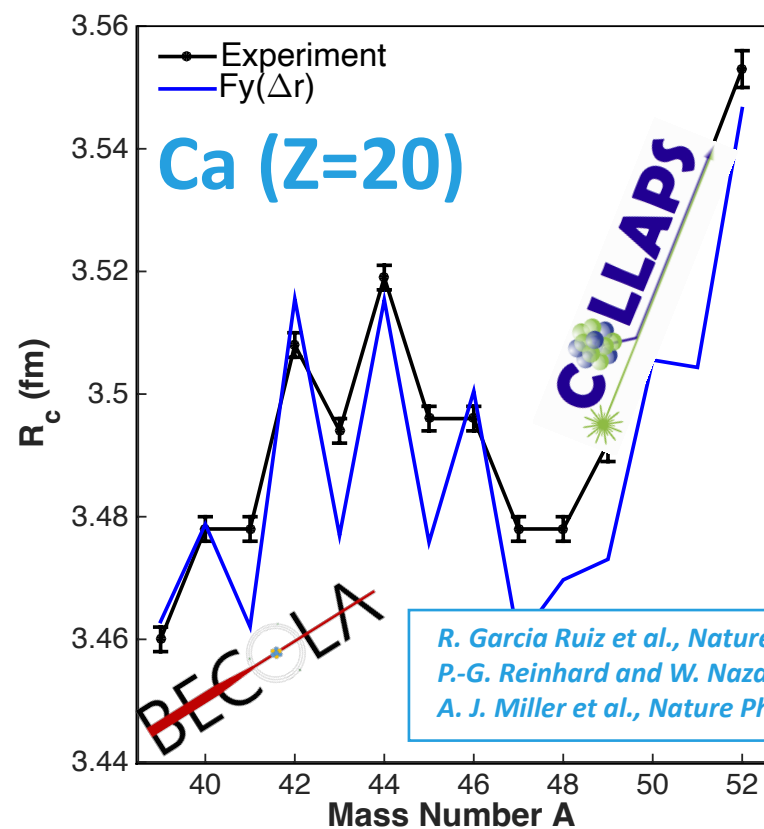
*M. L. Bissell et al., Phys. Rev. Lett. 113, 052502 (2014)*  
*Koszorús et al., Physics Letters B 819 (2021) 136439*

## Nuclear Matter



*Pineda et al., Phys. Rev. Lett. 127, 182503 (2021)*

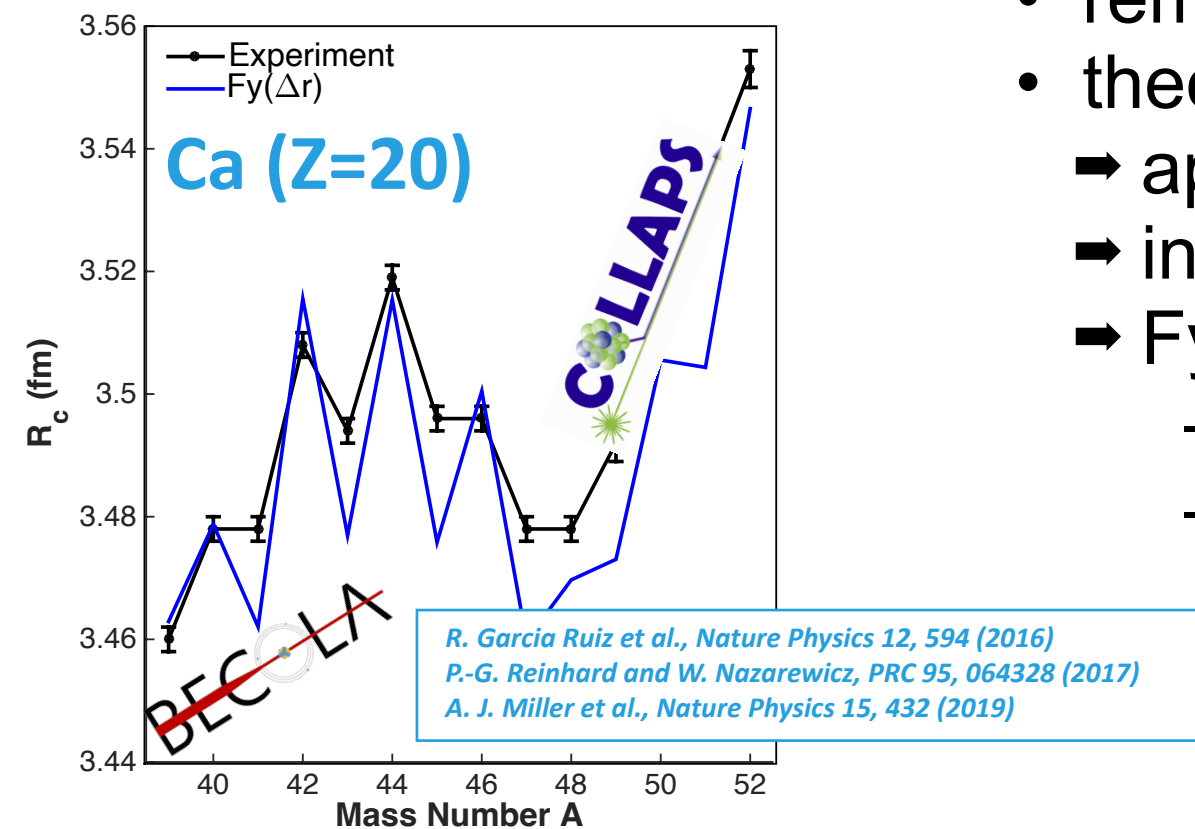
# Towards a 'universal' description of charge radii



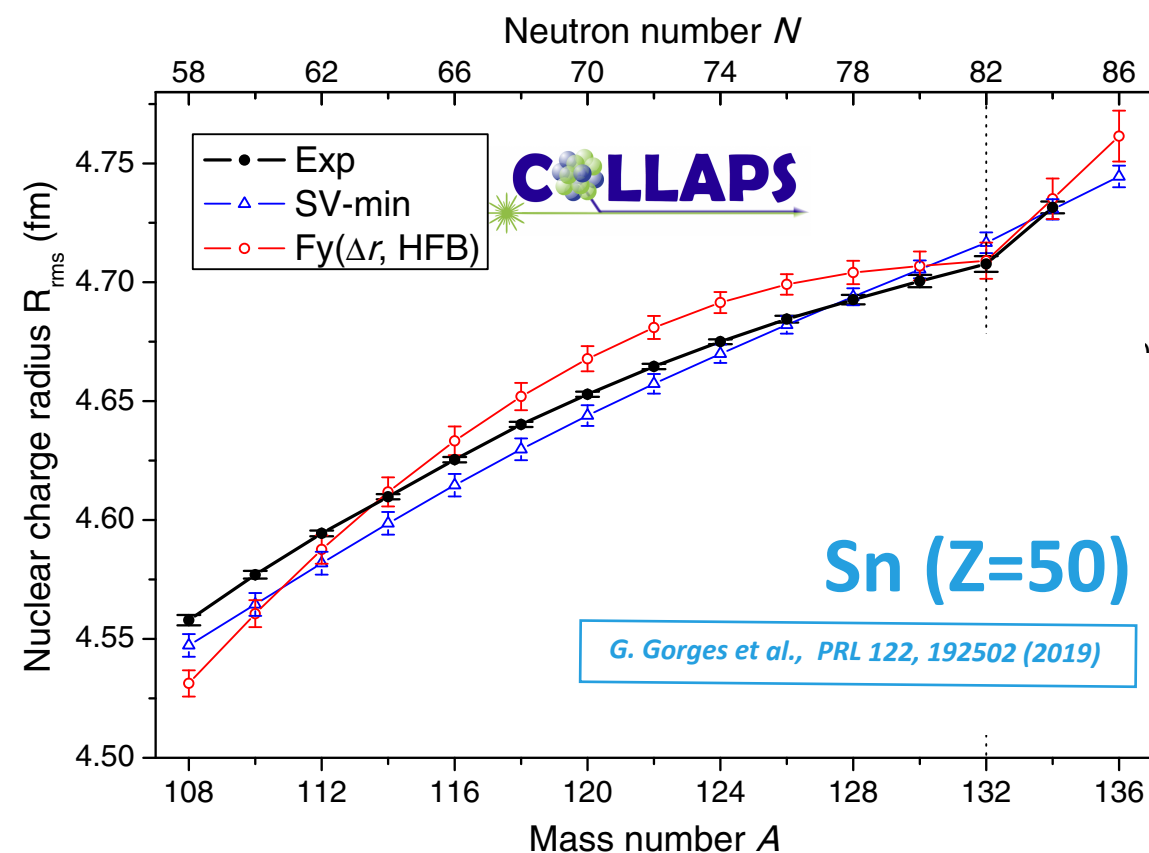
- remarkable progress in theory & experiment
- theoretical models:
  - ➔ applicable over wider mass range
  - ➔ including DFT and ab-initio methods
  - ➔  $Fy(\Delta r)$  excellent agreement to experiment
    - 'kink' at shell closures
    - odd-even staggering



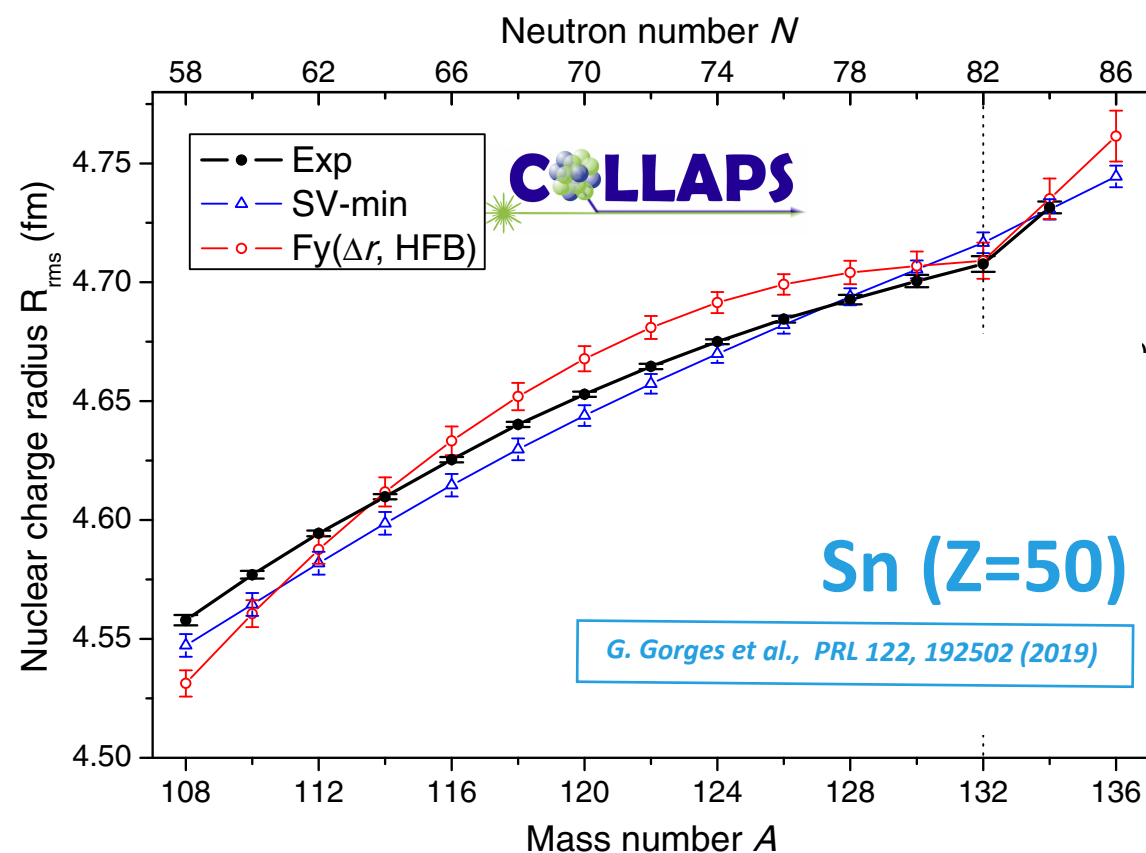
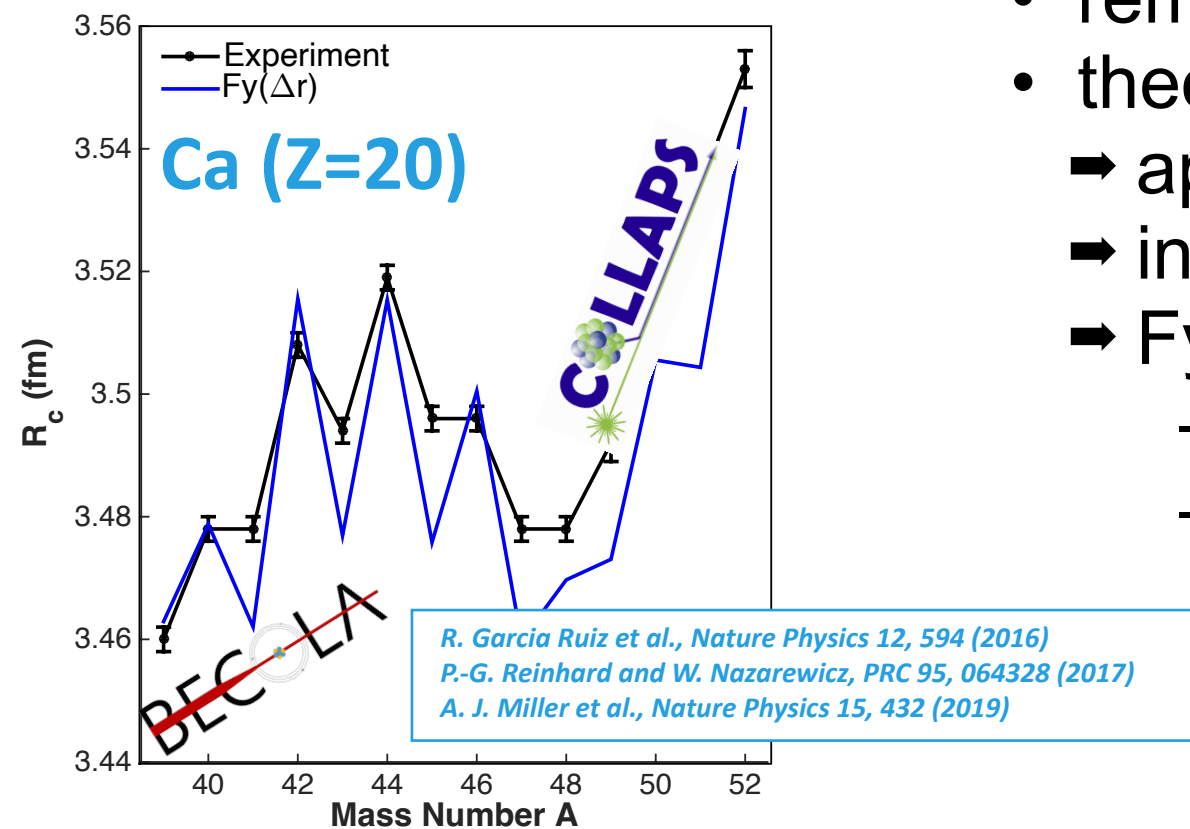
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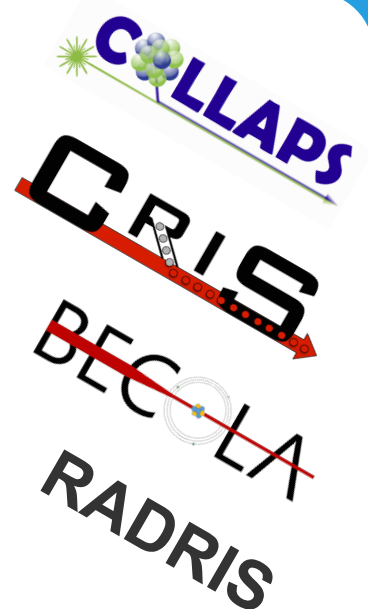
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    - odd-even staggering

8

## Exp. validations of Fayans DFT

| Element | Z   | Reference  |
|---------|-----|--|
| K       | 19  | Nature Physics 17, 439 (2021)                                  |
| Ca      | 20  | Nature Physics 12, 594 (2016)<br>Nature Physics 15, 432 (2019) |
| Fe      | 26  | Phys. Rev. Lett., 117, 252501 (2016)                           |
| Ni      | 28  | Phys. Rev. Lett., 128, 022502 (2022)                           |
| Cu      | 29  | Nature Physics 16, 620 (2020)                                  |
| Ge      | 32  | Physics Letters B, 856, 138867 (2024)                          |
| Cd      | 48  | Phys. Rev. Lett., 121, 102501, (2018)                          |
| In      | 49  | Nature Physics, 20, 1719 (2024)                                |
| Sn      | 50  | Phys. Rev. Lett., 122, 192502 (2019)                           |
| Fm      | 100 | Nature, 634, 1075 (2024)                                       |

**A = 36 - 134 ( plus 245-257 )**



# Towards a 'universal' description of charge radii

- remarkable progress in theory and experiment

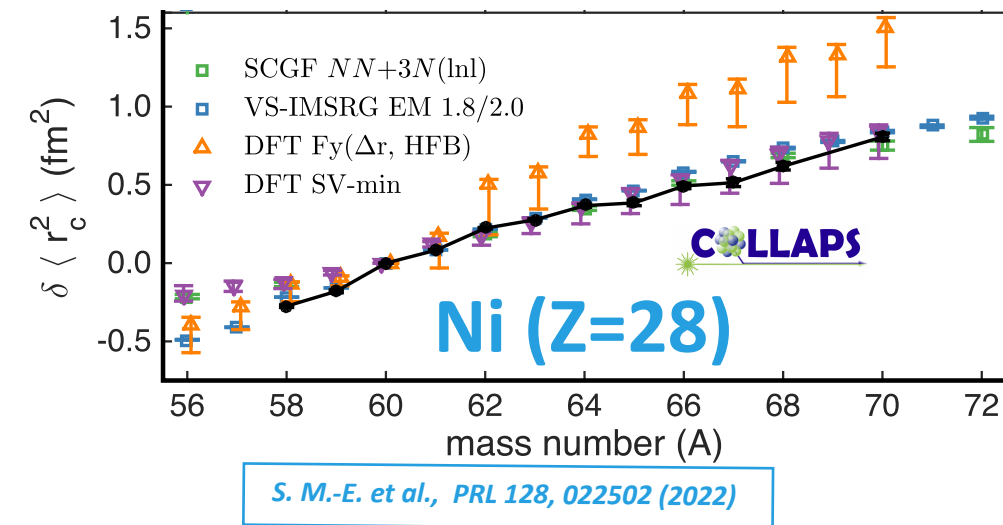
- theoretical models:

**But also shortcoming in Fayans functional  $Fy(\Delta r)$**

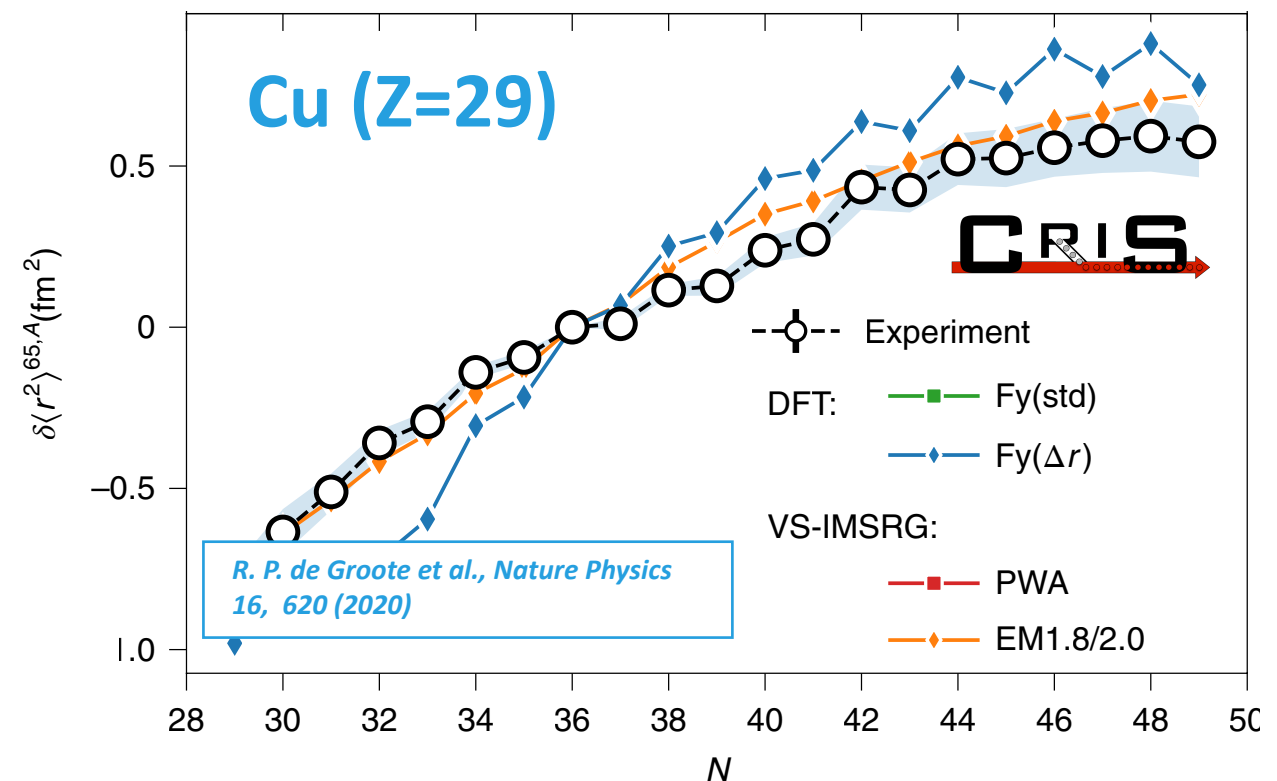
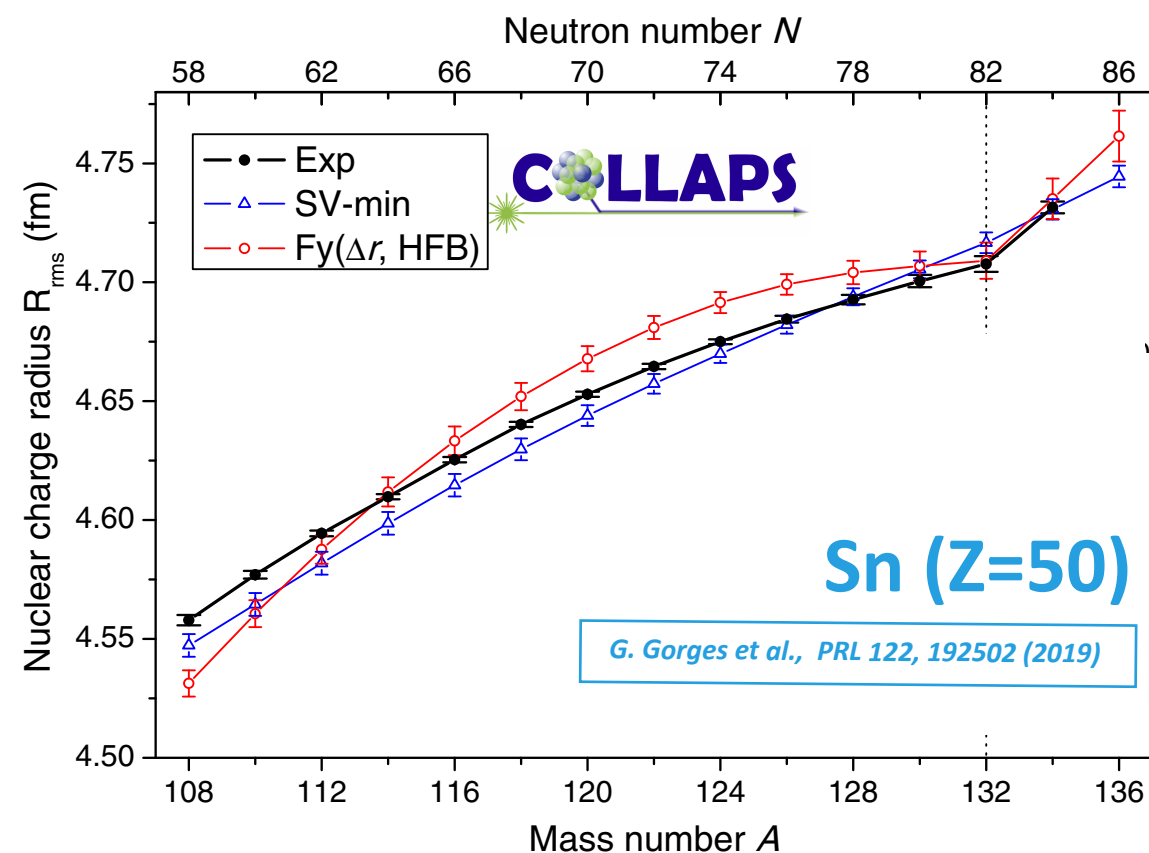
e.g. parabolic trend for Ni/Cu/Cd/Sn

→ newly-introduced isovector term

9



*Nature Physics, 20, 1719 (2024)*  
*Physics Letters B, 856, 138867 (2024)*



# Towards a 'universal' description of charge radii

- remarkable progress in theory and experiment

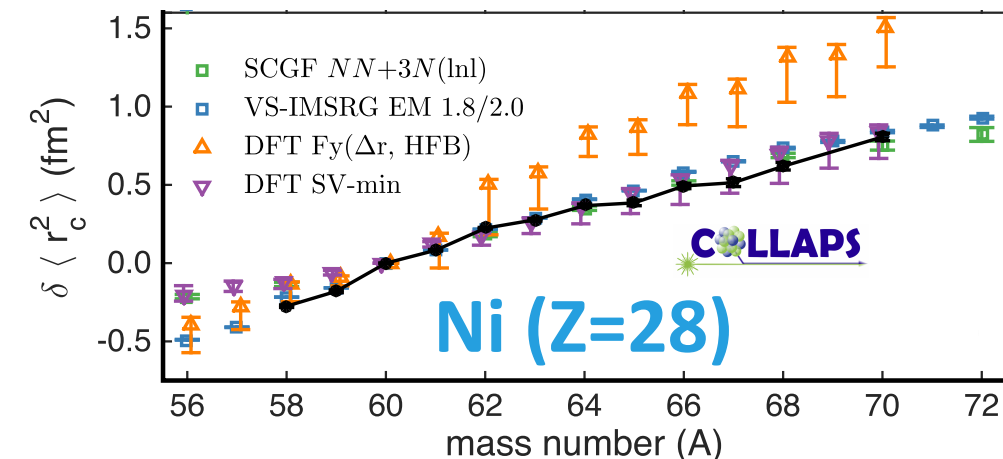
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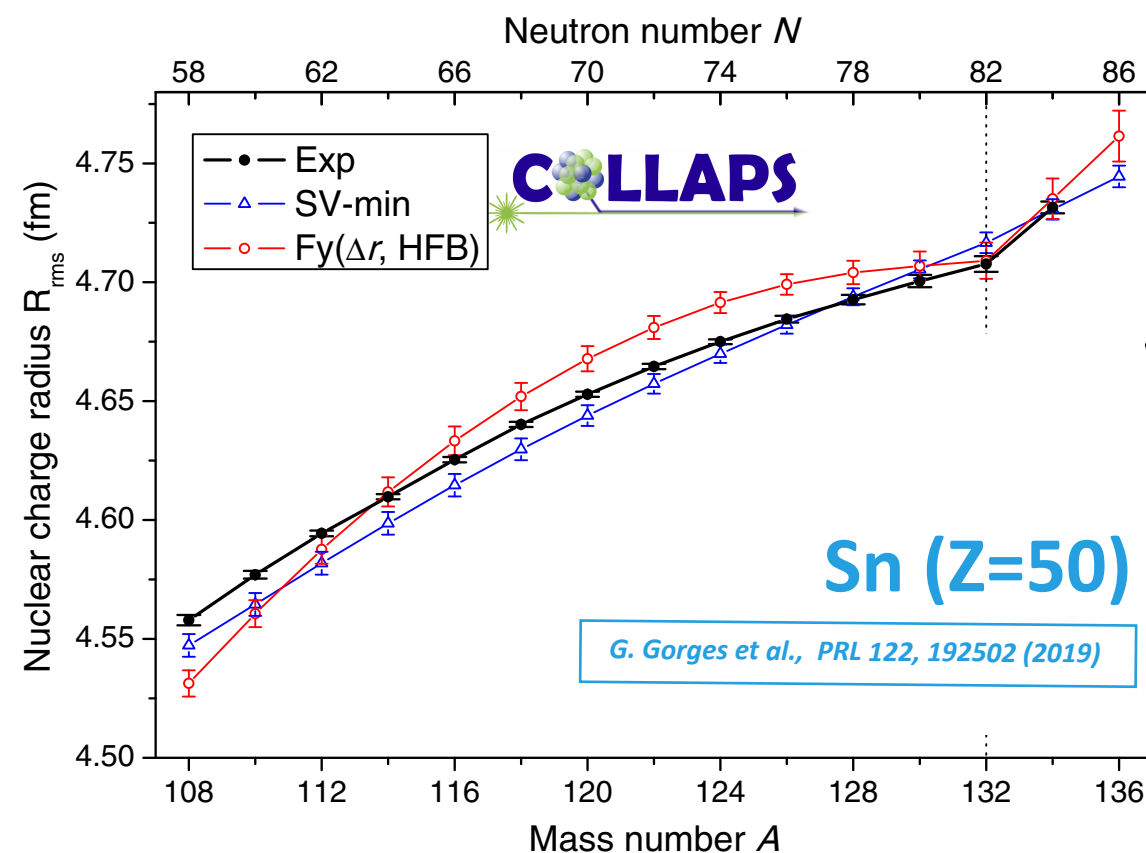
9



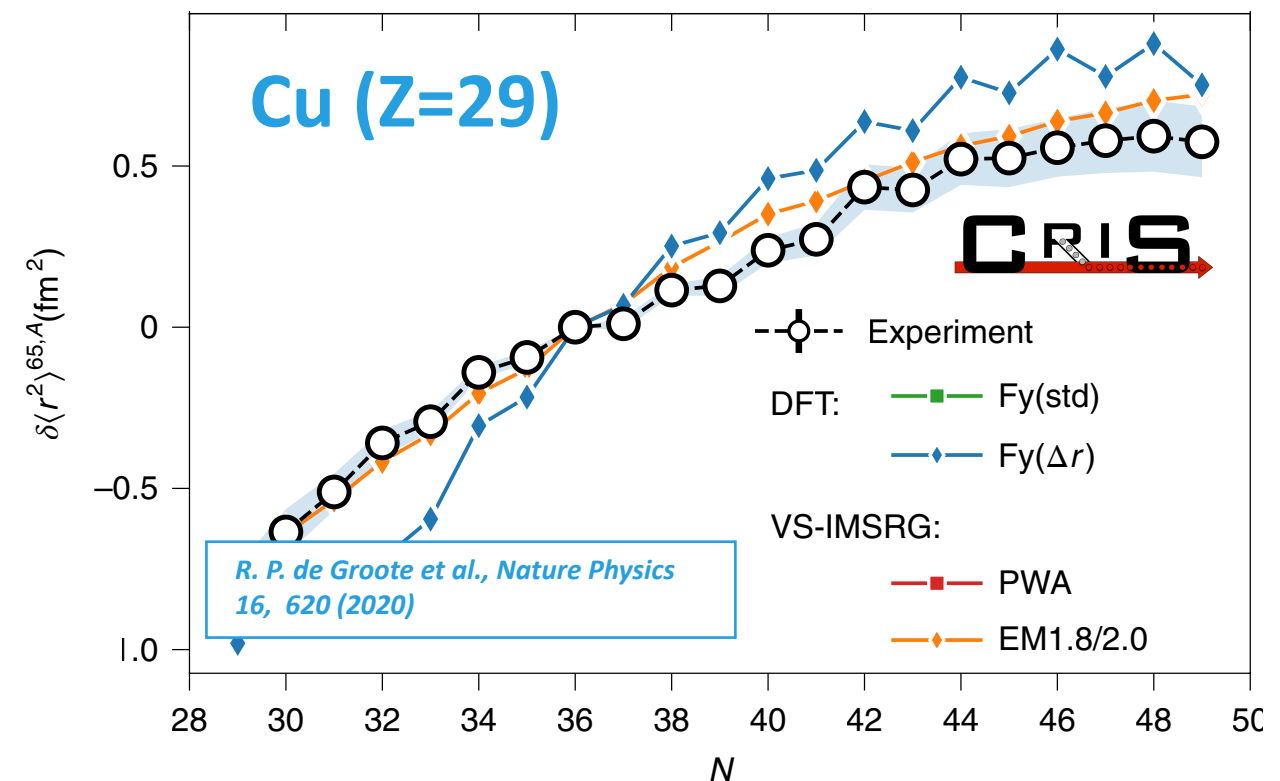
*S. M.-E. et al., PRL 128, 022502 (2022)*

*Nature Physics, 20, 1719 (2024)*  
*Physics Letters B, 856, 138867 (2024)*

⇒ new exp. benchmarks needed!



*G. Gorges et al., PRL 122, 192502 (2019)*



*R. P. de Groote et al., Nature Physics 16, 620 (2020)*

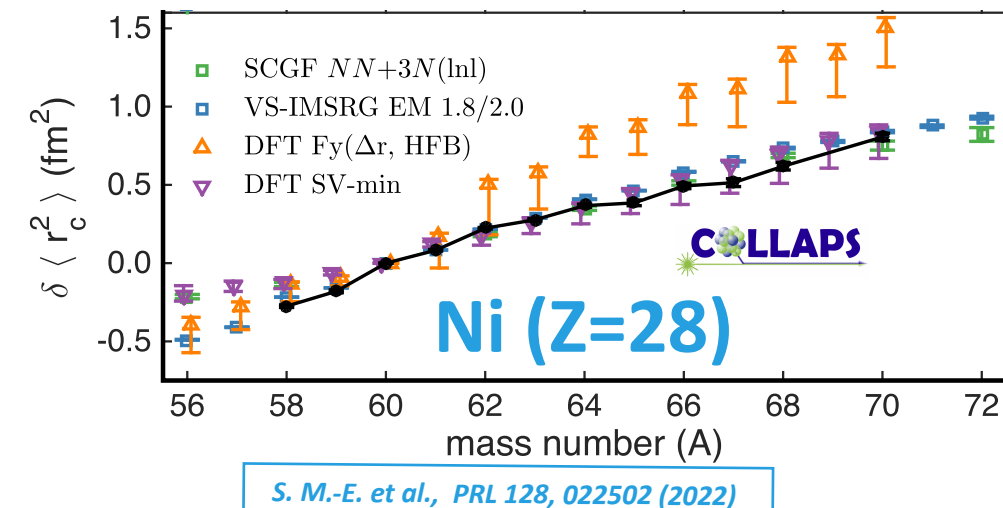


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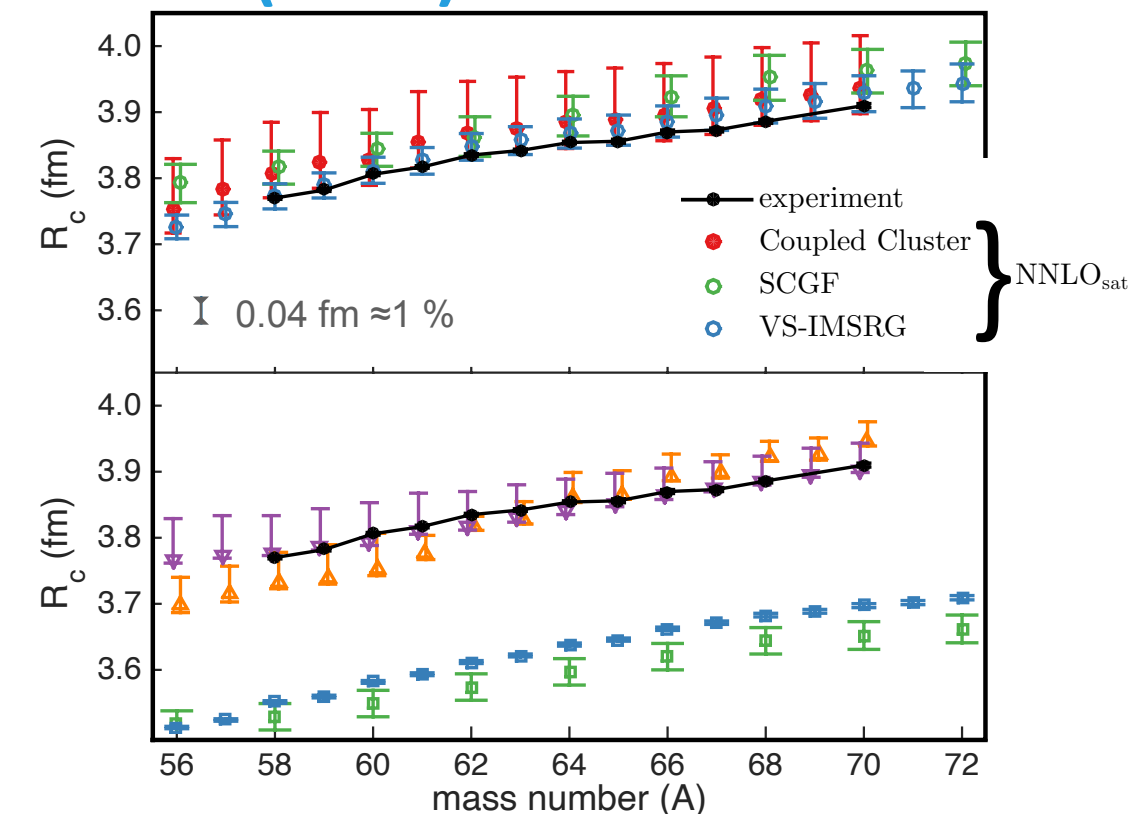
## Benchmark: ab initio calculations next to shell closure

- reproduces experiment at the  $\approx 1\%$  level
- agreement within *ab initio* methods  $\Rightarrow$  accuracy check
- importance of used nuclear potential: needs to capture relevant physics

10



## Ni (Z=28)



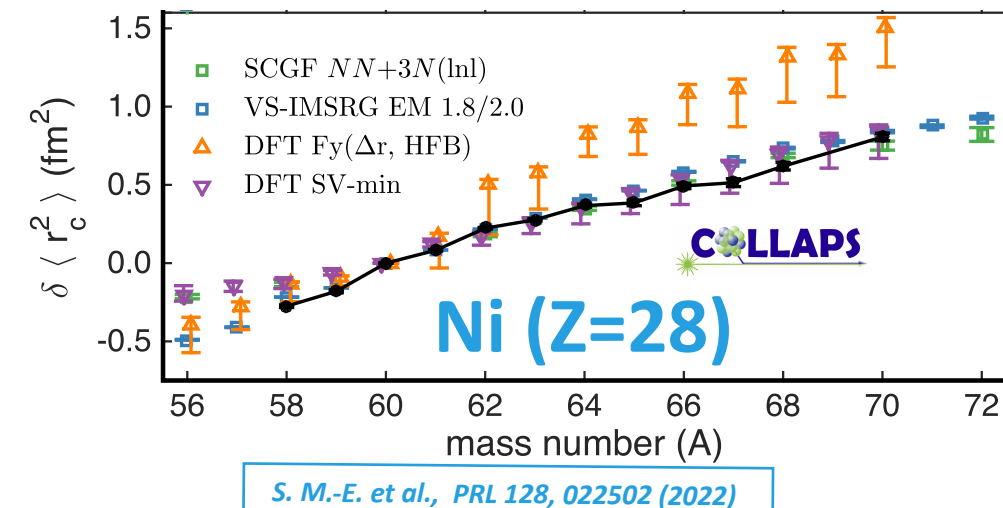
- SCGF NN+3N(lnl)
- VS-IMSRG EM 1.8/2.0

# Towards a 'universal' description of charge radii

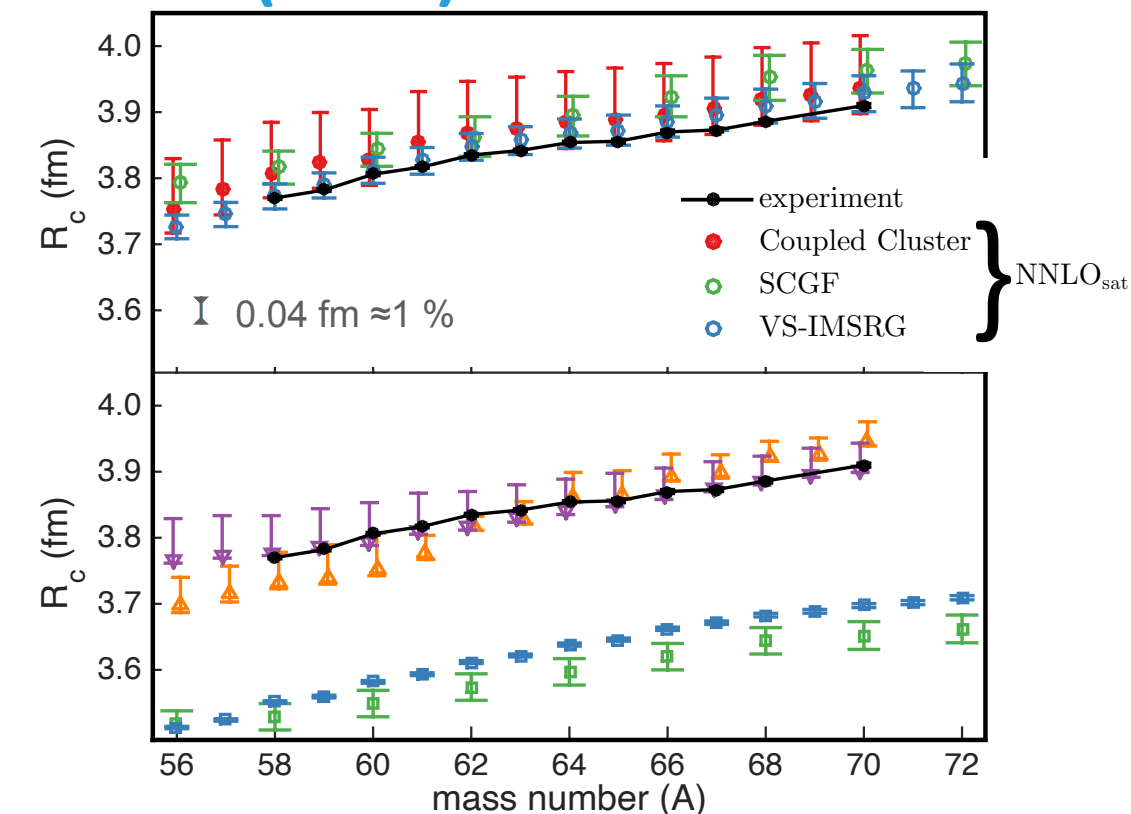
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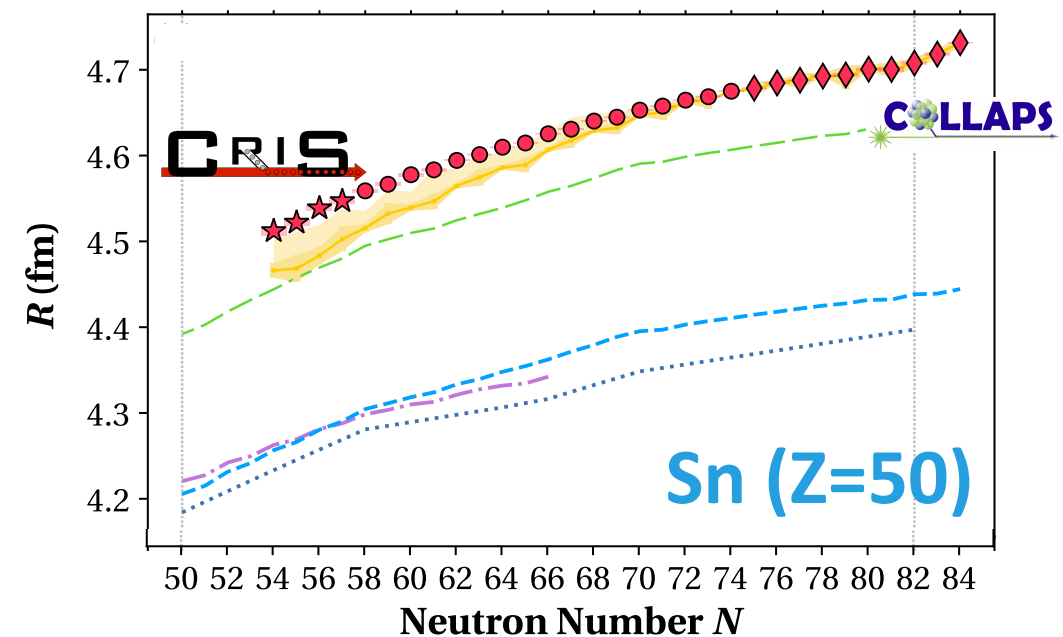


