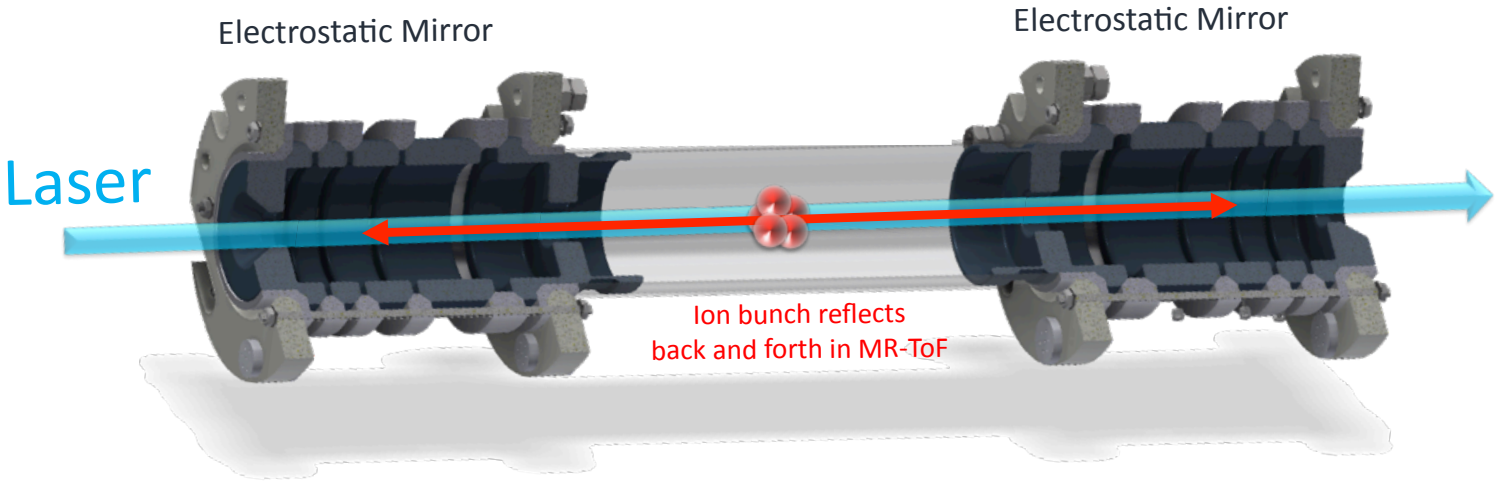
European Research Council

Building on MIRACLS:
radioactive molecules as novel, sensitive probes for electric dipole moments



MIRACLS

Stephan Malbrunot-Ettenauer
CERN research physicist

CERN

Outline

- **Motivation**
  ➡ (radioactive) molecules and EDM searches

- **Developments for highly sensitive collinear laser spectroscopy**

- **MIRACLS for radioactive molecules**

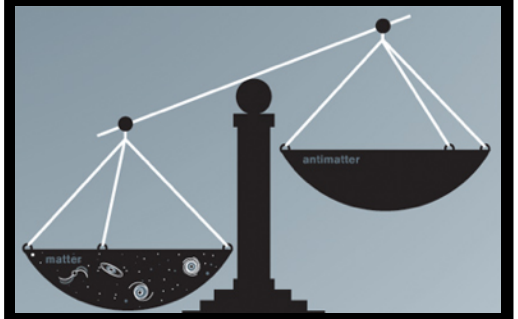
- **Summary**

The Hubble Space Telescope

where is all the antimatter?
The Hubble Space Telescope

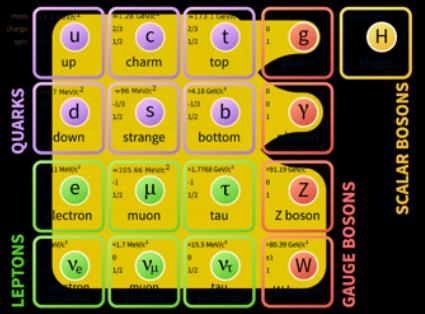
where is all the antimatter?

Macrocosmos



only matter

Microcosmos

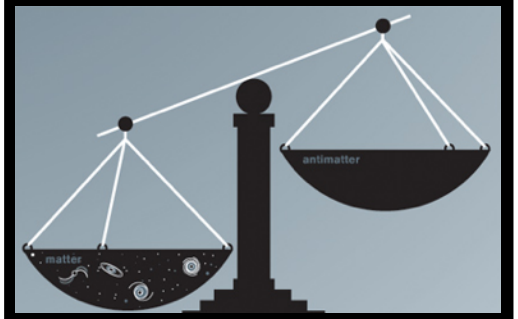


|matter| = |antimatter|

The Hubble Space Telescope

where is all the antimatter?

Macrocosmos



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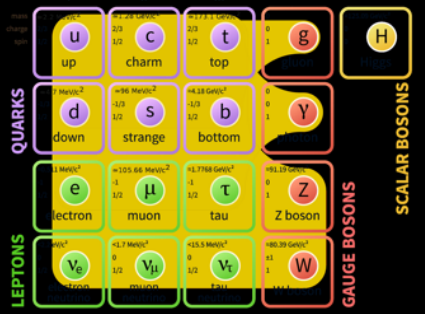
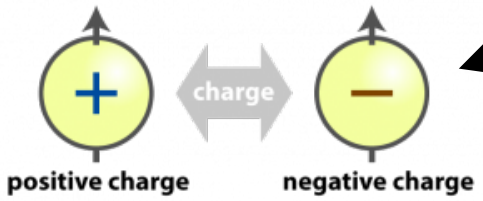
Microcosmos



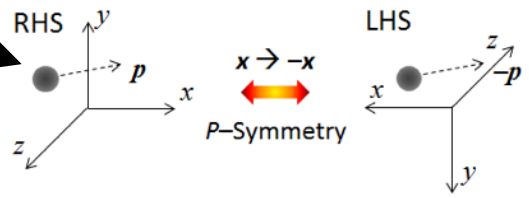
|matter| = |antimatter|

Sakharov, 1967

ingredient to resolve universe's matter-antimatter asymmetry:

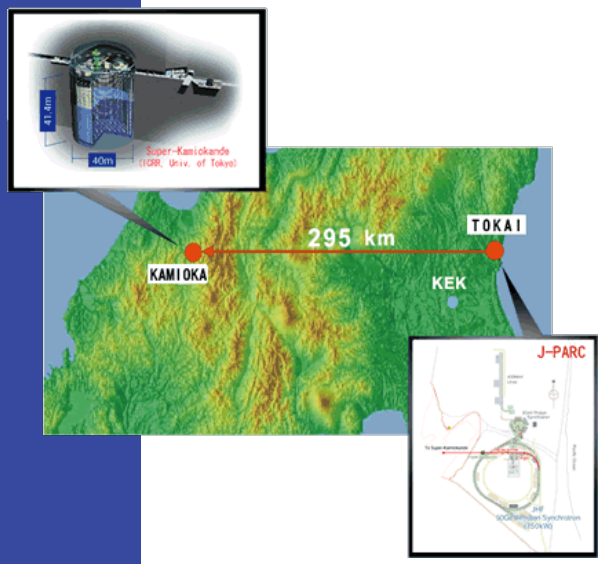


CP violation

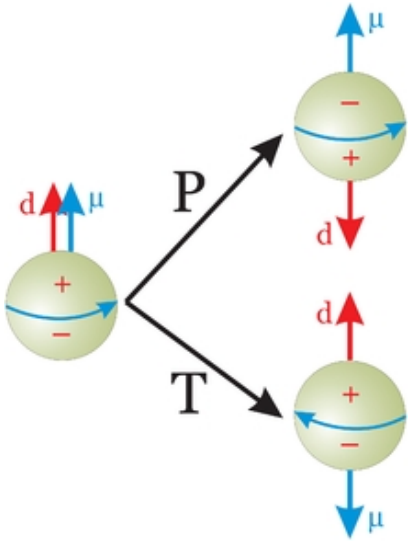


Searches for CP violation

T2K collaboration, Nature 580, 339 (2020)



AcMe collaboration, Nature 562, 355 (2018)
C. Abel et al., PRL 124, 081803 (2020)

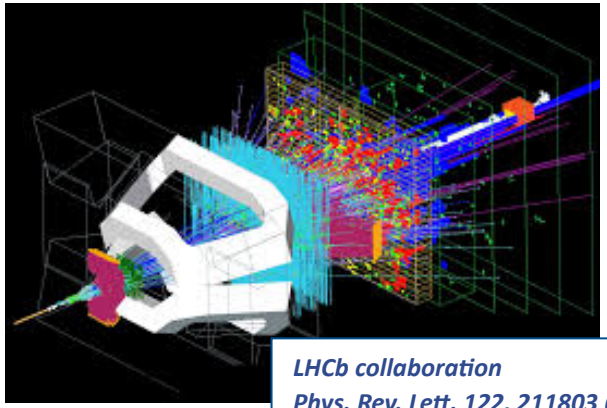


Intensity

Precision

CP

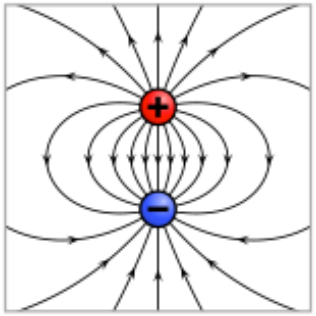
Energy



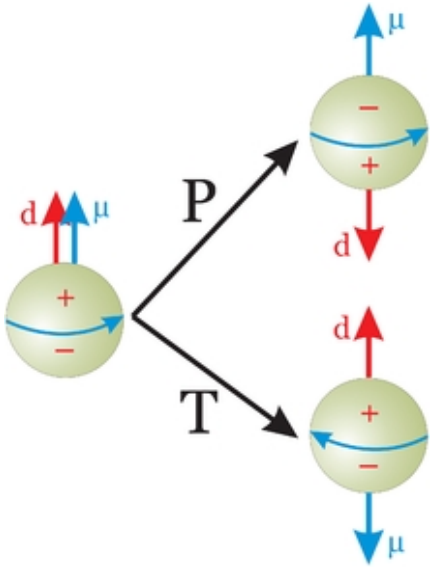
LHCb collaboration
Phys. Rev. Lett. 122, 211803 (2019)

Permanent electric dipole moment

- local separation of the electric charge along a particle's spin axis

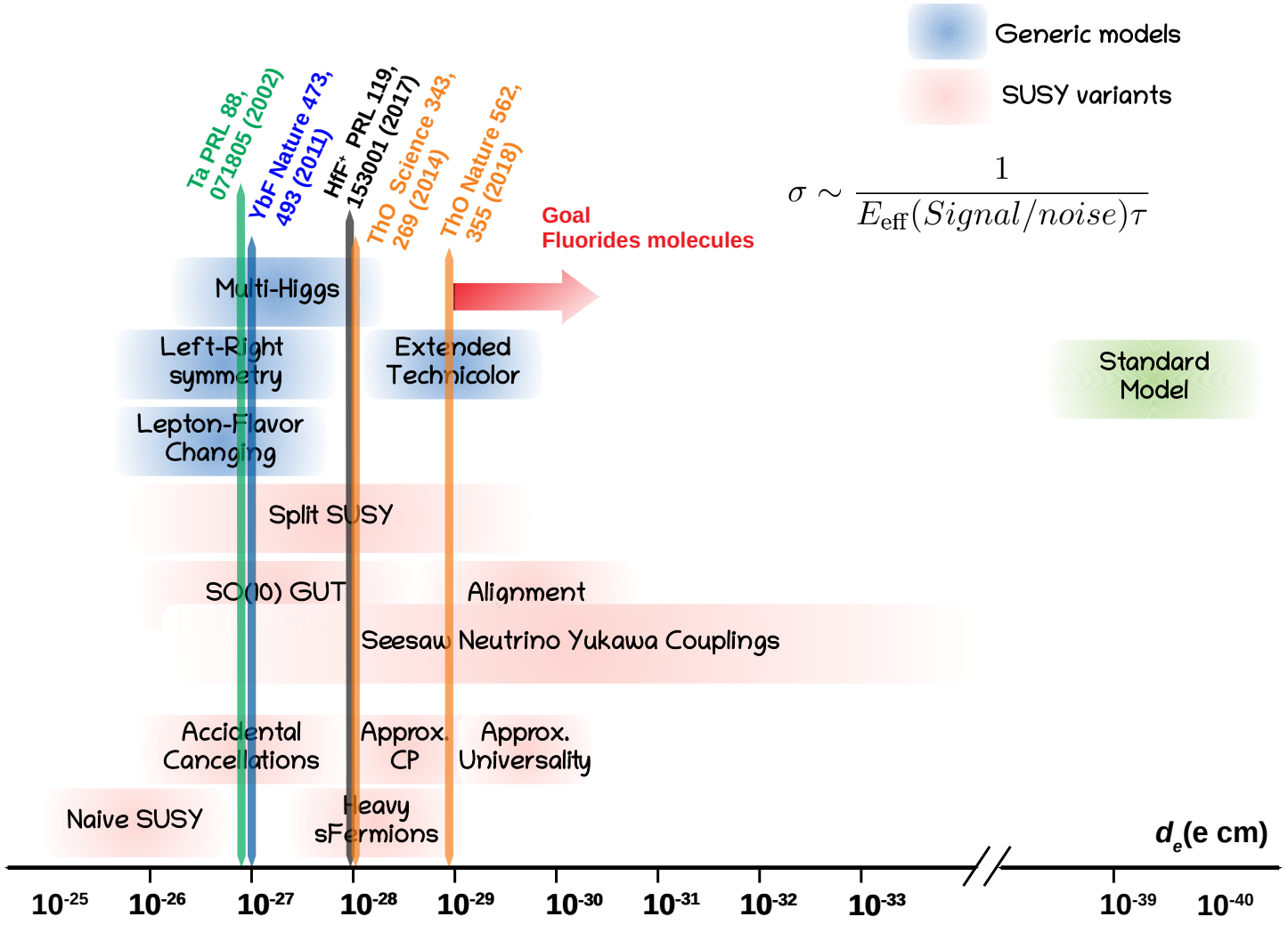


- implies time-reversal (T) violation ⇒ violation of CP symmetry (assuming CPT)

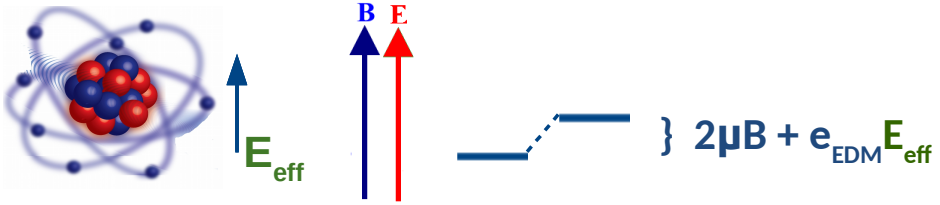


matter-antimatter asymmetry in the universe

electron EDM searches

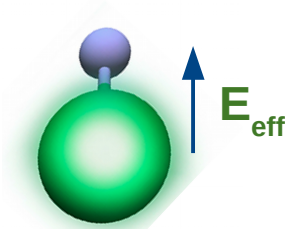


EDM searches in molecules



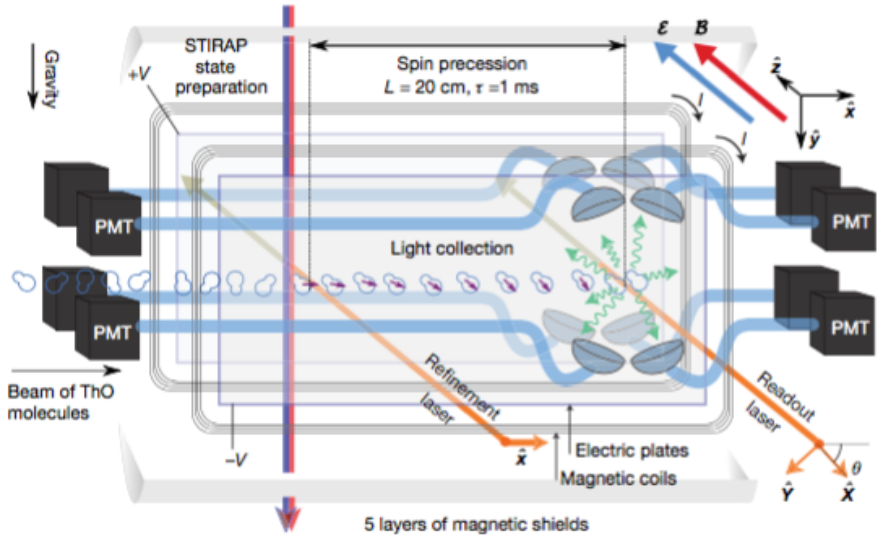
$$\sigma_d \sim \frac{1}{E_{\mathrm{eff}}\tau\sqrt{\dot{N}T}}$$

Molecules:

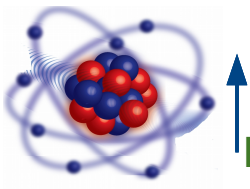


E_{ext} ~1 V/cm
E_{eff} ~80 GV/cm → • enhancement by 10^3 in sensitivity!

