

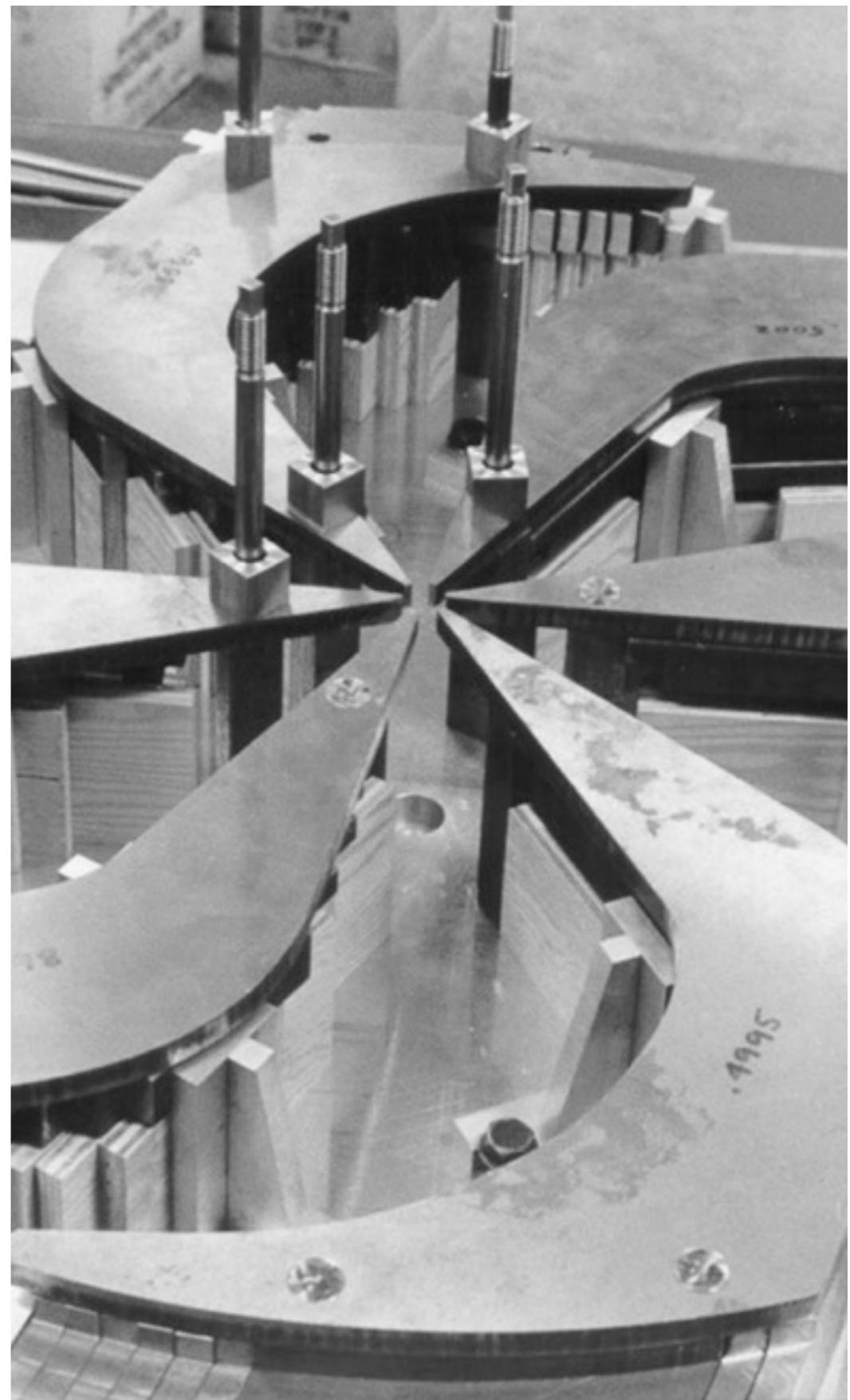


# Radioactive Molecules

## Novel Probes for New Physics

Stephan Malbrunot-Ettenauer  
TRIUMF, University of Toronto

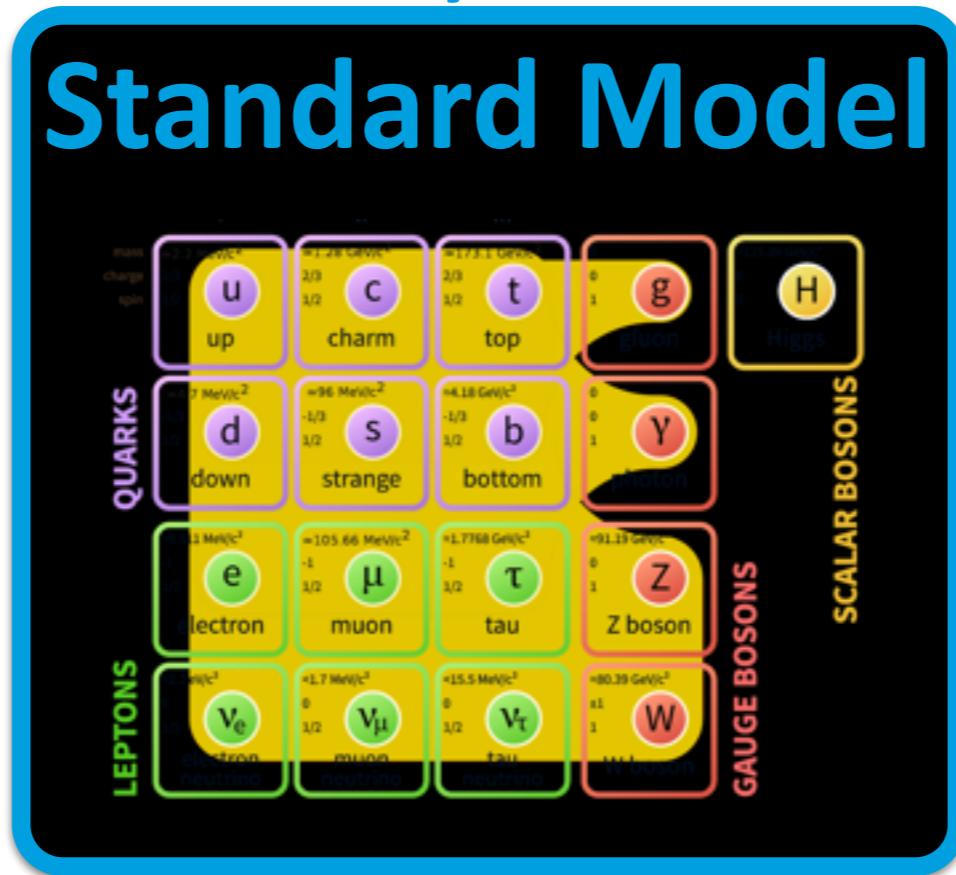
WNPPC 2022



# Fundamental symmetries

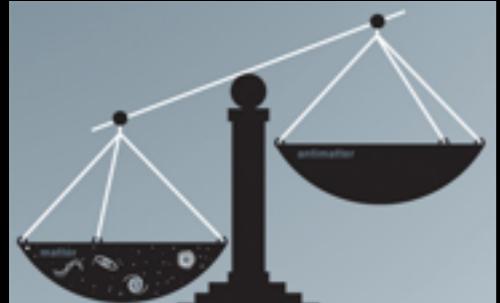
2

incredibly successful



# Fundamental symmetries

matter-antimatter asymmetry

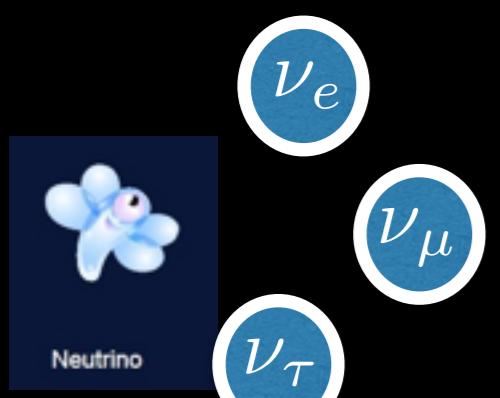


incredibly successful

## Standard Model

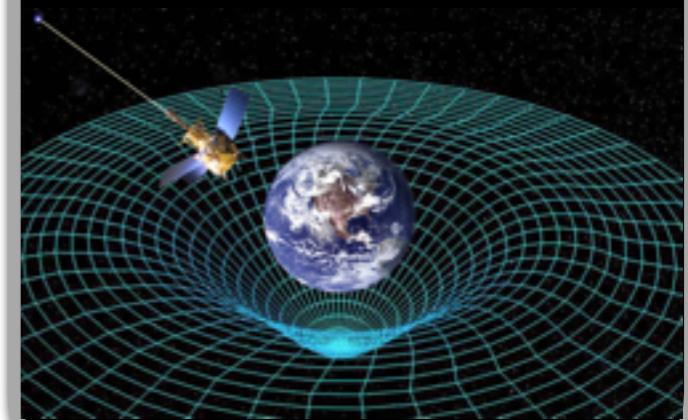
QUARKS		GAUGE BOSONS		SCALAR BOSONS	
u	c	t	g	H	
down	strange	bottom	gamma		
electron	muon	tau	Z boson		
$\nu_e$	$\nu_\mu$	$\nu_\tau$	W boson		

origin of neutrino masses

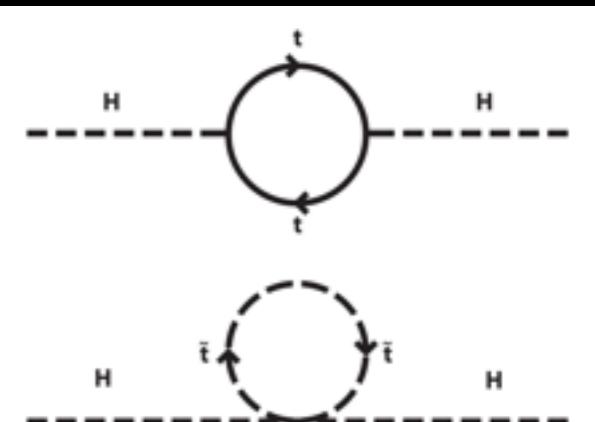


... yet incomplete

misses gravity



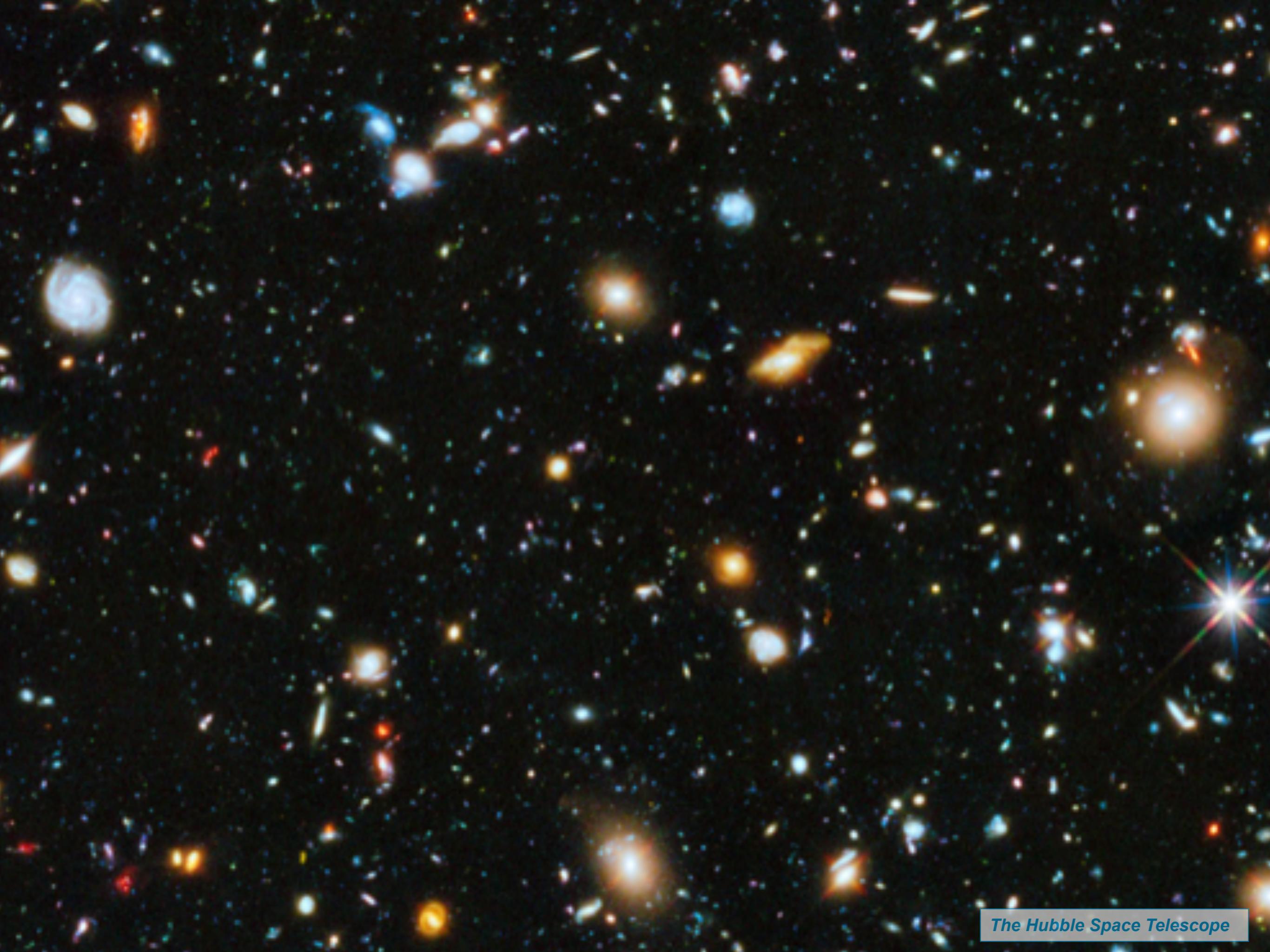
hierarchy problem



arbitrary constants:

$m_e, m_\mu, m_\tau, m_u, m_d, m_s, m_c, m_b, m_t, m_H,$   
 $\theta_{12}, \theta_{13}, \theta_{23}, \delta, g_1, g_2, g_3, \theta_{QCD}, v$

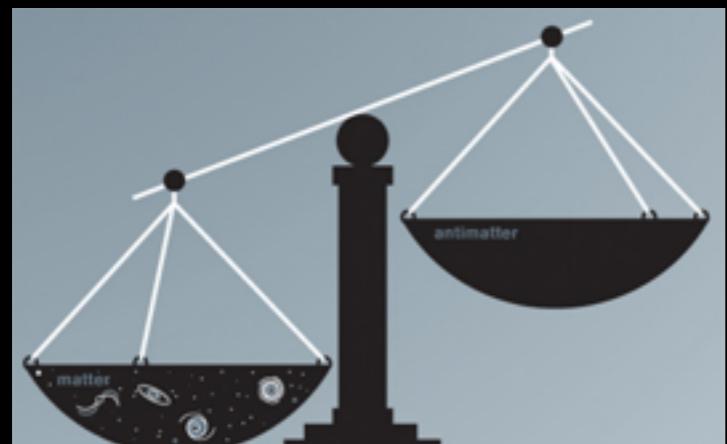
?  $m_{\nu_e}, m_{\nu_\mu}, m_{\nu_\tau}$   
?  $\theta_{12}, \theta_{13}, \theta_{23}, \alpha_1, \alpha_2$  ?