Ni (Z=28)



- SCGF  $NN+3N(\ln l)$
- VS-IMSRG EM 1.8/2.0

- ★ This Letter (CRIS)
- This Letter (CRIS+COLLAPS)
- ◆ This Letter (COLLAPS)
- ✦ Fy(IVP)
- 1.8/2.0(EM)[sph-HF]
- 1.8/2.0(EM)[<sup>100</sup>Sn]
- 1.8/2.0(EM)[<sup>80</sup>Zr]
- $\Delta N^2\text{LO}_{\text{GO}}(394)[^{100}\text{Sn}]$



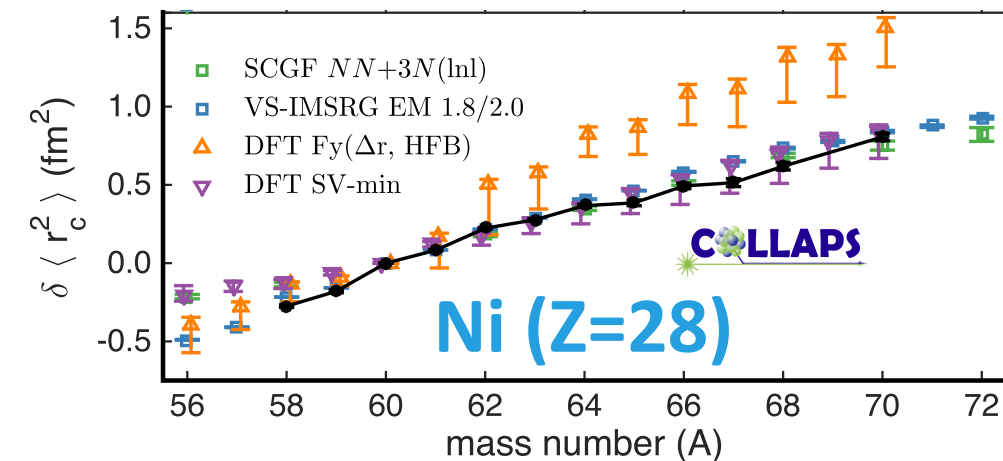
*F. P. Gustafsson et al. arXiv:2504.17060, accepted in Phys. Rev. Lett.*

# Towards a 'universal' description of charge radii

## Benchmark: ab initio calculations next to shell closure

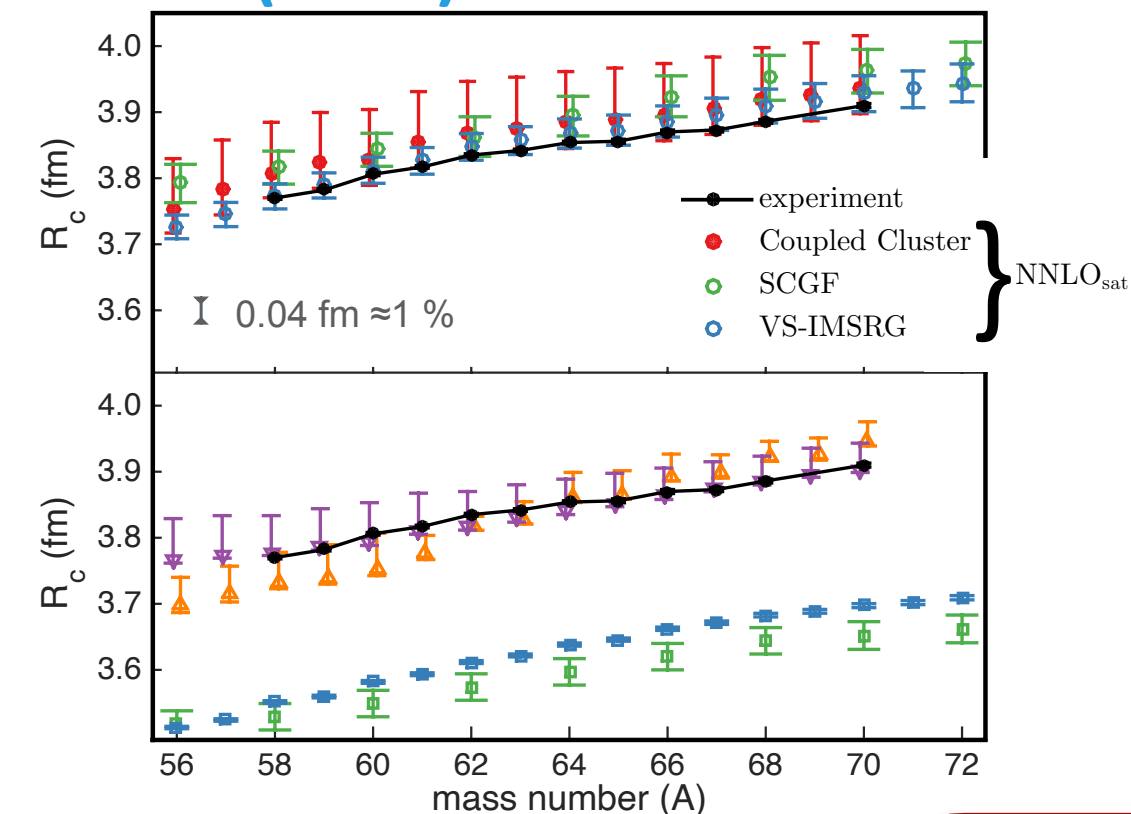
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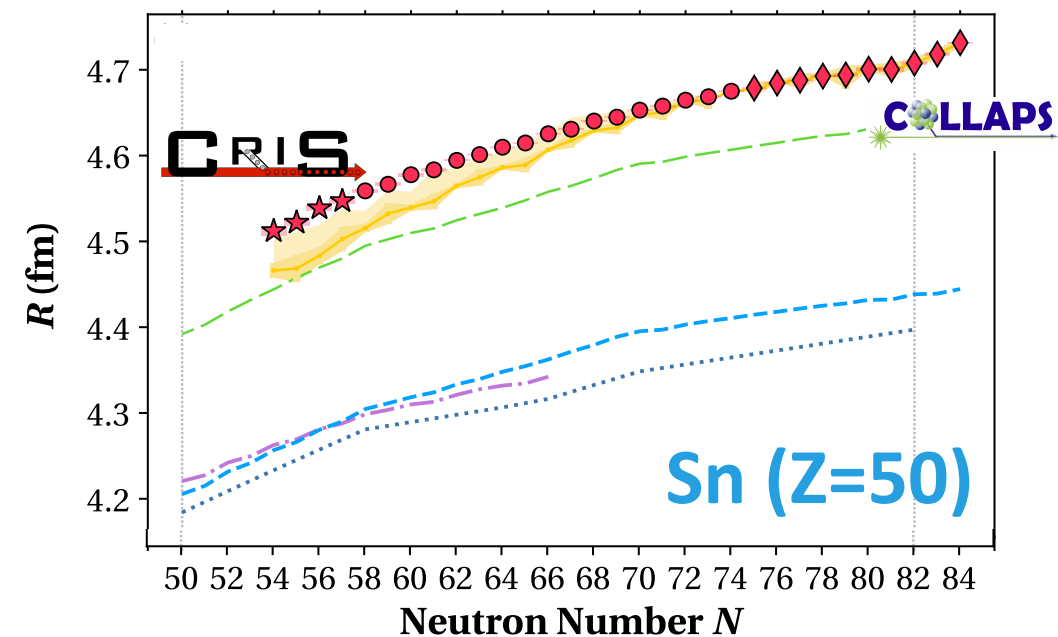
S. M.-E. et al., PRL 128, 022502 (2022)

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- 1.8/2.0(EM)[<sup>80</sup>Zr]
- $\Delta N^2\text{LO}_{\text{GO}}(394)[^{100}\text{Sn}]$



F. P. Gustafsson et al. arXiv:2504.17060, accepted in Phys. Rev. Lett.

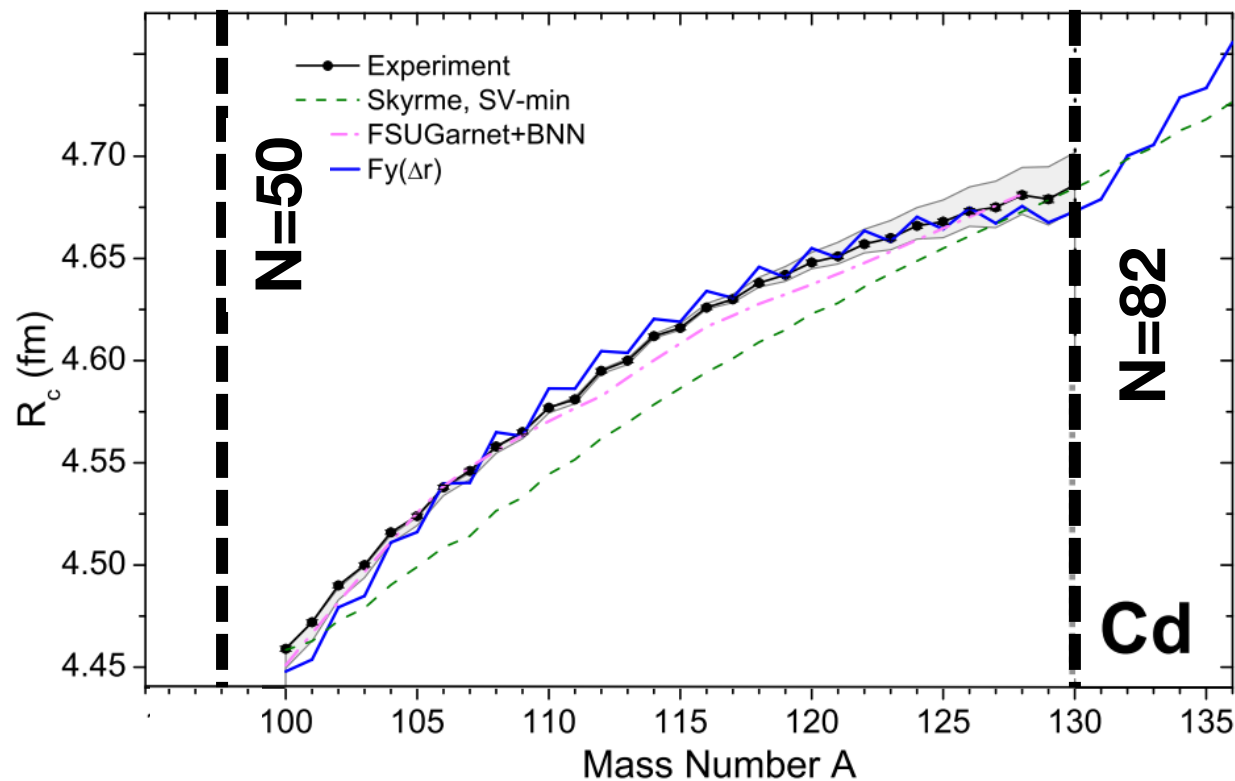
$\Rightarrow$  new benchmarks in mid-shell / deformed regions



# R<sub>c</sub>: new benchmarks...

## ....for Fayans DFT

- kink at N=82?
- curvature  $\Leftrightarrow$  isovector term in DFT

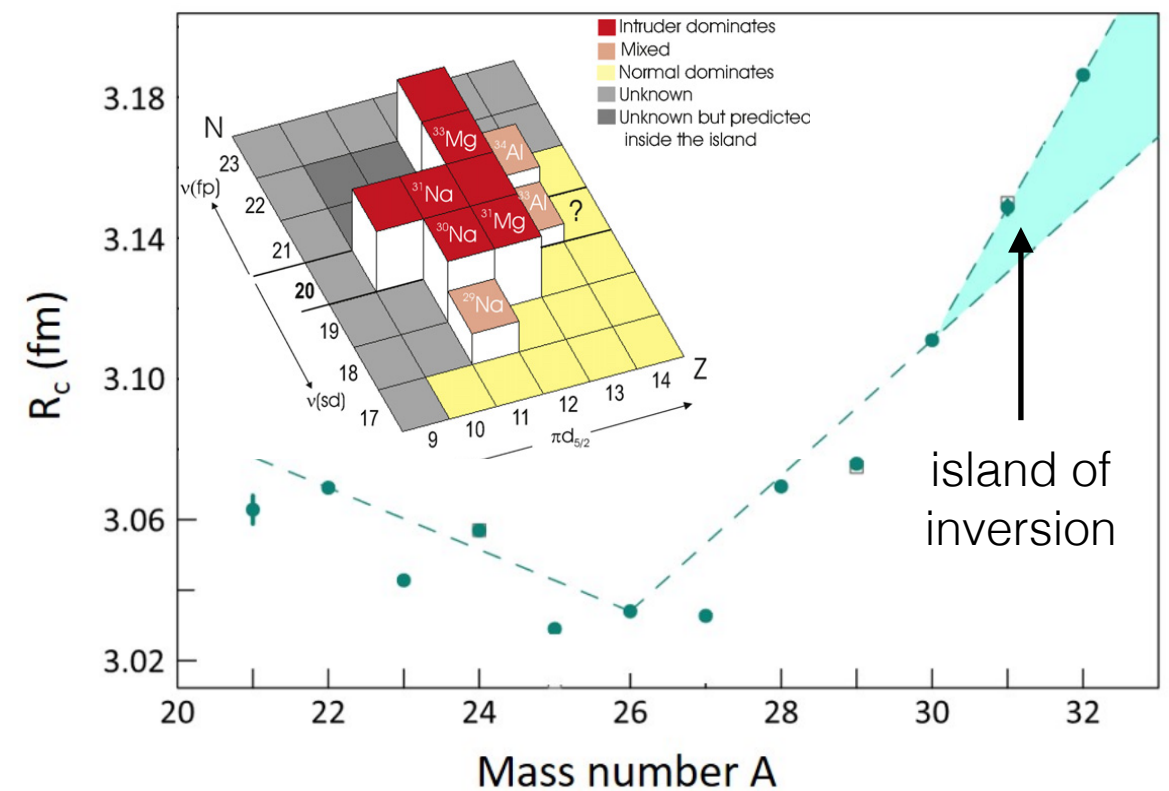


G. Gorges et al., PRL 122, 192502 (2019)

## ... for *ab initio* in deformed region

- island of inversion around  $^{32}\text{Mg}$
- shell evaluation

11

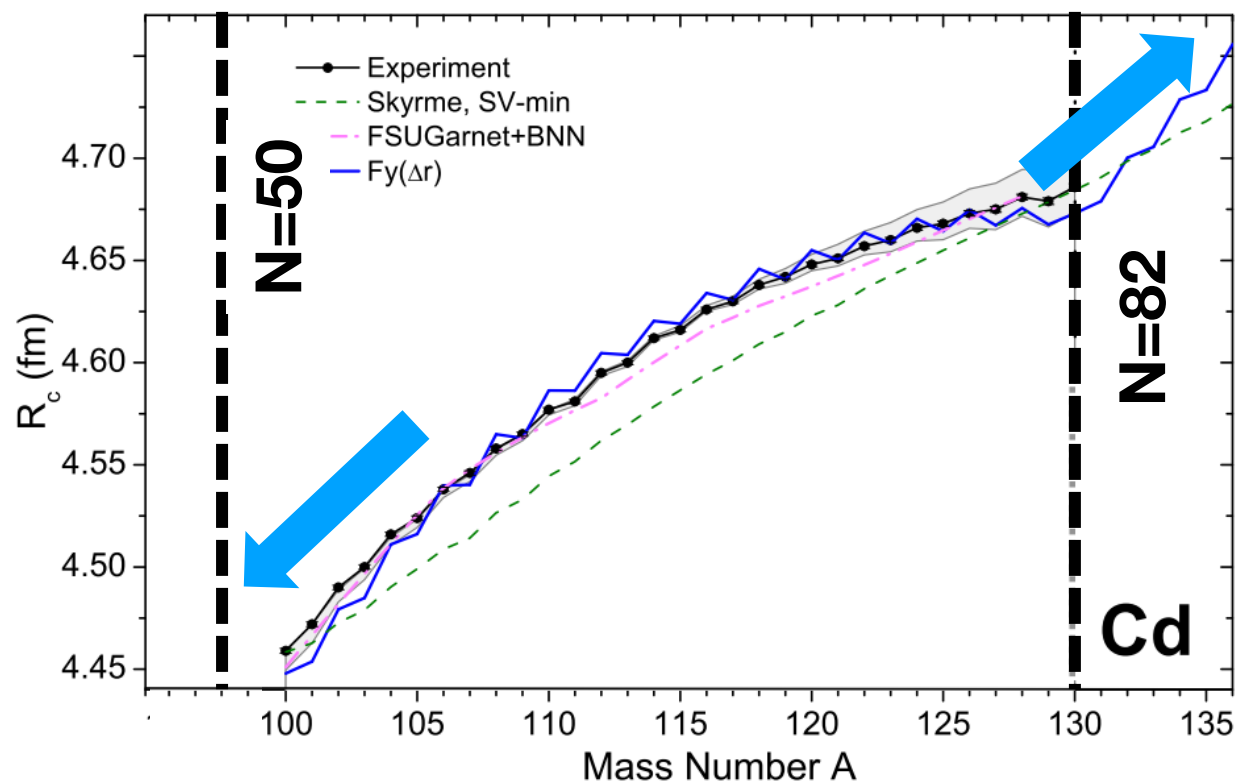


D. T. Yordanov et al., Phys. Rev. Lett. 108, 042504 (2012)

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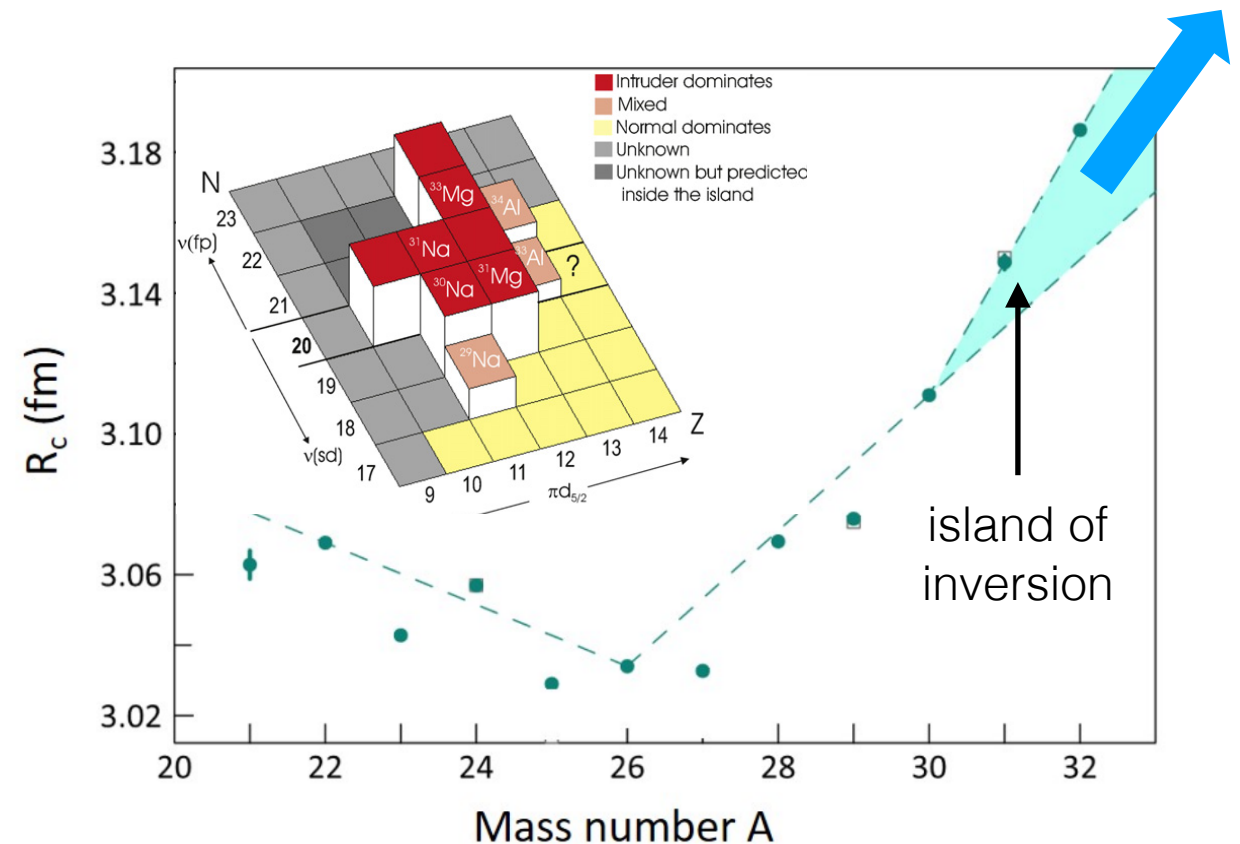
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G. Gorges et al., PRL 122, 192502 (2019)