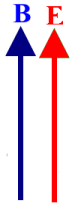
ThO *AcMe collaboration, Nature 562, 355 (2018)*



EDM searches in molecules



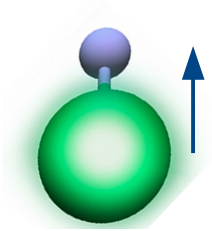
E_{eff}



} $2\mu B + e_{EDM} E_{eff}$

$$\sigma_d \sim \frac{1}{E_{\mathrm{eff}} \tau \sqrt{\dot{N} T}}$$

Molecules:



E_{eff}

E_{ext} ~1 V/cm

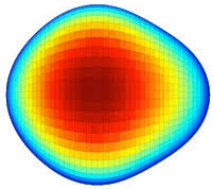
E_{eff} ~80 GV/cm → • enhancement by 10^3 in sensitivity!

ThO *AcMe collaboration, Nature 562, 355 (2018)*

• scales as $Z^2R(Z)$

⇒ opportunity for radioactive molecules

Schiff moment



nuclear-spin-dependent component

Radioactive molecules & EDMs

long coherence time

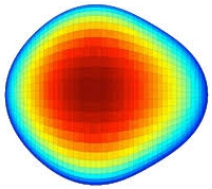
laser cooling

co-magnetometry / B-field insensitivity

close-lying opposite parity states

large E_{eff} $\propto Z^2 R(Z)$

Schiff moment deformed nuclei



same chemistry different physics sensitivity systematic spin dependence

nuclear-spin-dependent component & hadronic EDM

CP violating nuclear Magnetic Quadrupole Moment

nuclear-spin-independent component & electron EDM

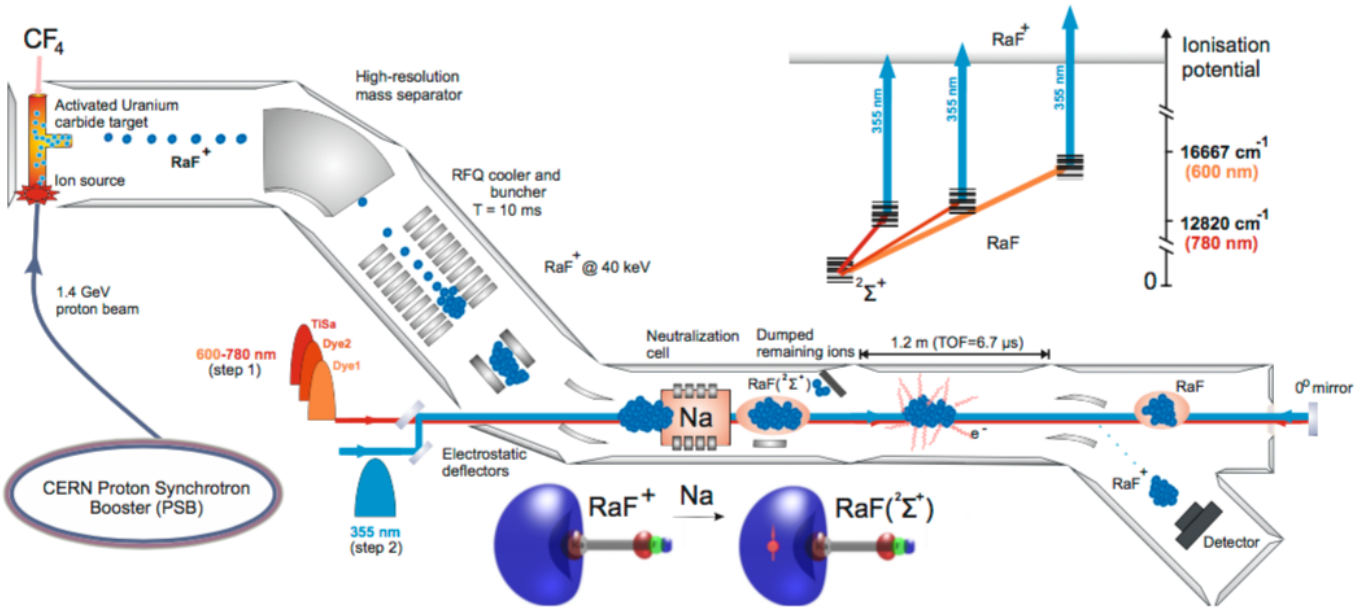
complementary physics

- parity violation
- quantum chemistry
- nuclear structure

radioactive molecules

	222Ra 38.0 S	223Ra 11.43 D	224Ra 3.6319 D	225Ra 14.9 D	226Ra 1600 Y	227Ra 42.2 M	228Ra 5.75 Y	229Ra 4.0 M	230Ra 93 M
Spin=	0	3/2	0	1/2	0	3/2	0	5/2	0

First spectroscopy of radioactive molecules



R. F. Garcia Ruiz et al., Nature 581, 396 (2020)

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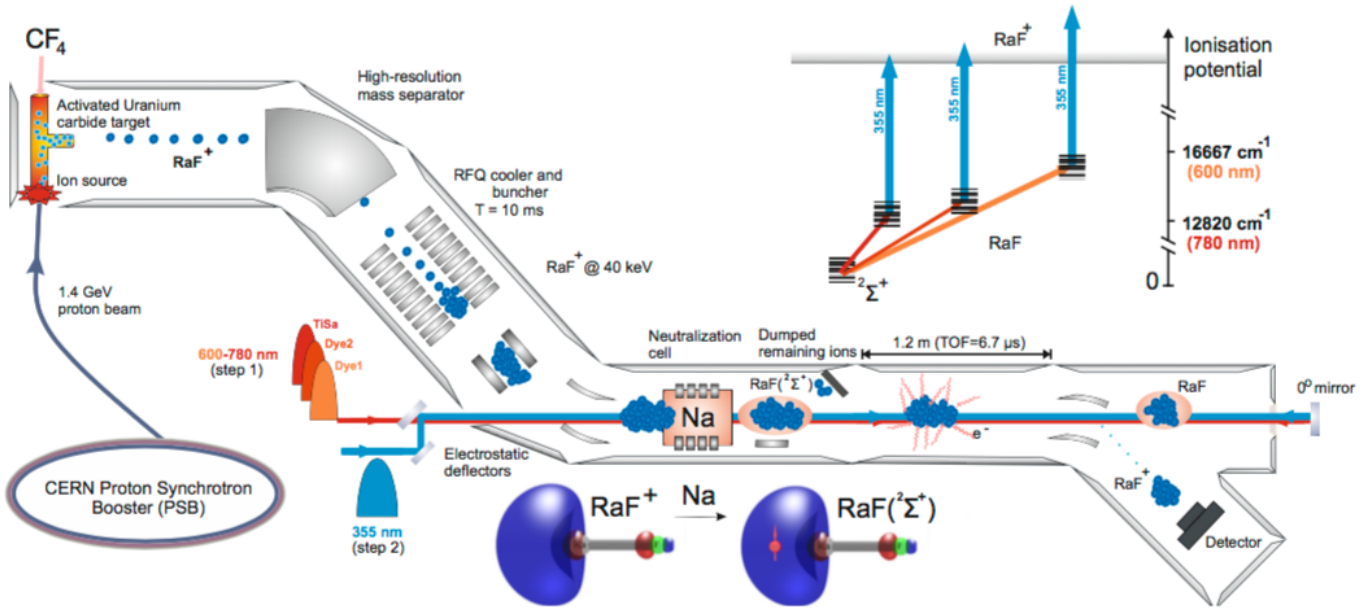
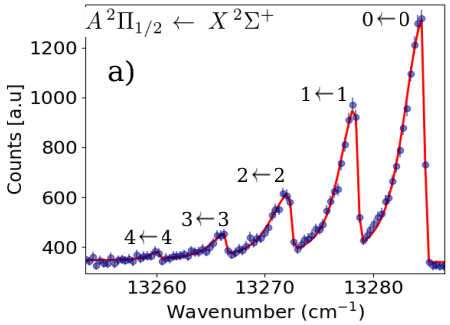
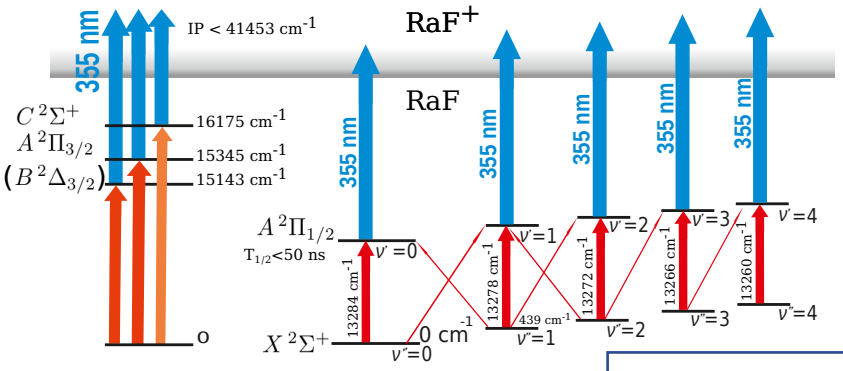






R. F. Garcia Ruiz et al., Nature 581, 396 (2020)

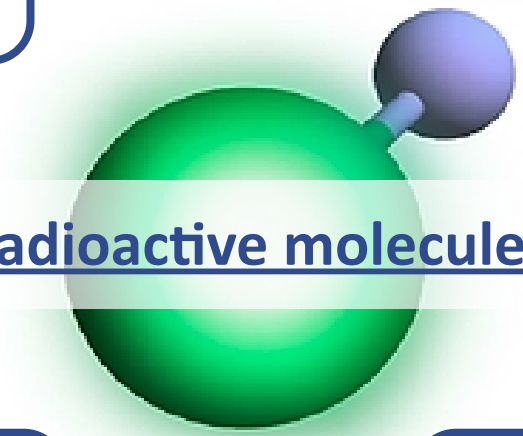
opens new research area

BSM interpretation

quantum chemistry

nuclear theory

molecular physics



RIB facility

radioactive molecules

EDM expertise

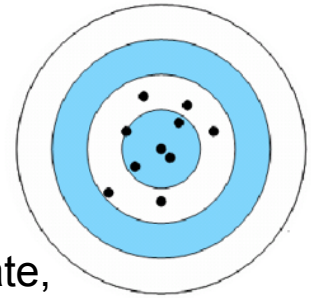
RIB techniques

atomic physics techniques

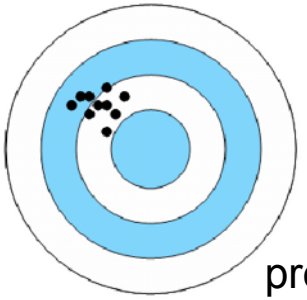
- dedicated reach programme and collaboration (S2068-LOI)
- identity most suitable radioactive molecule(s)

atomic physics techniques at RIB facilities

high **precision and accuracy**



accurate,
but not precise



precise,
but not accurate

ion traps
- masses
- RIB preparations
- in-trap decay

laser spectroscopy
- hyperfine structure
- isotope shifts
- optical pumping

atom traps
- in-trap decay
- laser spectroscopy
- APV

atomic physics techniques at RIB facilities

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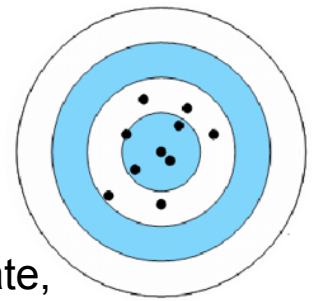

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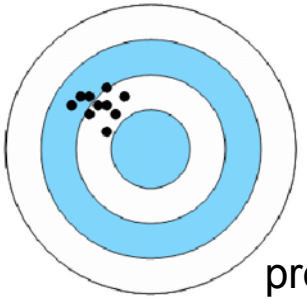

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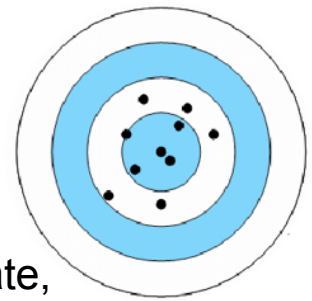

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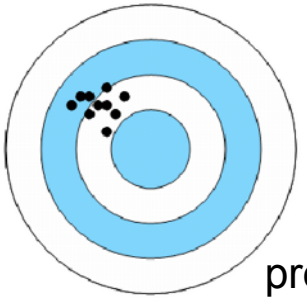

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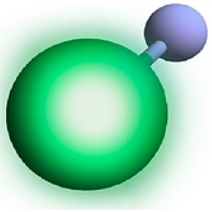
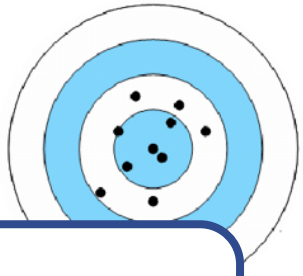
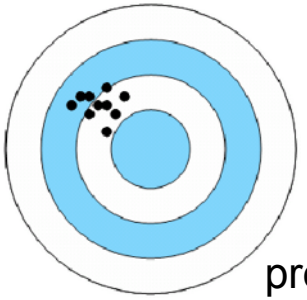


<u>radioactive molecules:</u>
advances in exp. techniques to link RIB science & molecular work

atomic physics techniques at RIB facilities

high **precision and accuracy**





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MIRACLS

ion traps
- masses
- RIB preparations
- in-trap decay

laser spectroscopy
- hyperfine structure
- isotope shifts
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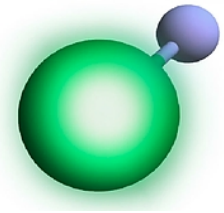
atom traps
- in-trap decay
- laser spectroscopy
- APV

Challenges

short half-lives **rarity** **contamination**



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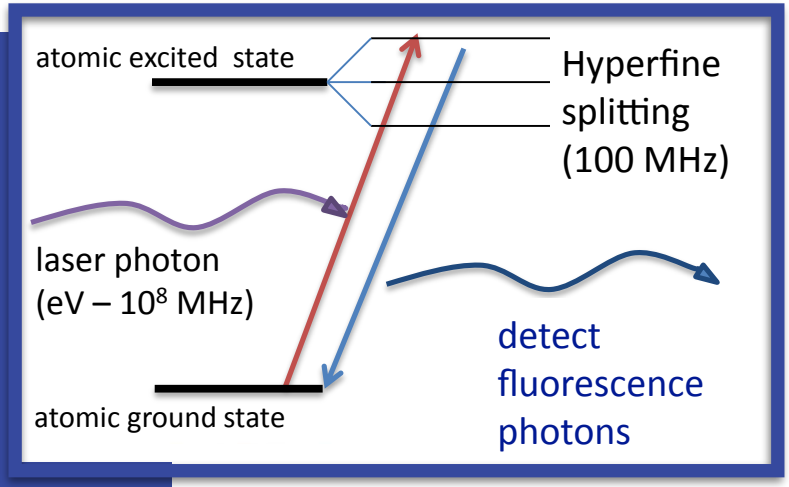
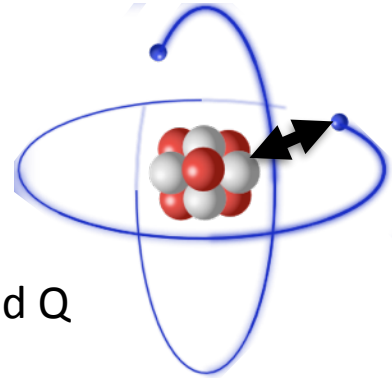
laser spectroscopy of radionuclides