The Hubble Space Telescope

# where is all the antimatter?

Macrocosmos



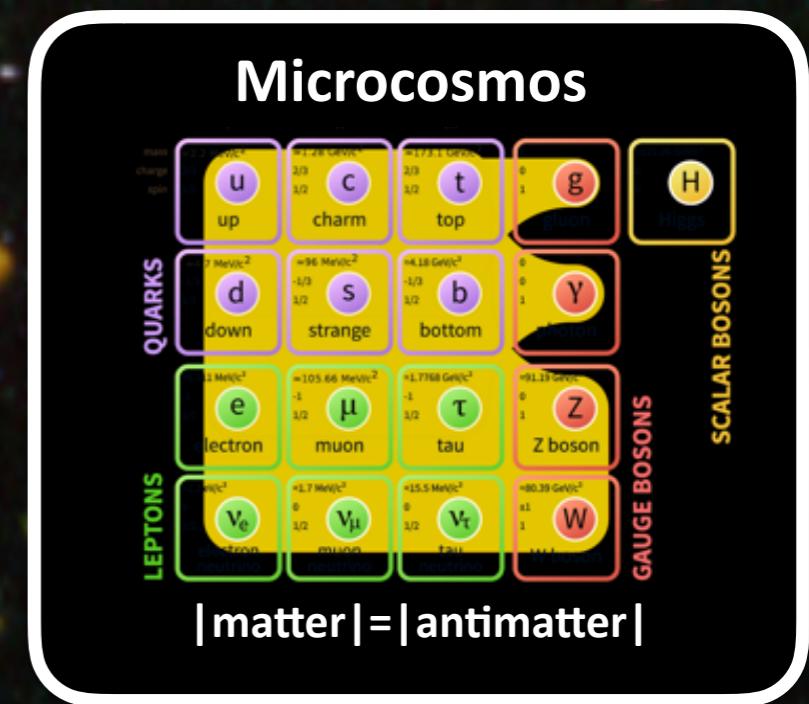
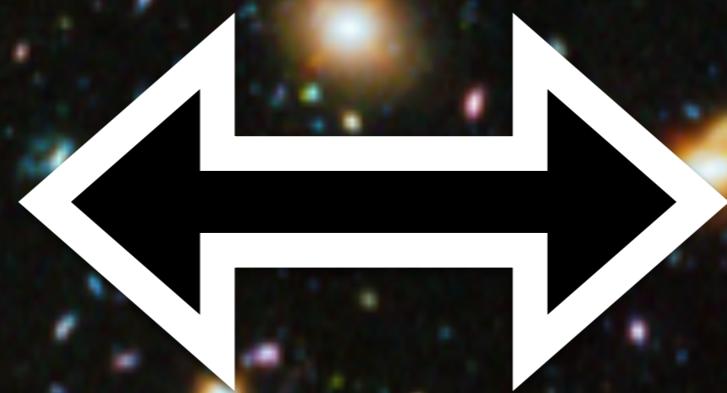
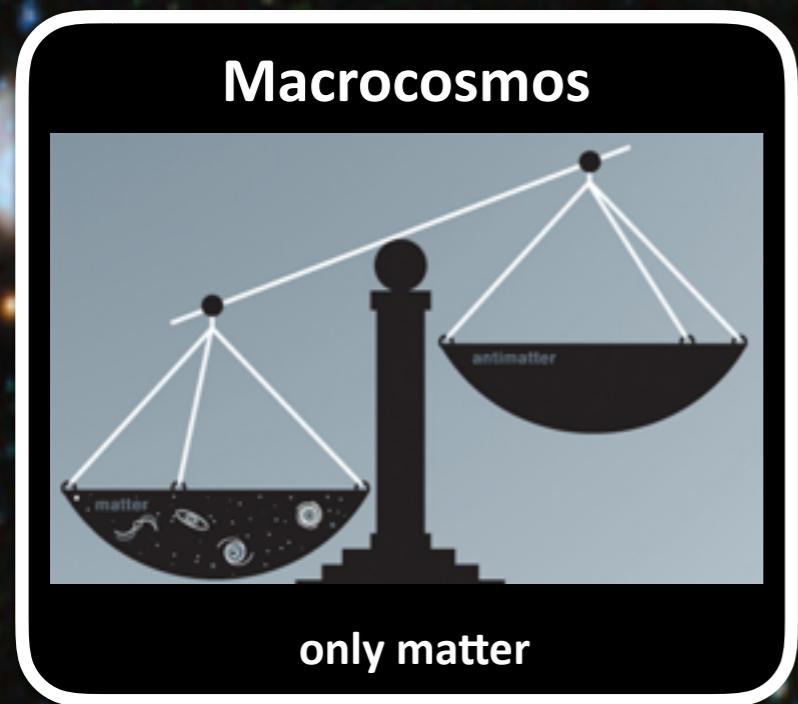
only matter

Microcosmos

mass charge spin	u	c	t	g	H
d	down	s	b	γ	W
e	electron	μ	τ	Z boson	GAUGE BOSONS
ν <sub>e</sub>	electron neutrino	ν <sub>μ</sub>	ν <sub>τ</sub>		
ν <sub>μ</sub>					
ν <sub>τ</sub>					

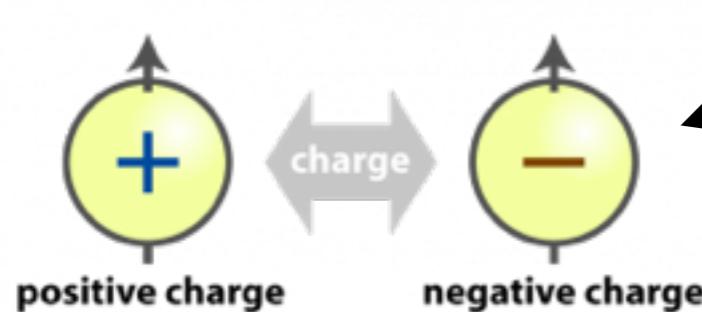
|matter|=|antimatter|

# where is all the antimatter?

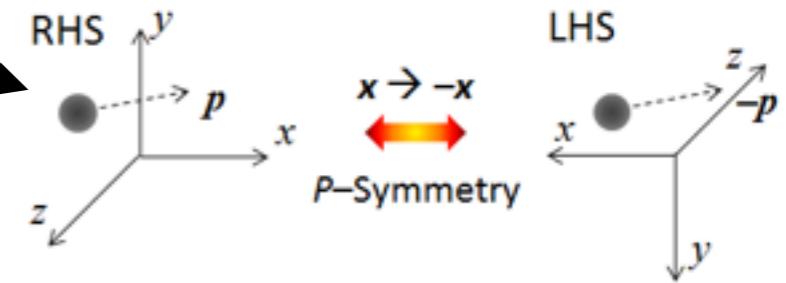


Sakharov, 1967

ingredient to resolve universe's matter-antimatter

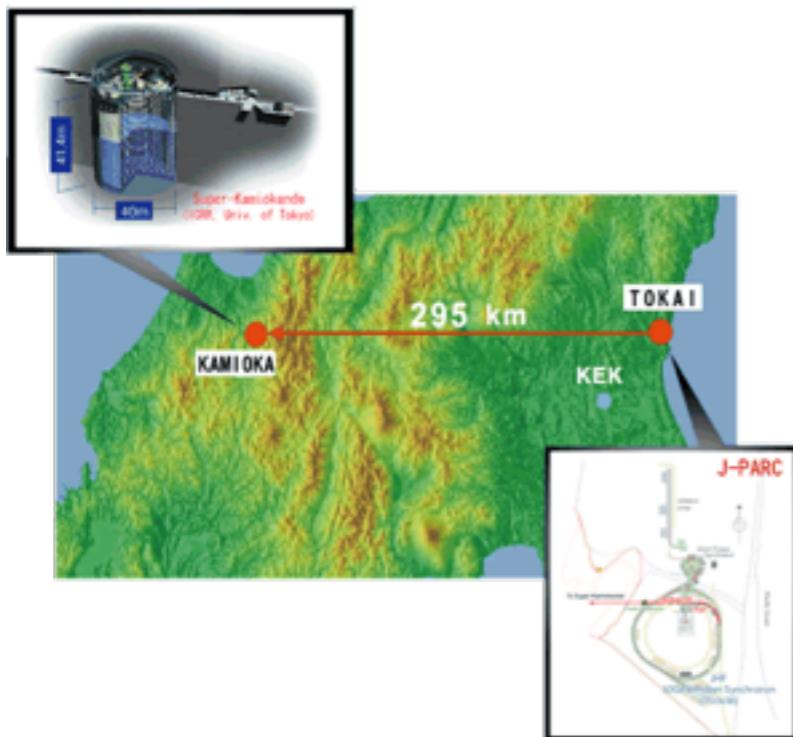


# CP violation

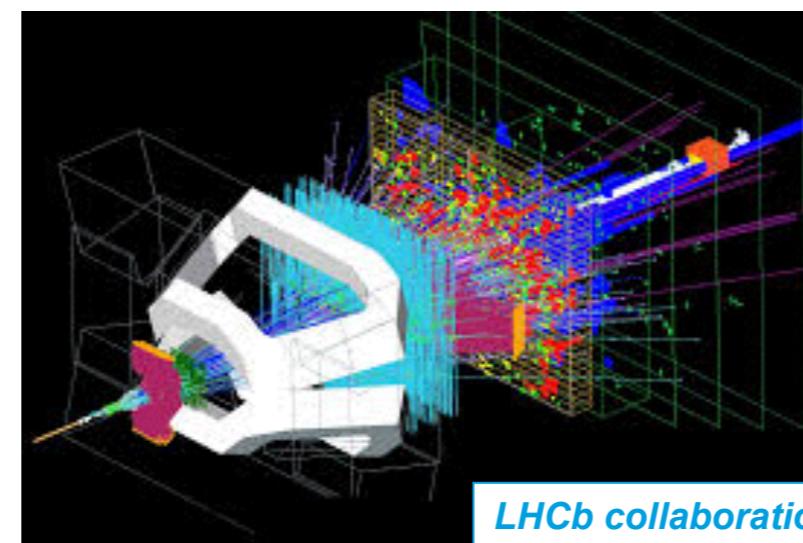
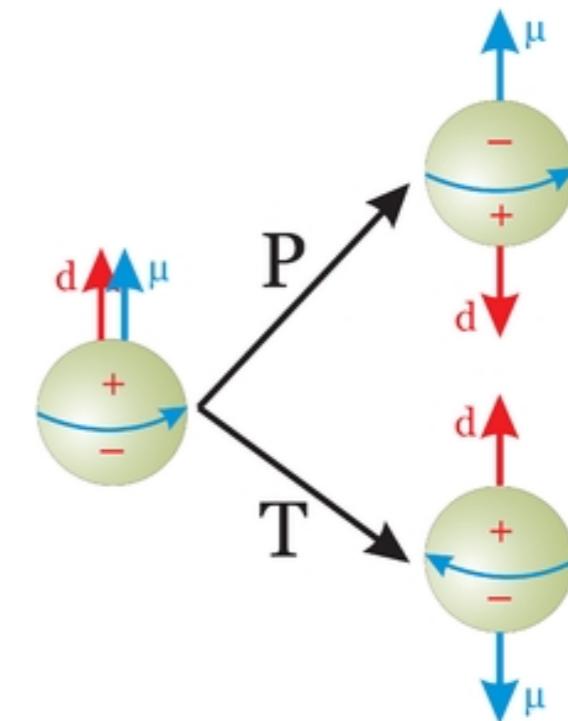


# Searches for CP violation

T2K collaboration, *Nature* 580, 339 (2020)



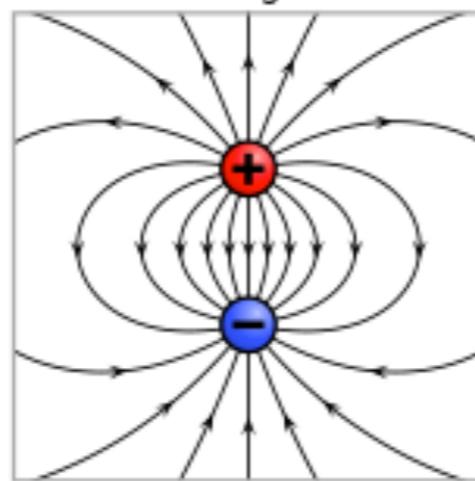
ACME collaboration, *Nature* 562, 355 (2018)  
C. Abel et al., *PRL* 124, 081803 (2020)



