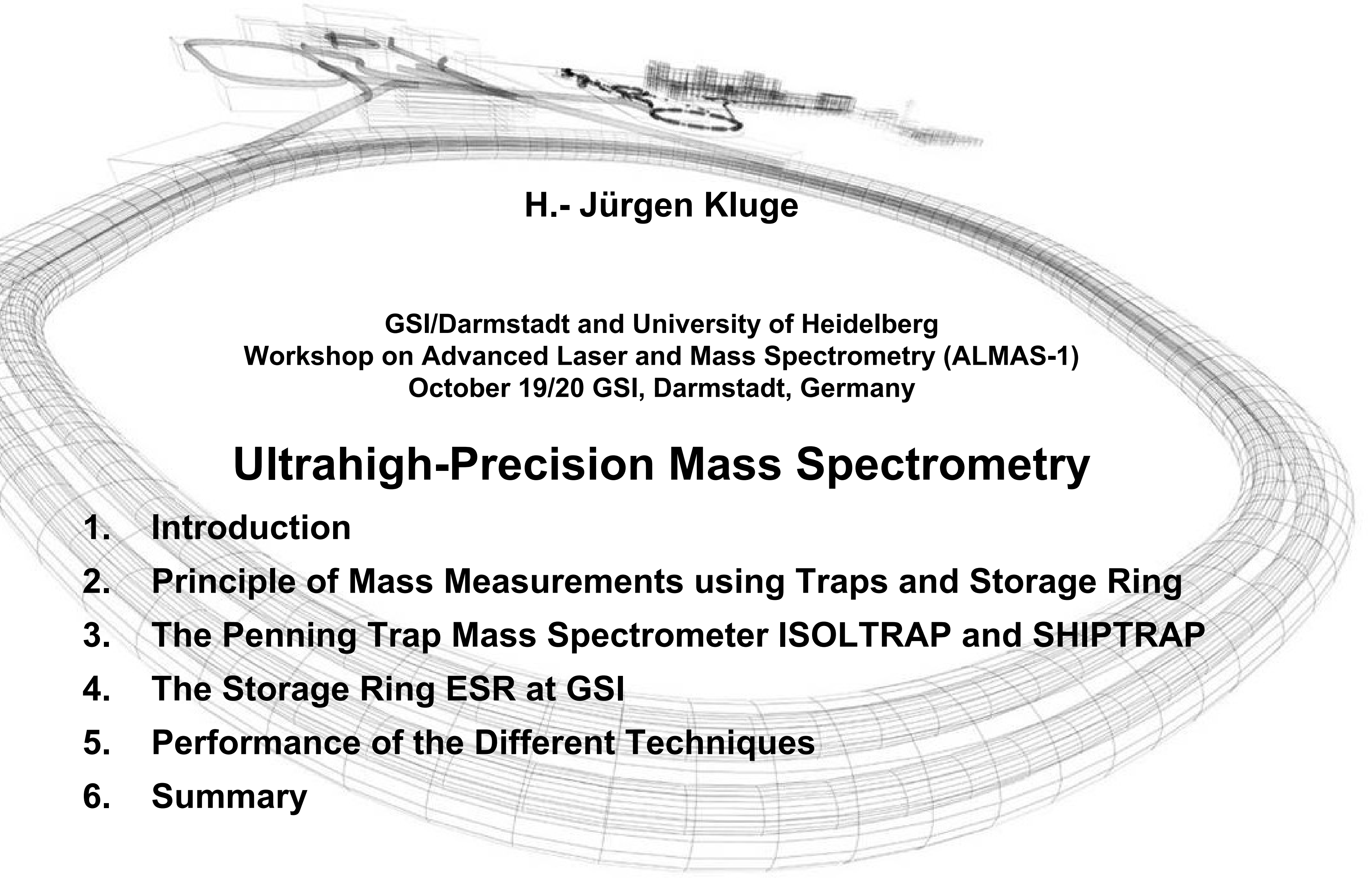


H.- Jürgen Kluge

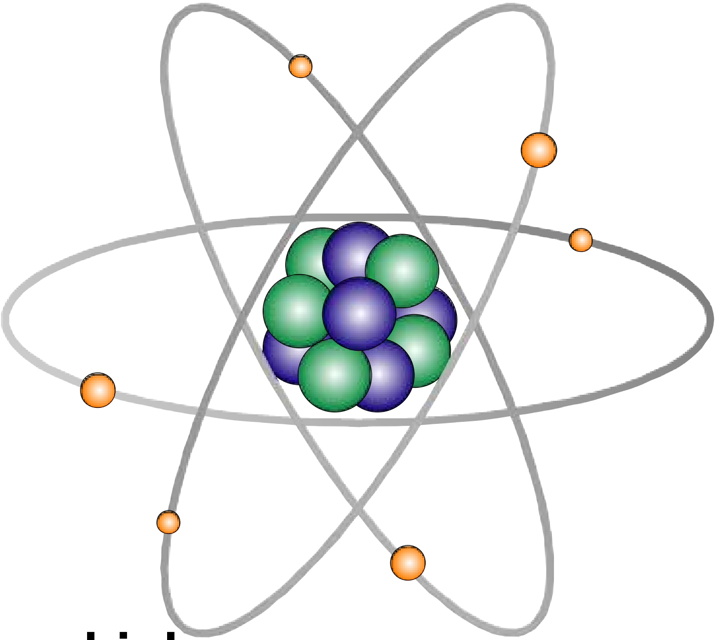
GSI/Darmstadt and University of Heidelberg
Workshop on Advanced Laser and Mass Spectrometry (ALMAS-1)
October 19/20 GSI, Darmstadt, Germany

Ultrahigh-Precision Mass Spectrometry

1. Introduction
2. Principle of Mass Measurements using Traps and Storage Ring
3. The Penning Trap Mass Spectrometer ISOLTRAP and SHIPTRAP
4. The Storage Ring ESR at GSI
5. Performance of the Different Techniques
6. Summary

Mass – Binding Energy – Forces

The atomic mass of an isotope is given by:



= *N* · 🟢 + *Z* · 🔵 + *Z* · 🟠

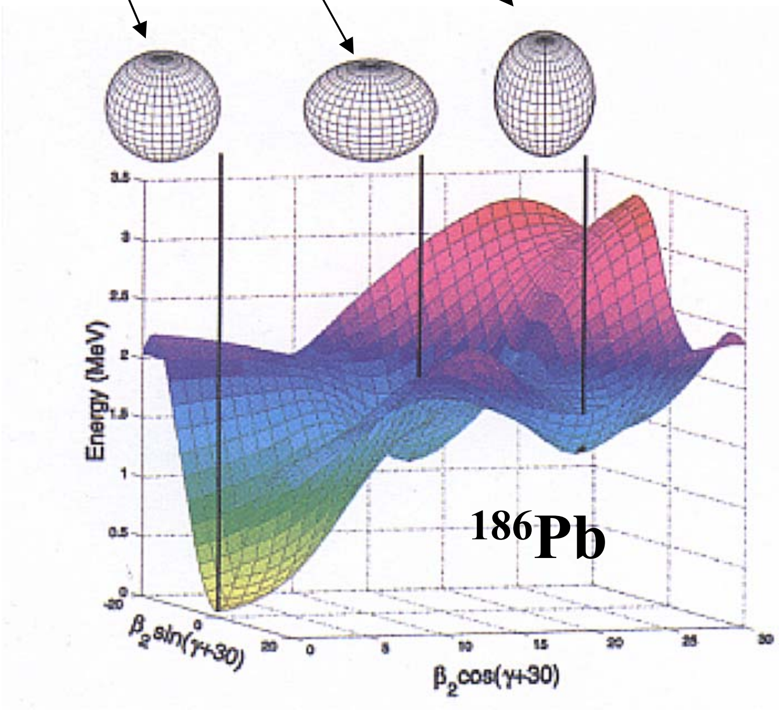
– binding energy

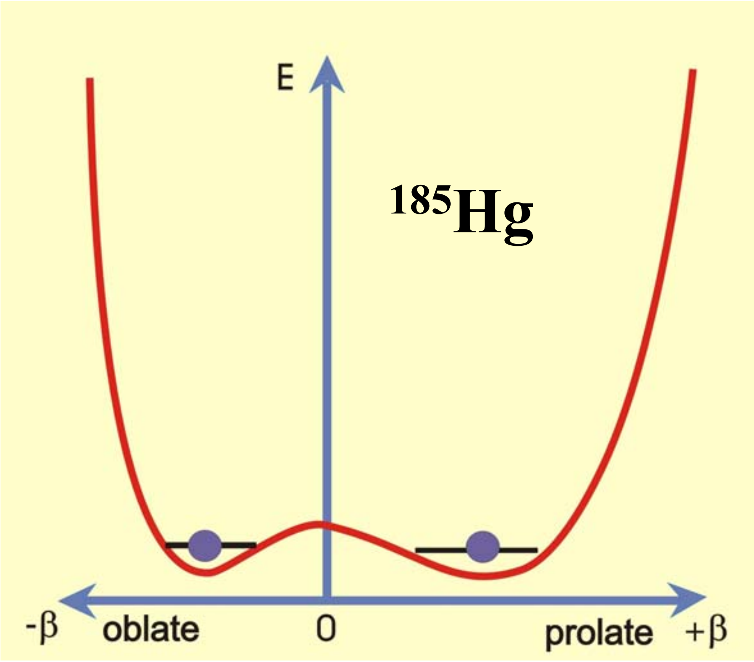
- ⇒ **high-accuracy mass measurements allow one to determine the atomic and nuclear binding energies**
- ⇒ **the nuclear binding energy reflects all forces (strong, electro-magnetic and weak interactions) acting in the nucleus**
- ⇒ **the atomic binding energy reflects the electro-magnetic force acting between the electrons and between the electrons and the nucleus.**

Information on Nuclear Ground State Properties by Atomic Physics Techniques

mass ⇒ depth of minima
isotope shift ⇒ position $|\beta^2|$
isomer shift ⇒ separation $\delta\,|\beta^2|$
quadrupole moment ⇒ sign of β
spin ⇒ single-particle properties
magnetic moment ⇒ single-particle properties