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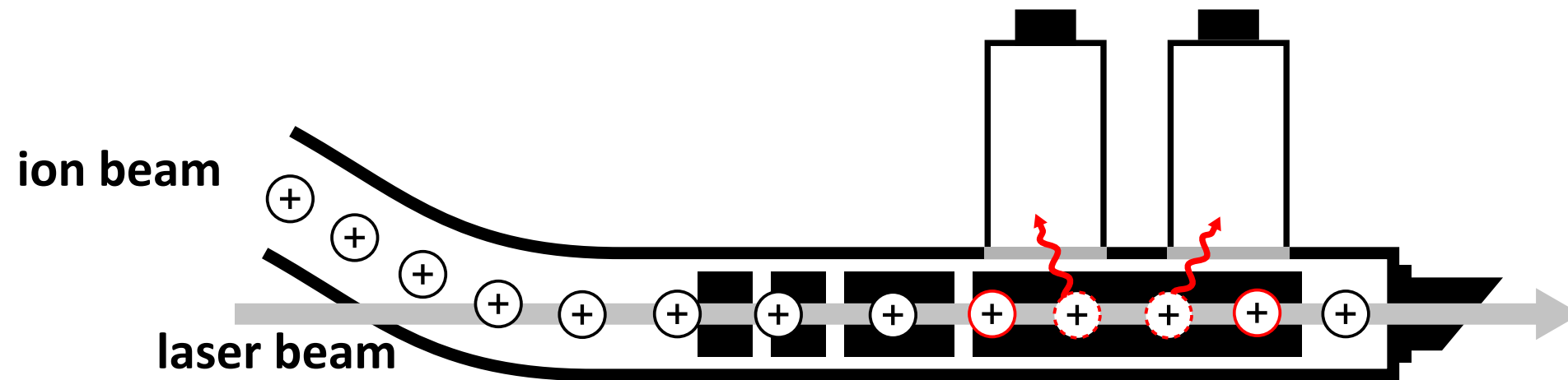
- island of inversion around  $^{32}\text{Mg}$
- shell evaluation



D. T. Yordanov et al., Phys. Rev. Lett. 108, 042504 (2012)

**Challenge: very low production yields <100 ions / sec**

# Strengths & Limitations of CLS



12

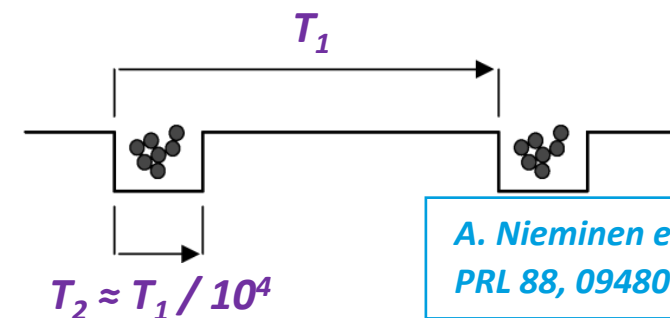


**beams of  $\geq 30$  keV**  
minimizes Doppler-broadening  
 $\Rightarrow$  high resolution

$$\delta\nu \propto \frac{\delta E}{\sqrt{E}}$$



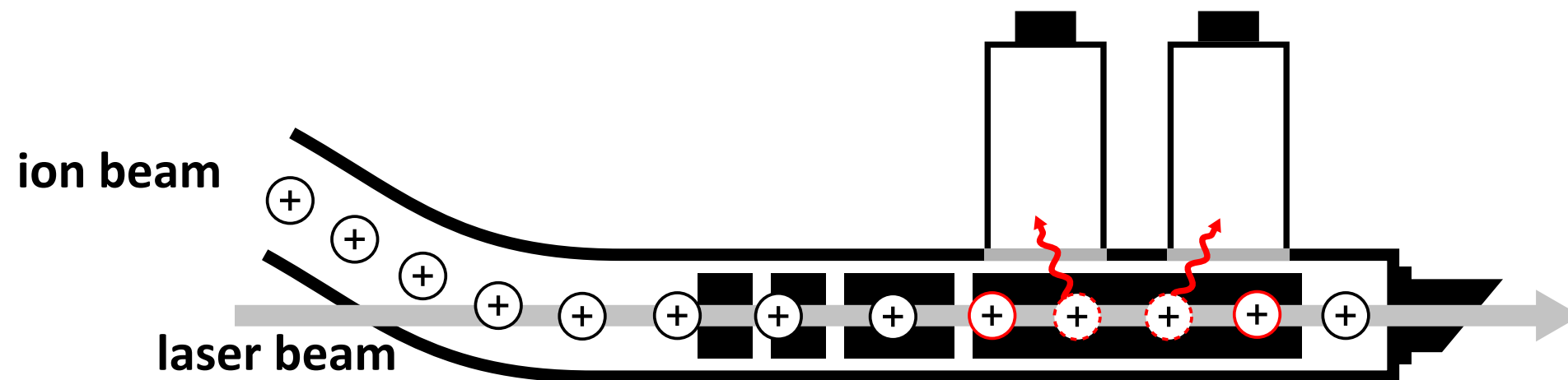
**Bunched beams:**  
reduce background  
by gating on bunch



*A. Nieminen et al.,  
PRL 88, 094801 (2002)*



# Strengths & Limitations of CLS



12

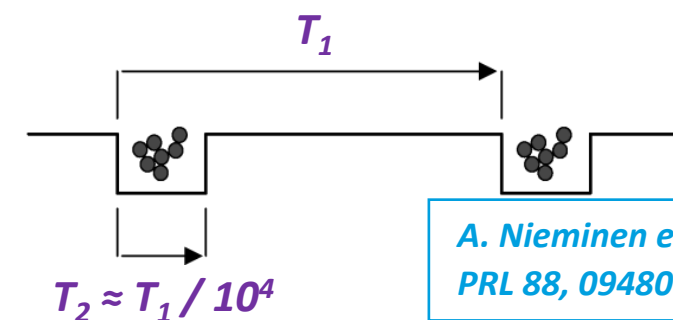


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$$\delta\nu \propto \frac{\delta E}{\sqrt{E}}$$



**Bunched beams:**  
reduce background  
by gating on bunch



*A. Nieminen et al.,  
PRL 88, 094801 (2002)*

**$T_{1/2}$  of accessible radionuclides:**  
5 ms to seconds

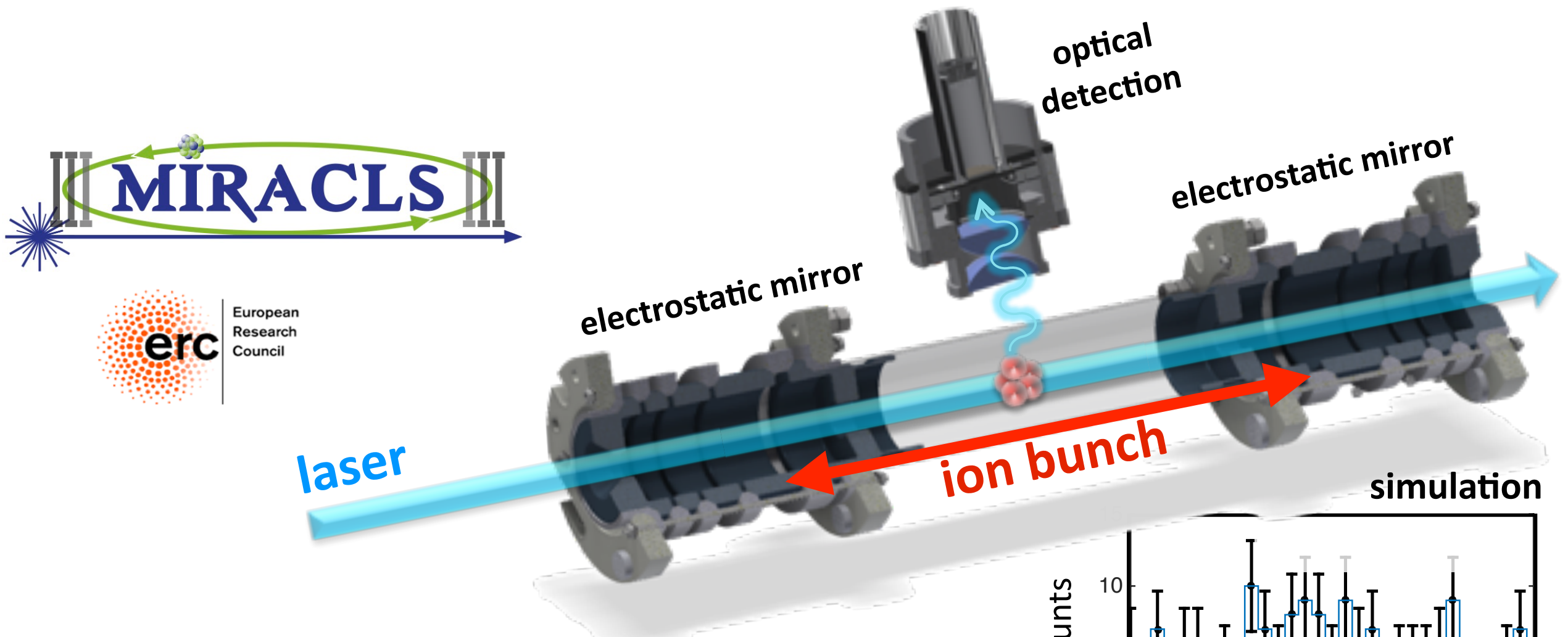
**effective use for CLS**  
100s of ns to a few  $\mu$ s

**! can one use exotic nuclides even more efficiently !**

# the Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy

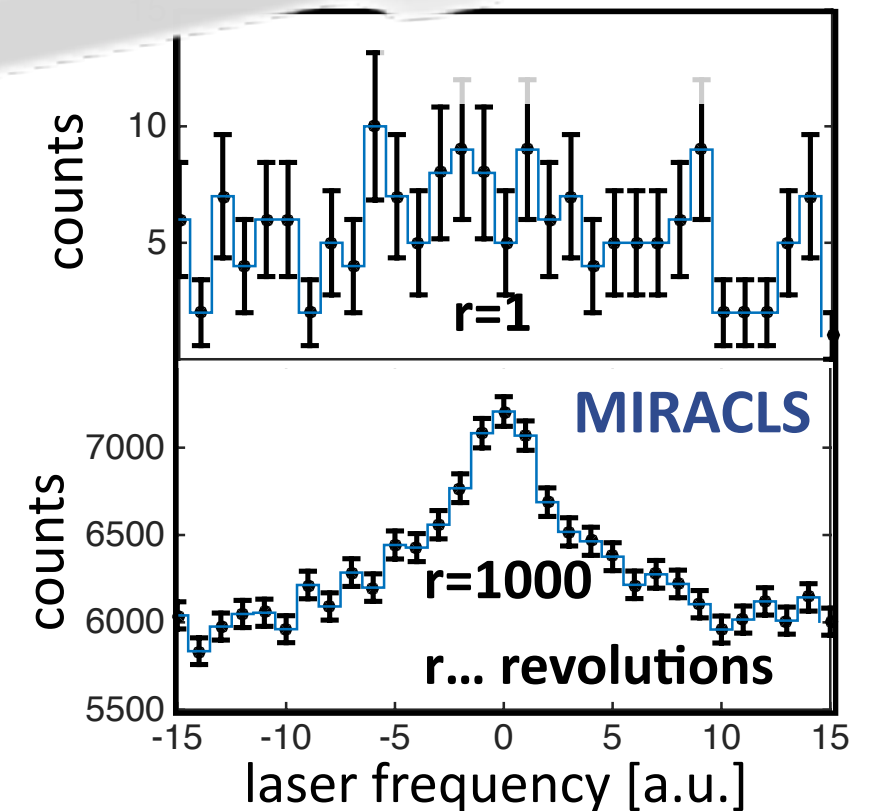
trap  $\Rightarrow$  long observation time  $\Rightarrow$  higher sensitivity  $\Rightarrow$  more exotic nuclides accessible

13



novel approach for collinear laser spectroscopy:

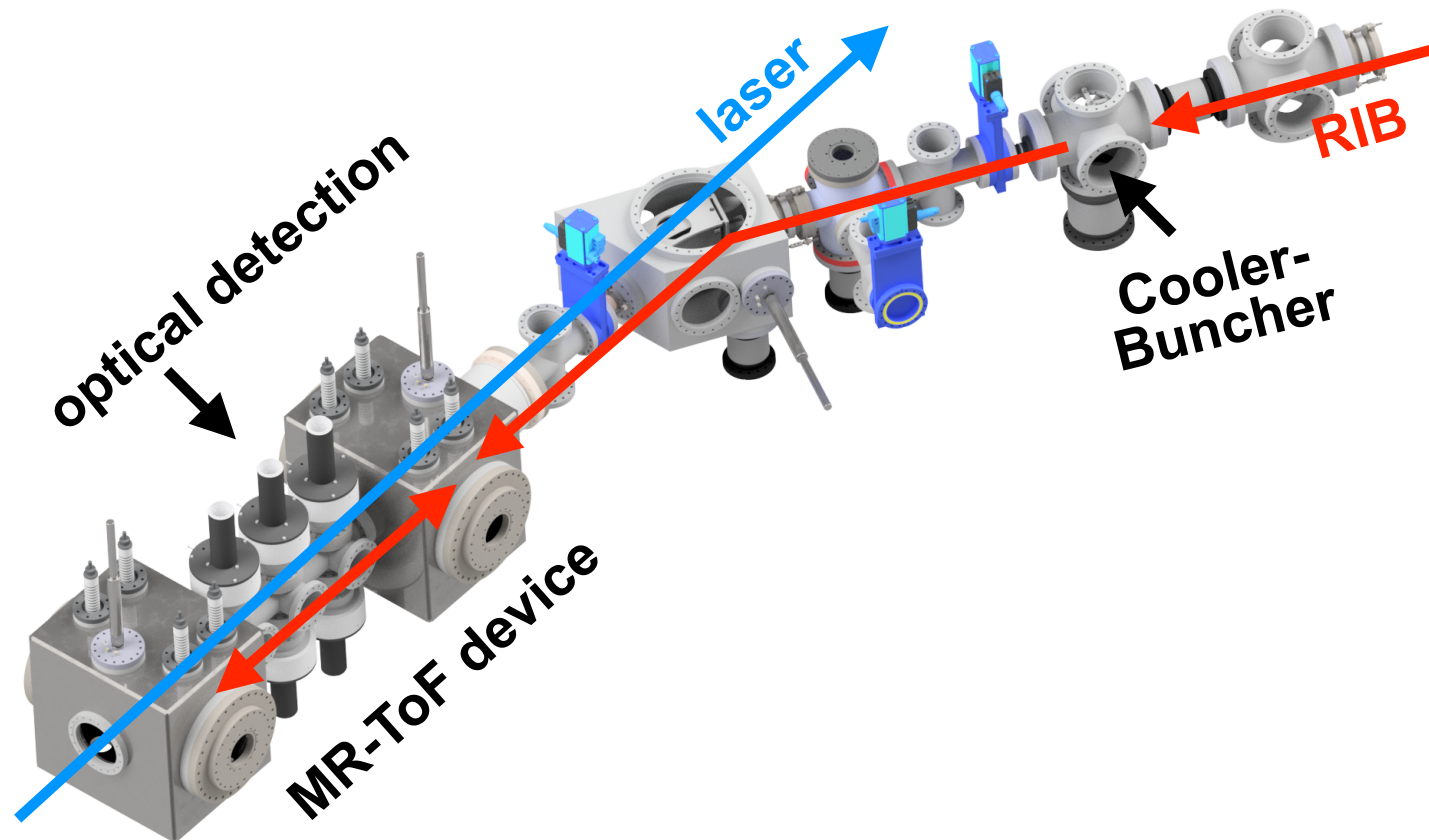
- ion trap  $\Rightarrow$  long observation time
- $>10$  keV beam  $\Rightarrow$  high resolution





setup at 

14



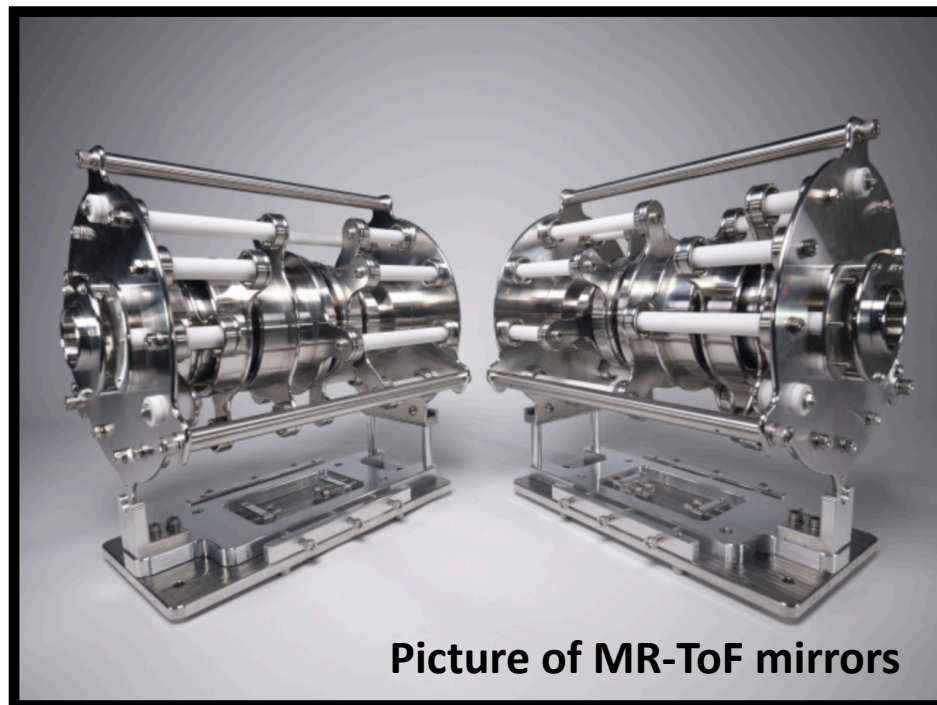
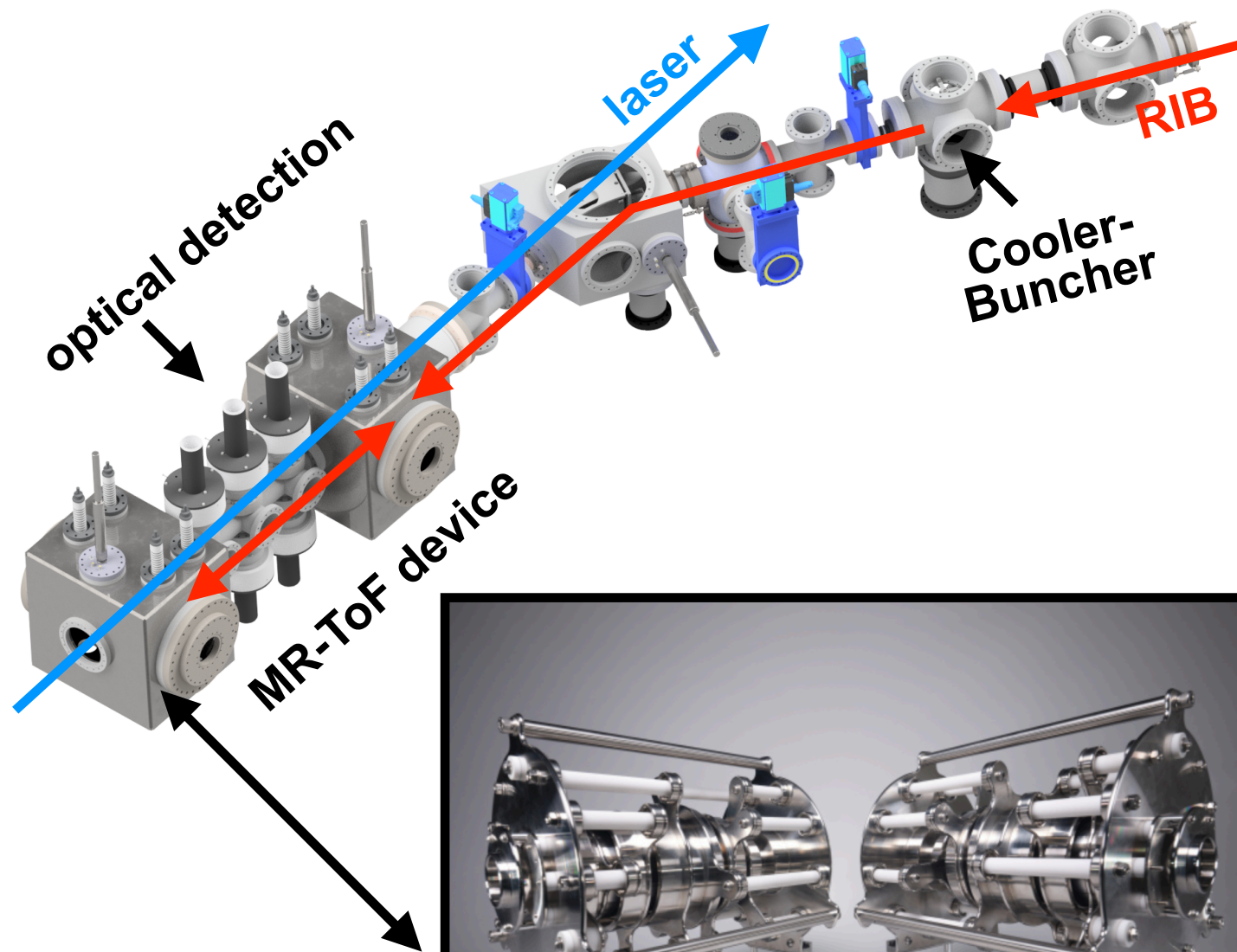