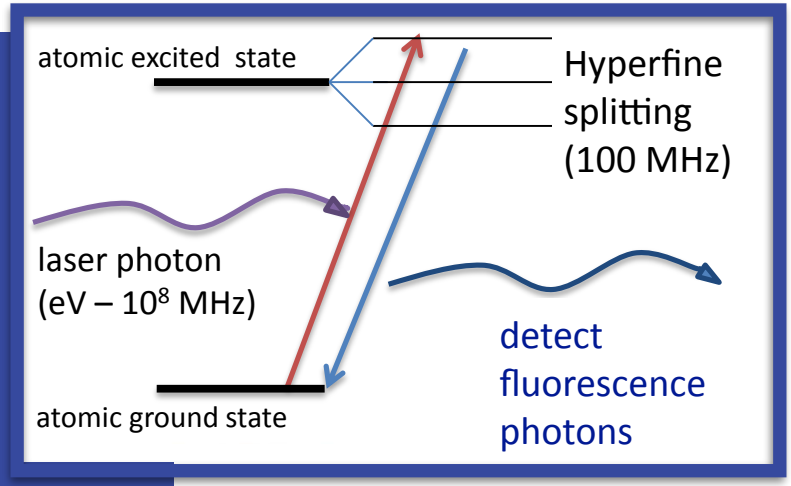
nucleus - electrons interaction

⇒ atomic hyperfine structure



- nuclear spin I
- electromagnetic moments μ and Q
- nuclear charge radius r_c

laser spectroscopy of radionuclides



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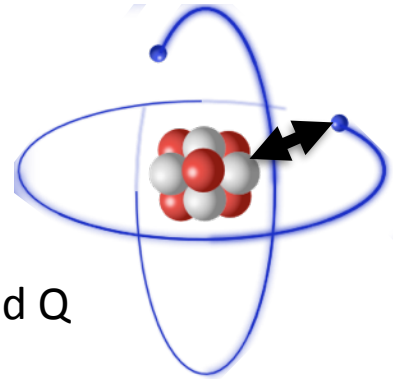
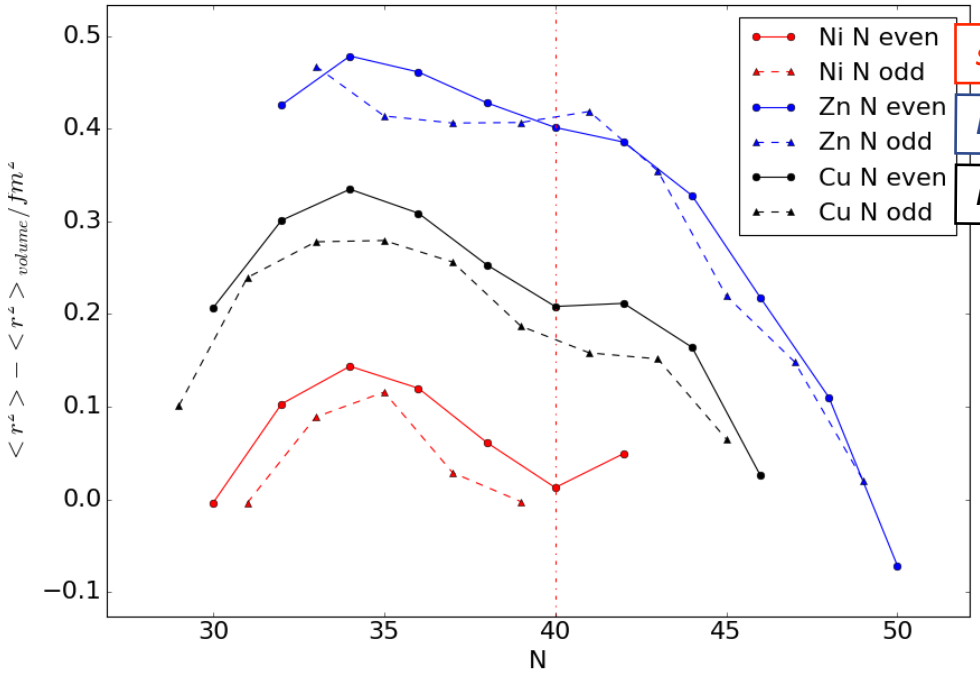




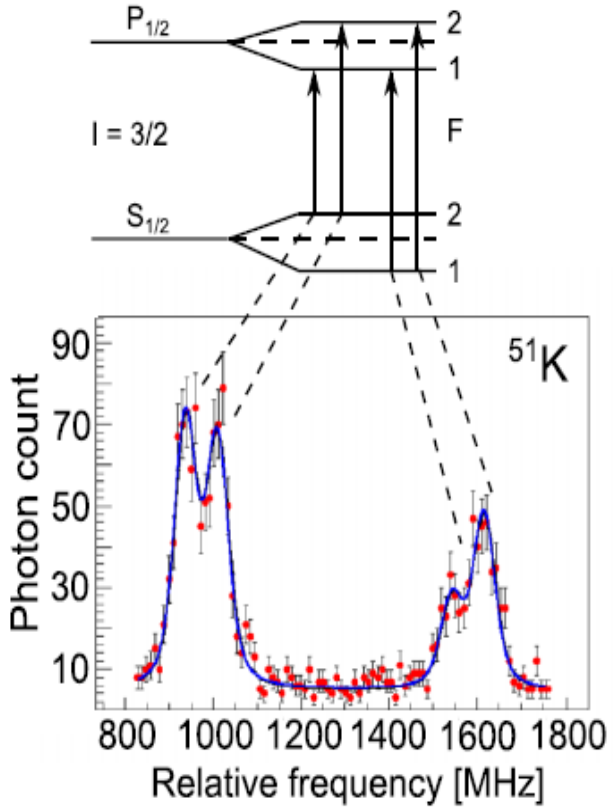
S. Malbrunot-Ettenauer + TRIUMF Theory et al., in prep. for Phys. Rev. Lett.

L. Xie et al., PLB 797, 134805 (2019)

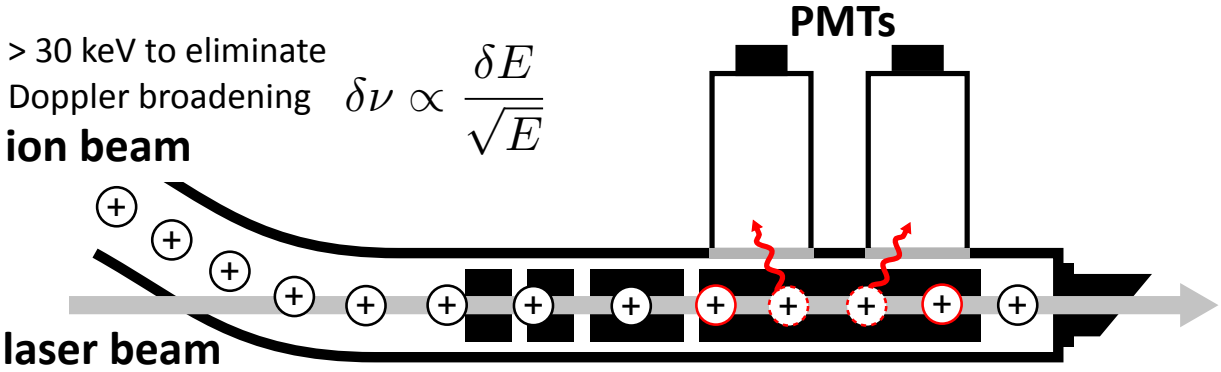
M. L. Bissell et al., PRC 93, 064318 (2016)

- **highlights changes in nuclear structure**
- **benchmark for modern nuclear theory**

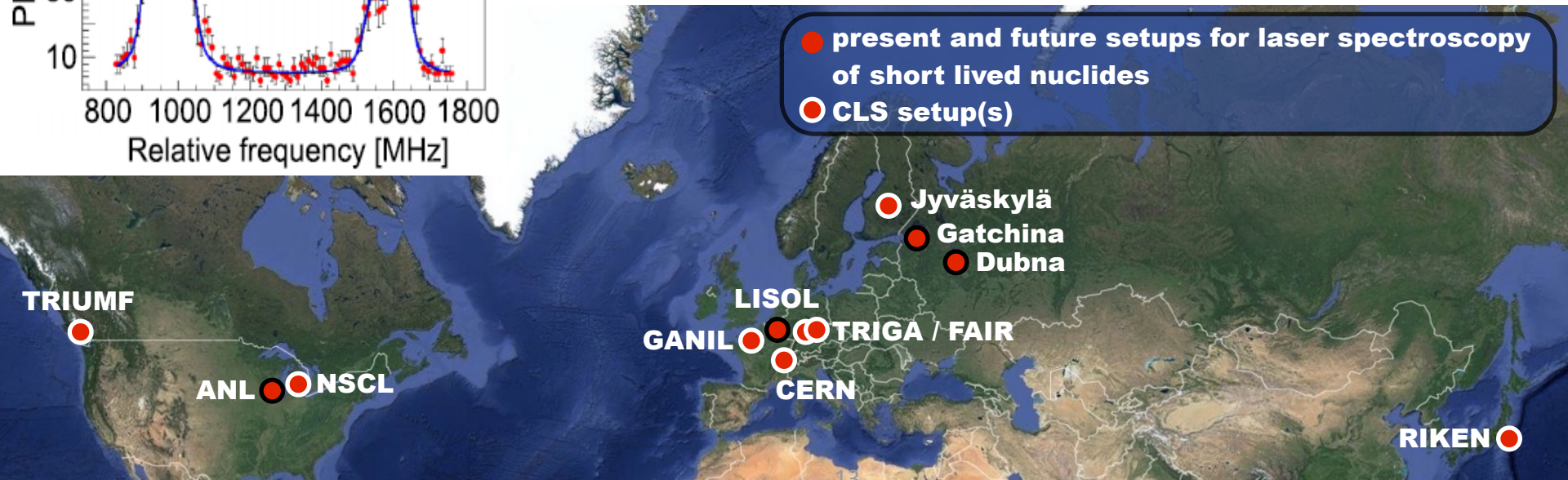
Collinear Laser Spectroscopy (CLS)



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



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)



Collinear Laser Spectroscopy (CLS)

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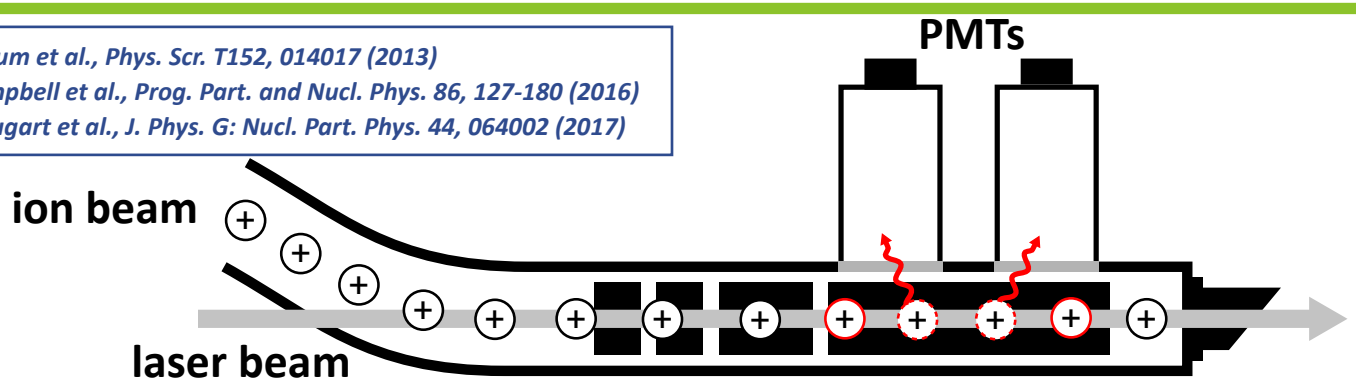
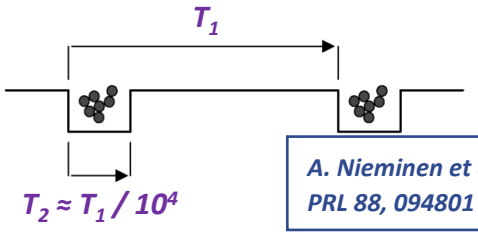


Fast beams (≥30 keV)
minimises Doppler-broadening

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

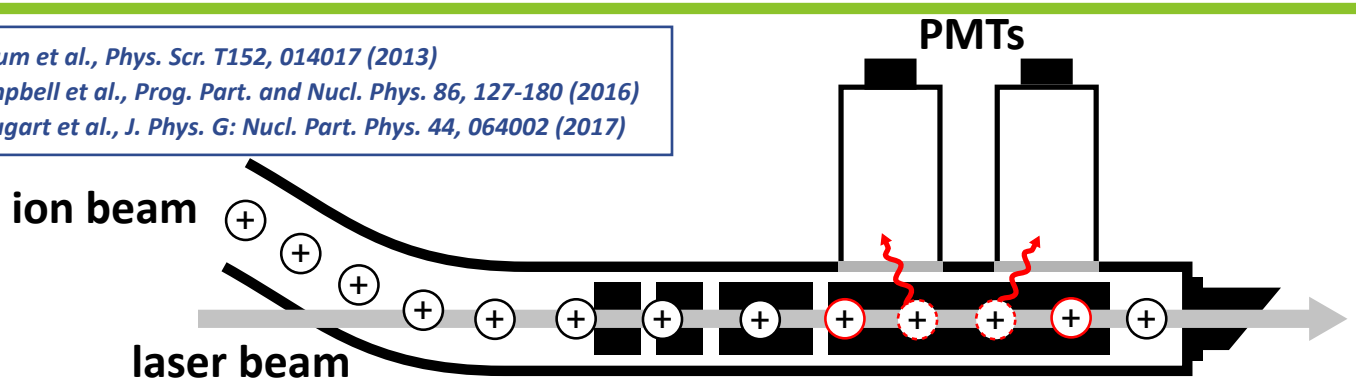
Bunched beams:
reduce background by gating on bunch



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

Collinear Laser Spectroscopy (CLS)

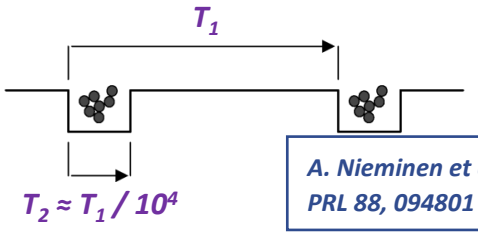
K. Blaum et al., Phys. Scr. T152, 014017 (2013)
P. Campbell et al., Prog. Part. and Nucl. Phys. 86, 127-180 (2016)
R. Neugart et al., J. Phys. G: Nucl. Part. Phys. 44, 064002 (2017)



Fast beams (≥30 keV)
minimises Doppler-broadening

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

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 µs

can one use exotic nuclides even more efficiently

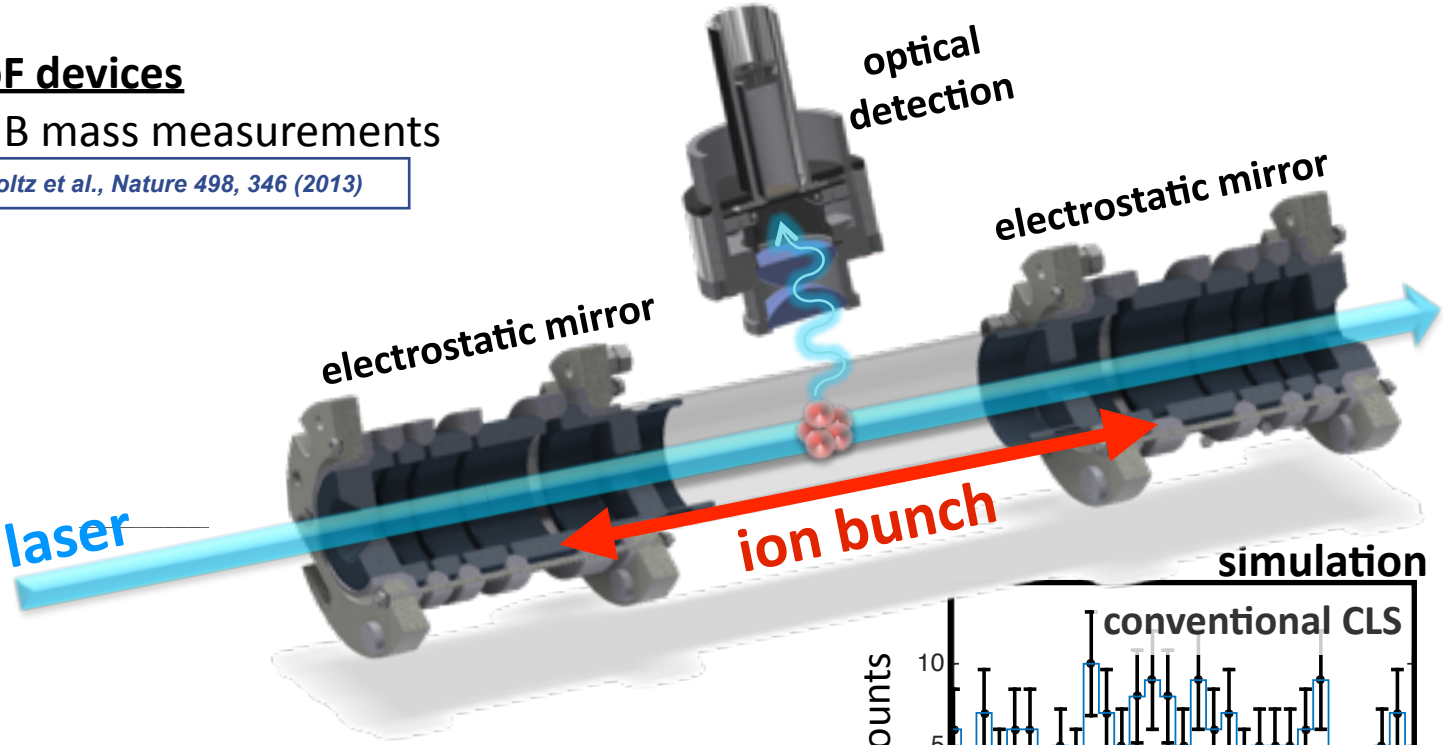
the Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy

trap ⇒ long observation time ⇒ higher sensitivity ⇒ more exotic nuclides accessible