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

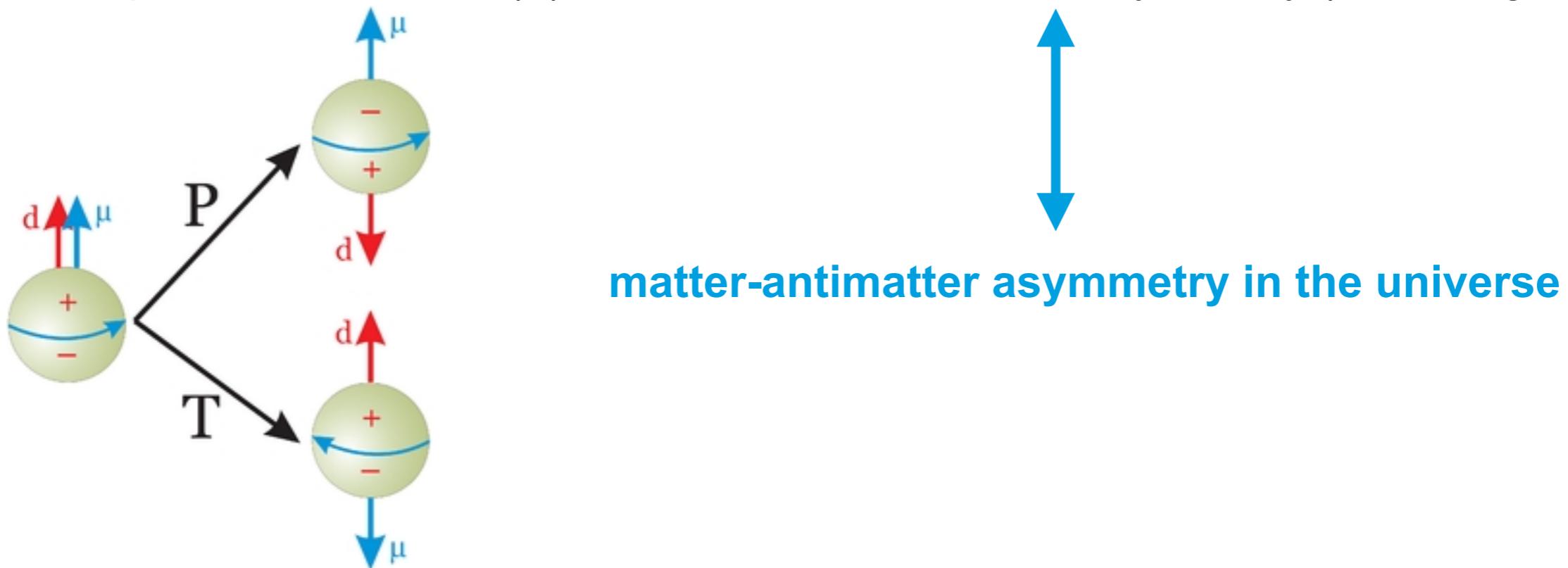
# Permanent electric dipole moment

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

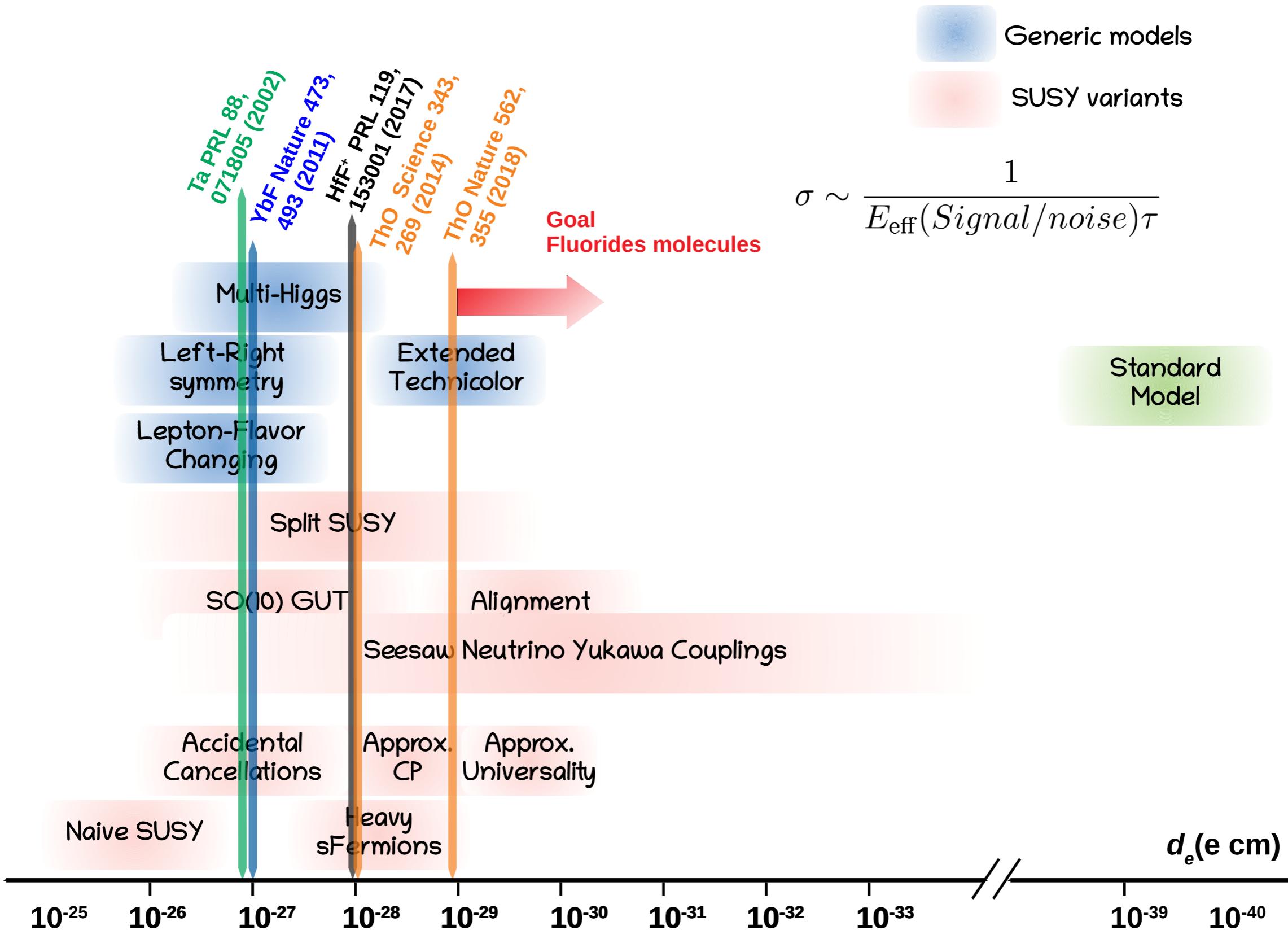


5

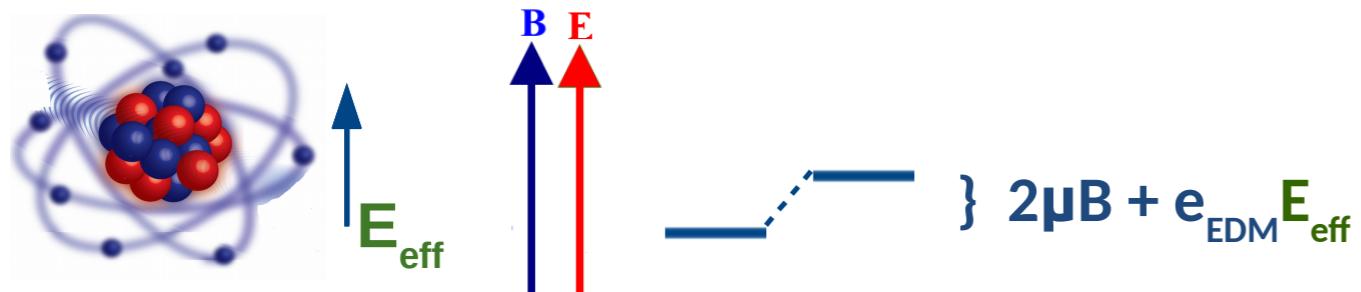
- implies time-reversal (T) violation  $\Rightarrow$  violation of CP symmetry (assuming CPT)



# Searches for an electron EDM

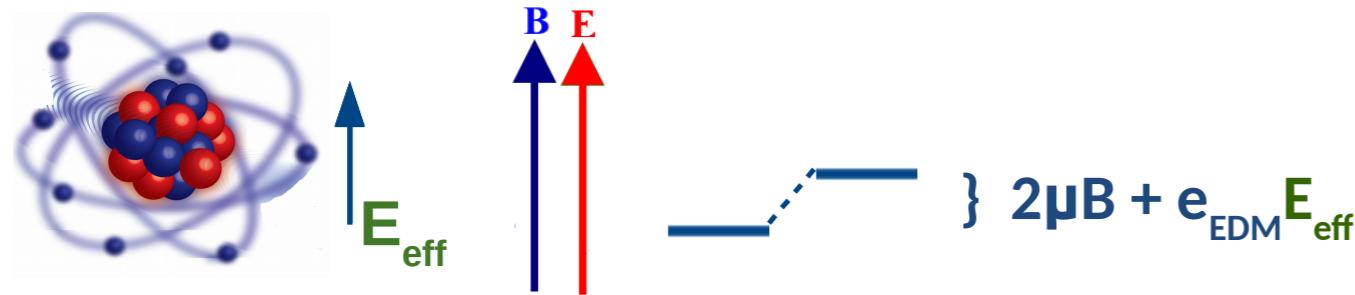


# EDM searches



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

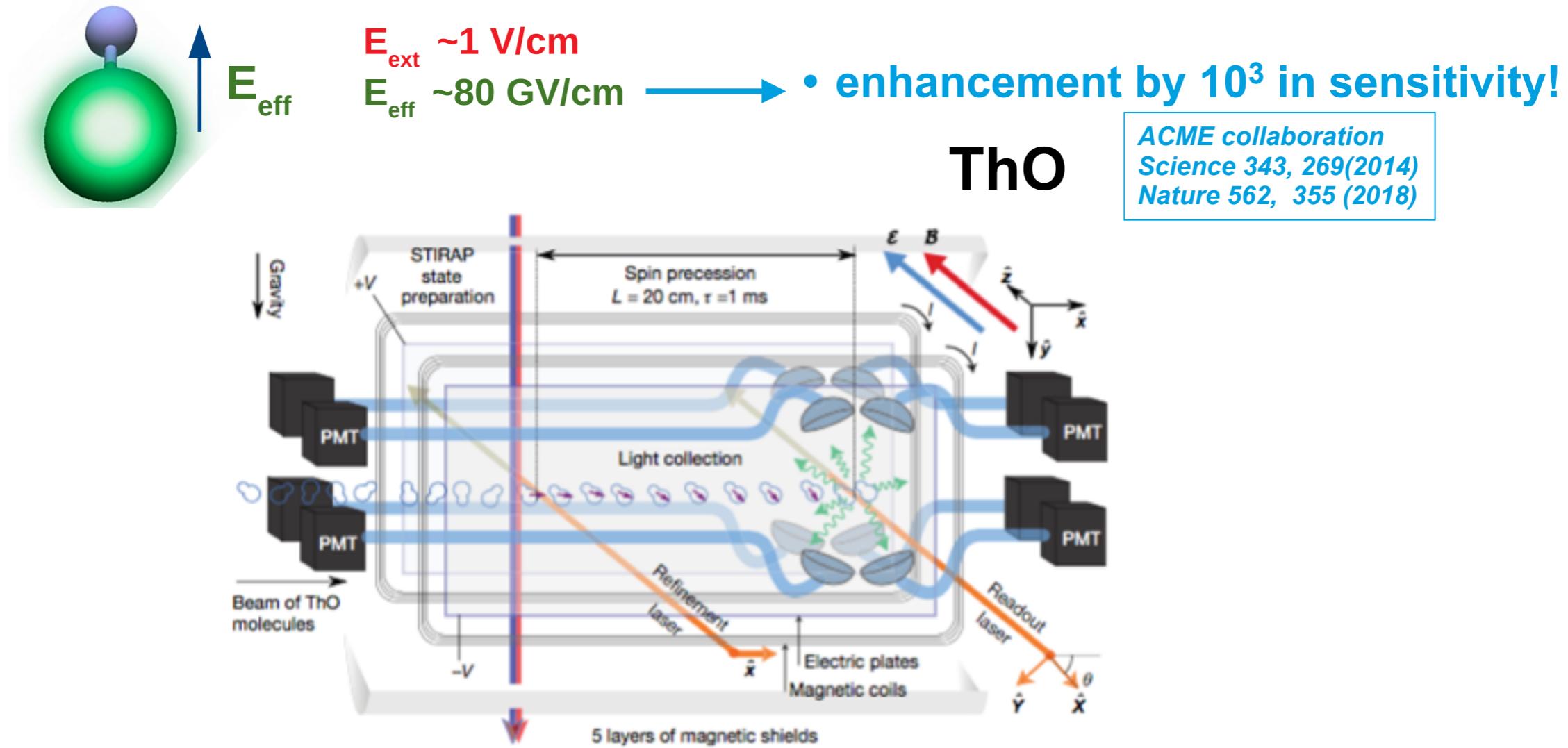
# EDM searches



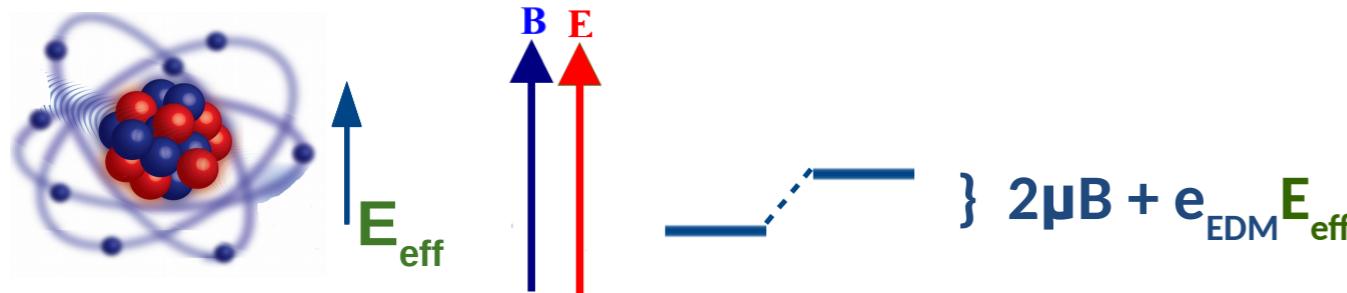
$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{\dot{N} T}}$$

7

## Molecules:



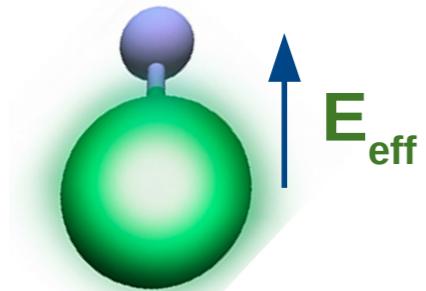
# EDM searches



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

7

## Molecules:



$$\begin{aligned} E_{\text{ext}} &\sim 1 \text{ V/cm} \\ E_{\text{eff}} &\sim 80 \text{ GV/cm} \end{aligned}$$

- enhancement by  $10^3$  in sensitivity!