setup at



14



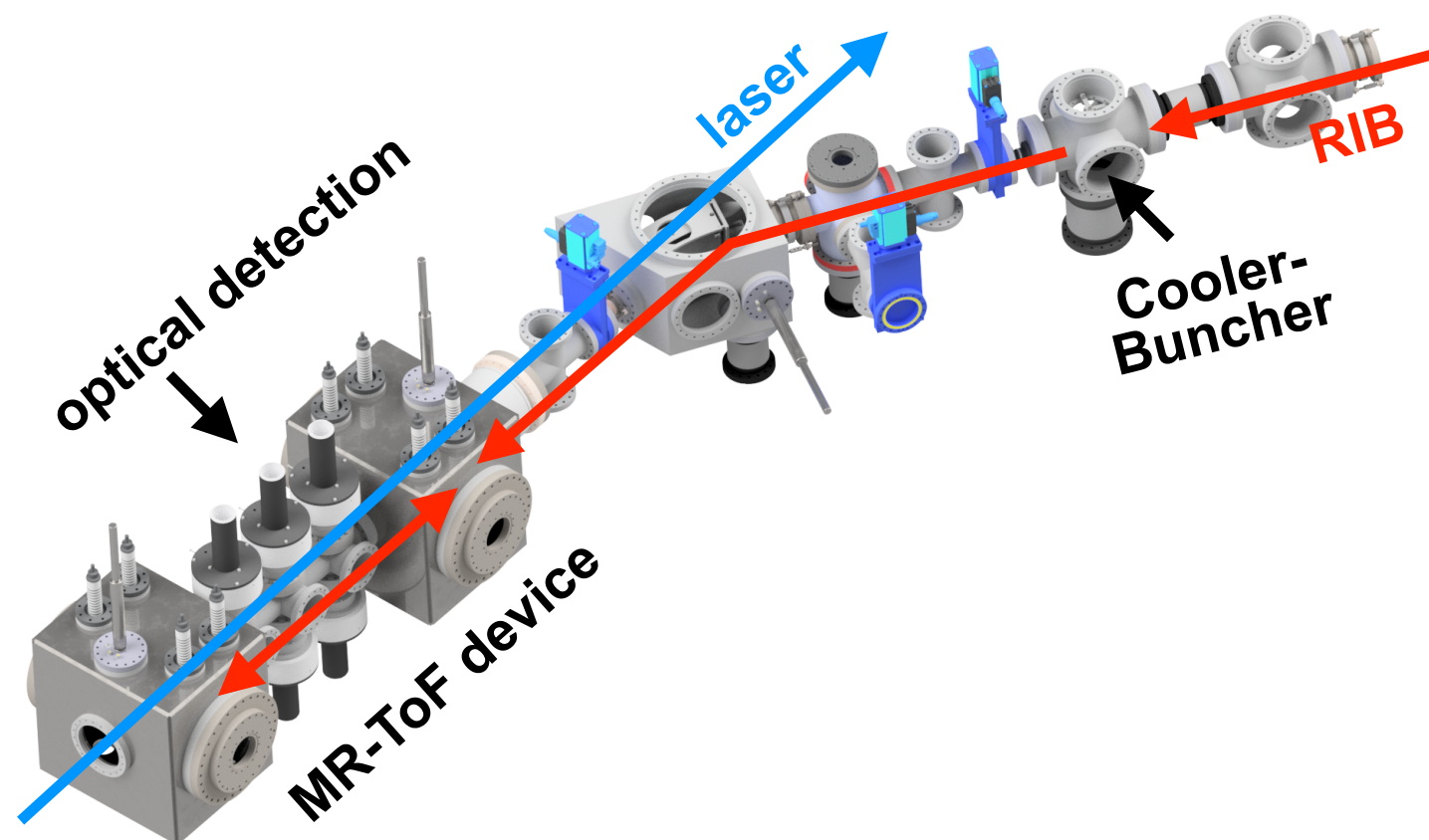
Picture of MR-ToF mirrors

**MR-ToF operation: 10-15 keV  
opportunity for RIB purification**

*F. Maier et al., Nuclear Instrum. Meth. A, 1056, 168545 (2023)*  
*F. Maier et al., Nuclear Instrum. Meth. A 1075, 170365 (2025)*

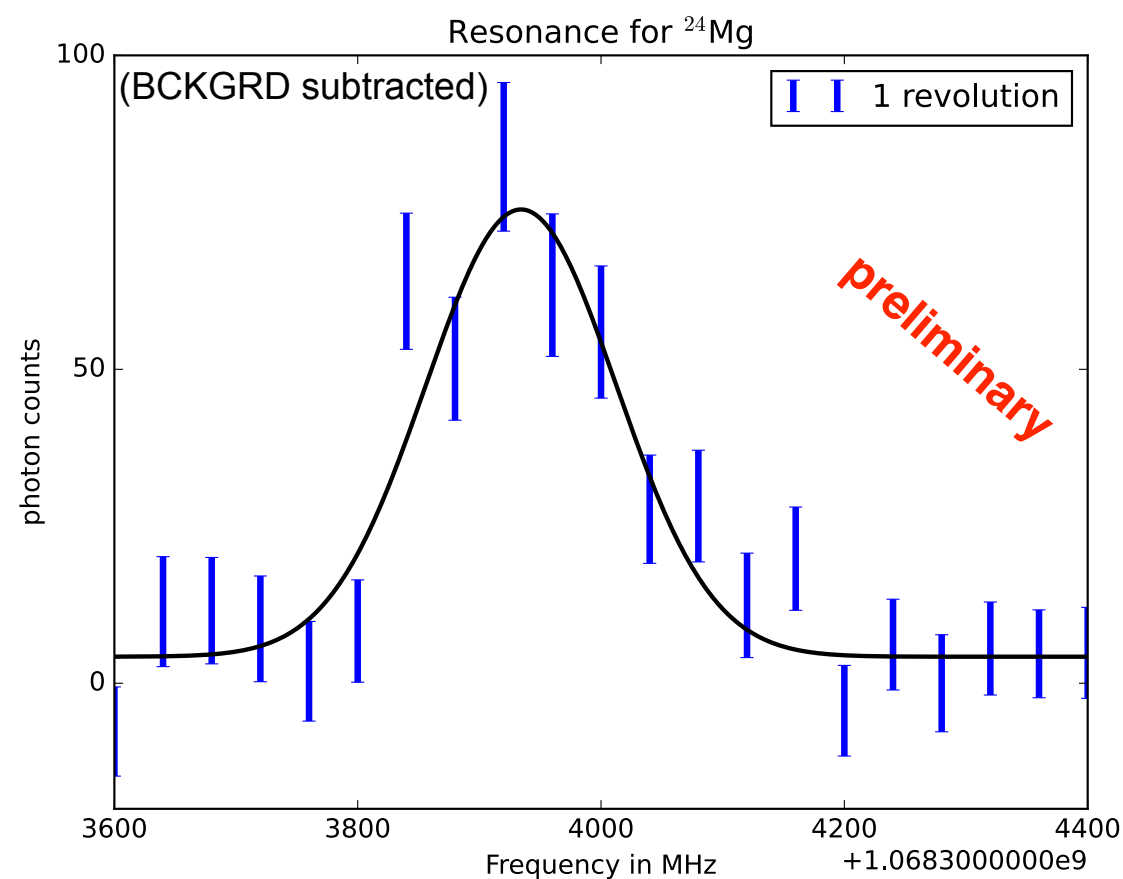


setup at



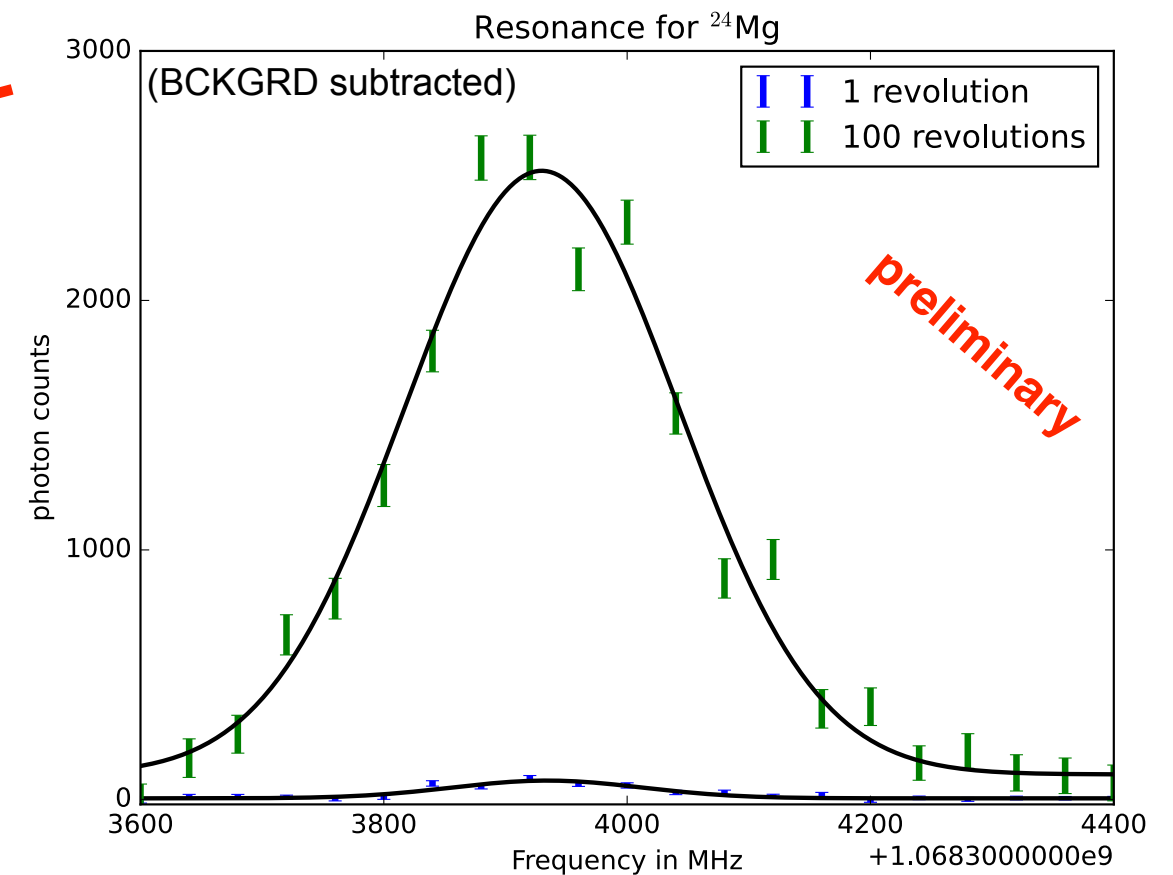
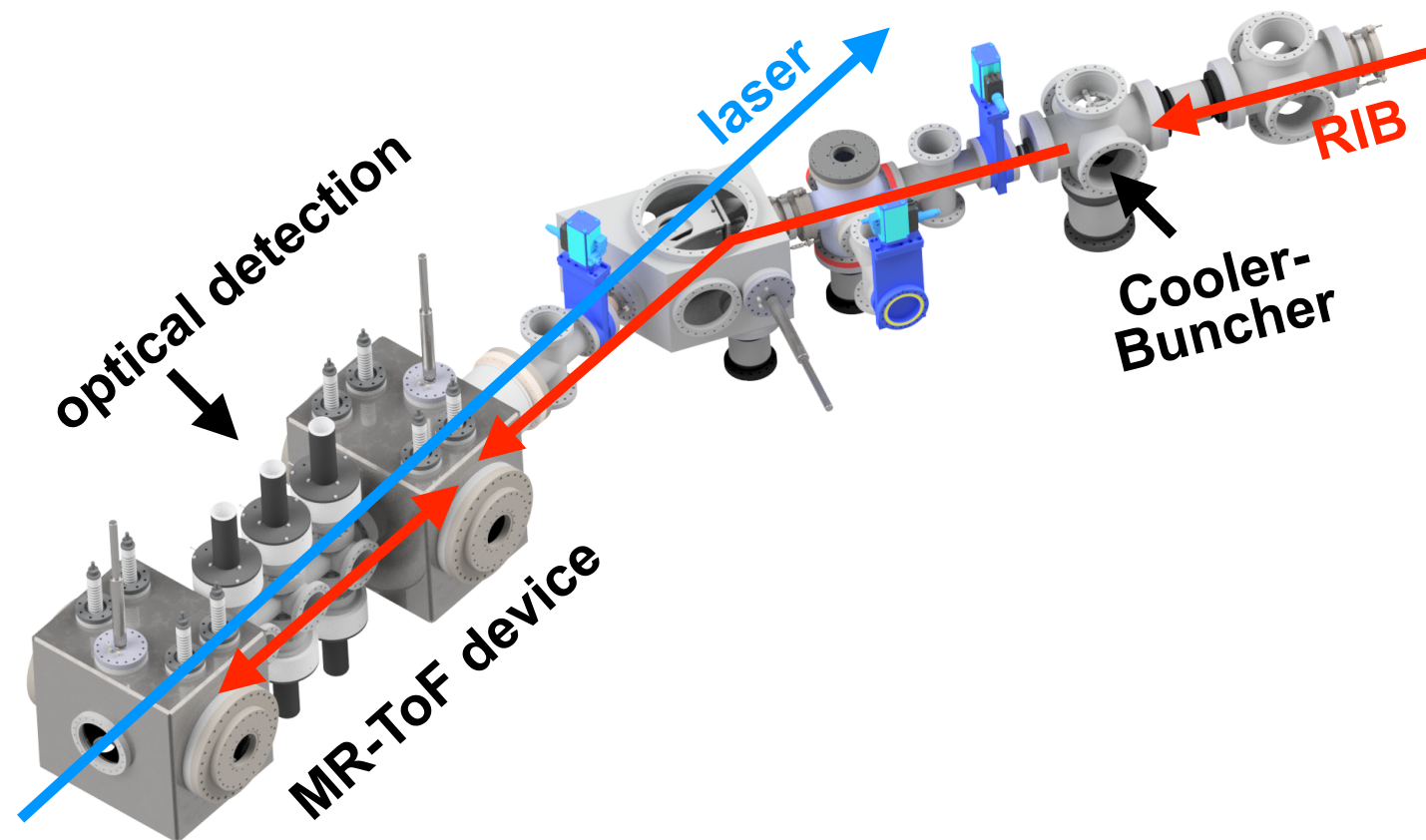
Single-passage = conventional CLS

14





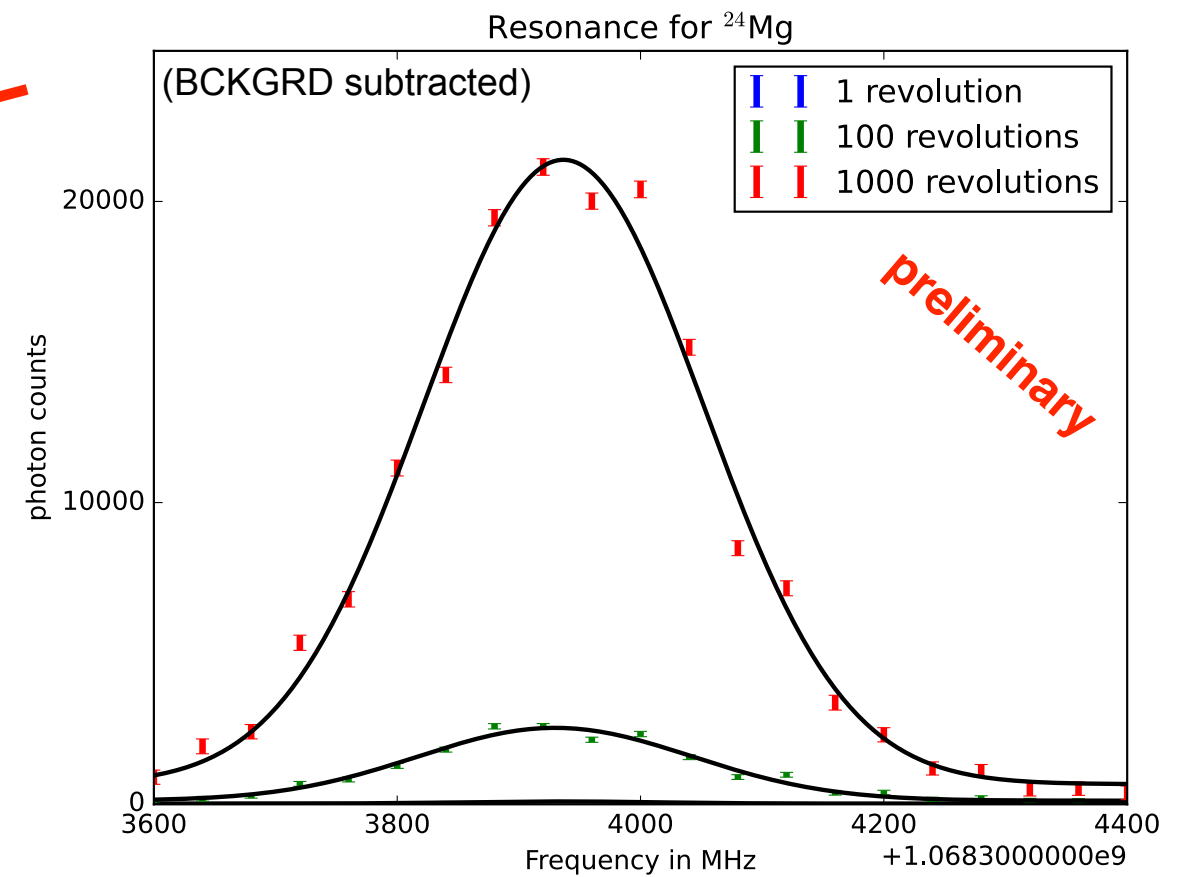
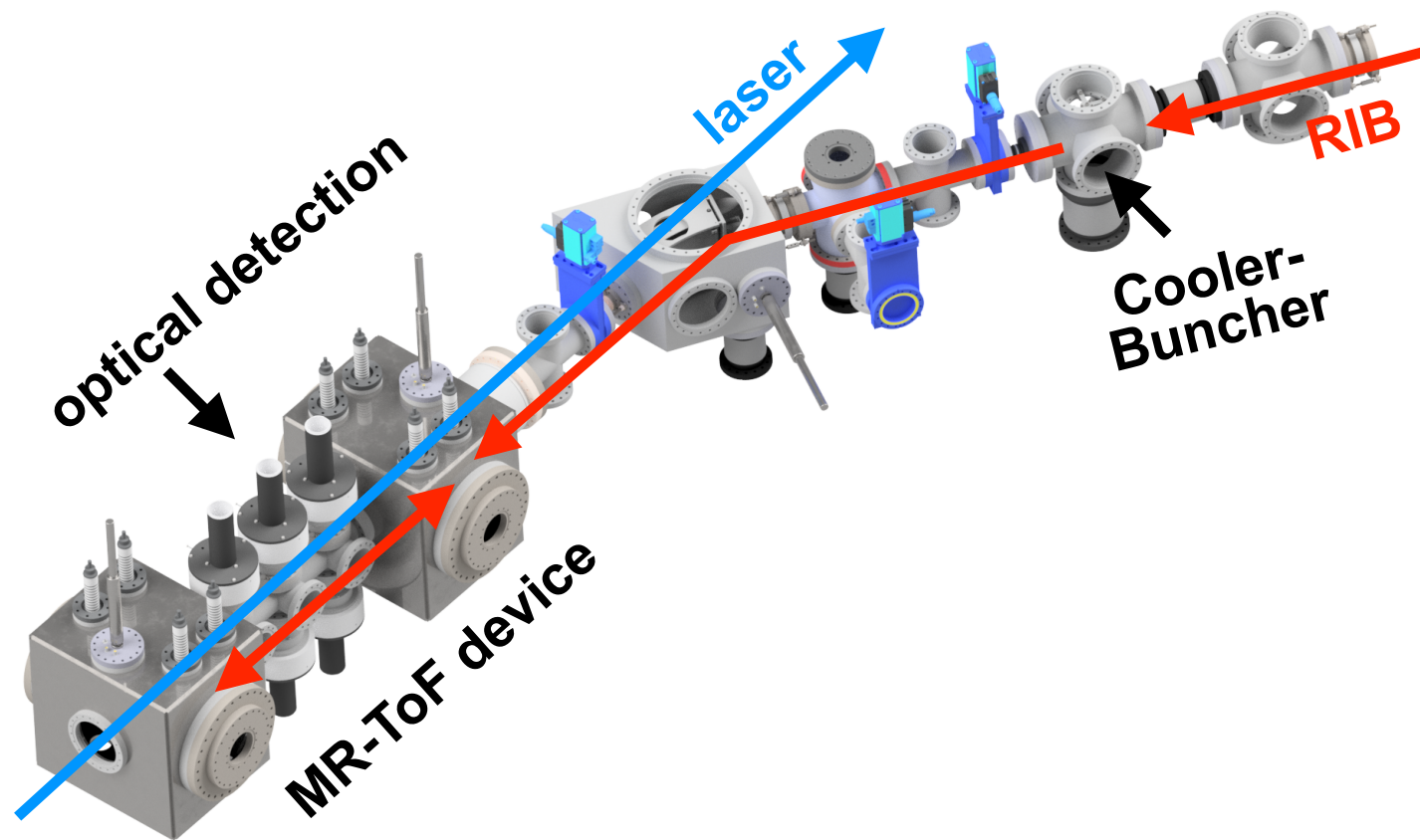
# demonstration