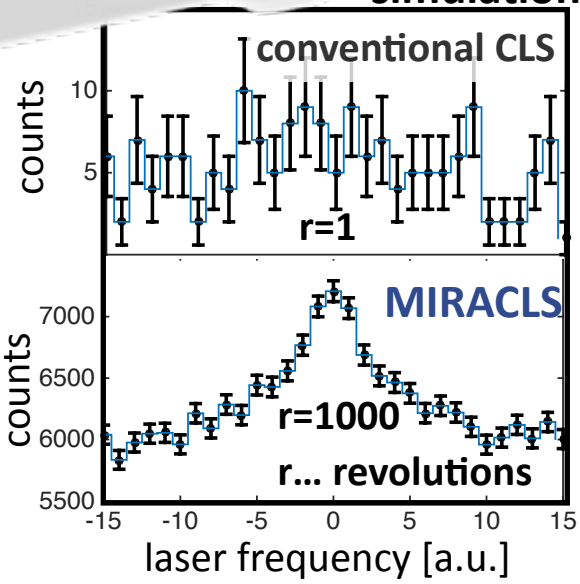
→ **MR-ToF devices**

first RIB mass measurements

F. Wienholtz et al., Nature 498, 346 (2013)



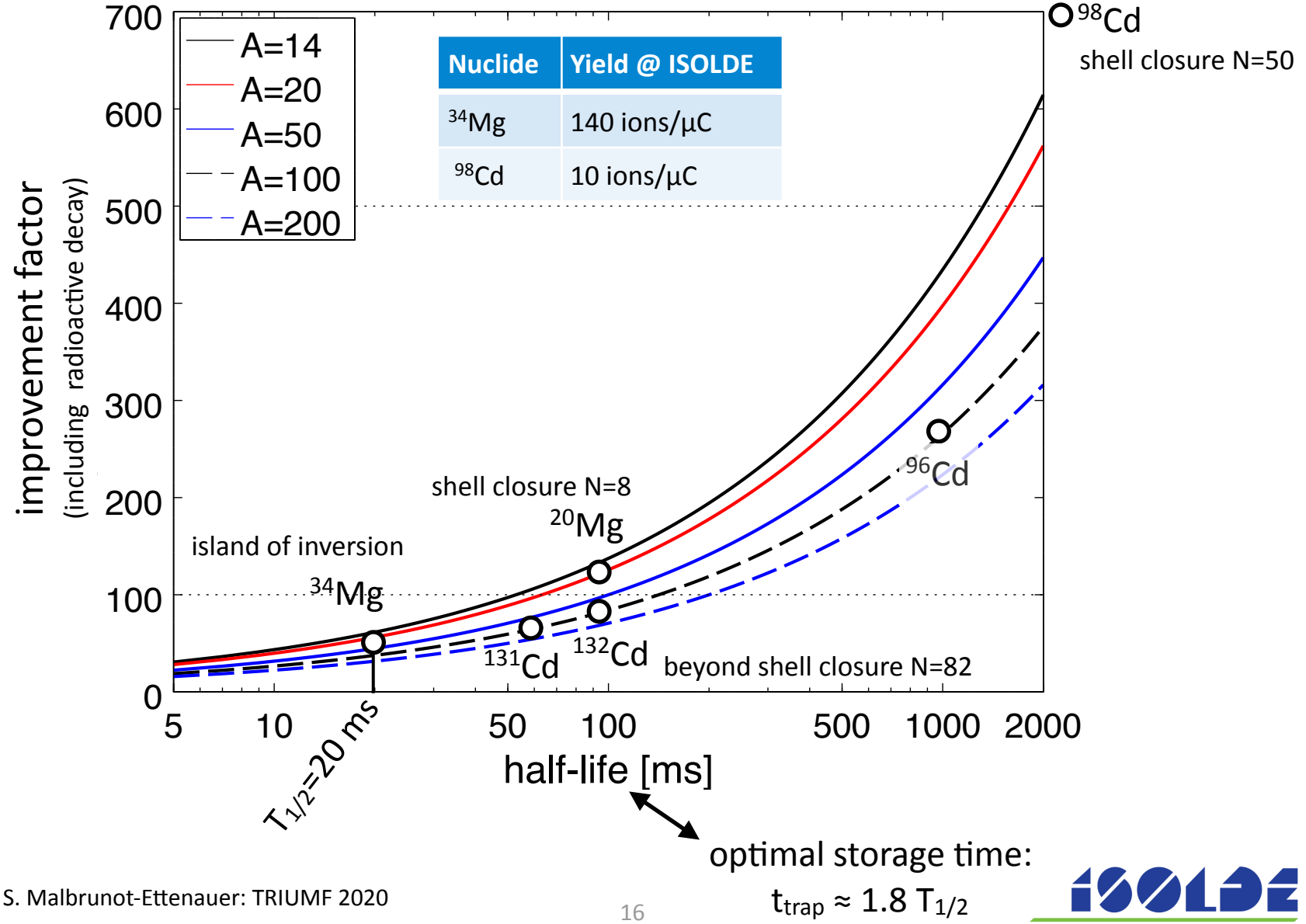
simulation



novel approach for collinear laser spectroscopy:

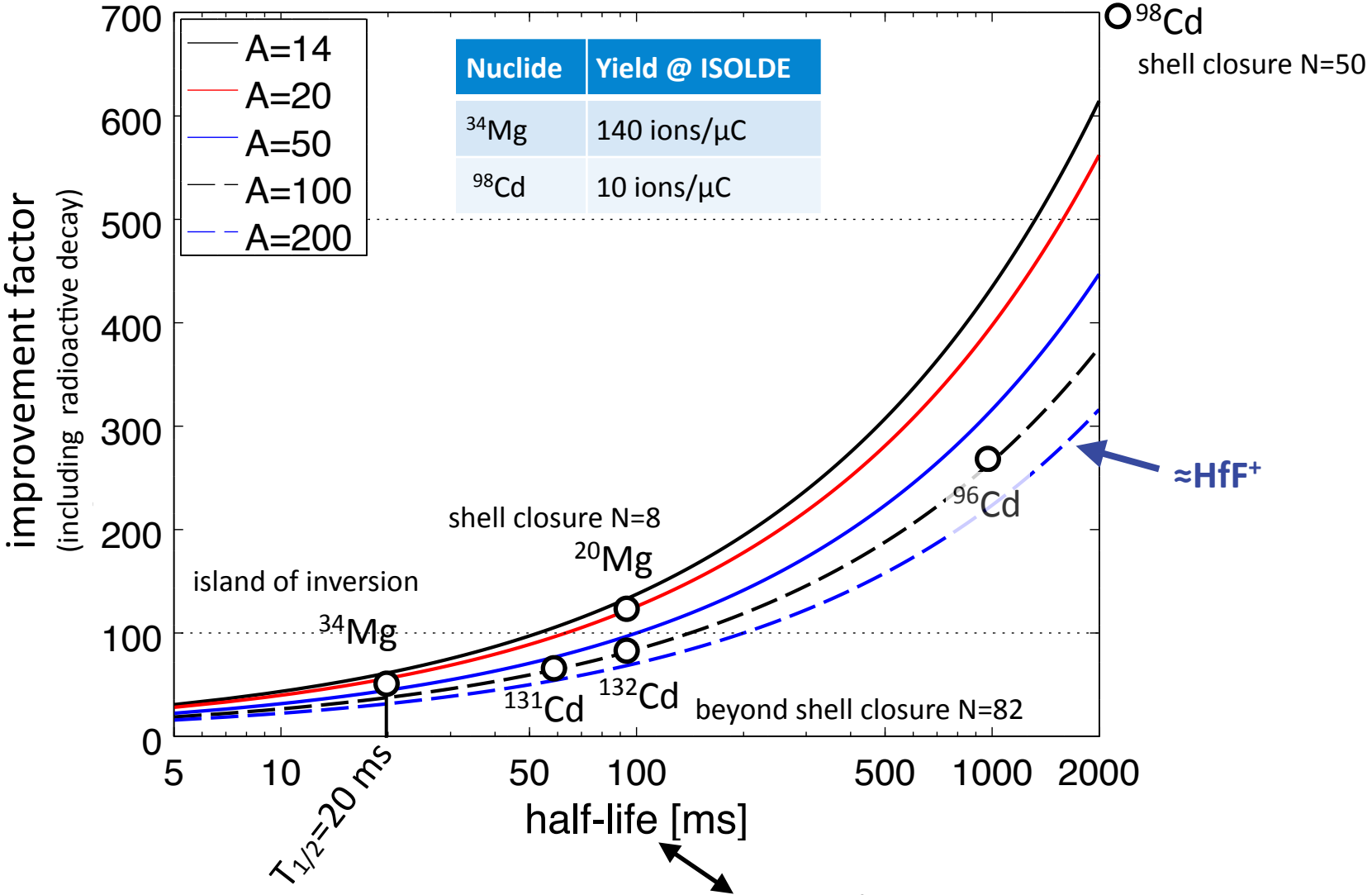
- ion trap ⇒ long observation time
- 30 keV beam ⇒ high resolution