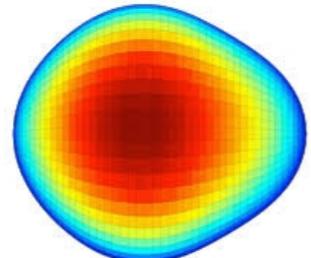
ThO

ACME collaboration  
Science 343, 269(2014)  
Nature 562, 355 (2018)

- scales as  $Z^2 R(Z)$

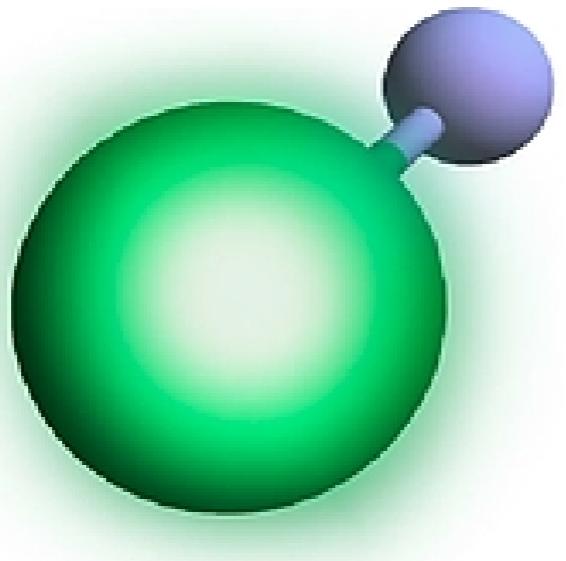
⇒ opportunity for radioactive molecules

Schiff moment



nuclear-spin-dependent component

# ‘Designer Molecules’



8

# Table of Elements

## 80 chemical elements (with stable nuclides)

<b>H</b>	Hydrogen	He	Helium
<b>Li</b>	Beryllium	<b>Be</b>	Neon
Lithium		Boron	
<b>Na</b>	Magnesium	<b>C</b>	Nitrogen
Sodium		Carbon	Oxygen
<b>K</b>	Calcium	<b>N</b>	Fluorine
Potassium		Nitrogen	
<b>Rb</b>	Sr	<b>O</b>	Neon
Rubidium	Sr	Oxygen	
<b>Cs</b>	Ba	<b>F</b>	Ar
Ceasium	Barium	Fluorine	Argon
<b>Fr</b>	Radium	<b>Ne</b>	Kr
Francium	Radium	Neon	Krypton
		<b>Br</b>	Xe
		Bromine	Xenon
		<b>In</b>	I
		Inert Gas	Iodine
		<b>Sn</b>	
		Tin	
		<b>Sb</b>	
		Antimony	
		<b>Te</b>	
		Tellurium	
		<b>I</b>	
		Iodine	
		<b>Po</b>	
		Poisonous	
		<b>At</b>	
		Astatine	
		<b>Rn</b>	
		Radon	
		<b>Og</b>	
		Oganesson	

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
Lanthanum	Cerium	Praseodymium	Nd	Neodymium	Sm	Samarium	Europium	Gadolinium	Terbium	Dysprosium	Holmium	Erbium	Thulium	Ytterbium	Lutetium
89	90	91	92	93	94	95	96	97	98	99	100	101	102	103	104
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	
Actinium	Thorium	Protactinium	Uranium	Neptunium	Plutonium	Americium	Curium	Berkelium	Californium	Einsteinium	Fermium	Mendelevium	Neobrium	Robotium	Lanthanum

# 'Designer Molecules'

Table of Isotopes

252 stable

≈90 naturally occurring radioisotopes

≈3000 short-lived radionuclides discovered

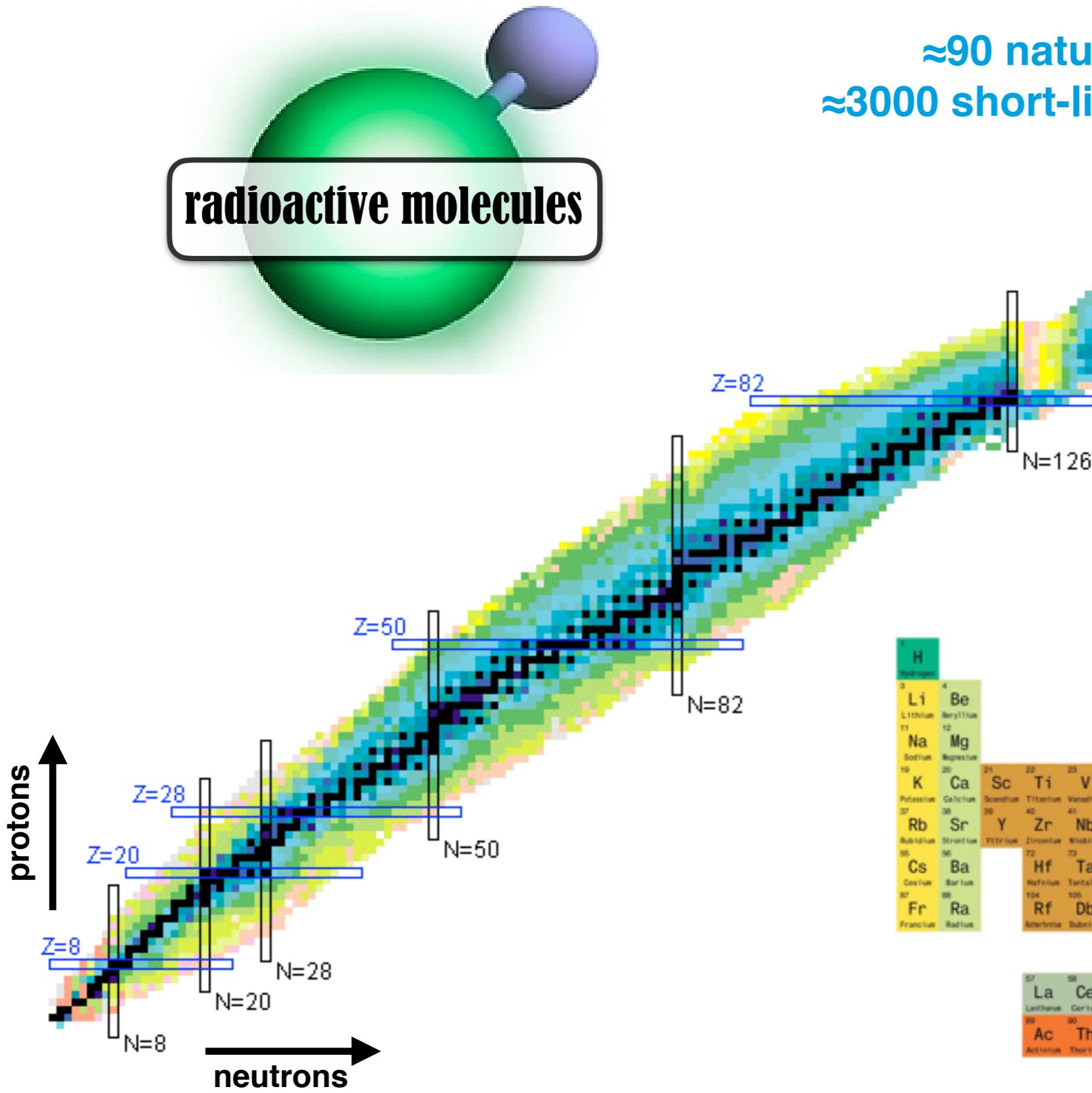


Table of Elements  
80 chemical elements  
(with stable nuclides)

H	He
Hydrogen	Helium
3 Li	4 Be
Lithium	Beryllium
11 Na	12 Mg
Sodium	Magnesium
19 K	20 Ca
Potassium	Calcium
37 Rb	38 Sr
Rubidium	Srtrontium
55 Cs	56 Ba
Cesium	Barium
87 Fr	88 Ra
Francium	Radium
5 B	6 C
Boron	Carbon
13 Al	14 Si
Aluminum	Silicon
21 Ga	22 Ge
Gallium	Germanium
31 In	32 Sn
Indium	Stannum
49 Tl	50 Pb
Thallium	Pbolidium
6 Ne	7 N
Neon	Nitrogen
10 Ar	11 O
Argon	Oxygen
17 Cl	18 F
Chlorine	Fluorine
35 Br	36 Kr
Bromine	Krypton
53 I	54 Xe
Iodine	Xenon
85 At	86 Rn
Astatine	Radon
95 Ts	96 Og
Tenesseine	Oganesson

57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
Actinium	Cerium	Praseodymium	Neodymium	Protactinium	Samarium	Europeum	Sodium	Terbium	Dysprosium	Holmium	Erbium	Thulium	Rutherfordium	Lanthanum
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
Thorium	Protactinium	Shankium	Neptunium	Plutonium	Americium	Curium	Berkelium	Cfcerium	Esmeralda	Fermium	Molecula	Nobellium	Lanthan	

# 'Designer Molecules'

Table of Isotopes

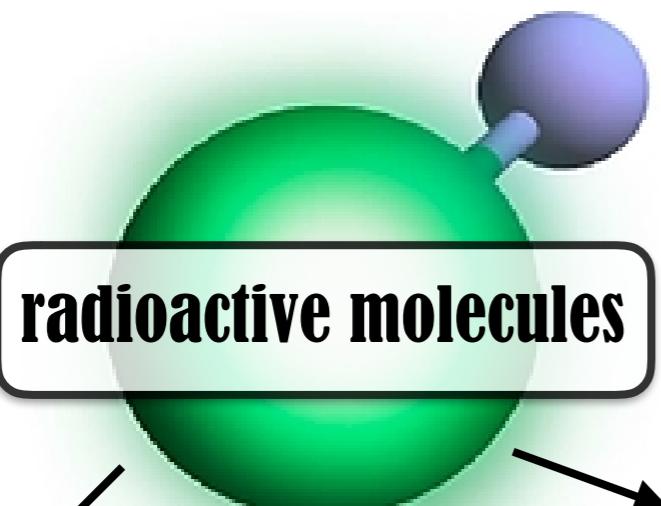
252 stable

≈90 naturally occurring radioisotopes

≈3000 short-lived radionuclides discovered

8

probes for  
new physics  
• EDM searches  
• P violation



Astrophysics

Atomic, molecular,  
optical physics

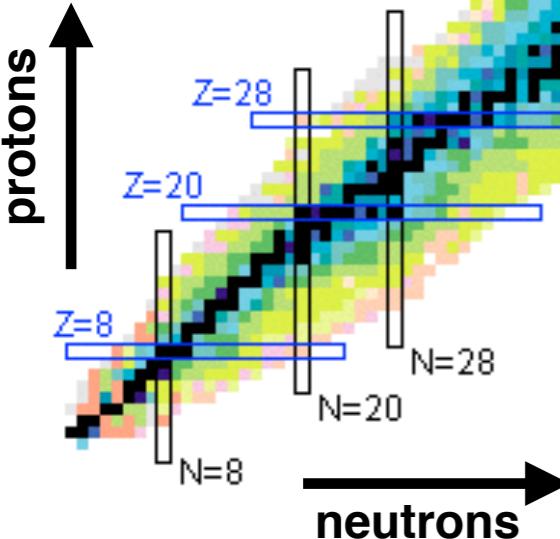
R. F. Garcia Ruiz et al., Nature 581, 396 (2020)  
S. M. Udrescu, et al. Phys. Rev. Lett. 127, 033001 (2021)  
Fan et al., Phys. Rev. Lett. 126, 023002 (2021)

Applied science  
• nuclear engineering  
• medicine

Quantum  
Chemistry

Nuclear physics

⇒ many, exciting science opportunities

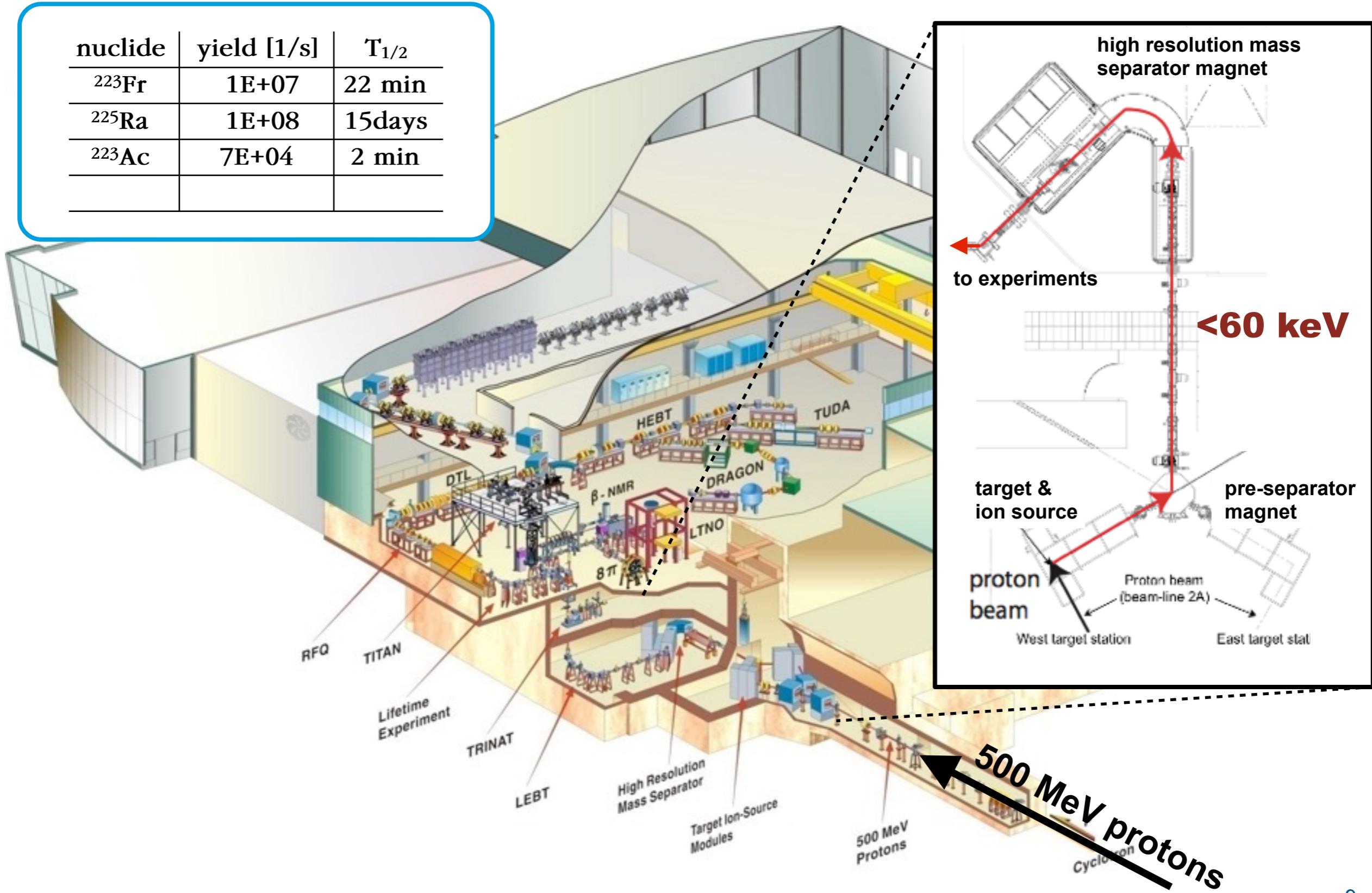
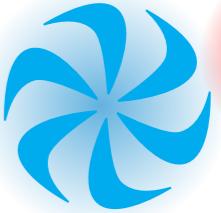