# demonstration

16

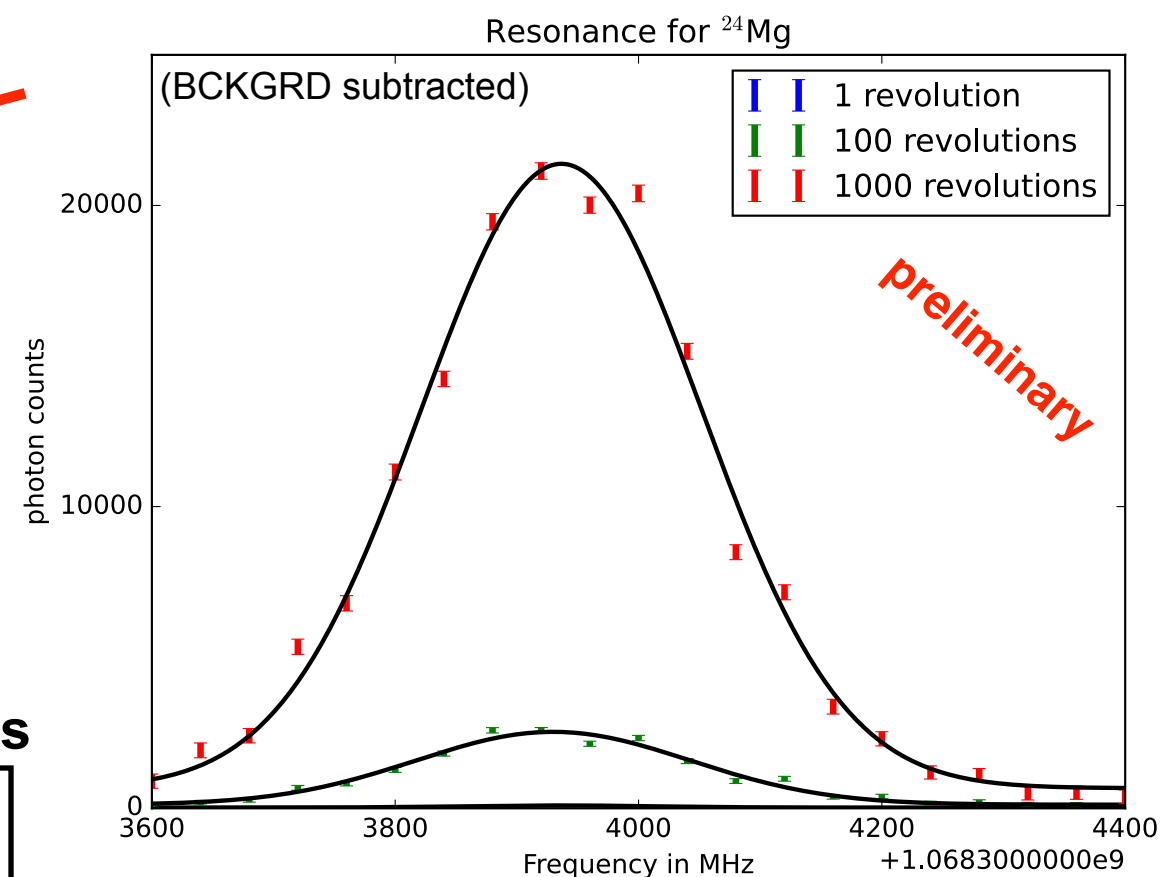
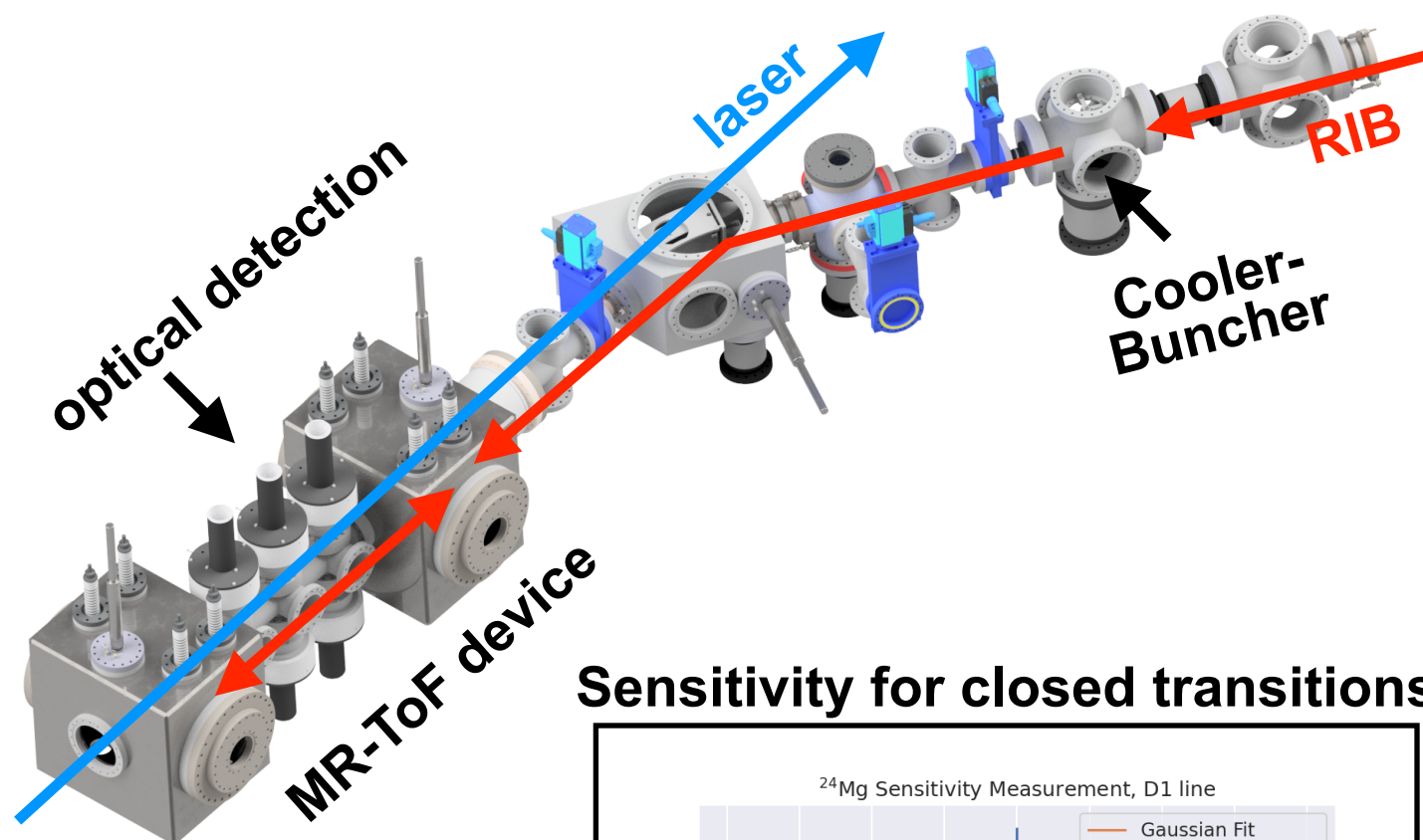




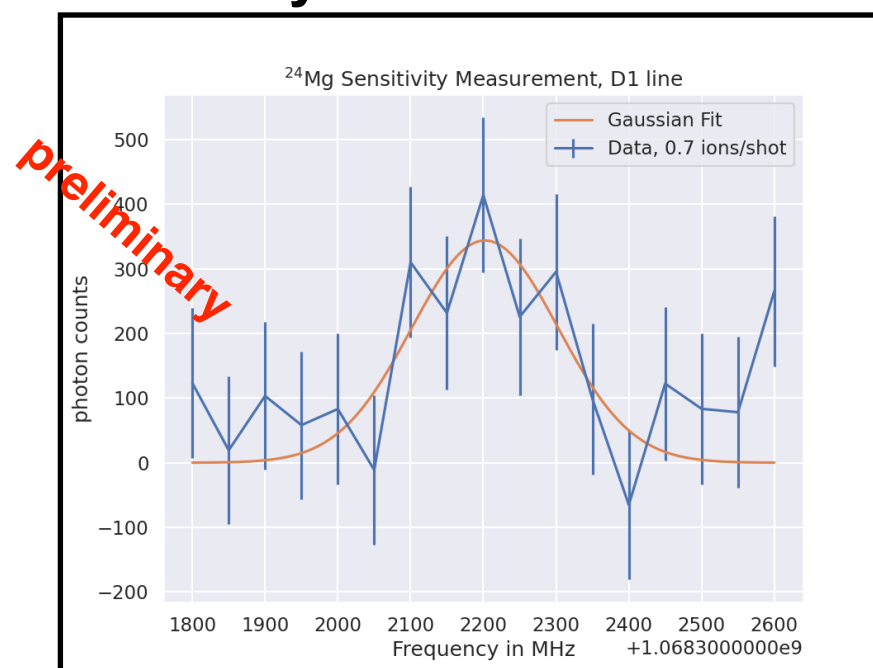


# demonstration

16



## Sensitivity for closed transitions

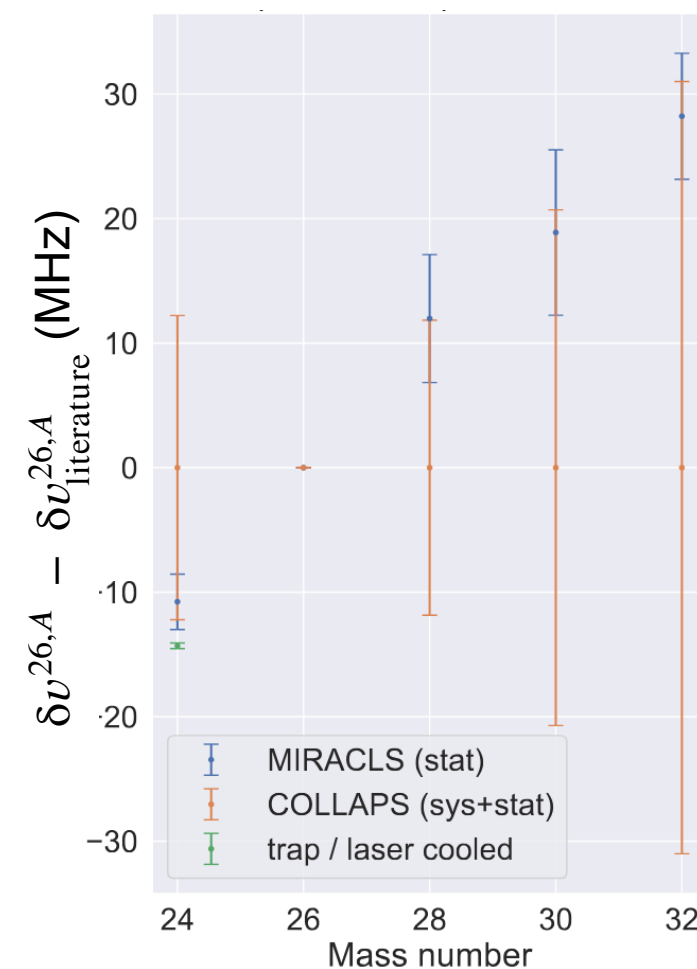
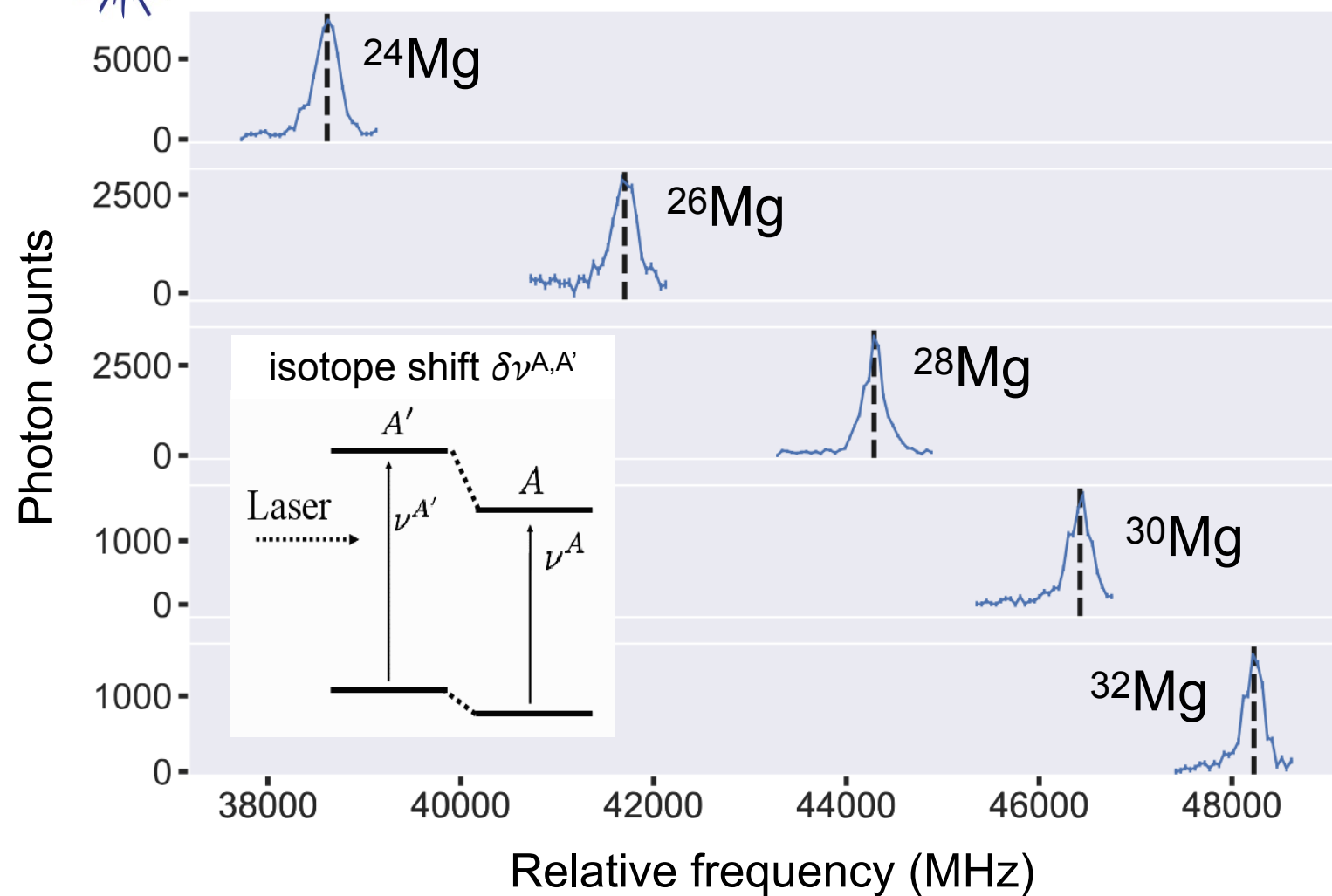


- 0.7 ions/cycle injected into MR-ToF
  - buncher efficiency 15-25 %
- ⇒ minimal ISOLDE yield: **3-5 ions / sec**



# online results

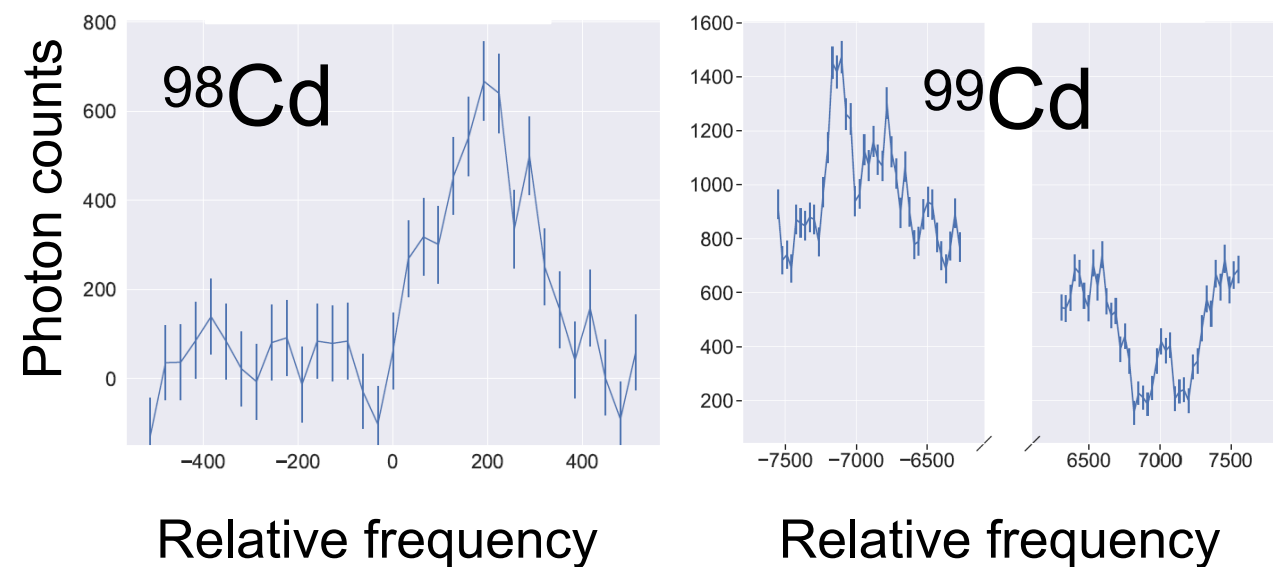
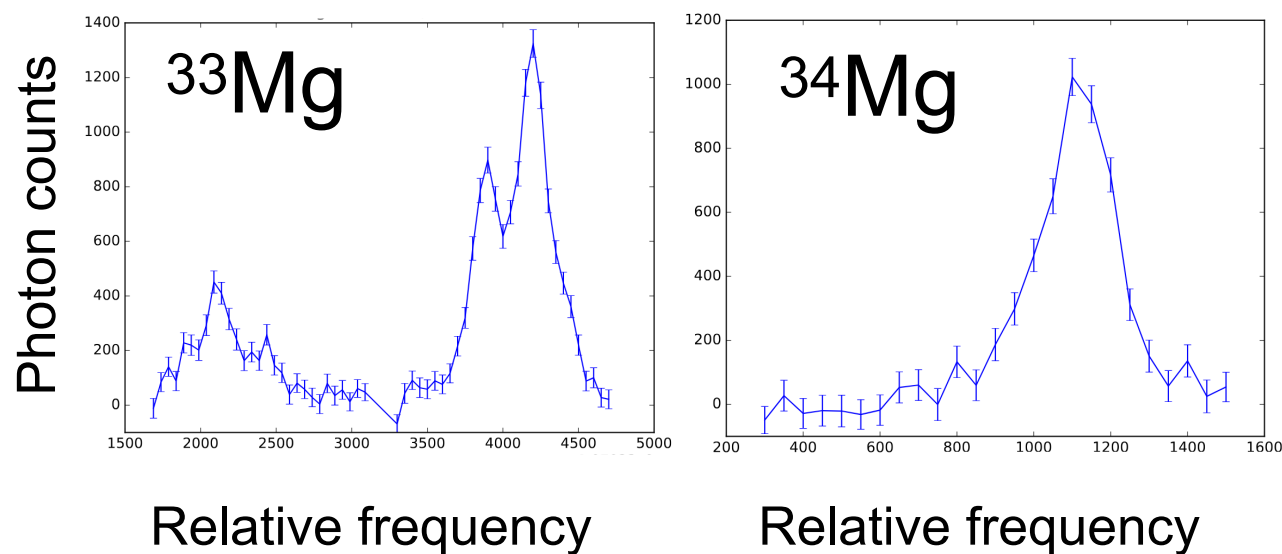
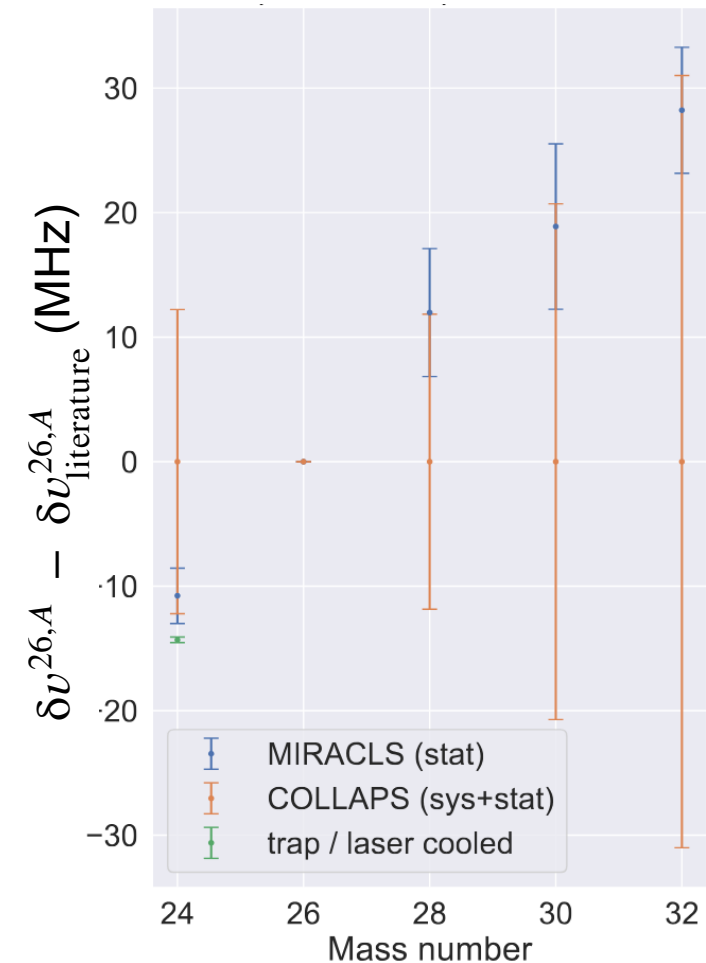
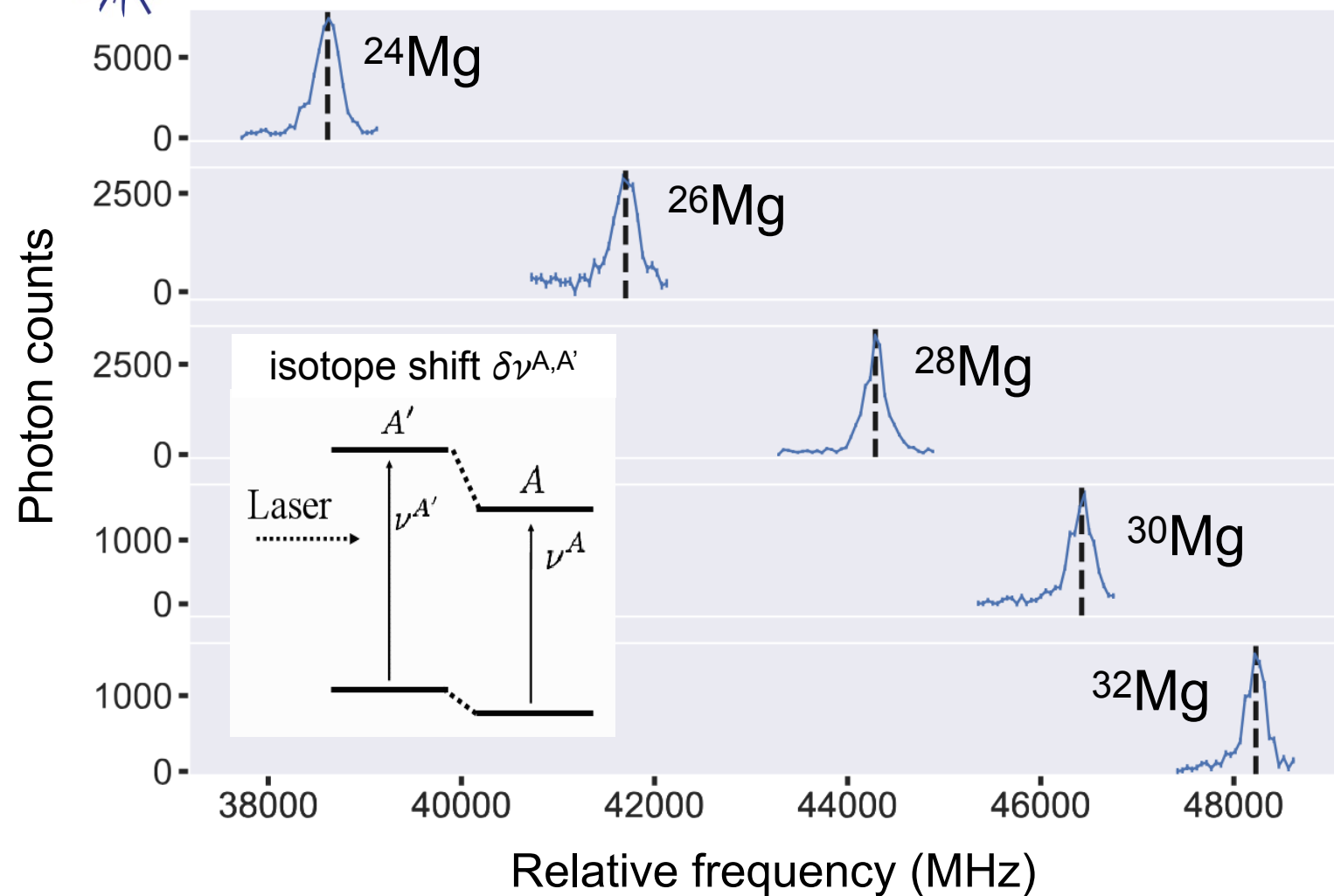
all preliminary