improvement factor & 1st science cases



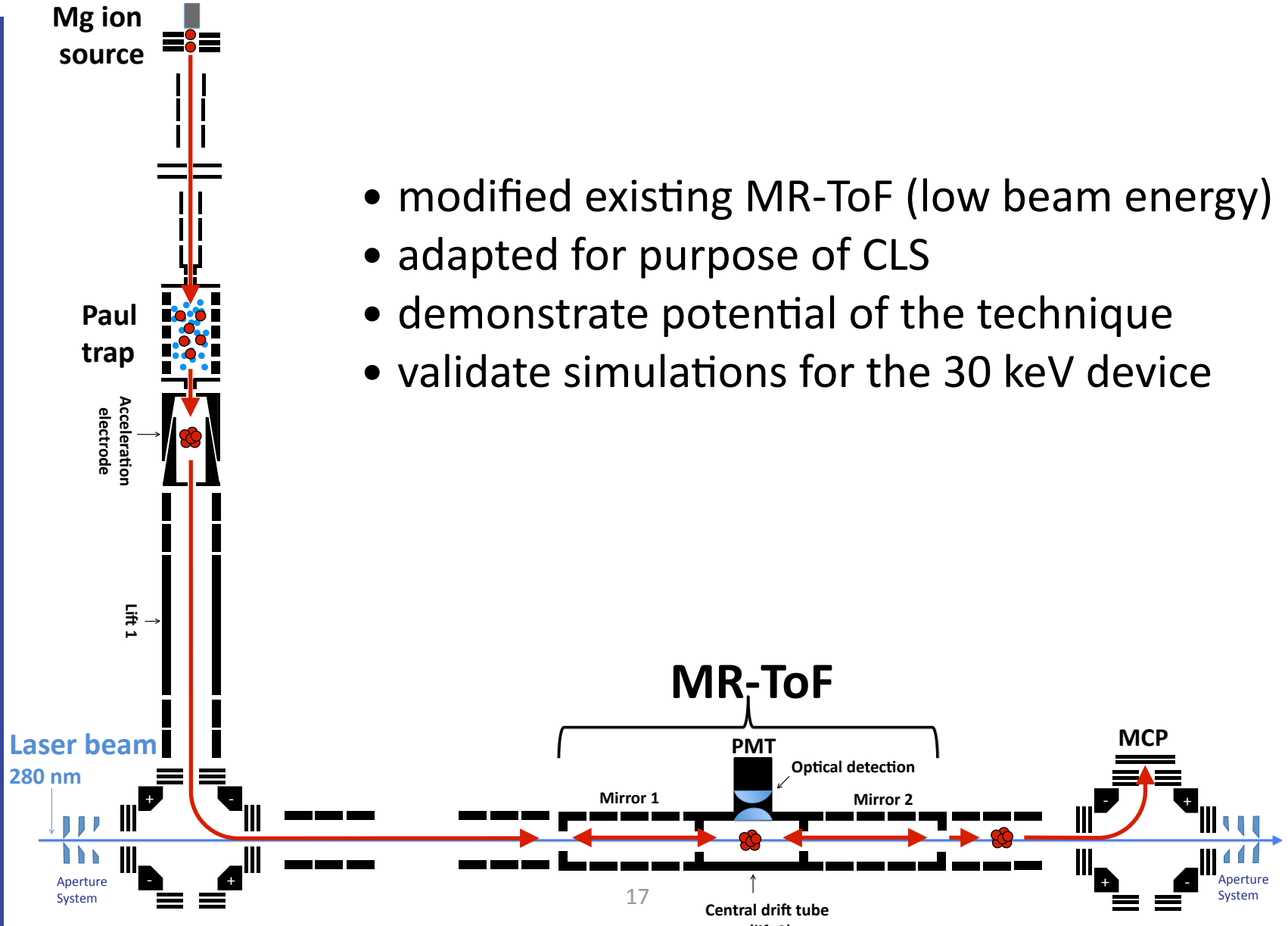
S. Malbrunot-Ettenauer: TRIUMF 2020

improvement factor & 1st science cases



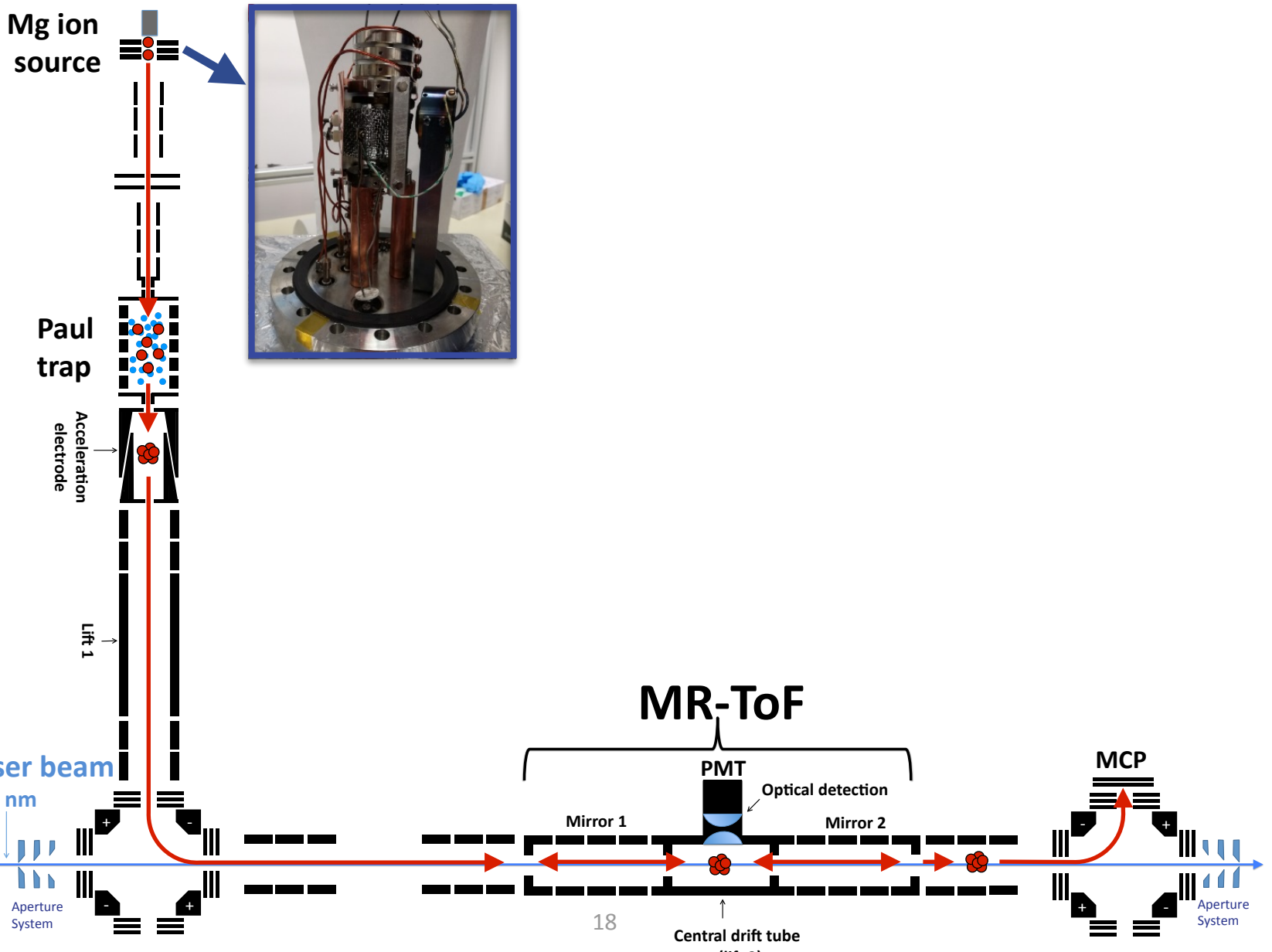
S. Malbrunot-Ettenauer: TRIUMF 2020

proof-of-principle experiment

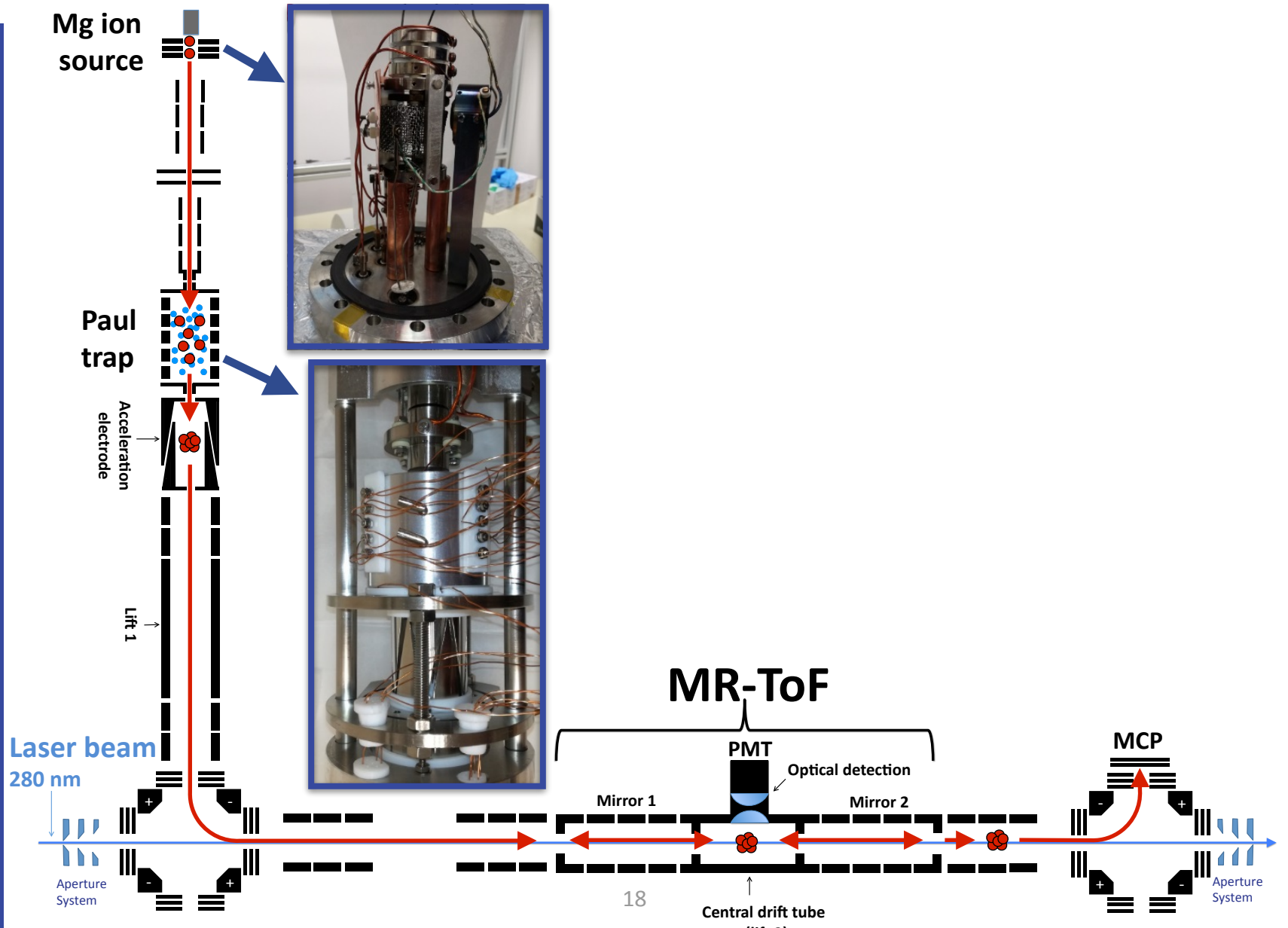


- modified existing MR-ToF (low beam energy)
- adapted for purpose of CLS
- demonstrate potential of the technique
- validate simulations for the 30 keV device

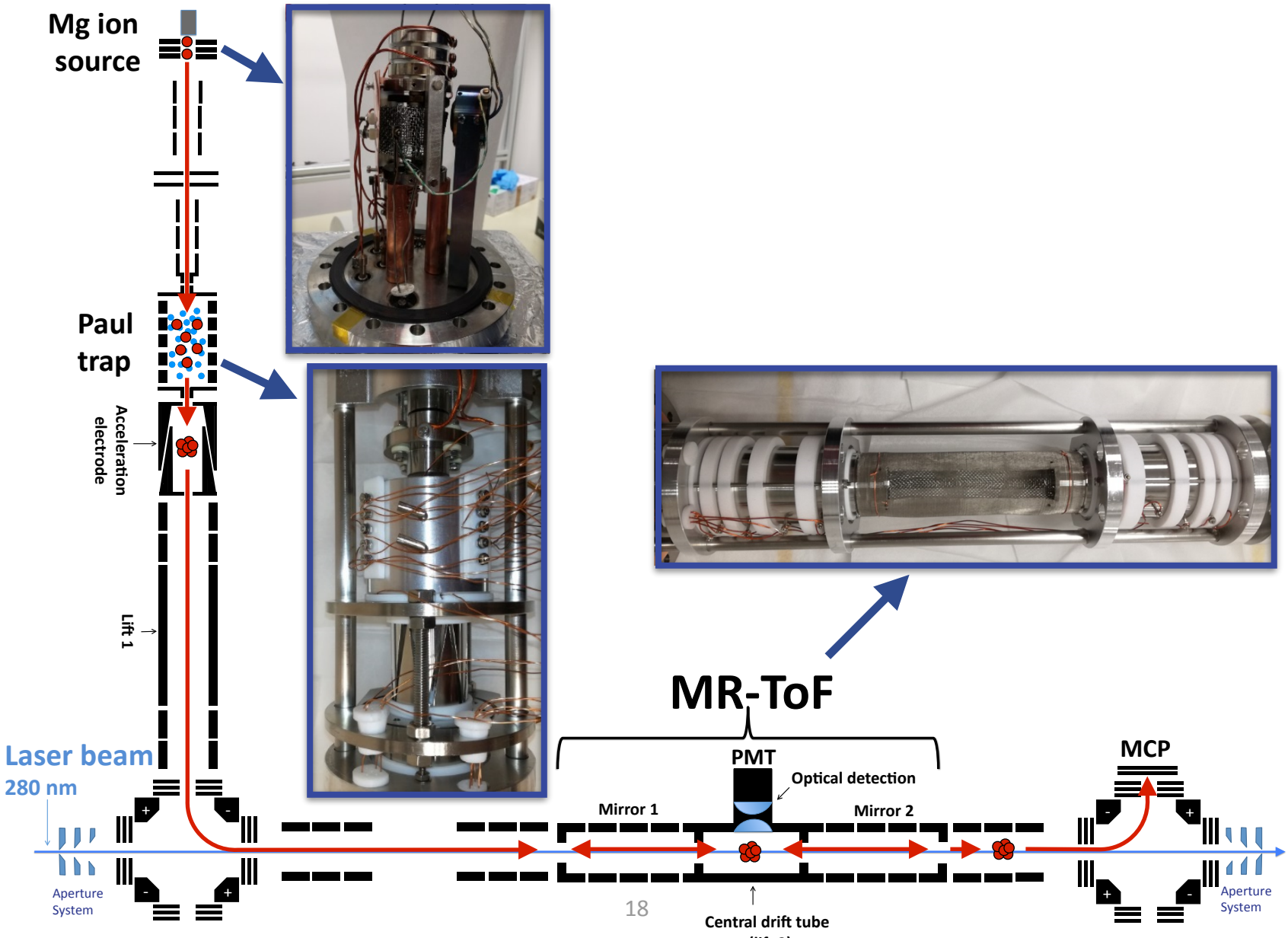
proof-of-principle experiment



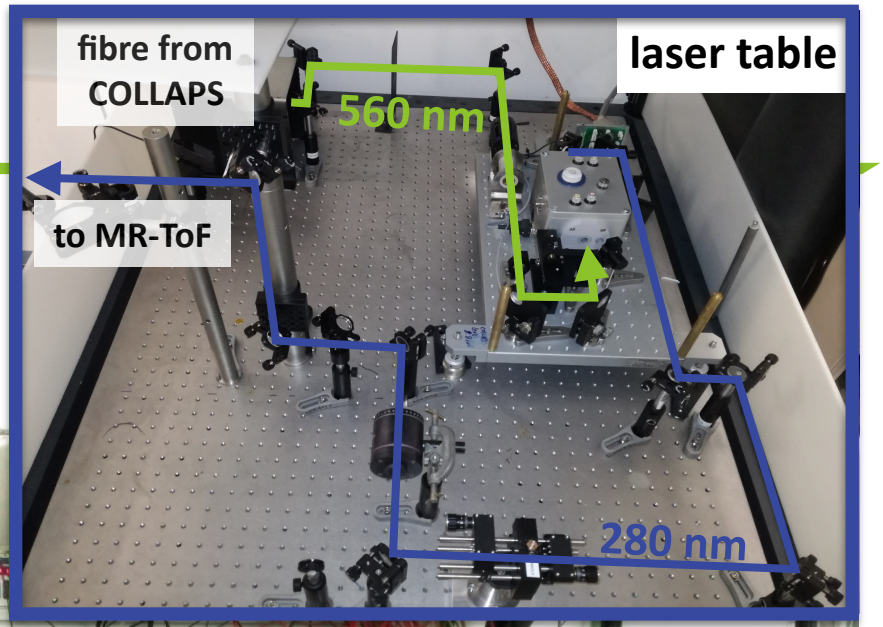
proof-of-principle experiment



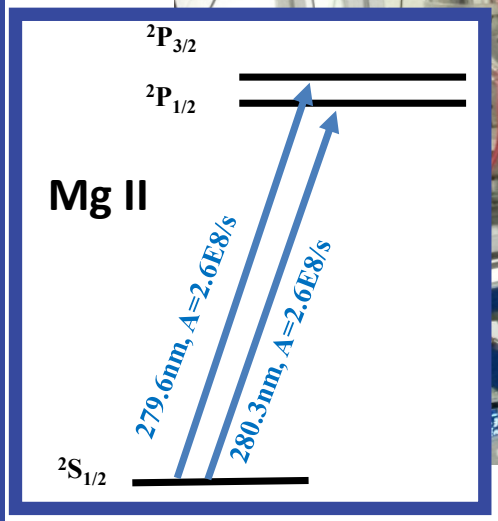
proof-of-principle experiment



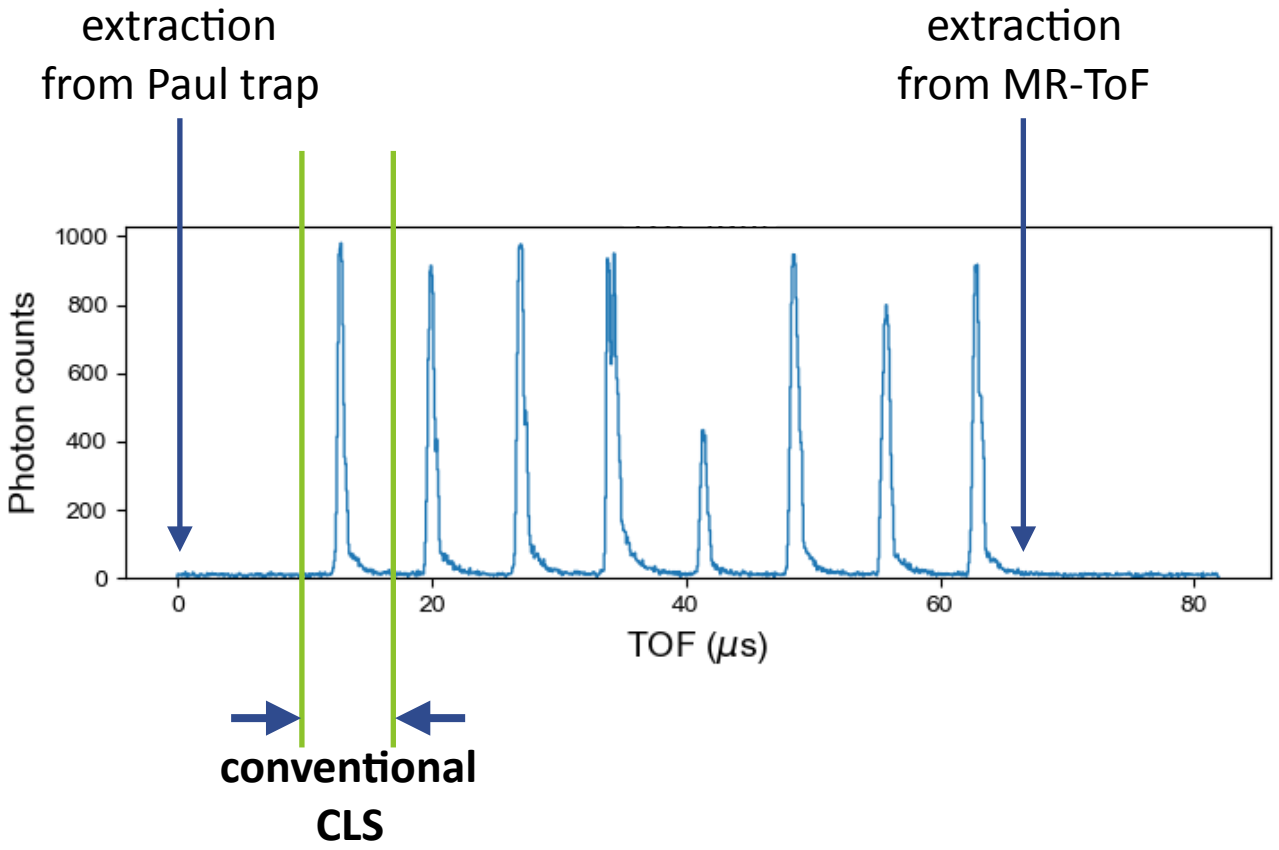
PoP setup



S. Sels et al., Nucl. Instr. Meth. B 463, 310 (2020)
F. Maier et al., Hyperfine Interact. 240, 54 (2019)
S. Lechner et al., Hyperfine Interact 240, 95 (2019)
V. Lagaki et al., Acta Phys. Pol. B 51, 571 (2020)
V. Lagaki et al., in preparation