H	Hydrogen
Li	Lithium
Be	Beryllium
Na	Sodium
Mg	Magnesium
K	Potassium
Ca	Calcium
Sc	Scandium
Ti	Titanium
V	Vanadium
Cr	Chromium
Mn	Manganese
Fe	Iron
Co	Cobalt
Ni	Nickel
Cu	Copper
Zn	Zinc
Rb	Rubidium
Sr	Stron튬
Y	Yttrium
Zr	Zirconium
Nb	Niobium
Mo	Molybdenum
Tc	Technetium
Ru	Ruthenium
Rh	Rhenium
Pd	Palladium
Ag	Argentium
Cd	Cadmium
Cs	Cesium
Ba	Barium
Hf	Hafnium
Ta	Tantalum
W	Tungsten
Re	Rhenium
Os	Osmium
Ir	Iridium
Pt	Platinum
Au	Gold
Hg	Mercury
Rf	Rutherfordium
Db	Dubnium
Sg	Singapore
Bh	Berillium
Hs	Hassium
Mt	Mithium
Ds	Destathium
Rg	Rutherfordium
Gn	Gopernium

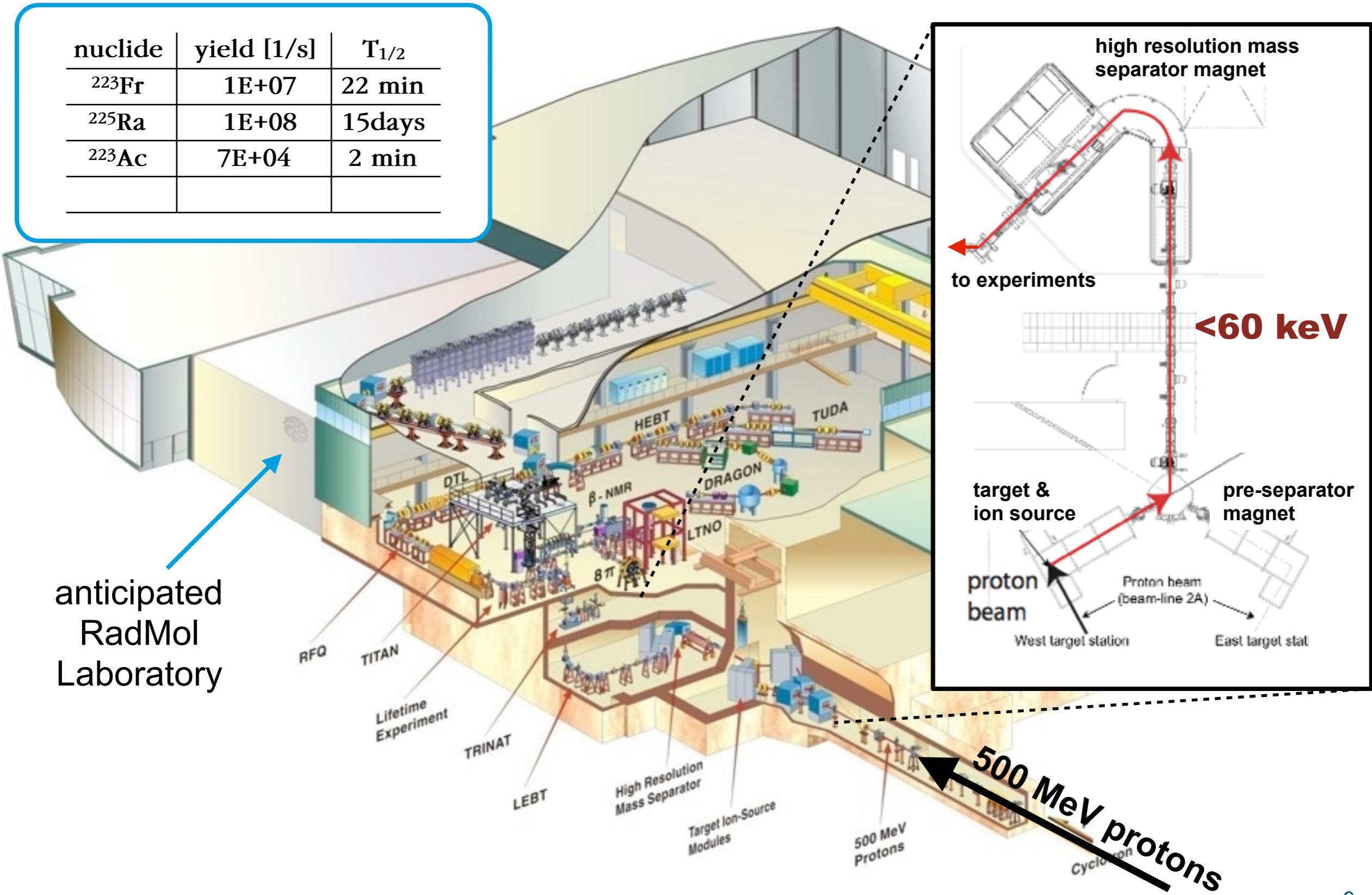
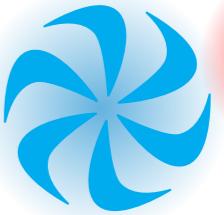
He	Helium
Ne	Neon
Ar	Argon
Kr	Krypton
Xe	Xenon
Br	Broine
I	Iodine
At	Atatine
Rn	Radon
Fr	Flerovium
Mc	Meitnerium
Lv	Liverso
Ts	Ts
Og	Oganesson
B	Boron
C	Carbon
N	Nitrogen
O	Oxygen
P	Phosphorus
S	Sulfur
Al	Aluminum
Si	Silicon
Ge	Germanium
As	Arseenic
Se	Selenium
In	Indiium
Sn	Stannium
Sb	Sbium
Te	Te
Bi	Bi
Po	Po
At	Atatine
Rn	Radon
Th	Thorium
Pa	Protactinium
U	Uranium
Np	Neptunium
Pu	Plutonium
Am	Americium
Cm	Cerium
Bk	Berkellium
Cf	Celferrium
Es	Einsteinium
Fm	Fermium
Md	Mendelevium
No	Nobelium
Lr	Lanthan

La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

# Radioactive Ions



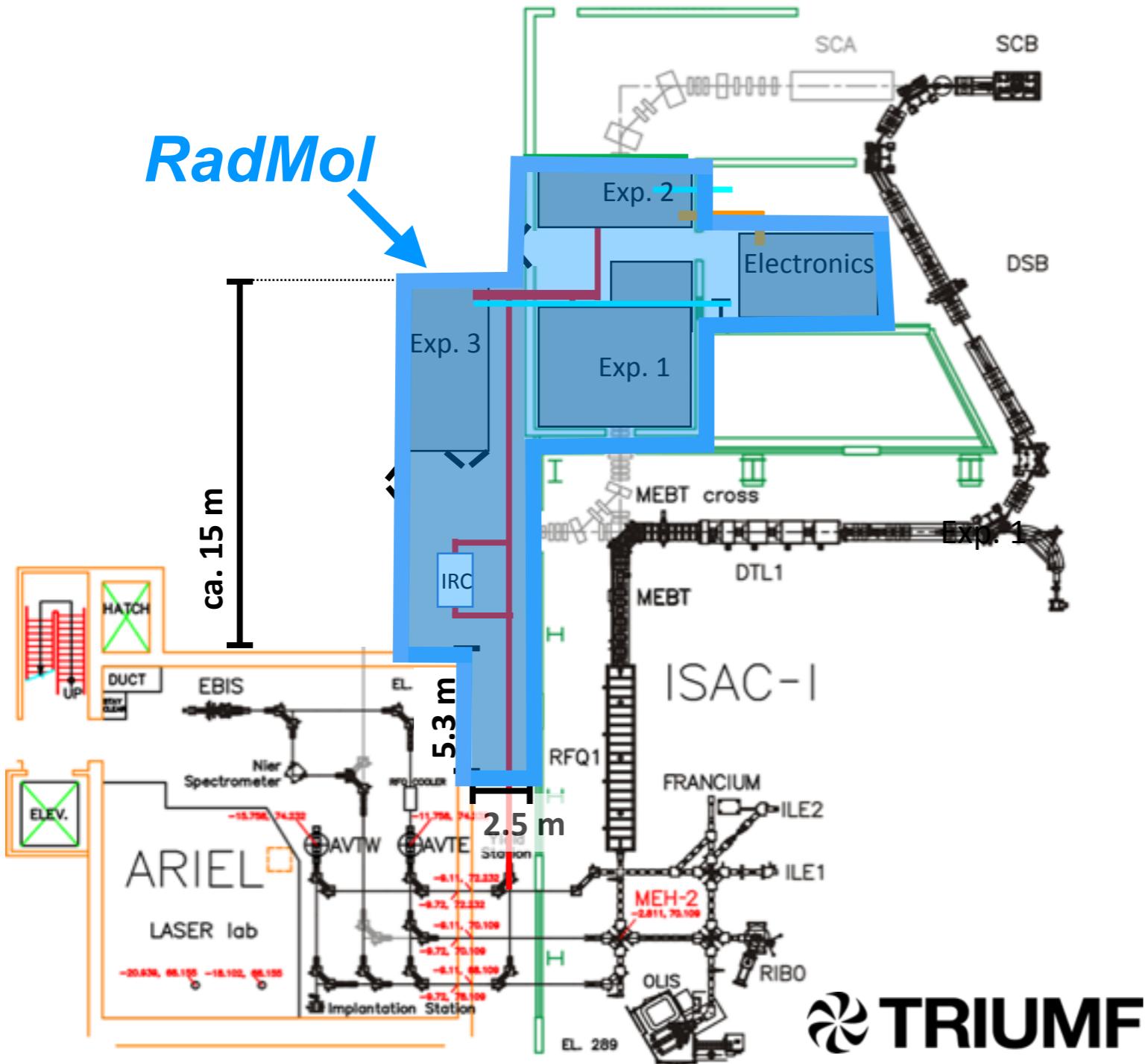
# Radioactive Ions



# RadMol



*a radioactive molecule lab for fundamental physics*



## Goals:

- world-wide unique laboratory for radioactive molecules
- precision studies for searches for new physics

## TRIUMF advantages

- large variety in radioactive ion beams (RIB)
- high beamtime availability (3 independent RIBs)
- existing laboratory space for large, multi-station program
- fast connection of RadMol lab to online facility

# Multidisciplinary

BSM interpretation

quantum chemistry

particle physics

molecular physics

nuclear theory

radioactive molecules

EDM expertise

RIB facility

atomic physics techniques

RIB techniques



UNIKASSEL  
VERSITÄT



McGill



uOttawa

MIT  
Massachusetts  
Institute of  
Technology

Philipps



Universität  
Marburg

OAK RIDGE  
National Laboratory

UNIVERSITY OF  
TORONTO

KU LEUVEN



University  
of Manitoba



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11



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VERSITÄT



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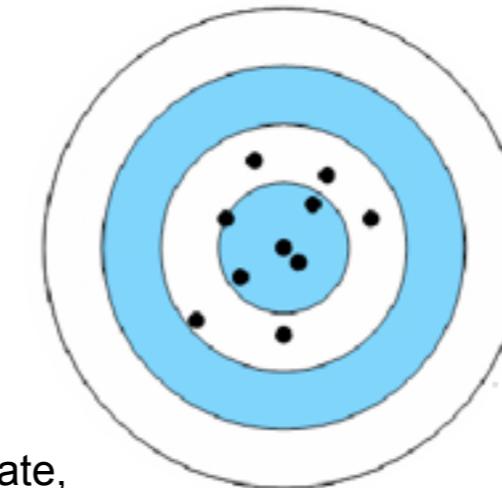
University  
of Manitoba