17



# online results all preliminary

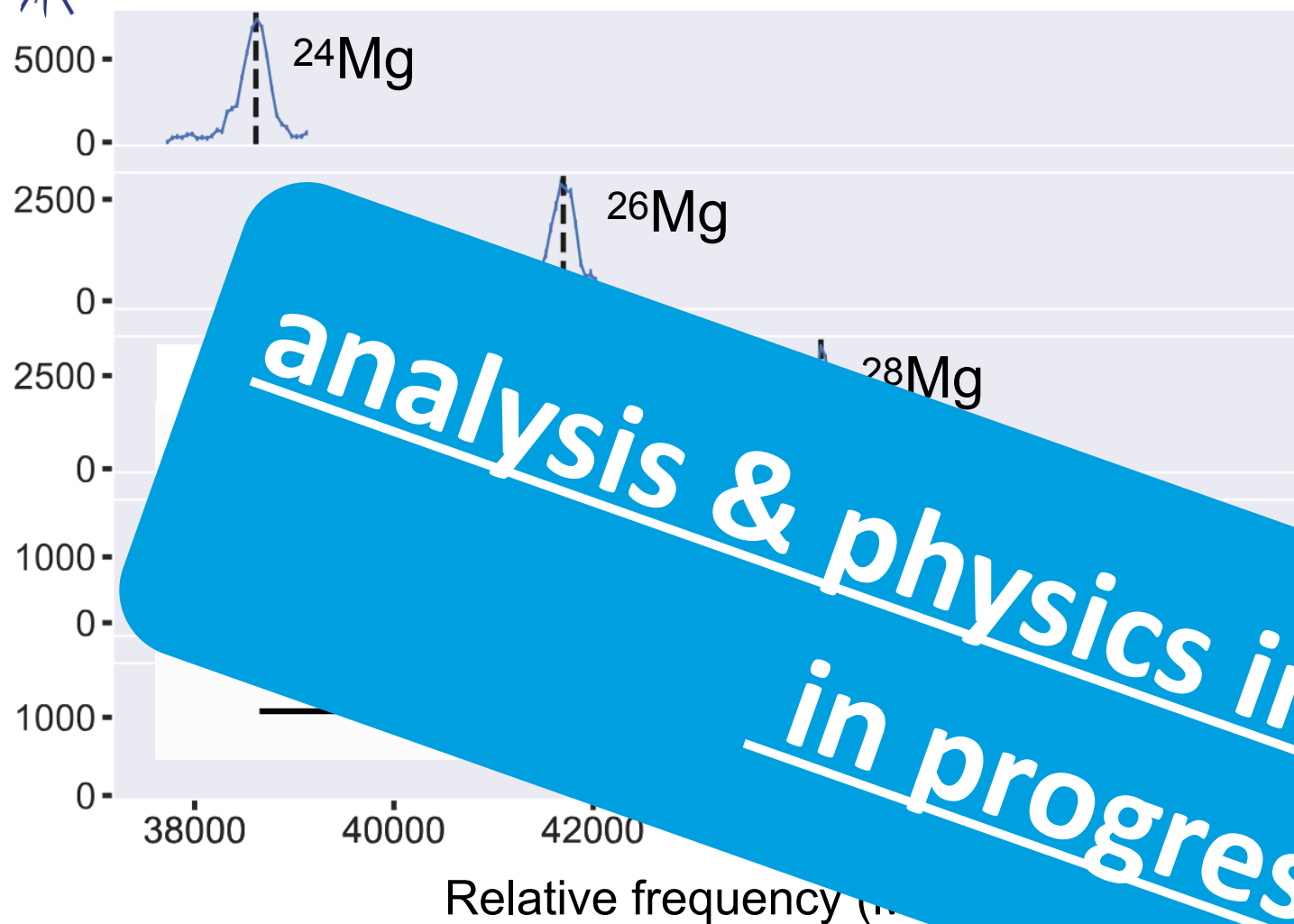




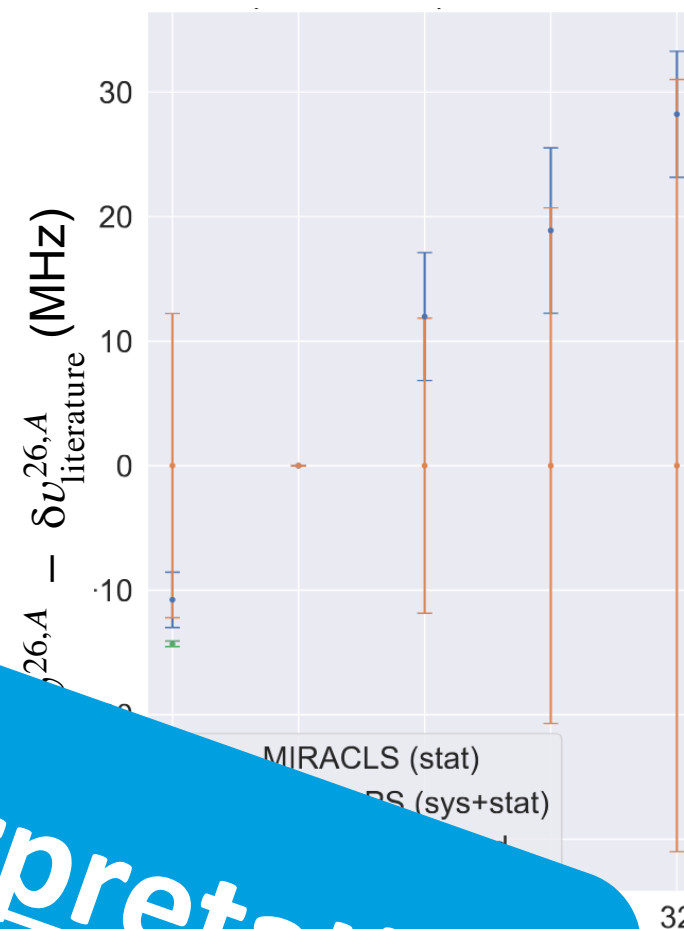
# online results

all preliminary

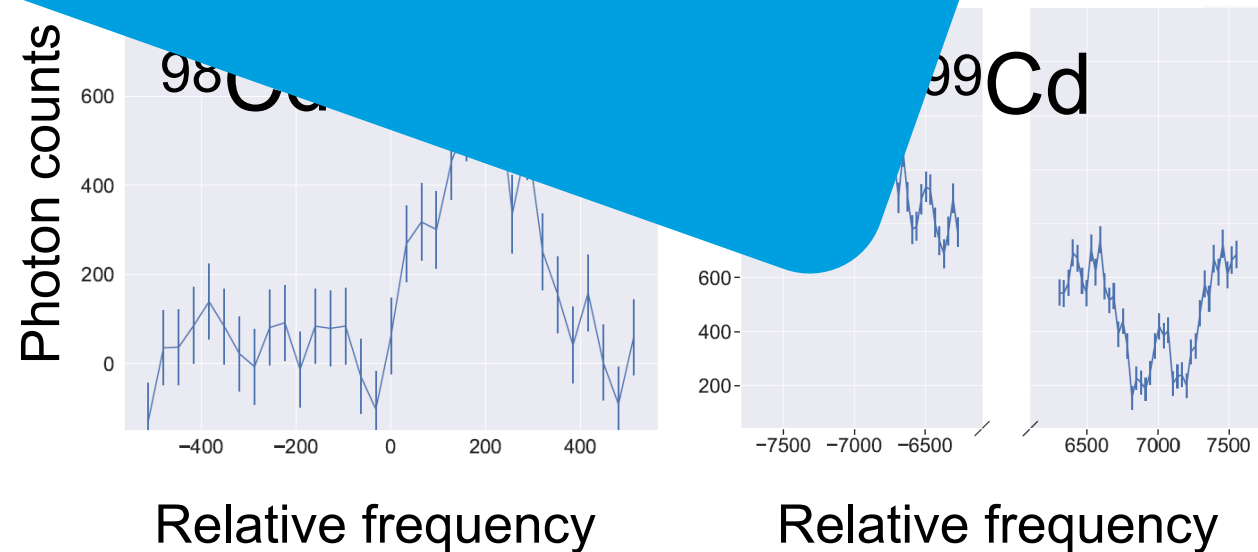
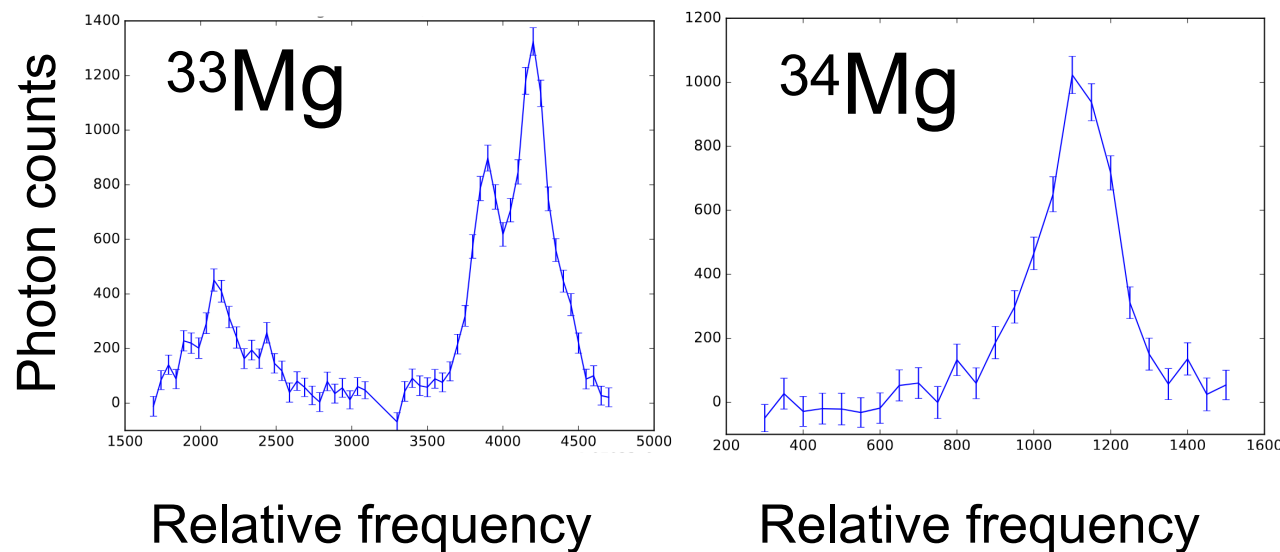
Photon counts



*analysis & physics interpretation  
in progress*



17





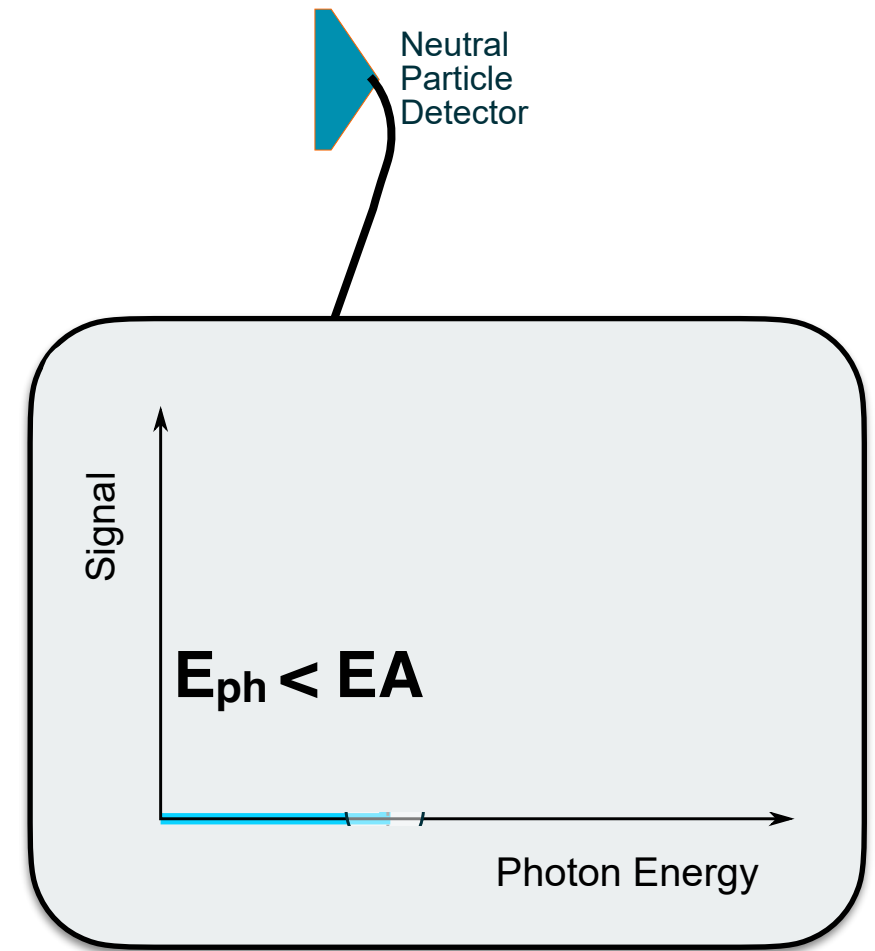
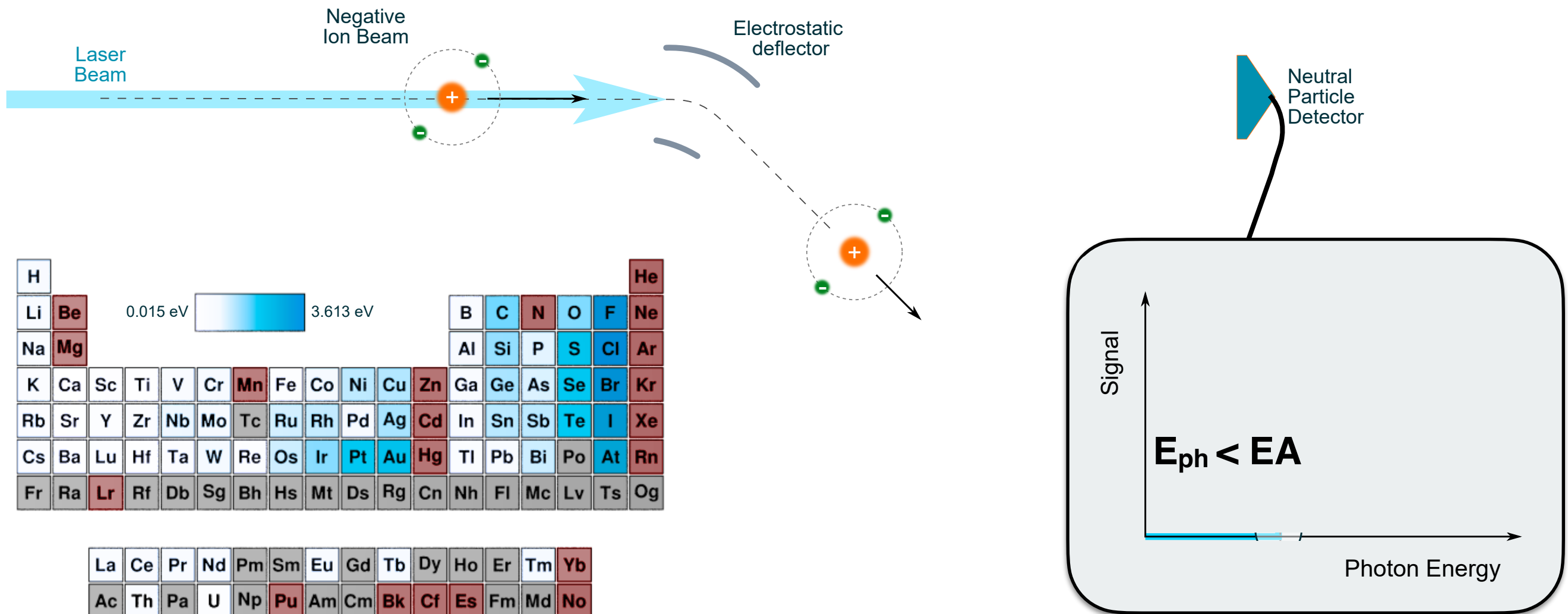
# CLS for Electron Affinity