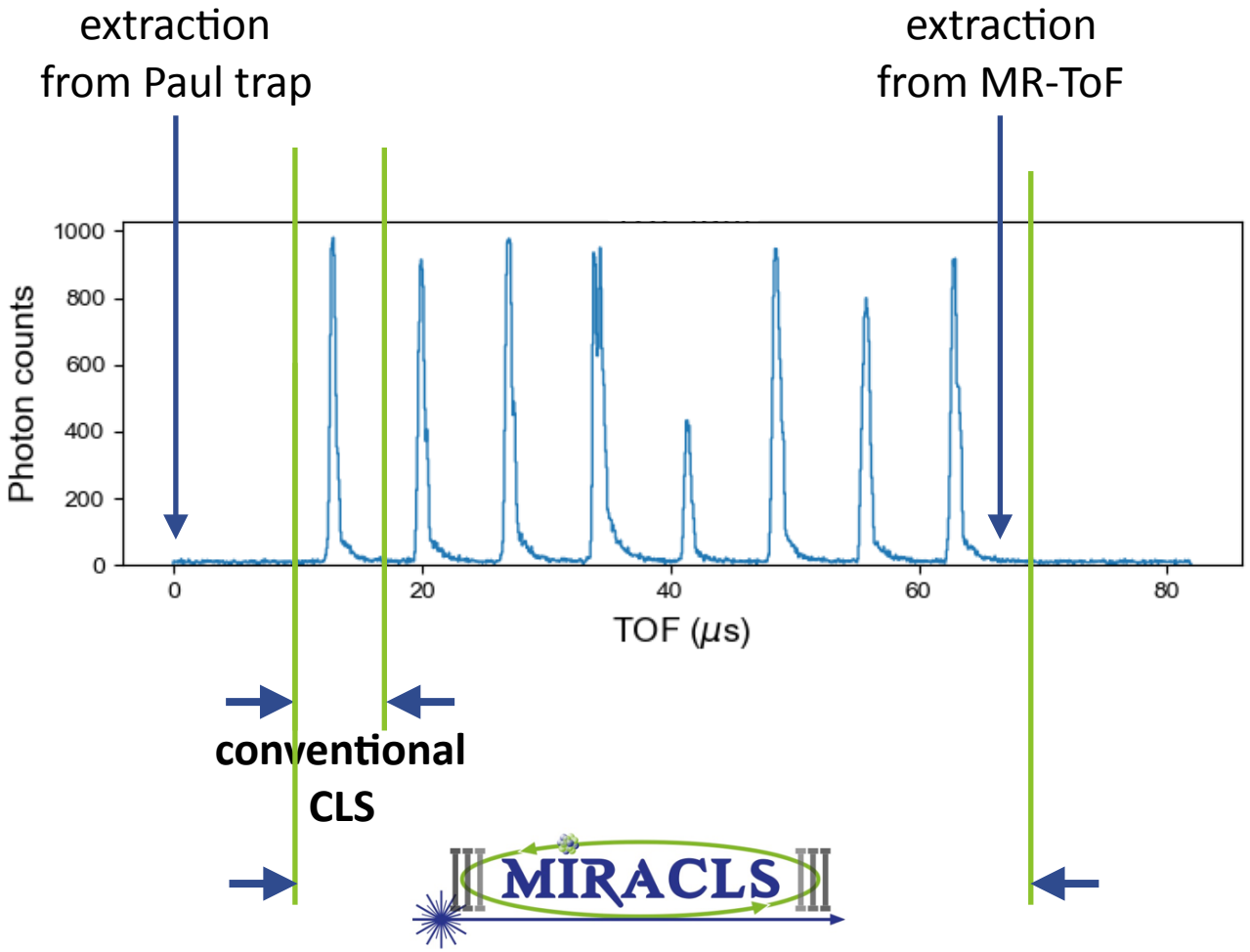


First CLS signals in an MR-ToF



S. Malbrunot-Ettenauer: TRIUMF 2020

First CLS signals in an MR-ToF



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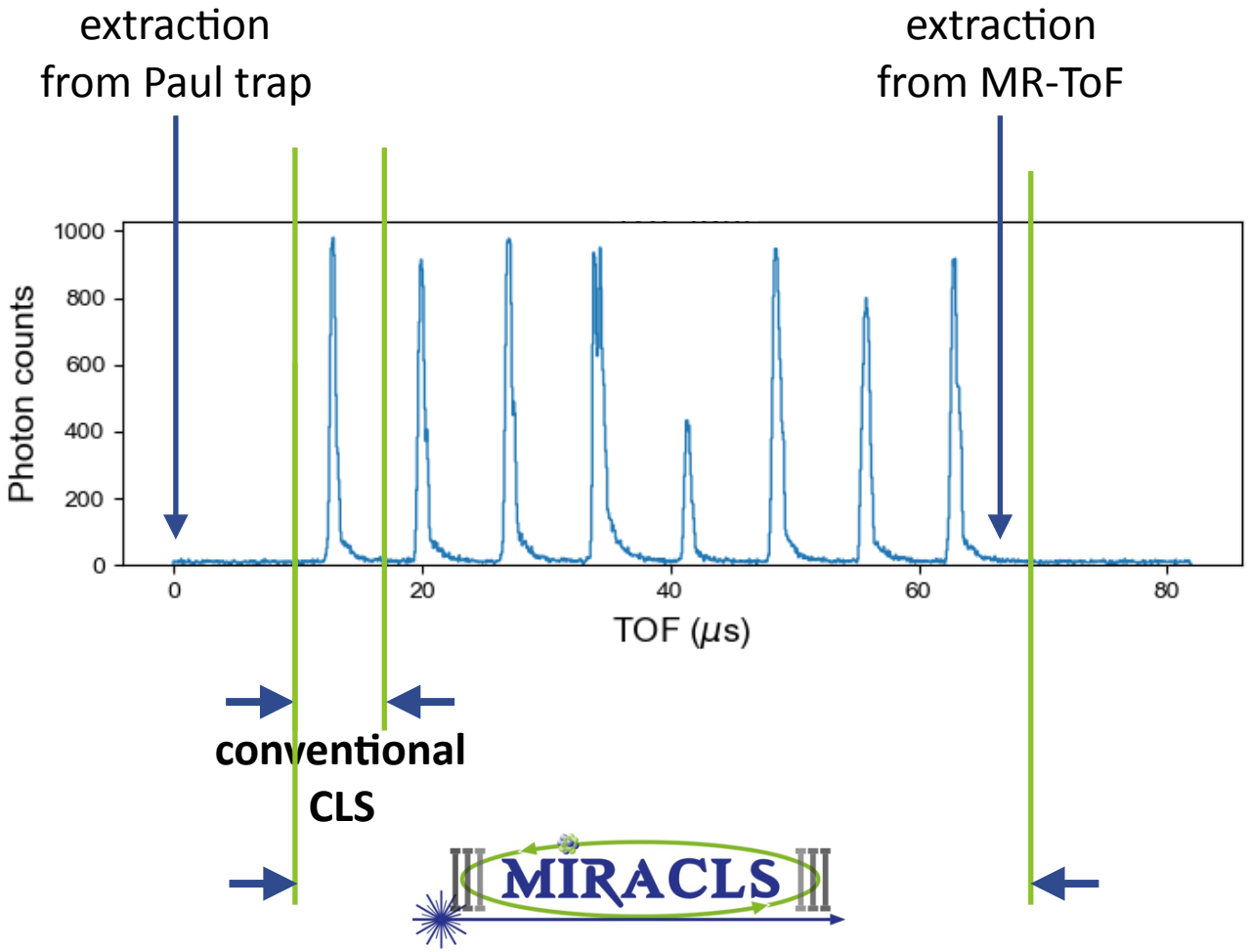
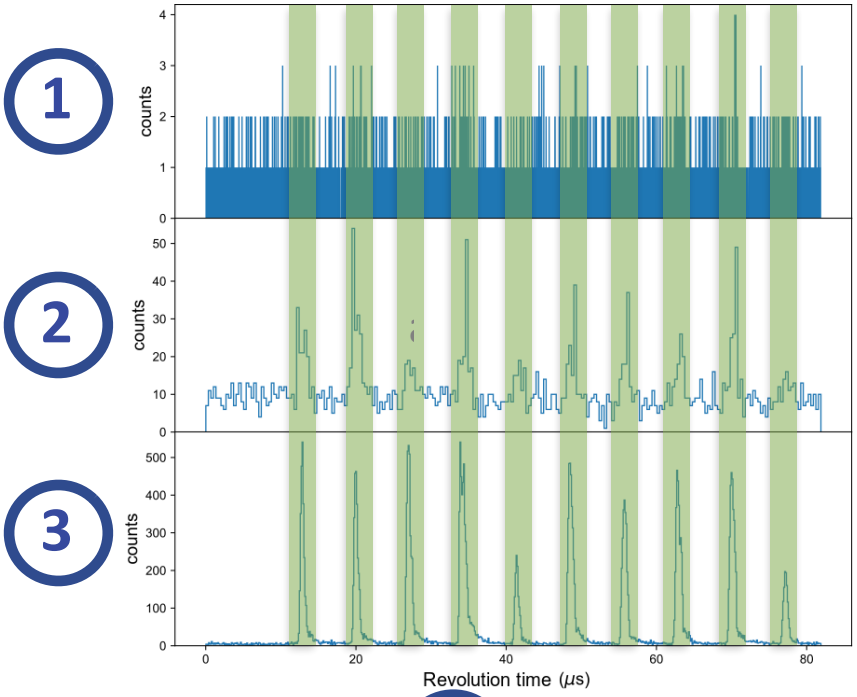
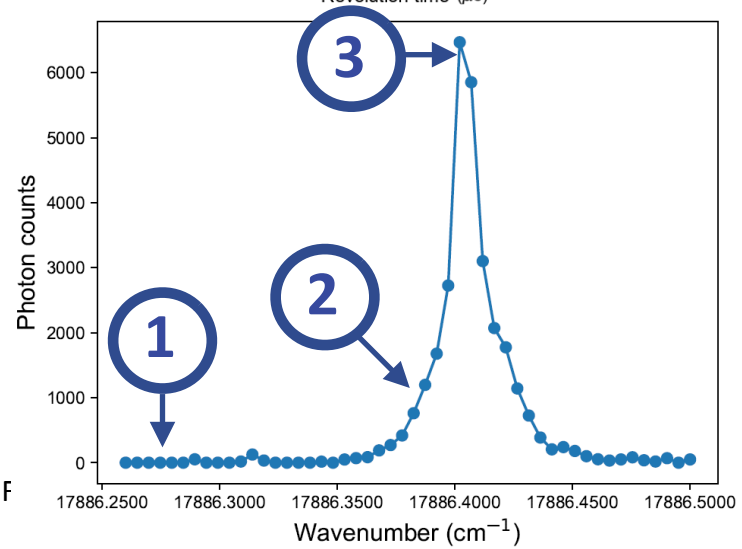


shown here only for 7 revolutions
demonstrated up to 5000 rev

First CLS signals in an MR-ToF

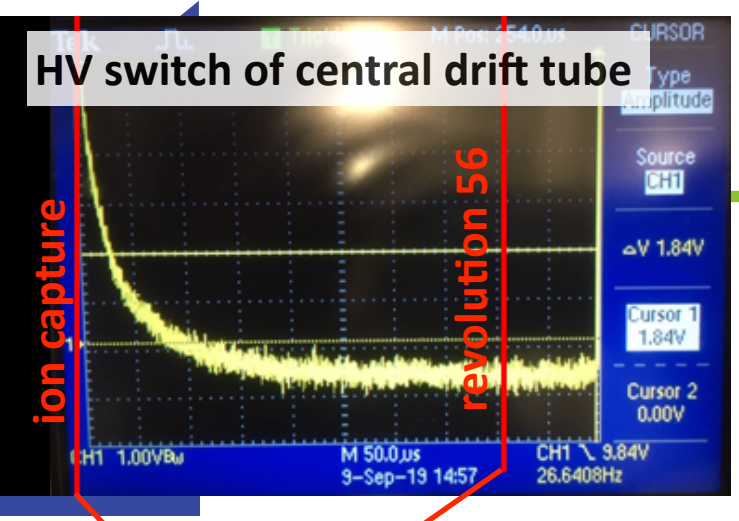


Counts for each
TOF peak
as function
of laser frequency

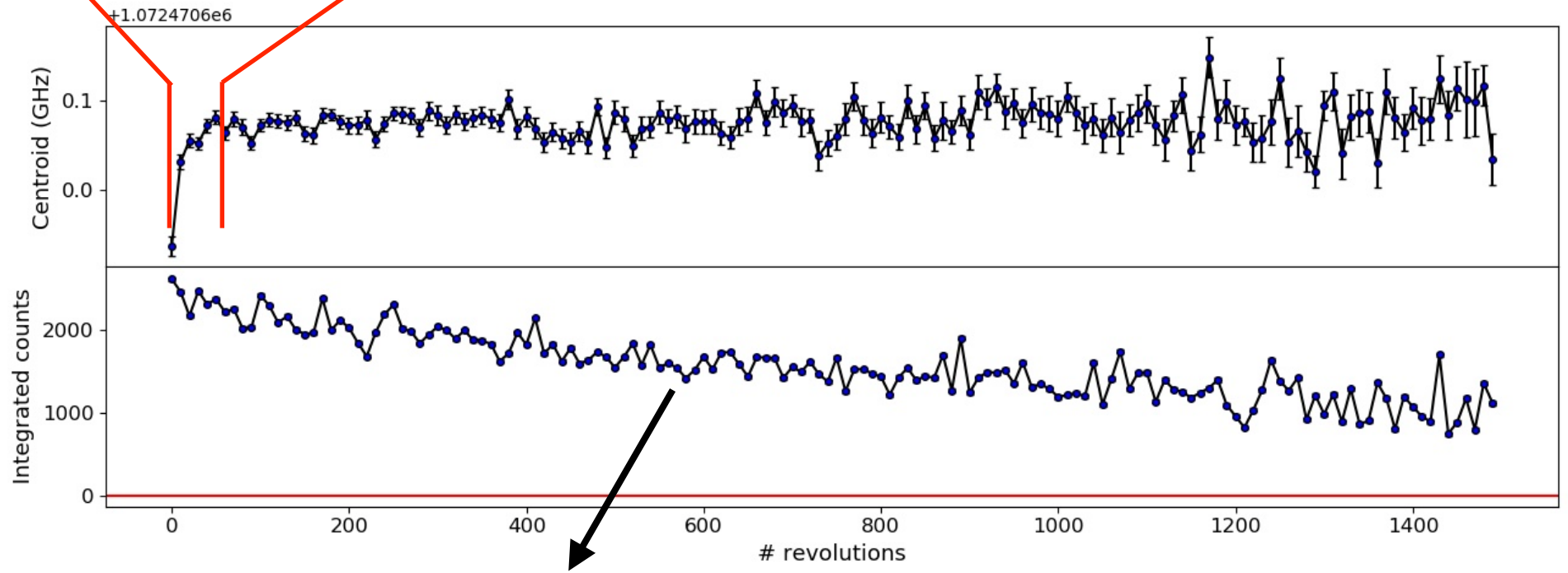


S. Malbrunot-Ettenauer: TRIUMF

CLS performance

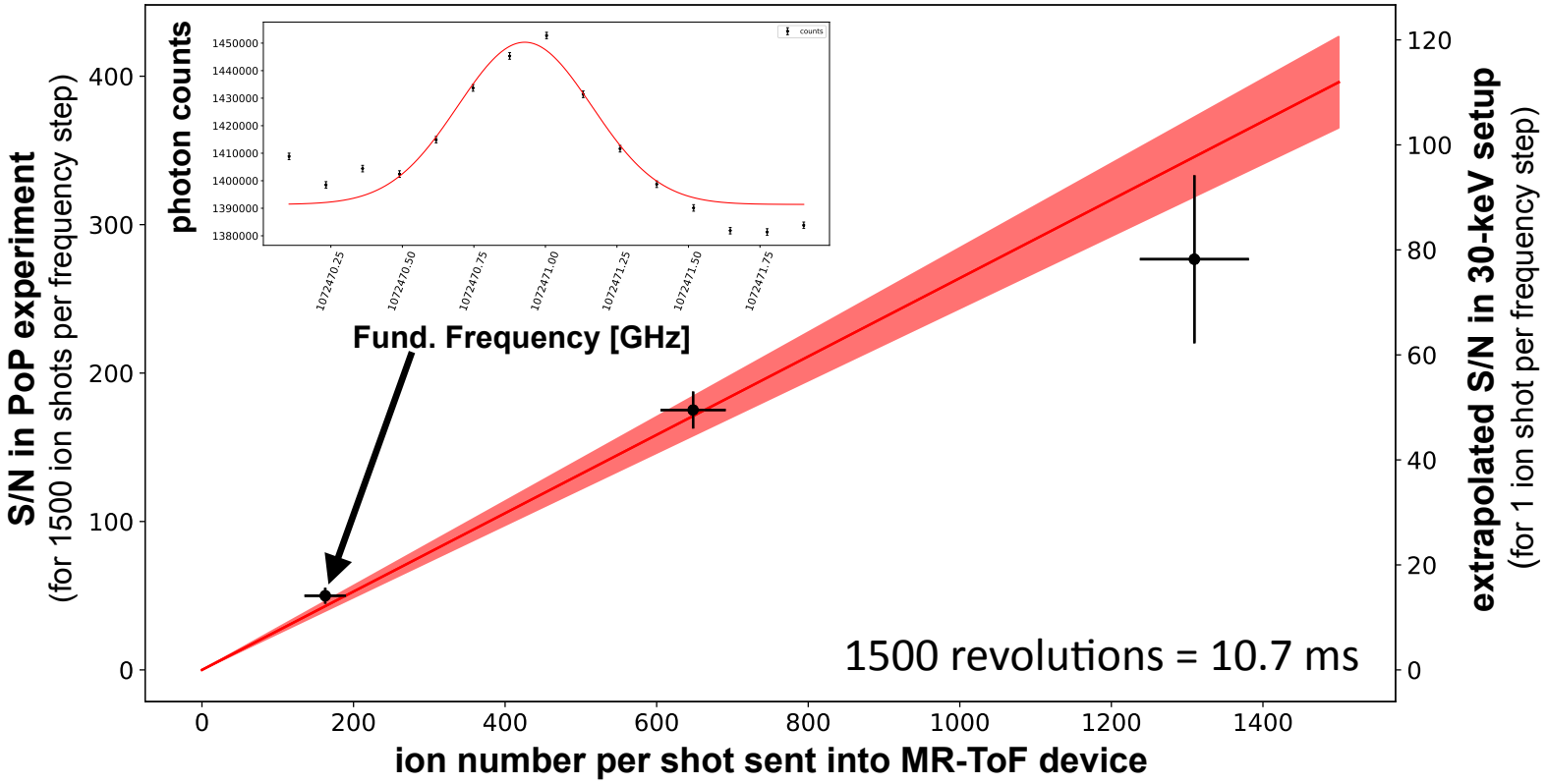


- excellent centroid stability after revolution ≈60
- initial drift due to HV switch



- CLS loses strongly linked to ion loses
- illustrates vacuum challenge specific for CLS in ion trap