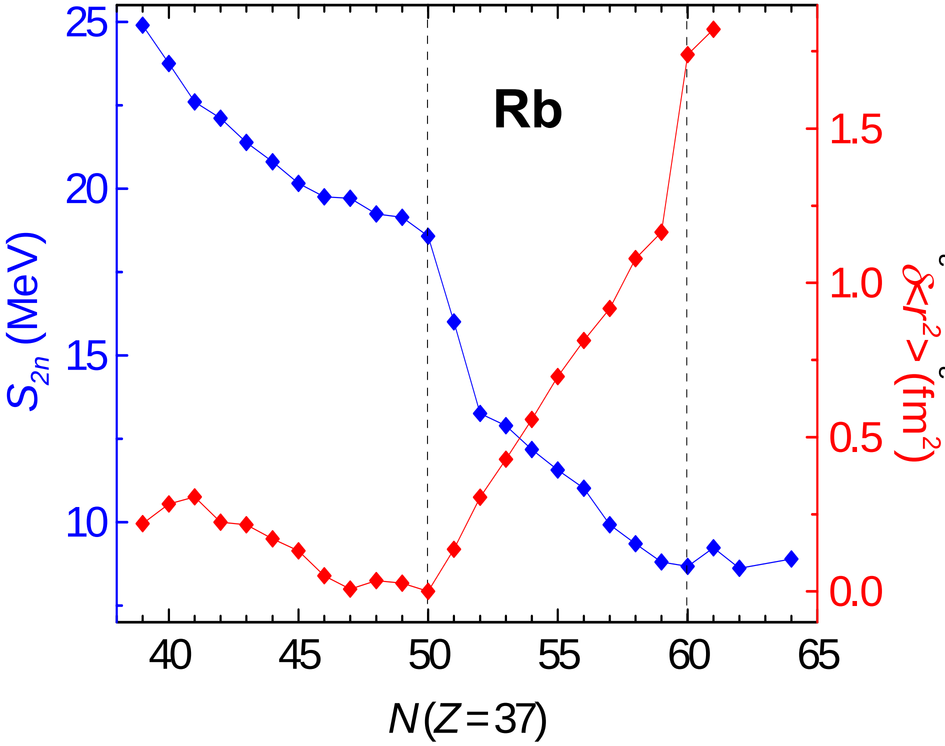
spherical oblate prolate

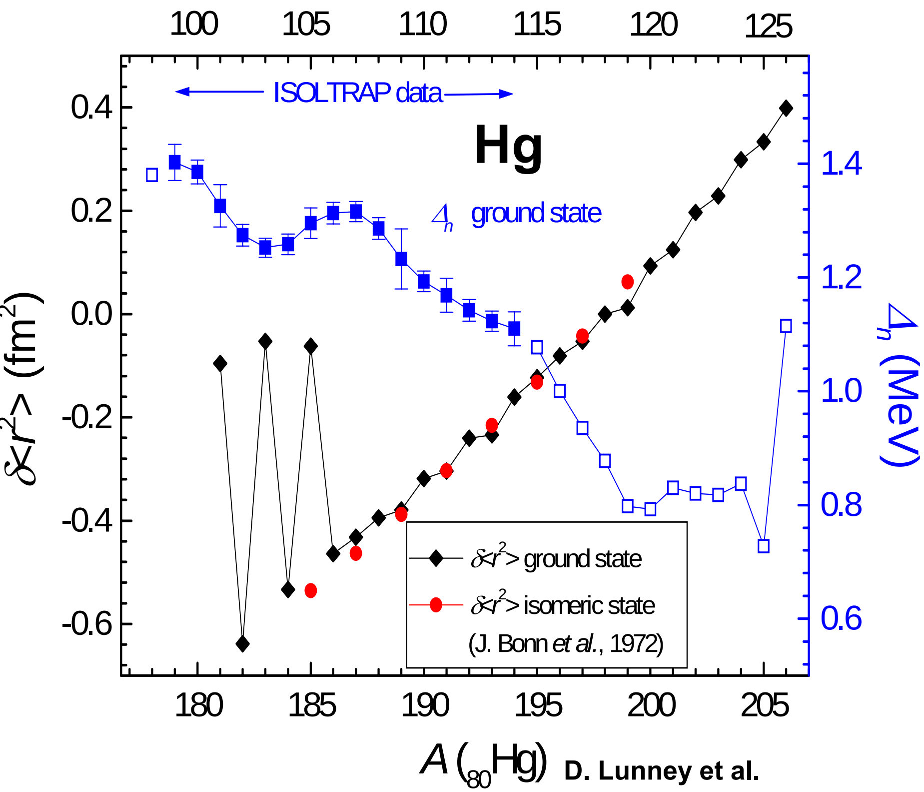




Comparison: Charge Radii – Nuclear Binding Energies

Is the mass more sensitive to nuclear structure effects than the information obtained by optical techniques (spin, moments, charge radii)?
Examples : Rb and Hg isotopes





D. Lunney et al.
Rev. Mod. Phys. 75 (2003) 1021

Nuclear forces maximize nuclear binding energy irrespective of shape or configuration.
Nuclear Binding: some MeV per nucleon (1 GeV) → 10^{-3}, $\delta E/E \leq 1\%$ → $\delta m/m \leq 10^{-5}$

Laser Spectroscopy ↔ Mass Spectrometry

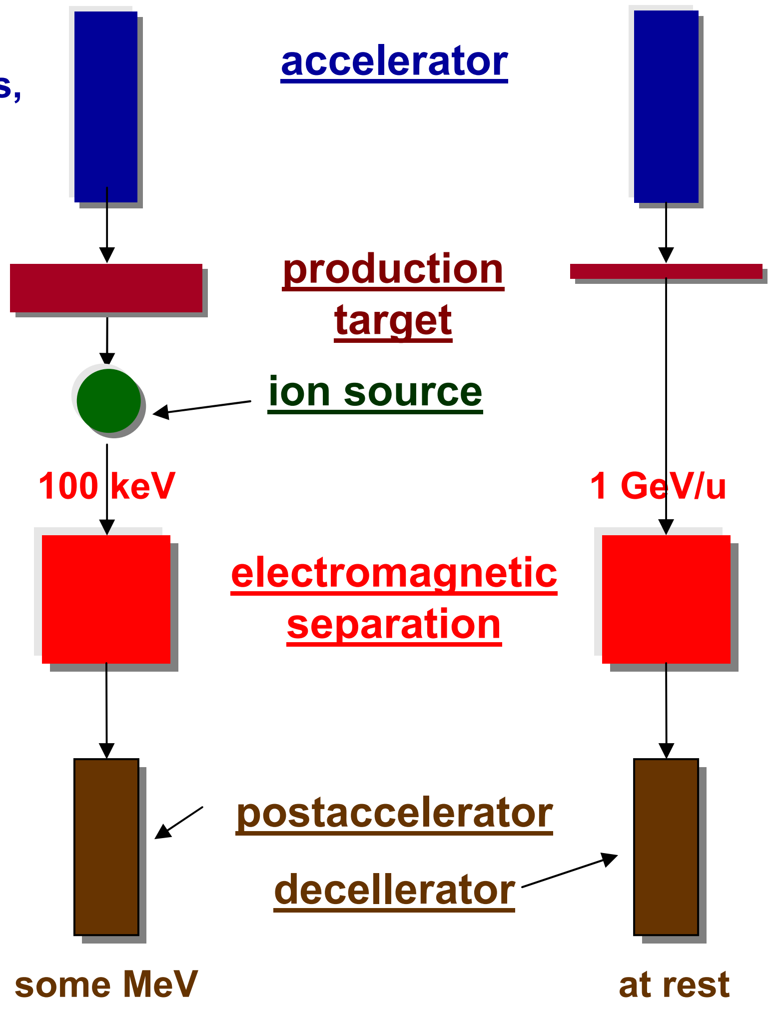
	Laser Spectroscopy	Mass Spectrometry
Techniques	many, often specialized: fluorescence, MOT; colinear, resonance ionization (mass) spectroscopy, ……	many, quite general: decays, reactions, Penning trap, storage ring, cyclotron, …….
Goal	spin, nuclear moments, charge radii	mass → nuclear & atomic binding energy
Calibration	well known transitions, frequency comb	well known masses, carbon cluster comb
Ultimate accuracy determined by	observation time → atomic lifetime	observation time → nuclear half-life
Sensitivity	≈ 1 atom/second	≈ 1 atom/second
Limitation	detection efficiency, charge radii of light isotopes	not resolved isomers, space charge by isobars