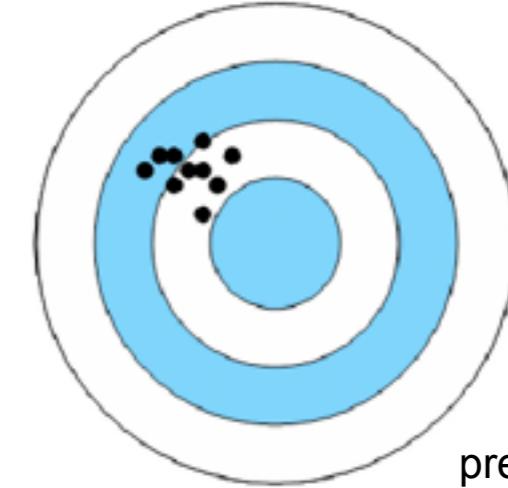
# Atomic physics techniques at RIB facilities

high precision and accuracy

K. Blaum, et al., *Phys. Scr. T152*, 014017 (2013)  
P. Campbell et al., *Prog. Part. and Nucl. Phys.* 86, 127-180 (2016)  
J. Dilling et al., *Annu. Rev. Nucl. Part. Sci.* 68, 45 (2018)



accurate,  
but not precise



precise,  
but not accurate

12

## ion traps

- masses
- RIB preparations
- mass separation
- in-trap decay

## laser spectroscopy

- hyperfine structure
- isotope shifts
- optical pumping

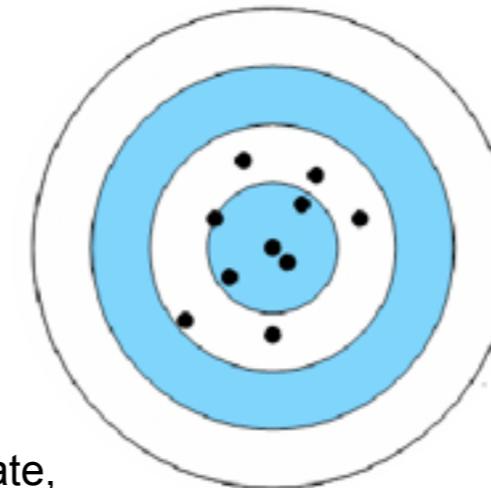
## atom traps

- in-trap decay
- laser spectroscopy
- APV

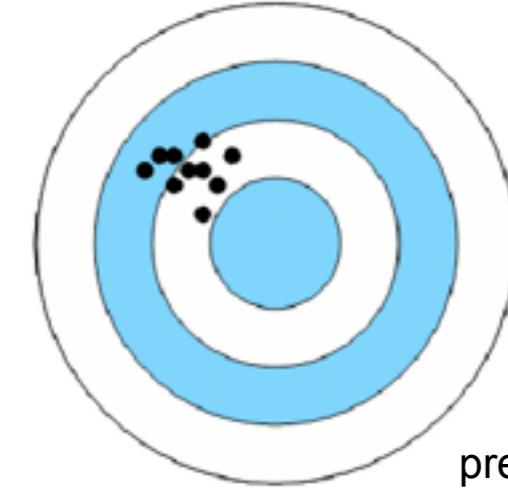
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### atom traps

- in-trap decay
- laser spectroscopy
- APV

#### short half-lives

$T_{1/2} < 10 \text{ ms}$   
 $(\Delta m/m = 6 \cdot 10^{-8})$

M. Smith et al., *PRL* 101, 202501 (2008)

#### low intensity

masses: 0.5 ions / h

M. Block et al., *Nature* 463, 785 (2010)  
E. Minaya Ramirez et al., *Science* 337, 1207 (2012)

#### Challenges

#### temperature

buffer gas cooling  
(selected cases of  
laser cooling)

#### purity

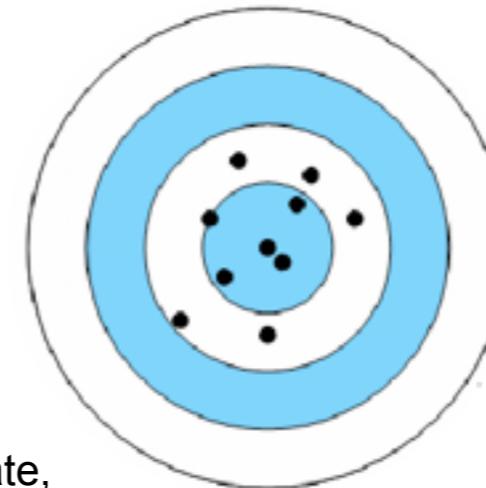
$R=m/\Delta m > 5 \cdot 10^6$   
limited ion capacity

S. Eliseev et al., *PRL* 110, 082501 (2013)

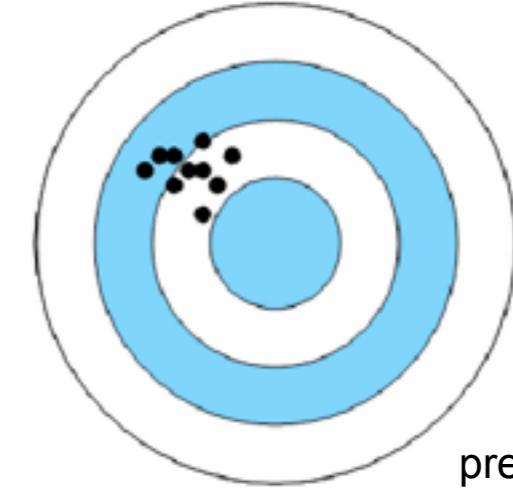
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## atom traps

- in-trap decay
- laser spectroscopy
- APV

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E. Minaya Ramirez et al., *Science* 337, 1207 (2012)

### temperature

buffer gas cooling  
(selected cases of  
laser cooling)

300 K

### purity

$R = m/\Delta m > 5 \cdot 10^6$   
limited ion capacity

S. Eliseev et al., *PRL* 110, 082501 (2013)

$\mu\text{K} - \text{mK} - \text{K}$

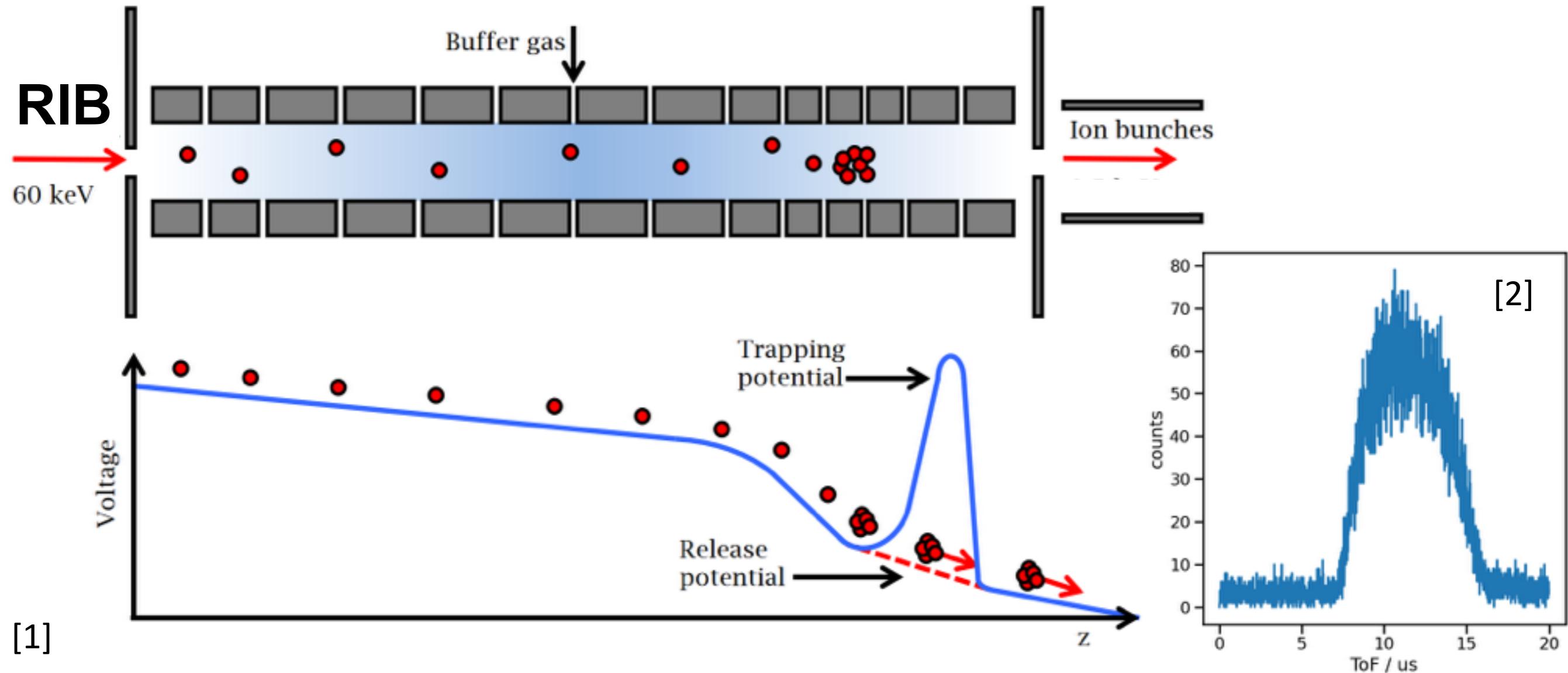


# Standard buffer gas cooling

cooler and bunchers at RIB facilities , operated at 300 K buffer gas

Cooling limit: 300 K

13

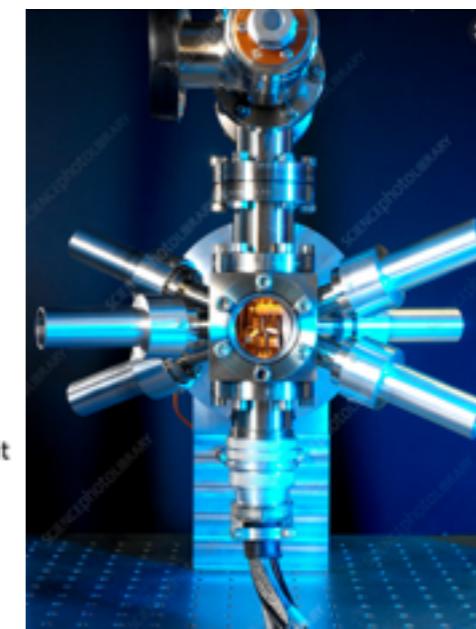
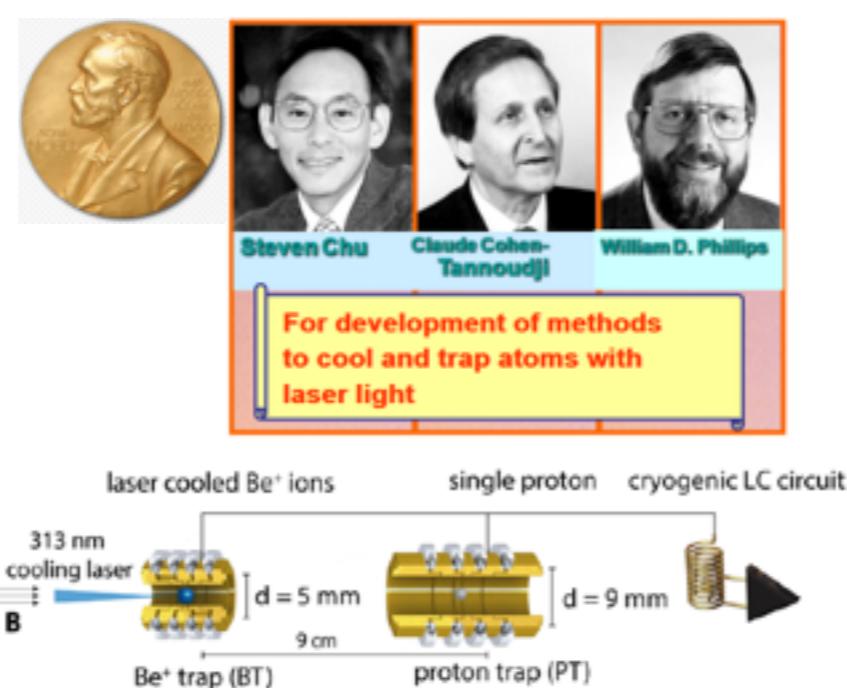


[1] K. Lynch, PhD thesis, University of Manchester, 2013.

[2] Sb run COLLAPS, 2018.

# Doppler Cooling

- Powerful technique to reach sub-K atom and ion temperatures [1]
- Standard tool for high-precision measurements: atomic clocks [2], quantum information science [3], physics beyond the standard model [4]

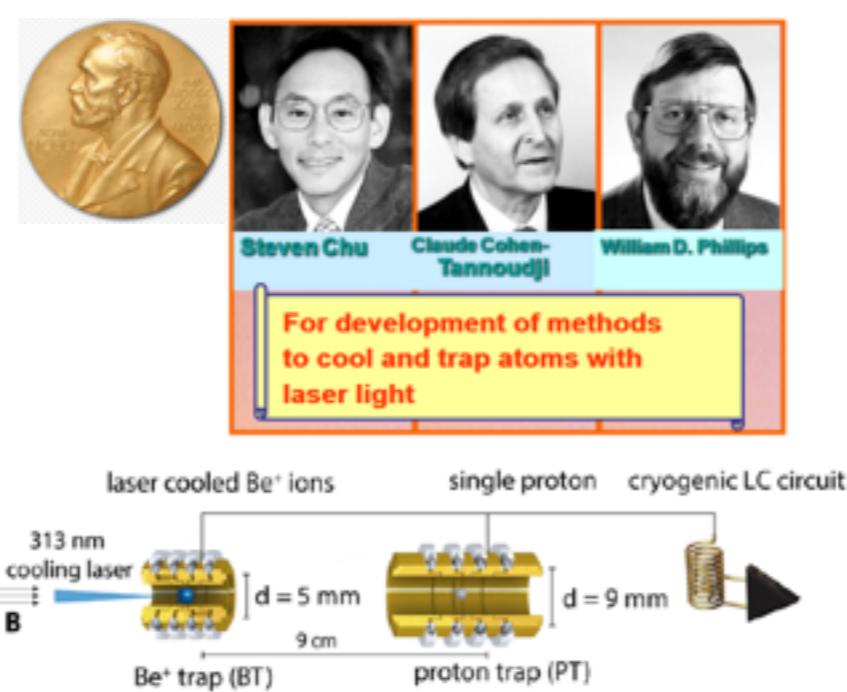


[1] T. Haensch and A. Schawlow, *Optics Communications* 13, 68 (1975).  
D. J. Wineland and W. M. Itano, *Phys. Rev. A* 20, 1521 (1979).  
J. Eschner et al., *J. Opt. Soc. Am. B* 20, 1003 (2003).

[2] D. Ludlow et al, *Rev. Mod. Phys.* 87, 637 (2015).  
[3] C. D. Bruzewicz et al, *Applied Physics Reviews* 6, 021314 (2019).  
[4] M. S. Safronova et al, *Rev. Mod. Phys.* 90, 025008 (2018).

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[4] M. S. Safronova et al, *Rev. Mod. Phys.* 90, 025008 (2018).

- Specific applications with RIBs

G. D. Sprouse and L. A. Orozco, *Annu. Rev. Nucl. Part. Sci.* 47, 429 (1997)  
J. A. Behr et al., *Phys. Rev. Lett.* 79, 375 (1997).  
M. Trinczek et al., *Phys. Rev. Lett.* 90, 012501 (2003).  
L. B. Wang et al., *Phys. Rev. Lett.* 93, 142501 (2004).