**EA:** energy needed to remove an electron from a negative ion

*“how much an element is prone to form chemical bonds by sharing electrons”*

18

Laser: probes the energy needed to dismantle the negative ion



**~1/3 of EAs in the Periodic Table are unknown**

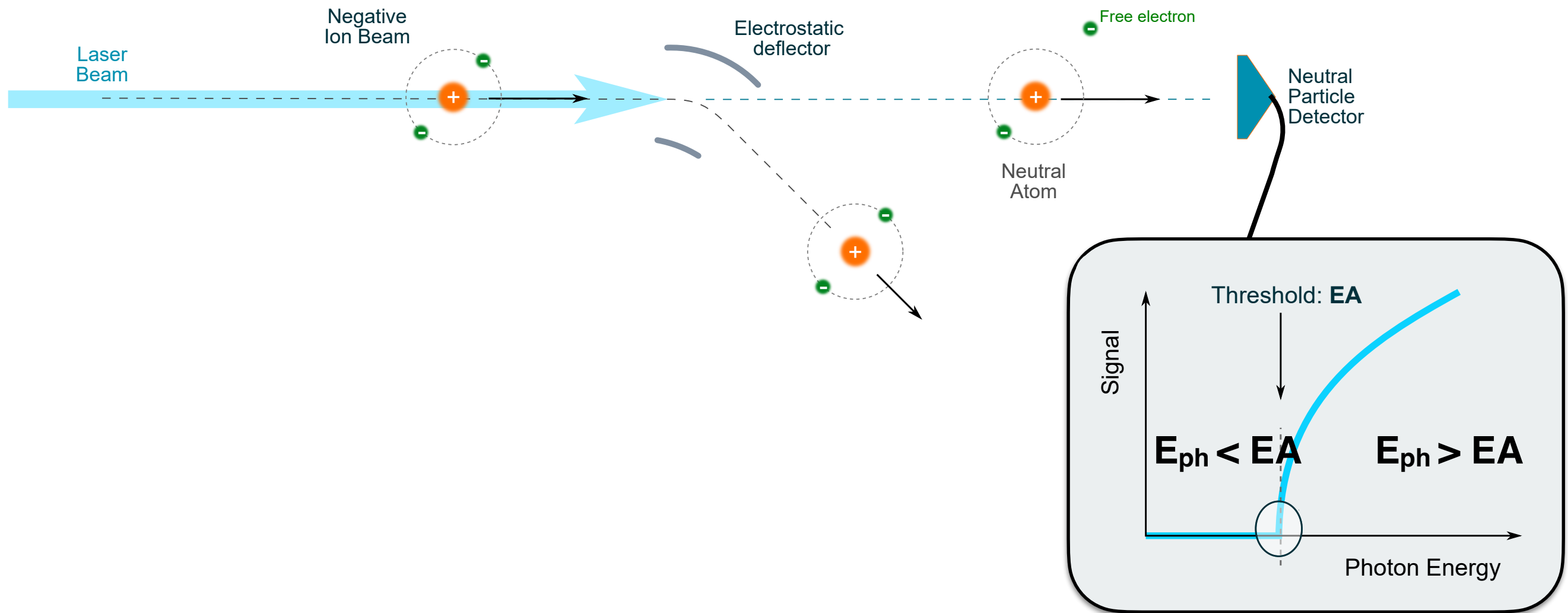
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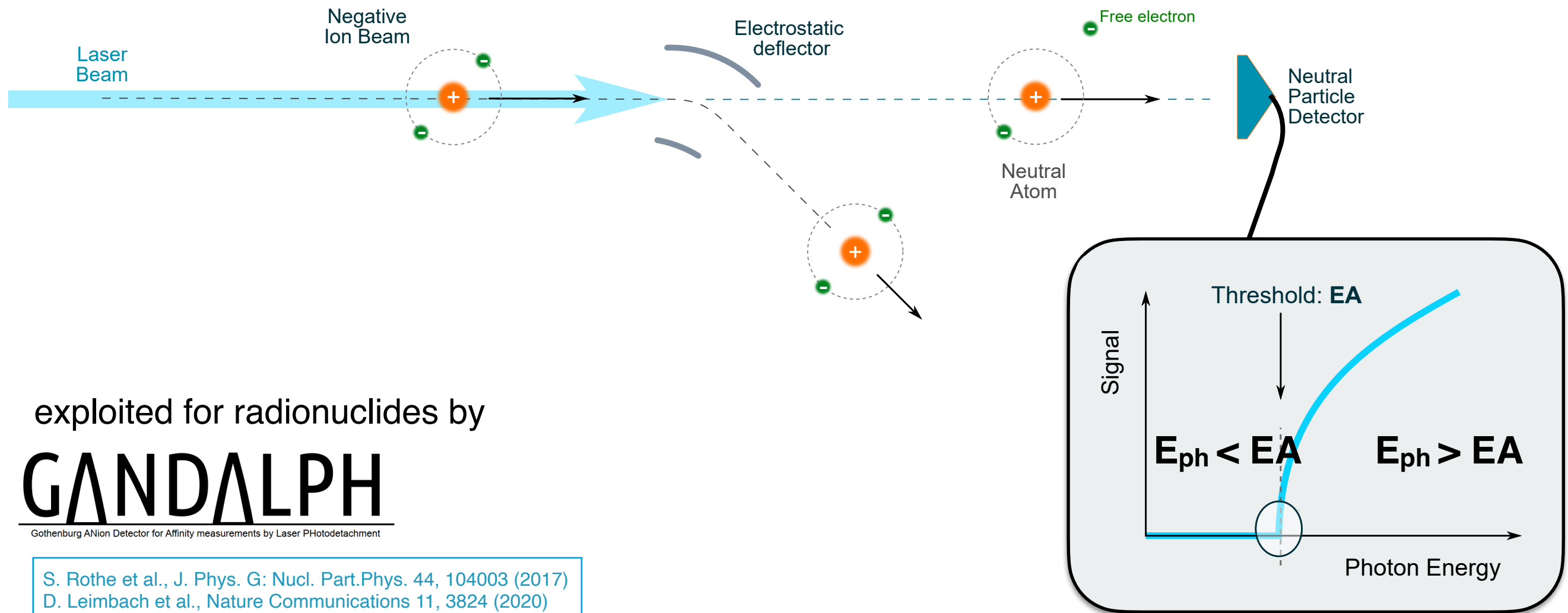
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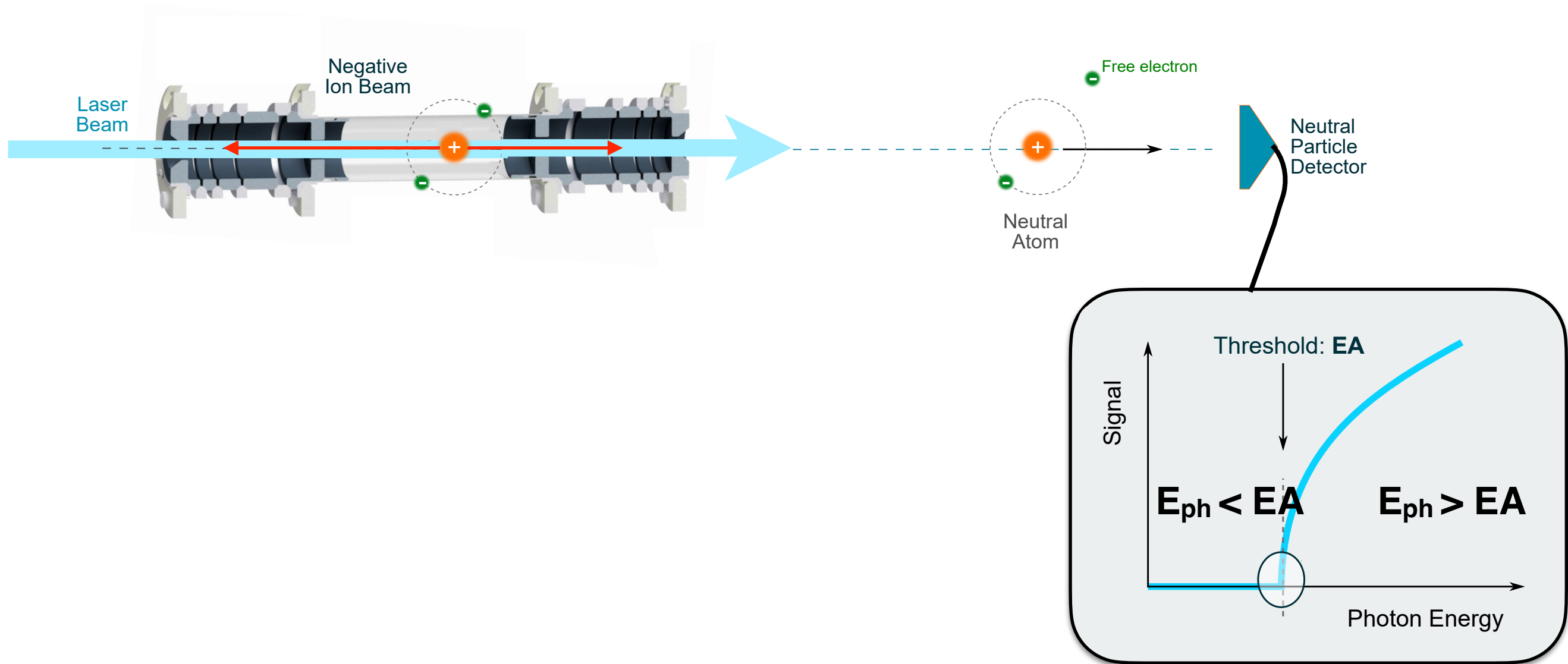
# EA studies with



## Increase exposure to laser beam by ion-beam confinement in MR-ToF

long exposure time  $\Rightarrow$  less particles for same signal  $\Rightarrow$  gain in sensitivity

20





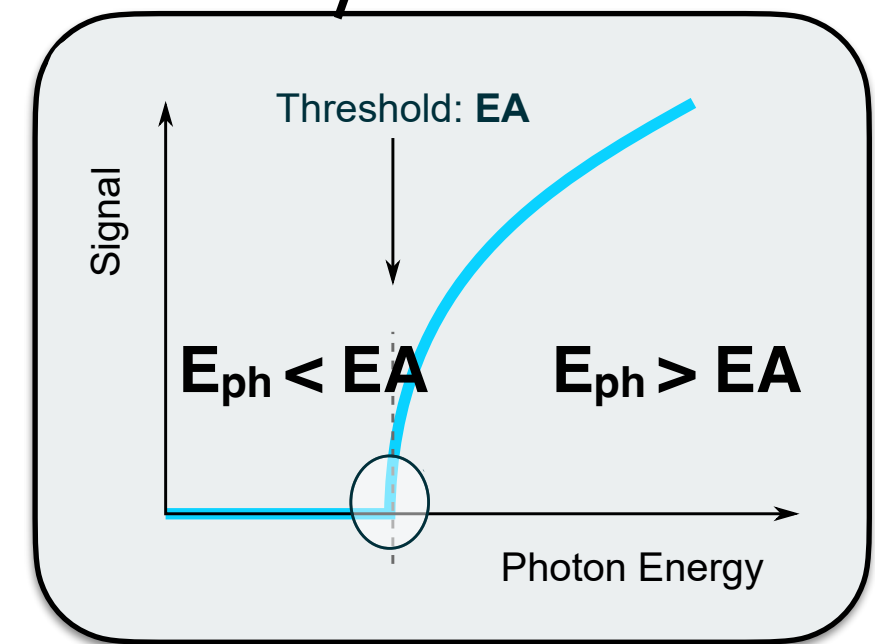
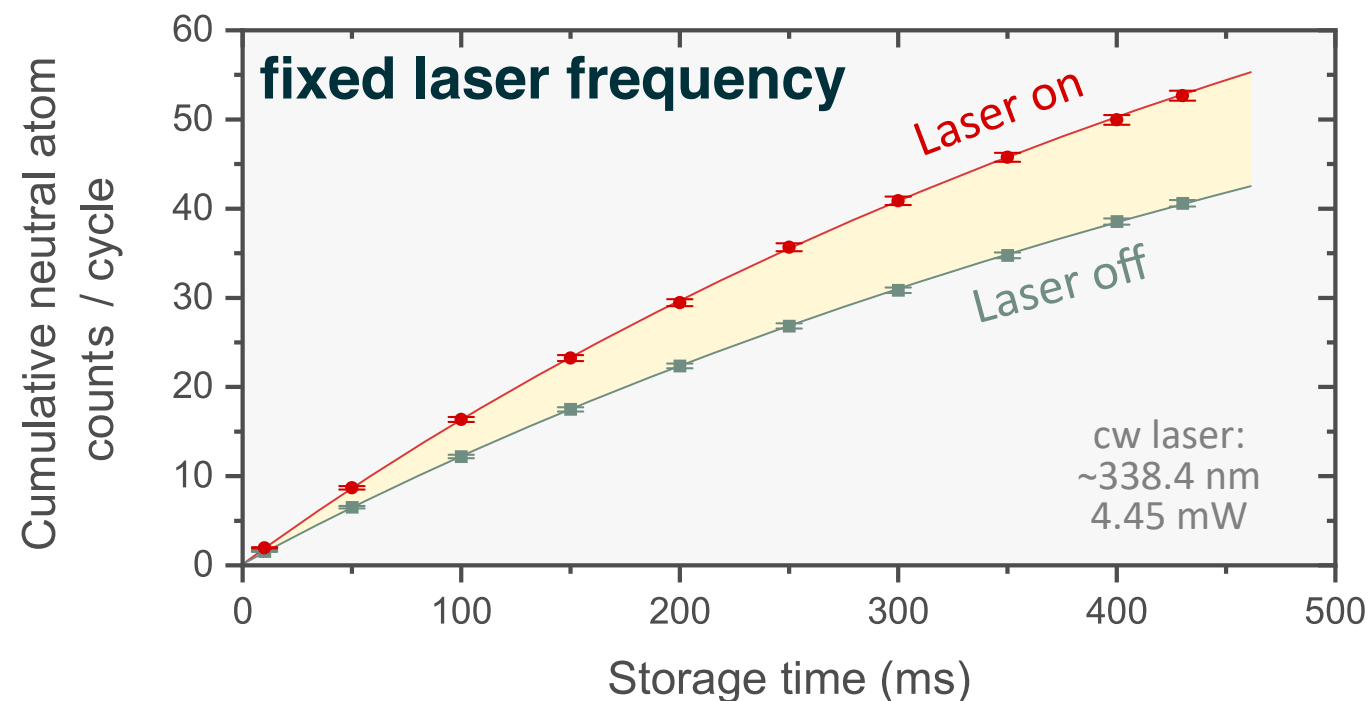
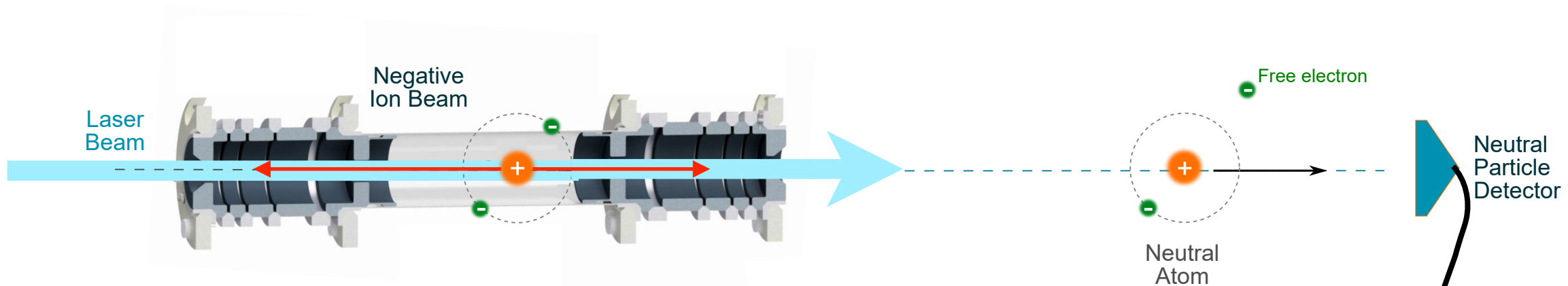
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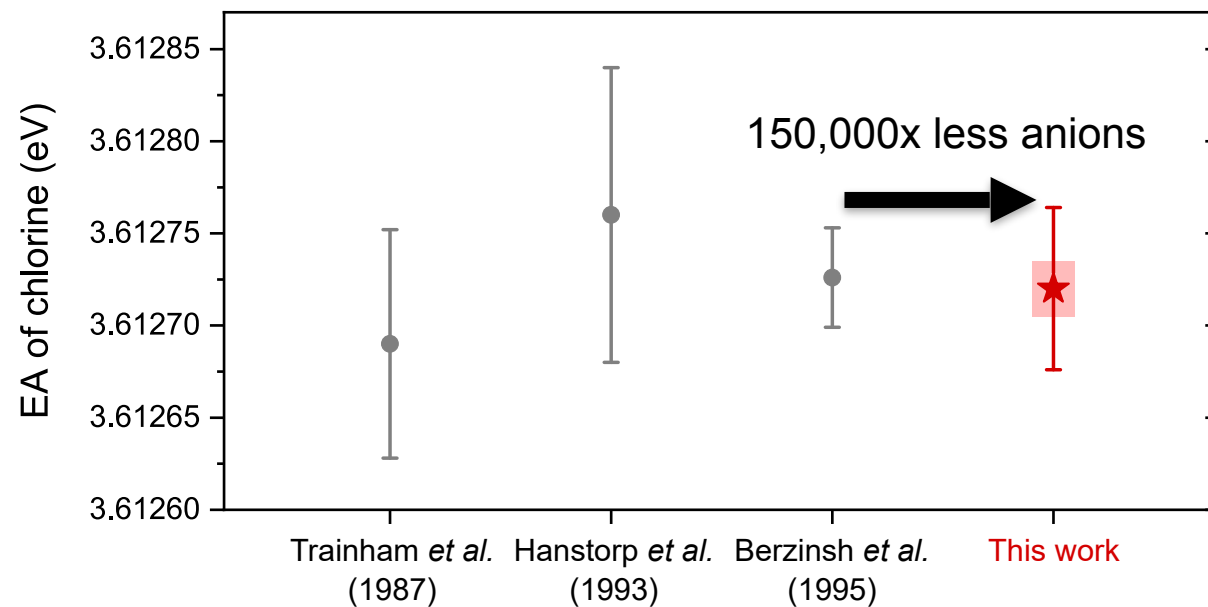
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20



# EA sensitivity gain with



## Results:

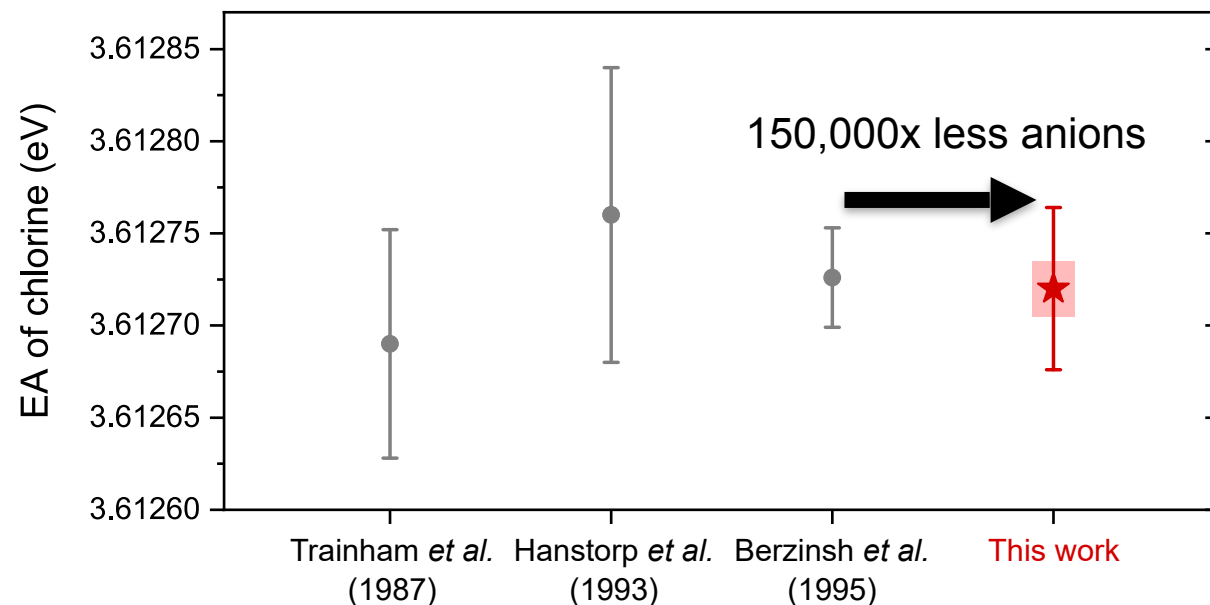
- (improved EA of Cl)
- same precision with  $\approx 10^5$  fewer anions

## Opportunities:

- further improvements to be implemented  
⇒ atom-at-a-time sensitivity
- EA isotope shifts
- EA measurements of (super)heavy elements

F. Maier et al., Nature Communications, 16, 9576 (2025)

# EA sensitivity gain with



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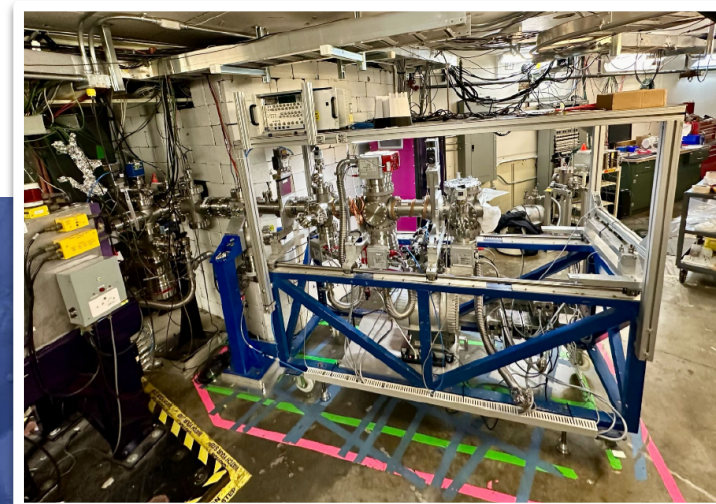
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- EA measurements of (super)heavy elements

F. Maier *et al.*, *Nature Communications*, 16, 9576 (2025)

## Advanced Electrostatic Trap for Heavy Element Research (AETHER) @ BGS / 88" cyclotron

MIRACLS low-energy system sent to LBNL to seed the new infrastructure



# Summary & Conclusions

- collinear laser spectroscopy (CLS) powerful tool to access nuclear ground-state properties

new CLS beamlines



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- charge radii: excellent benchmarks for nuclear theory
  - ➔ towards a 'universal' description of charge radii
  - ➔ DFT Fayans: odd-even staggering + kinks at shell closures
  - ➔ ab initio: 1 % accuracy in Ni
  - ➔ importance of new experimental data
- sensitivity challenge:

**CRIS**



- ➔ novel ion-trap system for highly sensitive CLS
- ➔ successful online measurements of Mg and Cd
- ➔ highly-sensitive EA measurements: towards (super-)heavies
- ➔ new opportunities, e.g., for RIB purification





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TECHNISCHE  
UNIVERSITÄT  
DARMSTADT



23

collaboration:

## MIRACLS team and participants

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## MIRACLS EA team

F. M. Maier, E. Leistenschneider, M. Au, U. Berzins, Y. N. Vila Gracia, D. Hanstorp, C. Kanitz, V. Lagaki, S. Lechner, D. Leimbach, P. Plattner, M. Reponen, L. V. Rodriguez, S. Rothe, L. Schweikhard, M. Vilen, J. Warbinek, S. Malbrunot-Ettenauer

funding:



European  
Research  
Council



Medical  
Applications  
Funds



<https://miracsls.web.cern.ch>