Requirements for Mass Spectrometry in Different Research Areas

	δm/m	required resolving power
General physics & chemistry	$< 10^{-5}$	10^{4}
Nuclear structure physics - separation of isobars	10^{-6}	10^{5}
Astrophysics - separation of isomers	$< 10^{-7}$	10^{6}
Weak interaction studies	10^{-8}	10^{7}
Metrology - fundamental constants	$< 10^{-9}$	10^{8}
CPT Tests	$< 10^{-10}$	10^{9}
QED in highly-charged ions - separation of atomic states	$\leq 10^{-11}$	10^{10}

For radioactive isotopes, a resolving power of > 10^6 is required in order to discriminate between ground and isomeric states.

The Two Ways: ISOL Fragmentation



high intensities for long-lived isotopes

limitation at $T_{1/2} < 10$ ms

high beam quality

no refaractory elements

contamination by isobars

singly-charged ions

low intensities

limitation only at $T_{1/2} < 1\mu$s

low beam quality

all elements

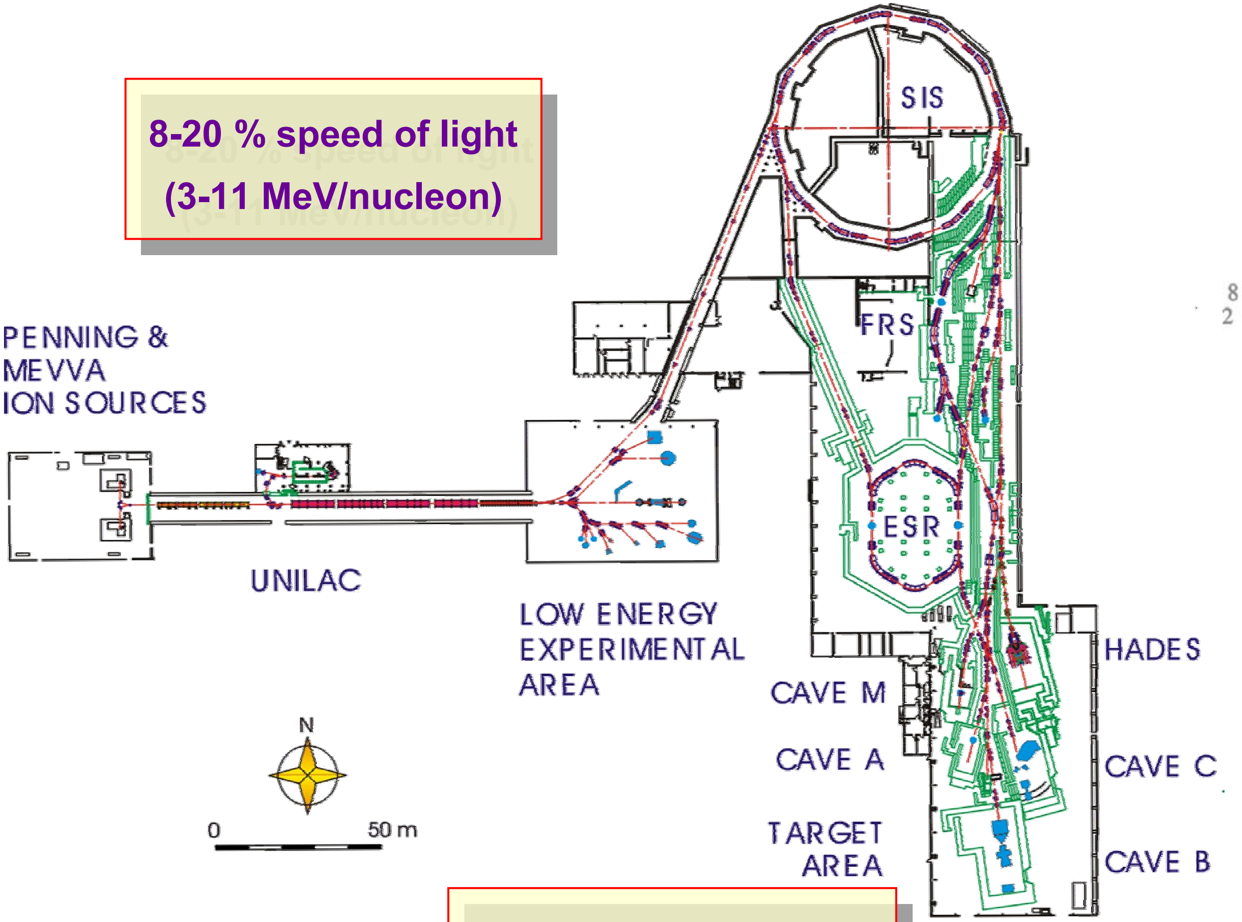
rejection of isobars by q/m identification