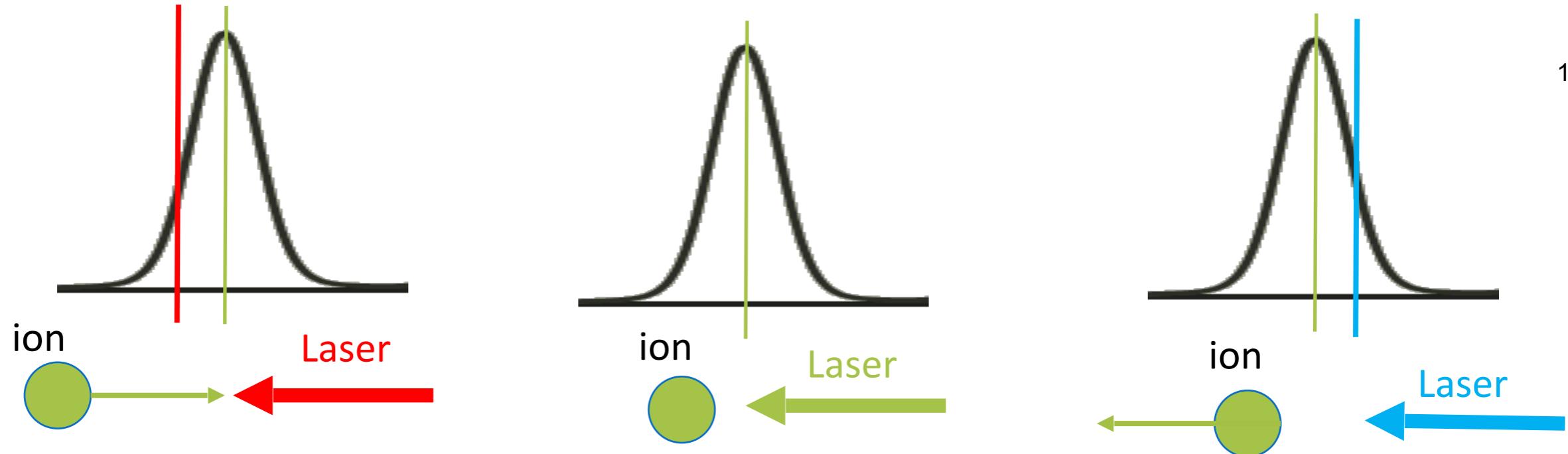
P. A. Vetter et al., *Phys. Rev. C* 77, 035502 (2008).  
J. R. A. Pitcairn et al., *RRC* 79, 015501 (2009).  
A. Takamine et al., *Phys. Rev. Lett.* 112, 162502 (2014).  
B. Fenker et al., *Phys. Rev. Lett.* 120, 062502 (2018)

- unexplored as cooling technique to deliver high quality (molecular) RIBs

**Goal: provide ultra-cold (molecular) RIBs**

- ... compatible with short half-lives
- ... universally applicable (via sympathetic cooling)

# Doppler Cooling principle

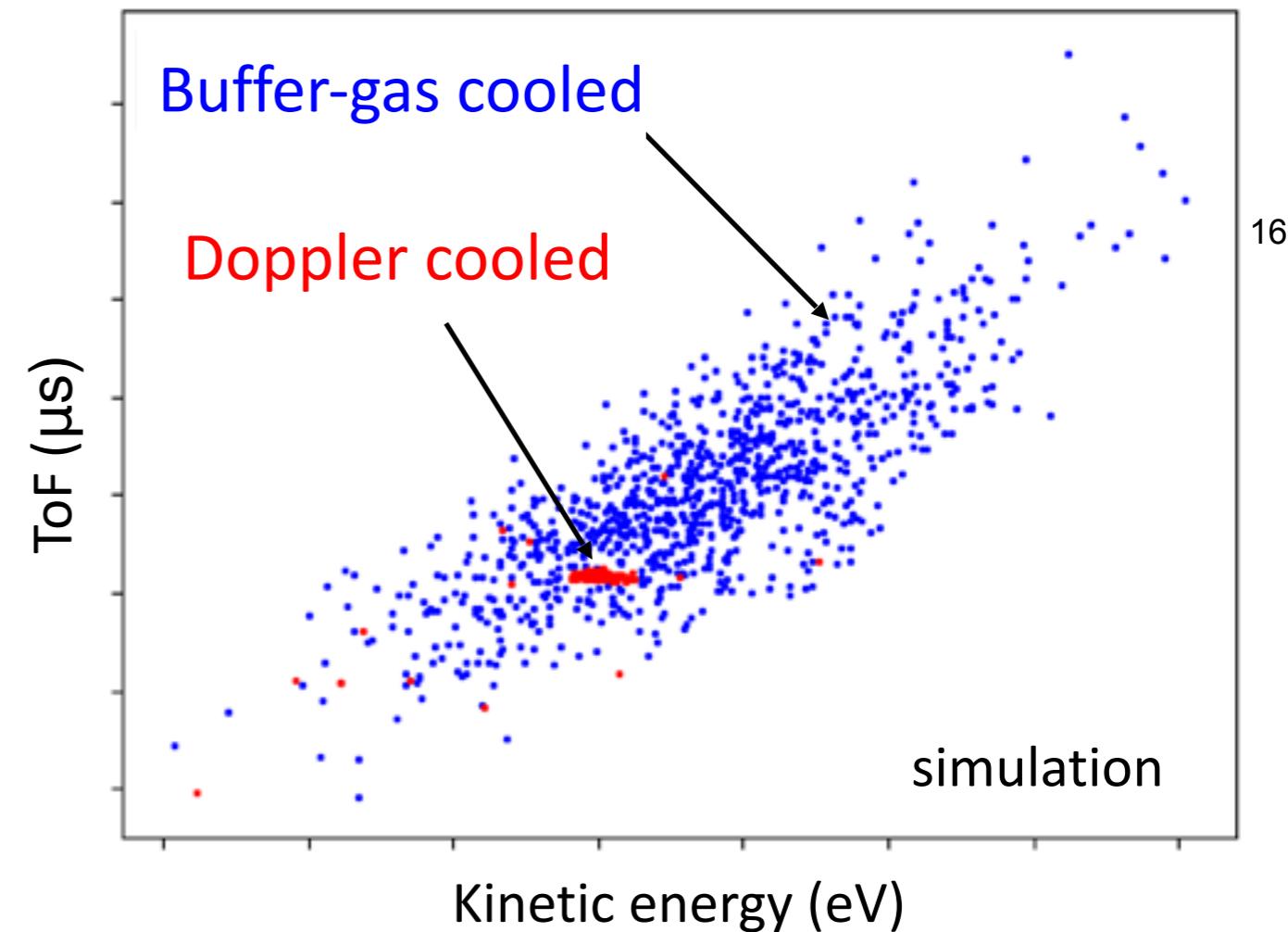
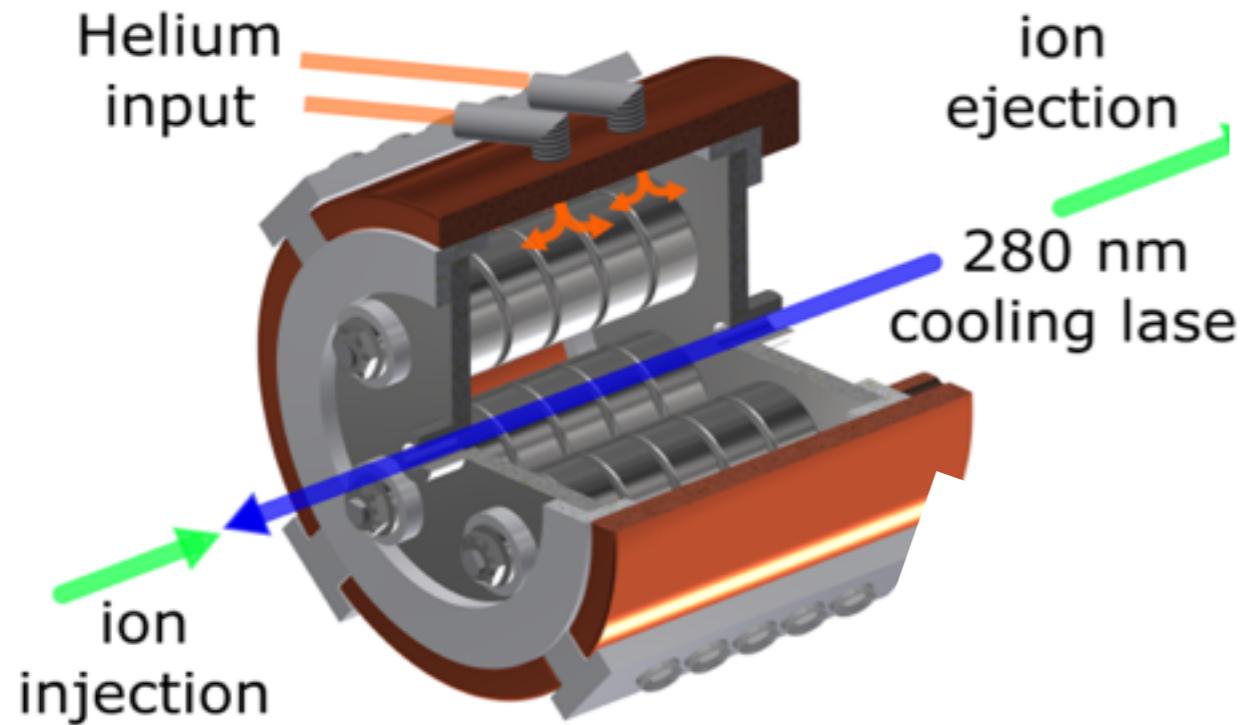


- Moving ions observe Doppler shift in laser frequency
- Absorption of photon in one direction
- Spontaneous emission of photon in random direction
- Net-cooling or heating effect since photon momentum is **subtracted from/added to** the Mg ion momentum
  - Red-detuning: cooling, blue detuning: heating