MIRACLS sensitivity in $^{24}Mg^{+}$

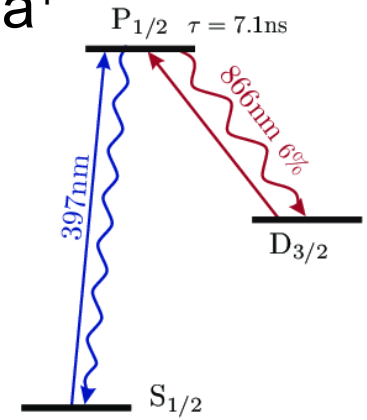


online measurements with *O*(10) ions/sec possible

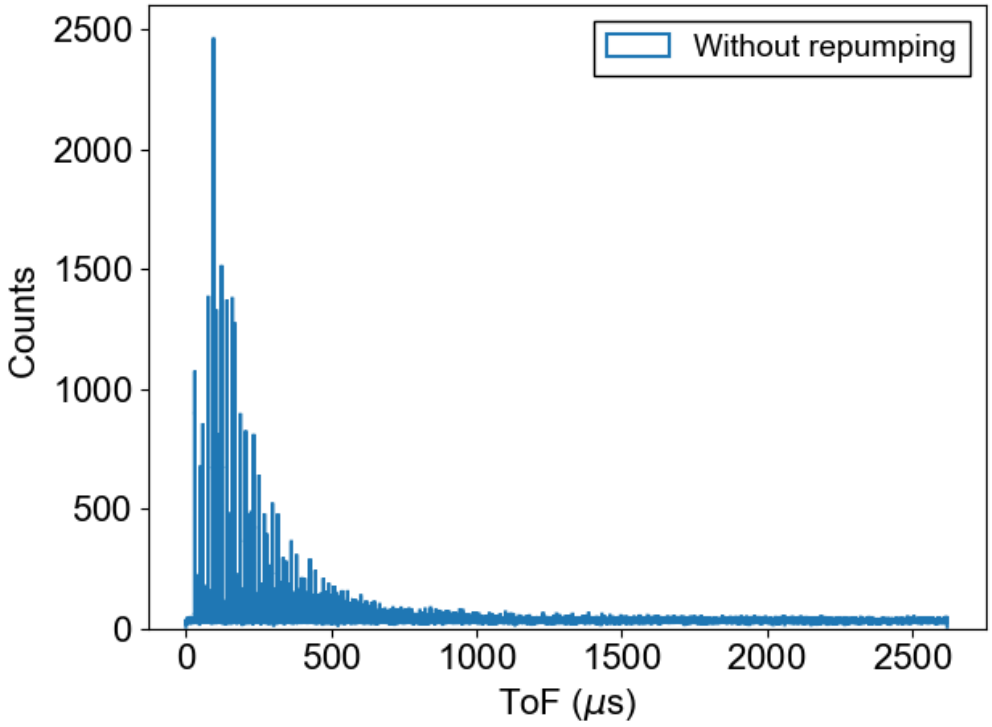
S. Malbrunot-Ettenauer: TRIUMF 2020

$^{40}Ca^{+}$: towards non-closed level systems

$^{40}Ca^{+}$

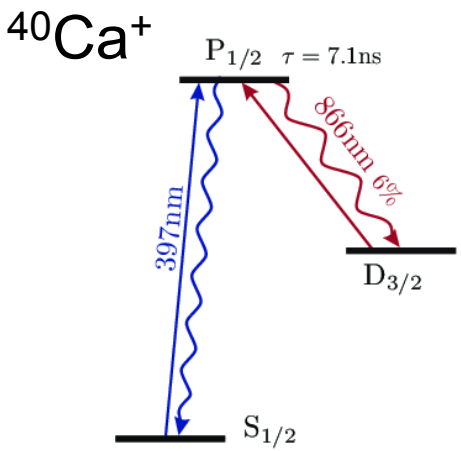


397 nm in collinear
866 nm in anticollinear mode



Without re-pumping: lose signal after 40 revolutions (still an improvement!!)

$^{40}Ca^{+}$: towards non-closed level systems



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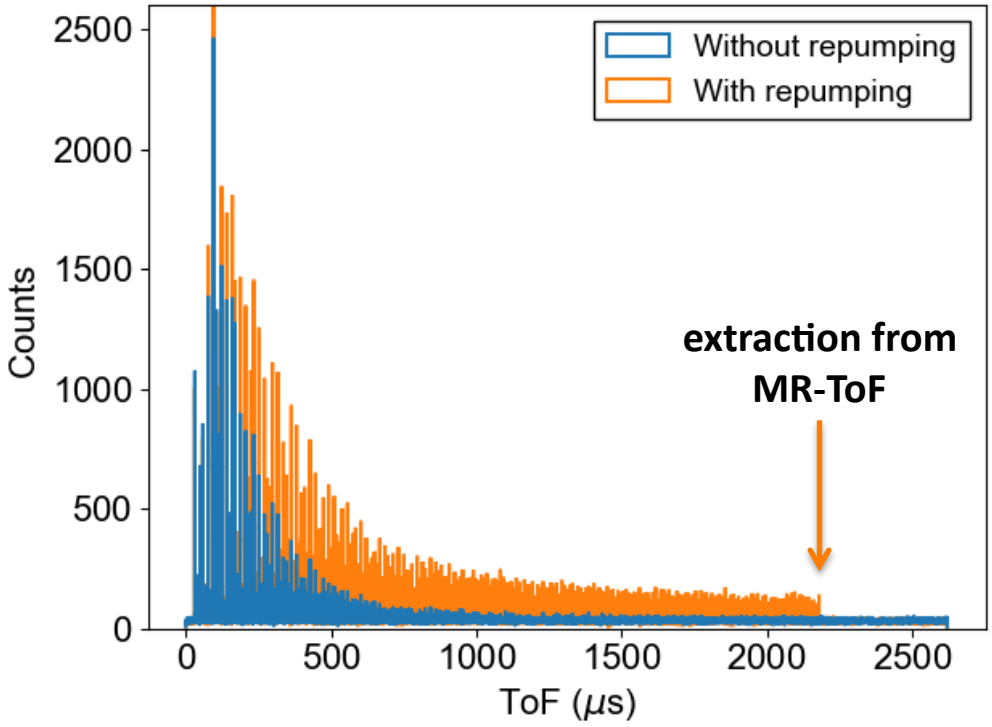
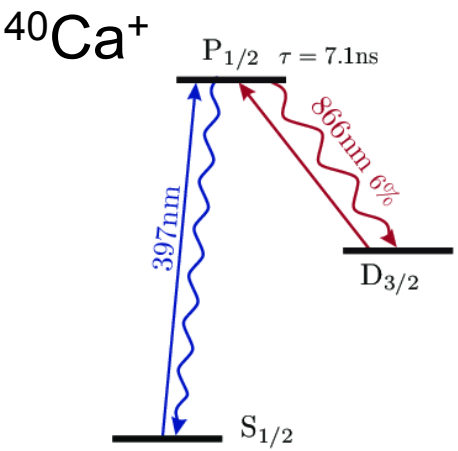


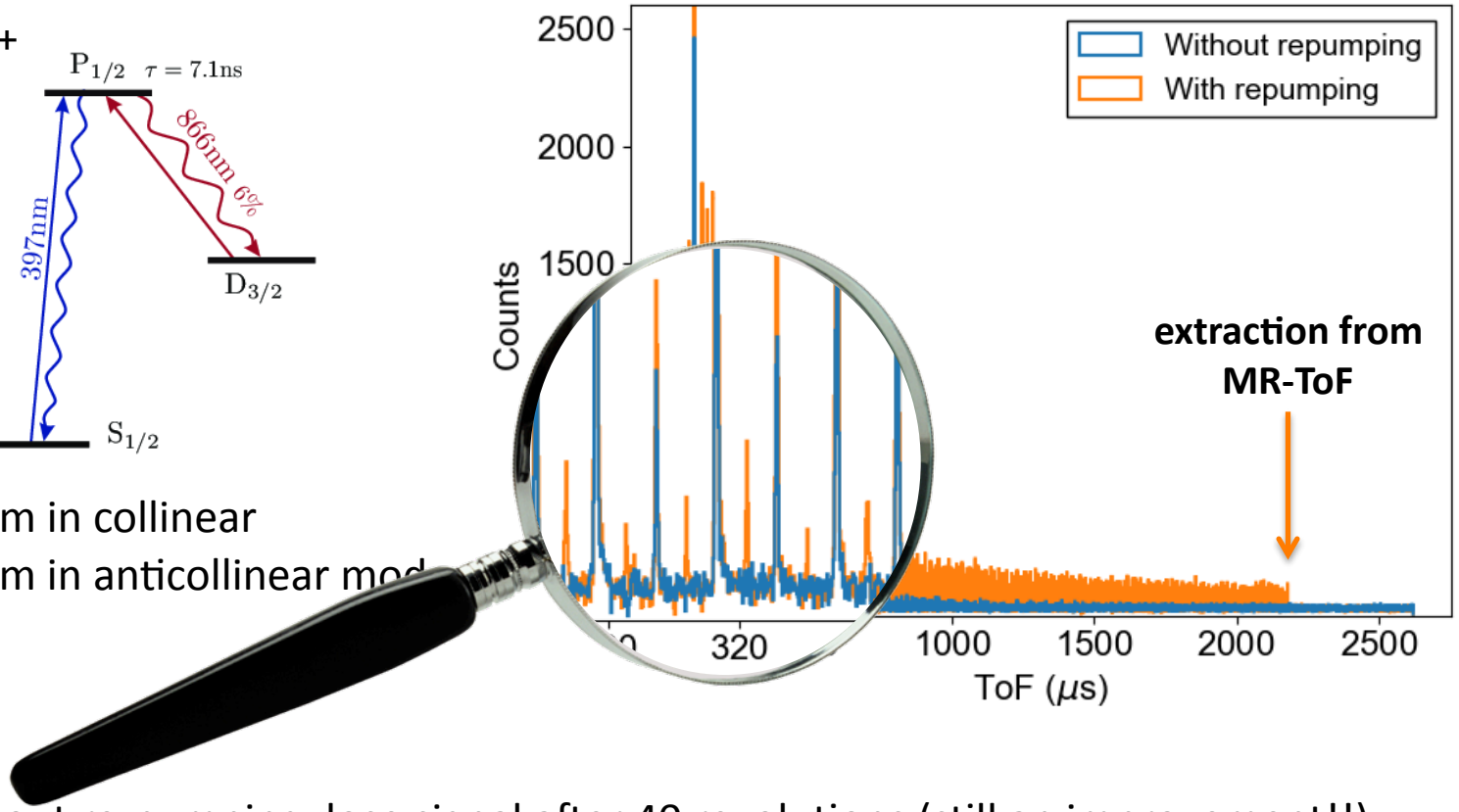
Without re-pumping: lose signal after 40 revolutions (still an improvement!!)

With re-pumping: signal visible until extraction

Proof of recycling of 'lost' ions via re-pumping form a dark state

$^{40}Ca^{+}$: towards non-closed level systems





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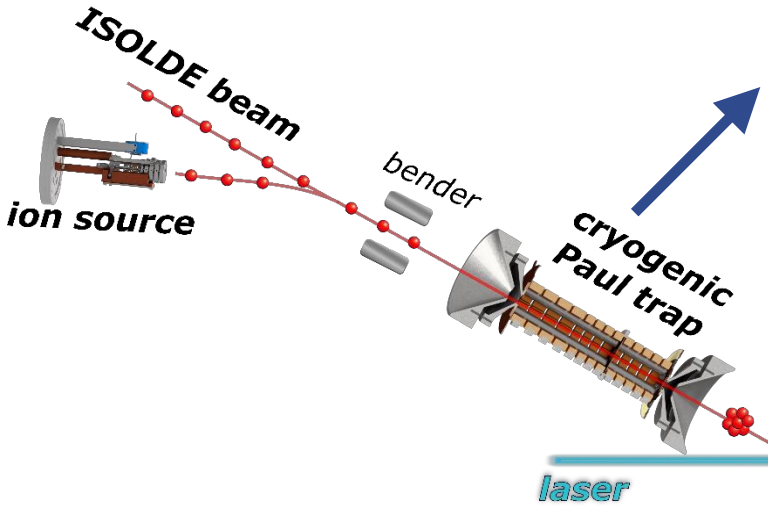
Proof of recycling of 'lost' ions via re-pumping form a dark state

looking for re-pump signal: did turn off blue laser for higher sensitivity!!

S. Malbrunot-Ettenauer: TRIUMF 2020

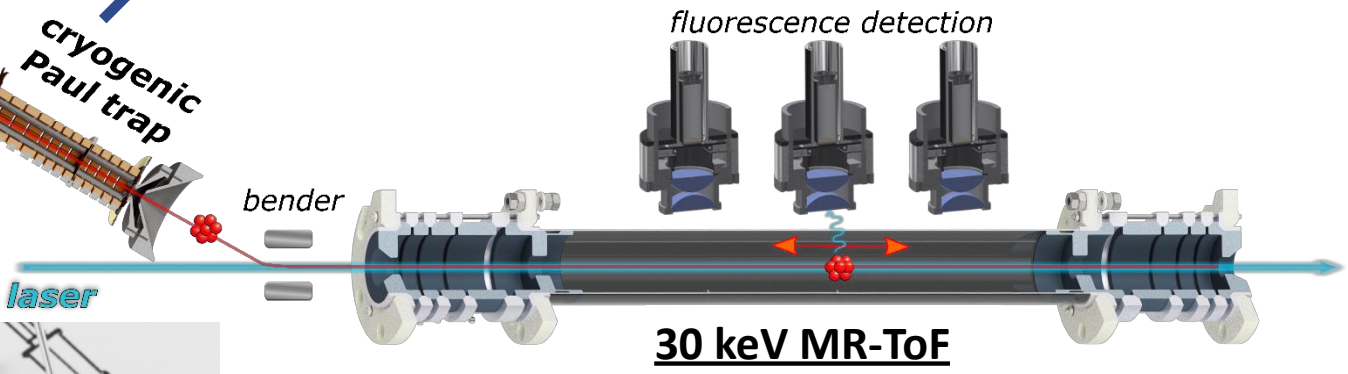
MIRACLS 30-keV setup

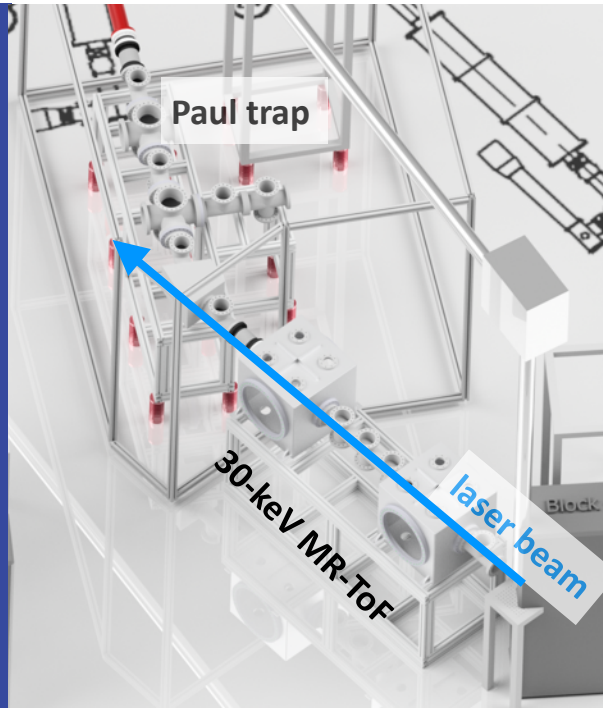


unique linear Paul trap (4 K)

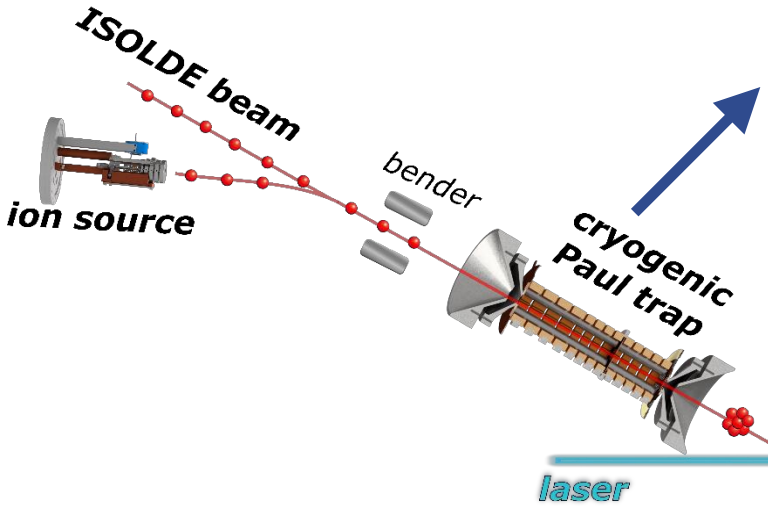
beam cooling to cryogenic temperature

➡ good time focus and energy spread





MIRACLS 30-keV setup



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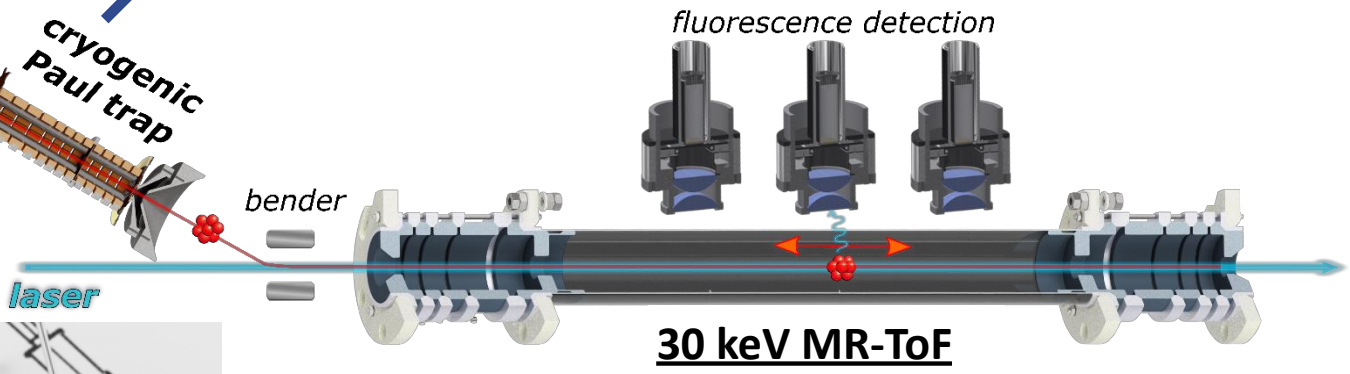
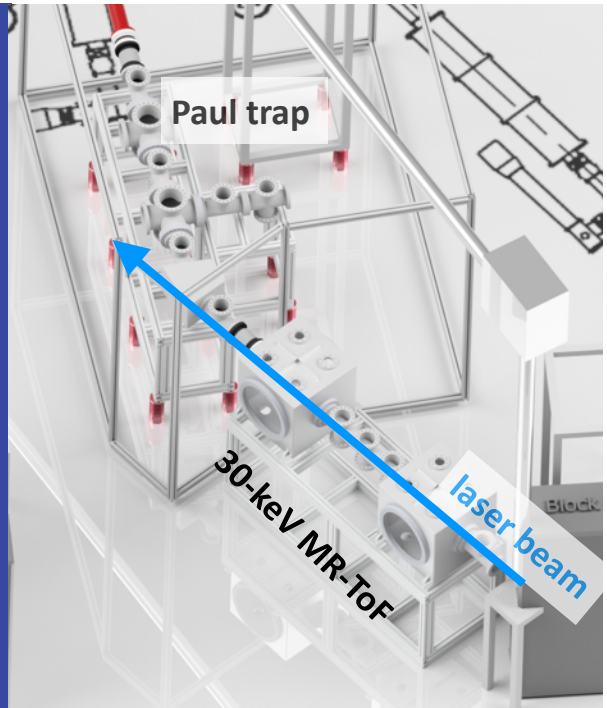