# Experimental Demonstration at



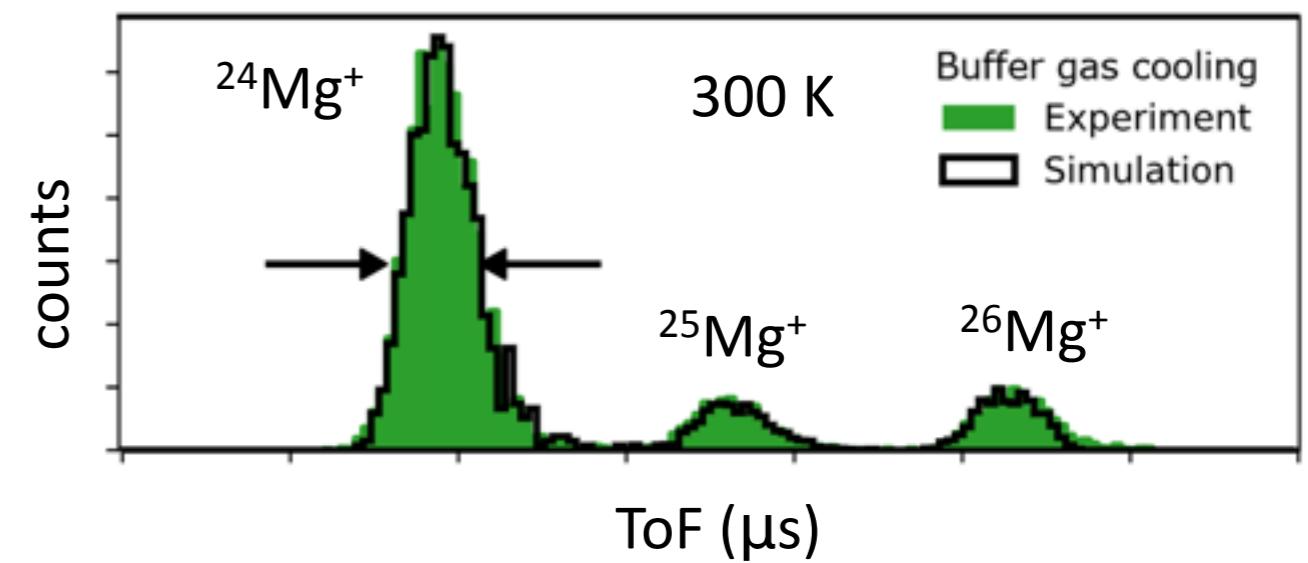
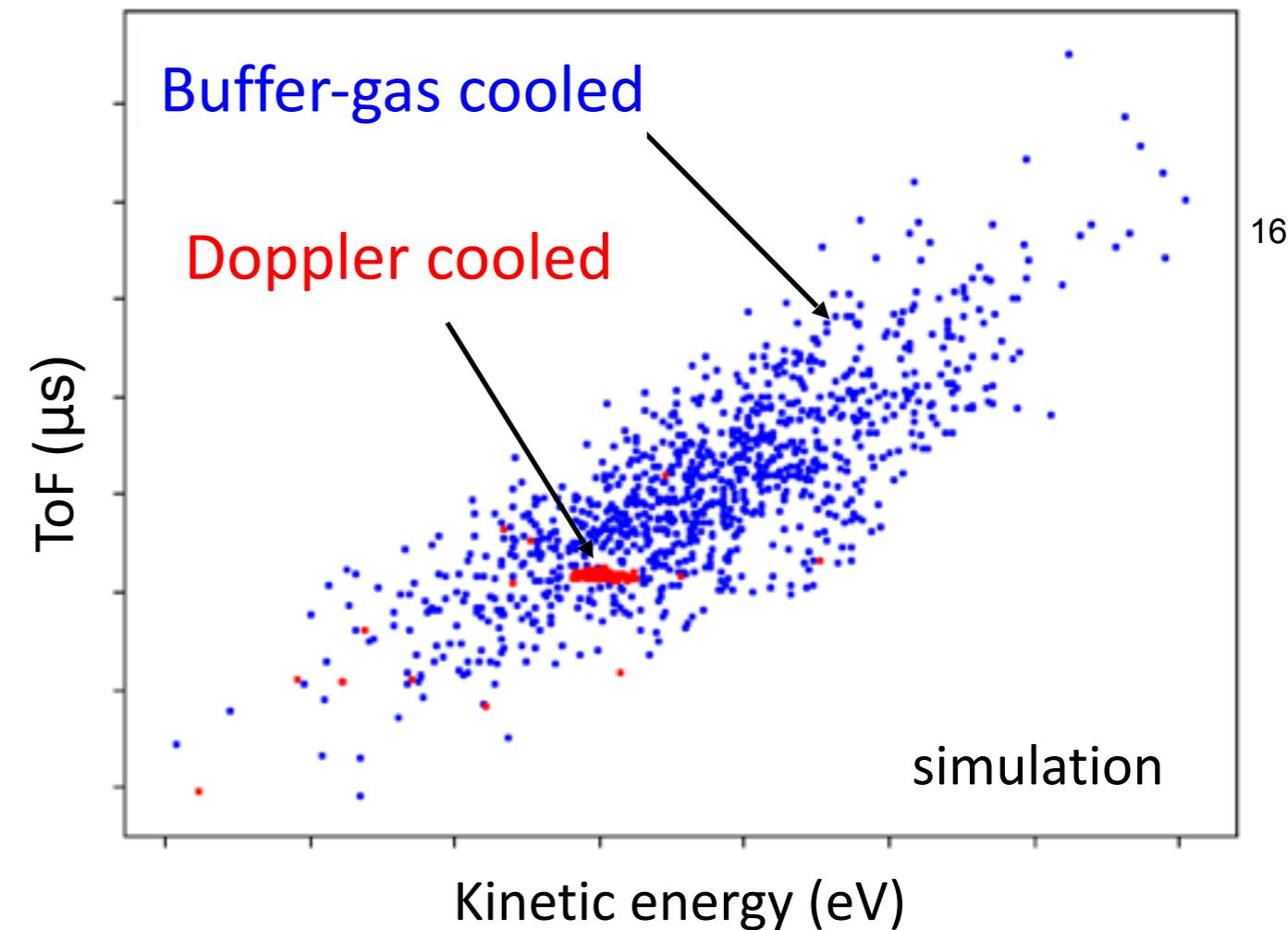
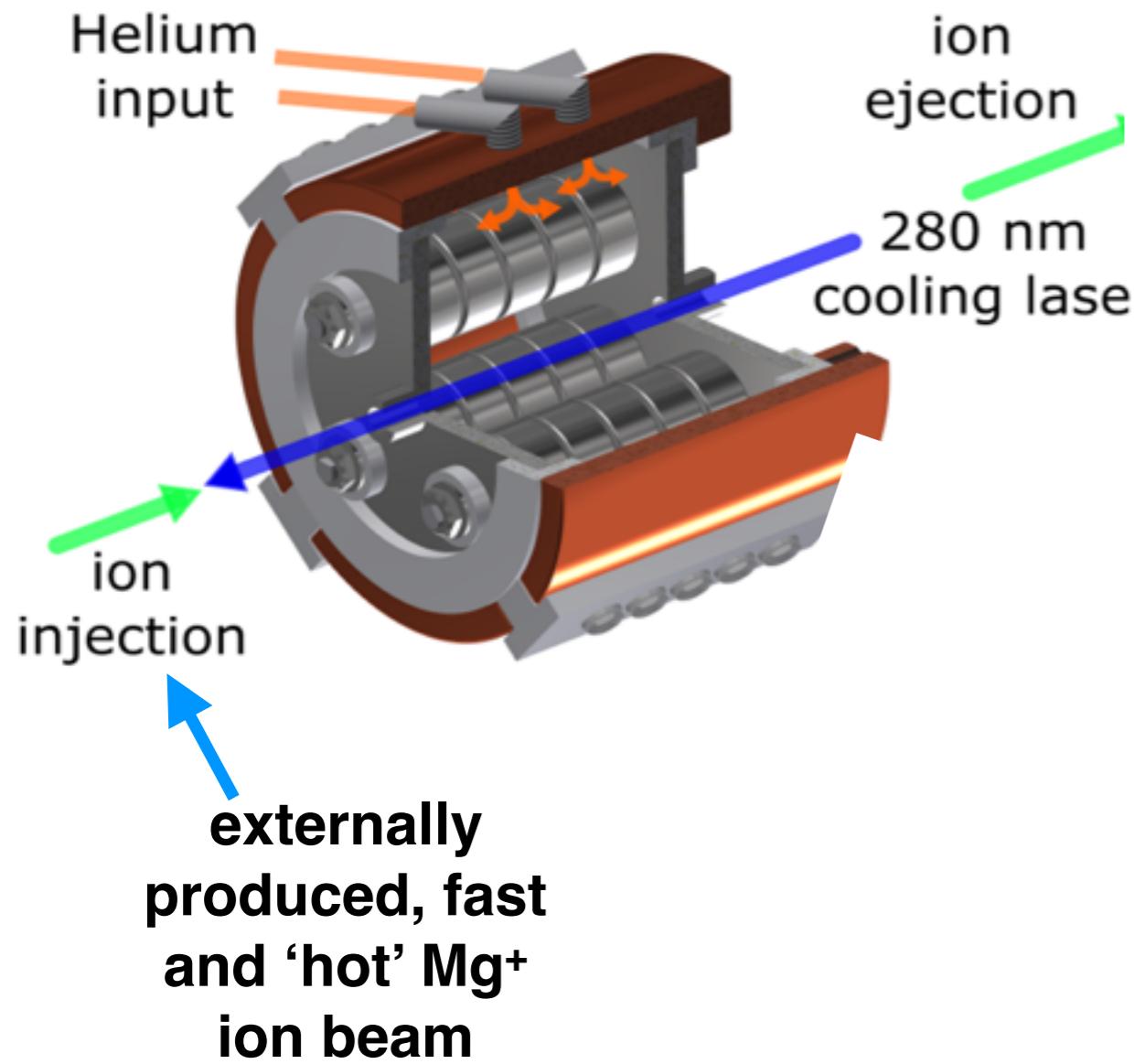
## Paul trap:



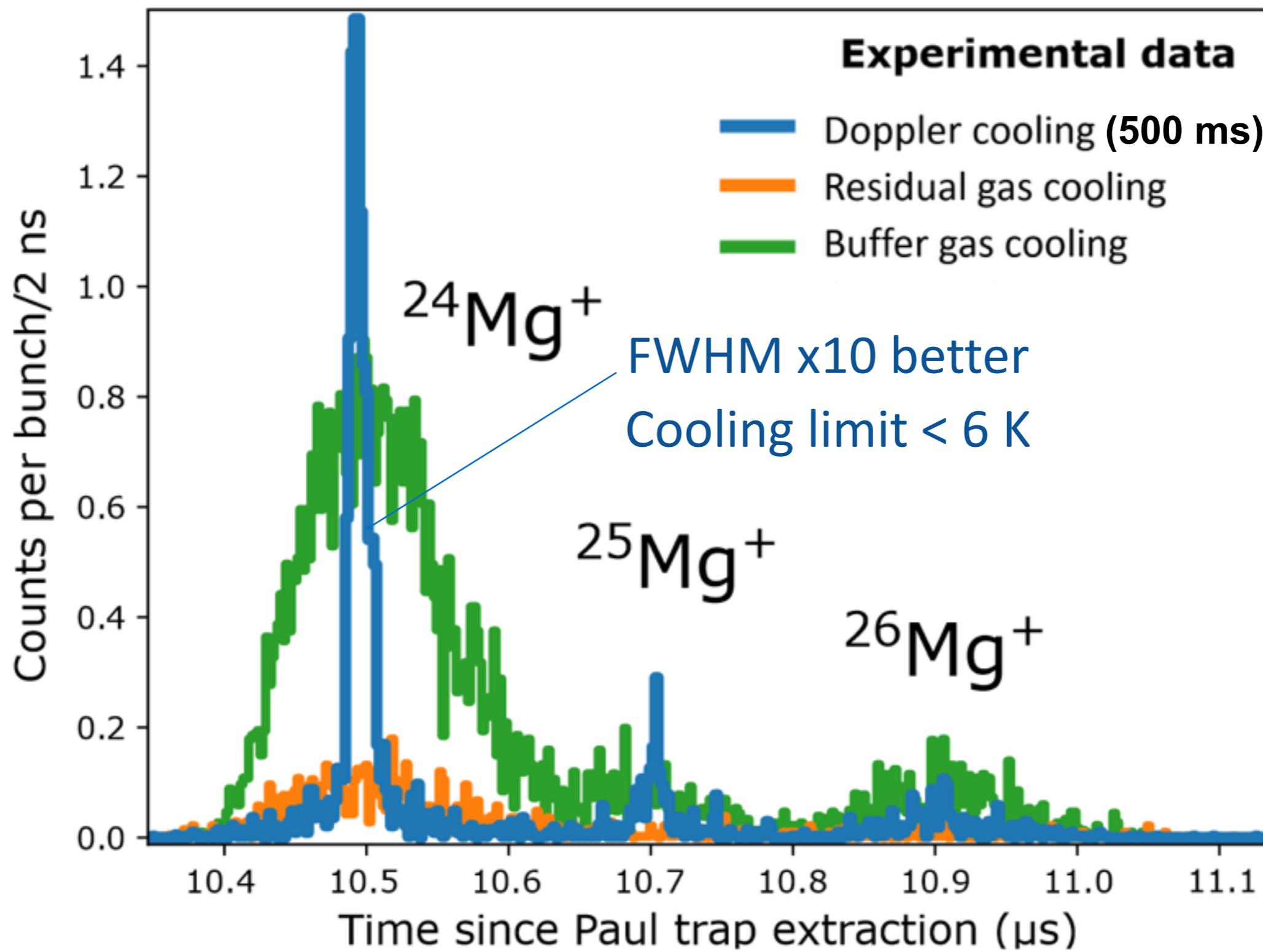
# Experimental Demonstration at



## Paul trap:



# Experimental results

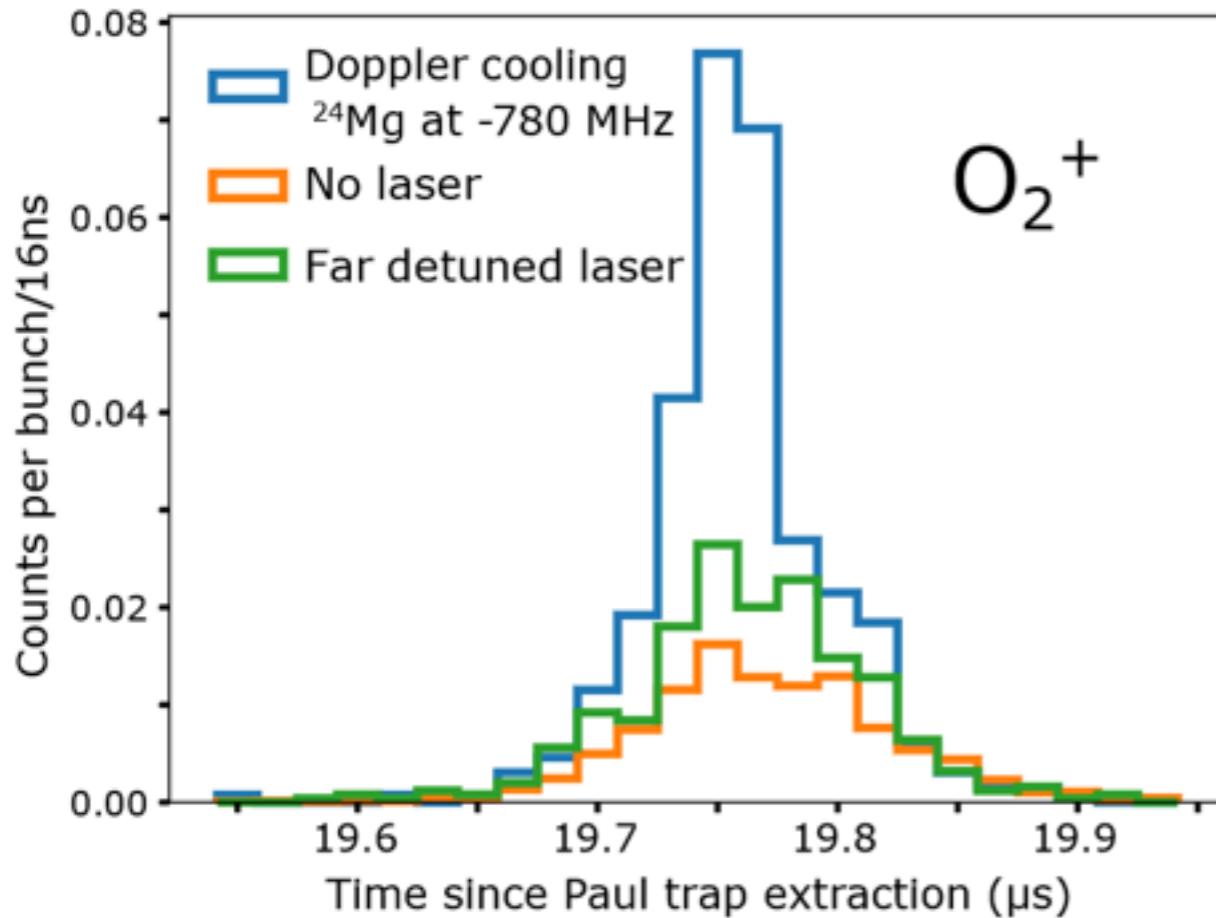


# Sympathetic cooling



- ‘universal’ availability of cold ion ensembles
- including ionic systems which cannot be directly laser-cooled

18



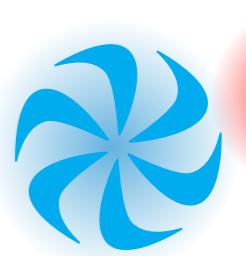
opportunity for cold molecular RIBs

	$\text{O}_2^+$
Peak width residual-gas or buffer-gas cooling	113(5) ns
Sympathetic cooling	58(4) ns
Improvement in countrate	Factor 2.6

Can be done better analogous to existing work, e.g. [1],[2]

*J. Wuebbena et al, Phys. Rev. A 85, 043412, 2012.*  
*[2] M. Guggemos. New Journal of Physics 17, 103001, 2015.*

# Summary



19

- **Radioactive Molecules**
  - entirely new science path
  - intriguing&unexplored **probes for New Physics**
- **RadMol**
  - dedicated laboratory for radioactive molecules & precision studies at TRIUMF
  - designed to master experimental challenges
- **Cold radioactive, molecular beams**
  - Doppler + sympathetic cooling

Thank you  
Merci

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