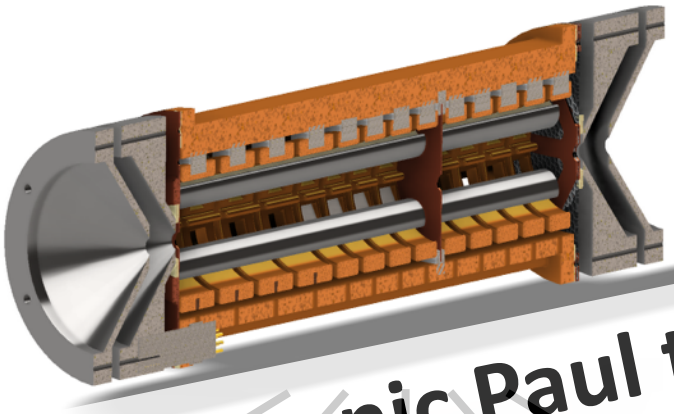


new opportunities

- fast & selective mass separation
  (fundamental physics & applications)
- spectroscopy of negative ions
- excellent emittance (PUMA, emission channeling,…)
- fundamental physics with radioactive molecules
  (e.g. EDM searches)

applications beyond CLS at MIRACLS

cryogenic Paul trap



cryogenic Paul trap

PUMA
antiproton
+
radio-
nuclide

fundamental physics with radioactive molecules

all techniques with high emittance requirements
(e.g. laser and trap experiments)

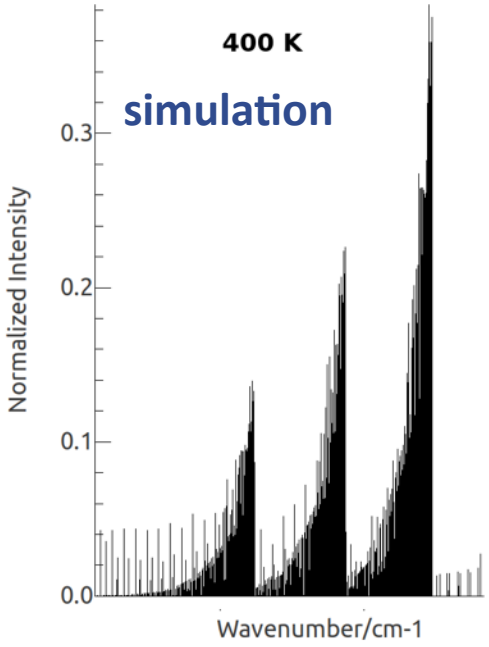
SSP: Emission Channeling

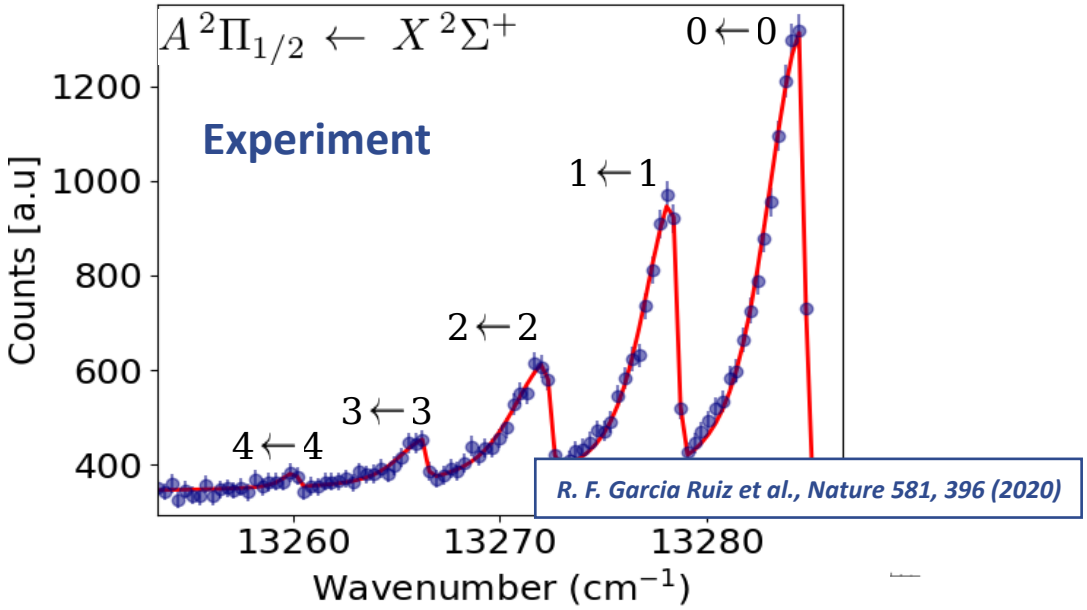
prerequisite for high flux MR-ToF

laser spectroscopy of molecules

- cooling of internal degrees of freedom (especially vibrations)
  - ➡ higher population of the low-lying states
  - ➡ simpler spectra ⇒ more easily identification
- buffer-gas cooling in cryogenic Paul trap:
  - ➡ overall the gain could be more than x100 in scanning time.
  - ➡ enables efficient initial state preparation for later EDM searches

RaF spectra





R. F. Garcia Ruiz et al., Nature 581, 396 (2020)

simulation by R. F. Garcia Ruiz

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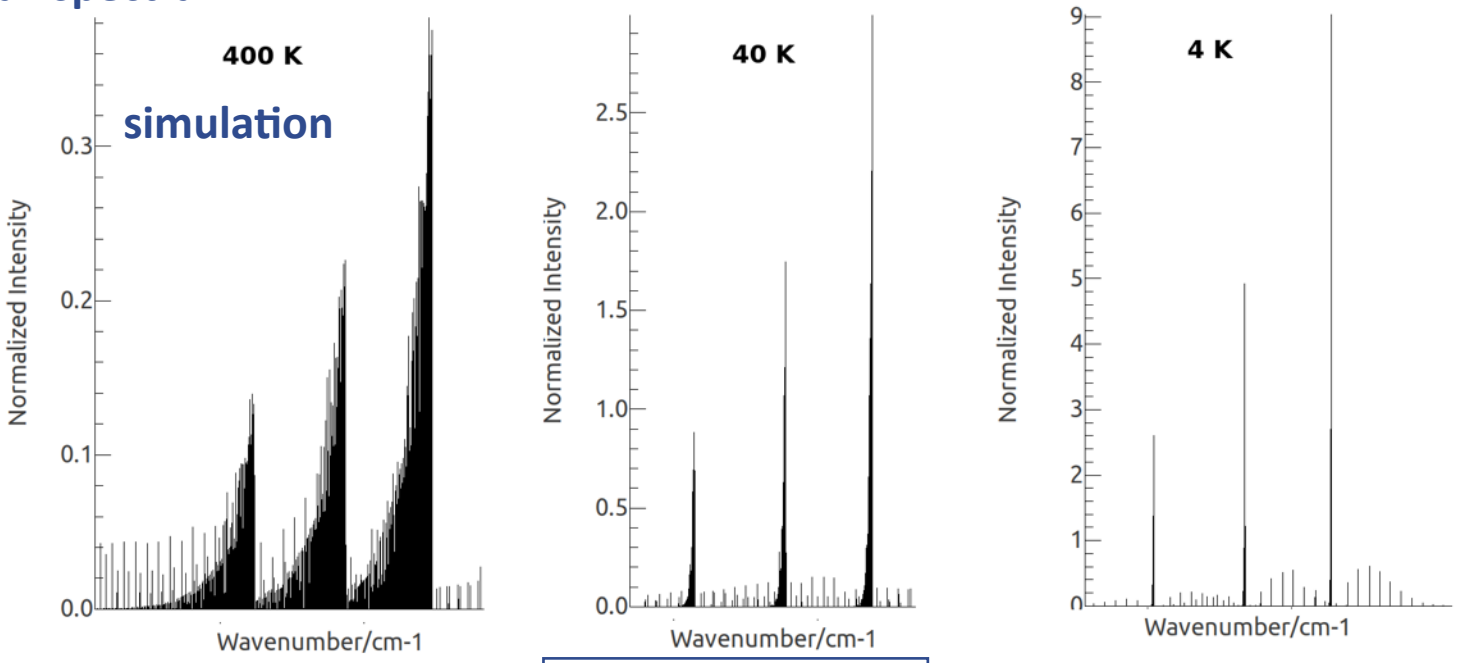
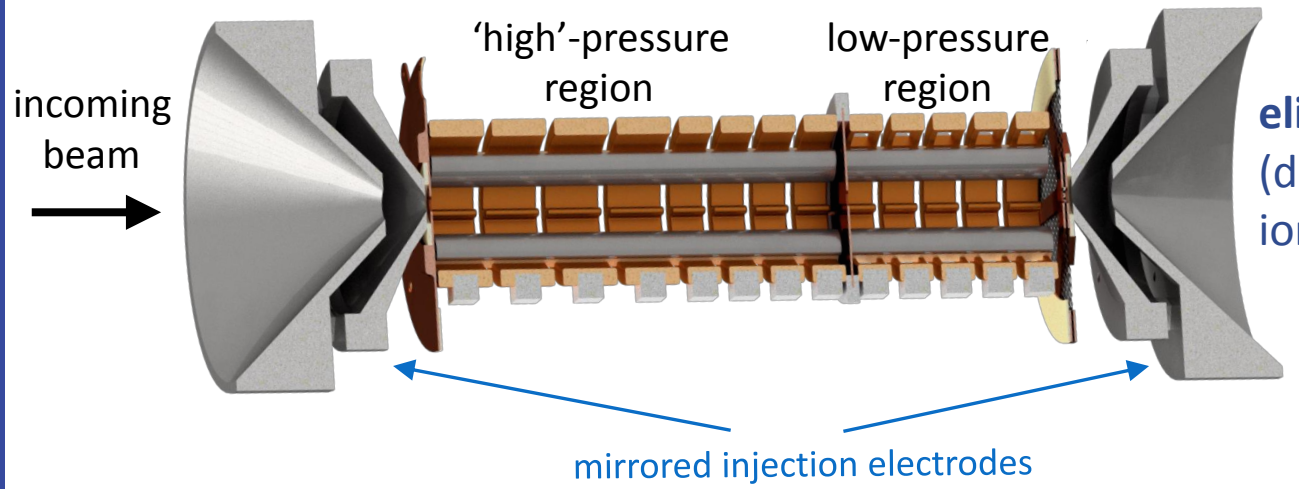
RaF spectra

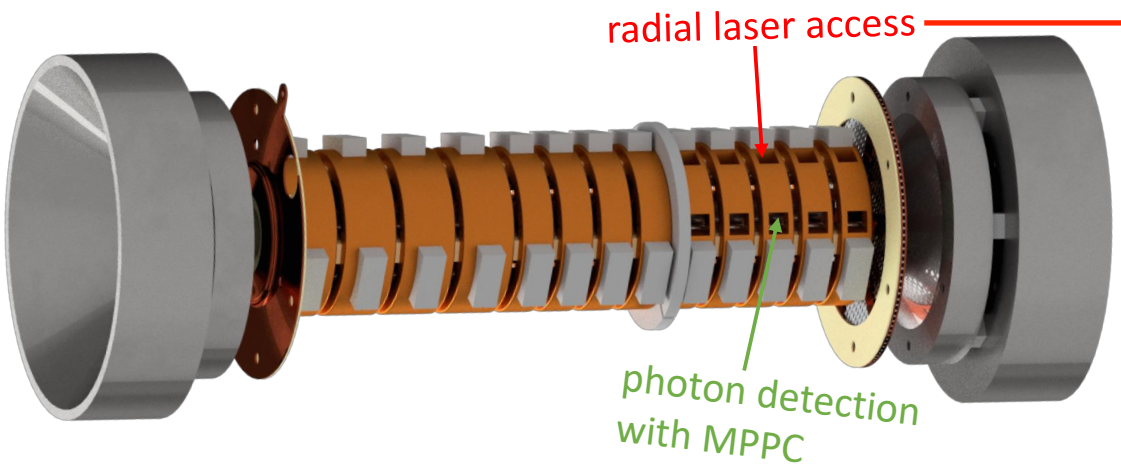


simulation by R. F. Garcia Ruiz

MIRACLS' cryogenic Paul trap



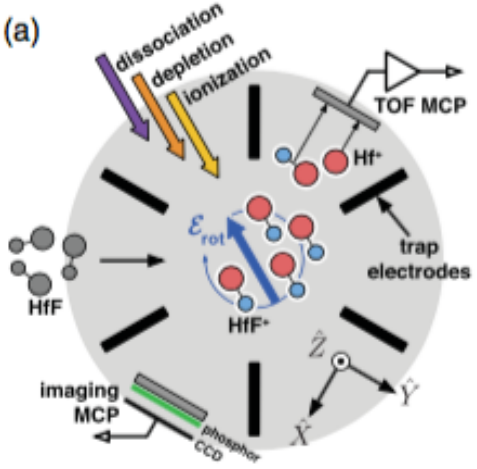
eliminates re-heating (during extraction and ion acceleration)



for state preparations of molecules

ionic (radioactive) molecules

- (stable) $^{180}Hf^{19}F^{+}$



$$\sigma_d \sim \frac{1}{E_{\mathrm{ef}} \tau \sqrt{\dot{N} T}}$$

trapped ionic molecules

W. B. Cairncross et al., PRL 119, 153001 (2017)

- spectroscopy of ionic radioactive molecules at MIRACLS

  ➡ higher population of the low-lying states
  ➡ high experimental sensitivity for trapped ions
  ➡ discussions with theorist to identify best candidates

Summary & Conclusions

- **EDM searches**
  - ➡ implies CP violation ⇒ **matter-antimatter asymmetry in the universe**
  - ➡ molecules sensitive probes for EDM
  - ➡ **radioactive molecules** unexplored but highly sensitive

- MIRACLS
  - ➡ novel approach for highly sensitive CLS
  - ➡ successful proof-of-principle experiment
  - ➡ **radioactive molecules: spectroscopy and towards EDM searches**
    - ➡ cooling of inner degrees of freedom
    - ➡ spectroscopy of ionic molecules in MR-ToF
    - ➡ RIB prepration: excellent emittance
    - ➡ highly selective mass spectrometry for identification of molecules
    - ➡ negative ions (of molecules)

MIRACLS

collaboration:

CERN

UNIVERSITÄT GREIFSWALD
Wissen lockt. Seit 1456

TECHNISCHE UNIVERSITÄT DARMSTADT

team members:

P. Fischer, **V. Lagaki, S. Lechner, F. Maier, P. Plattner**,
H. Heylen, M. Rosenbusch, **S. Sels**, F. Wienholtz, **M. Vilen**,
R. Wolf, W. Nörtershäuser, L. Schweikhard,
S. Malbrunot- Ettenauer (Spokesperson)

MIRACLS Alumni:
F. Hummer (2019), **L. M. Bartels** (2018),
F. Maier (2018), **L. Fischer** (2017)
F. Stabel (2017), **S. Sailer** (2017)

students under my supervision
PhD students
MSc students
BSc students

my post-docs

funding:

erc European Research Council

ATTRACT

CERN **Medical Applications Funds**

https://miracls.web.cern.ch

ISOLDE