highly-charged ions

ISOL and fragmentation facilities are complementary

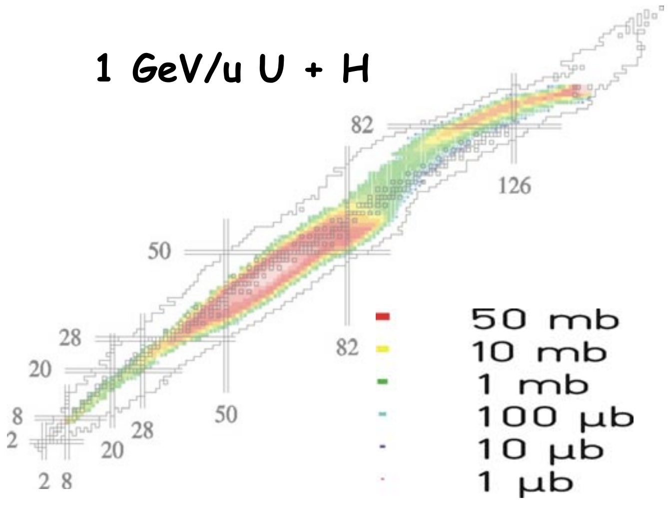
The Present GSI Heavy-Ion Accelerator Facility

8-20 % speed of light
(3-11 MeV/nucleon)

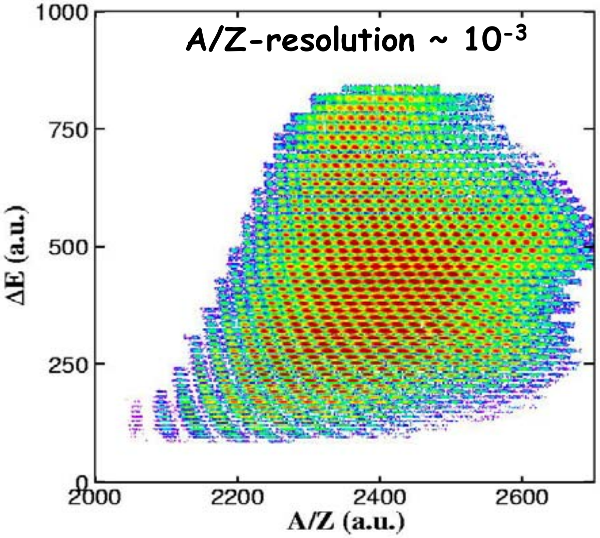


90 % speed of light
(2 GeV/nucleon)

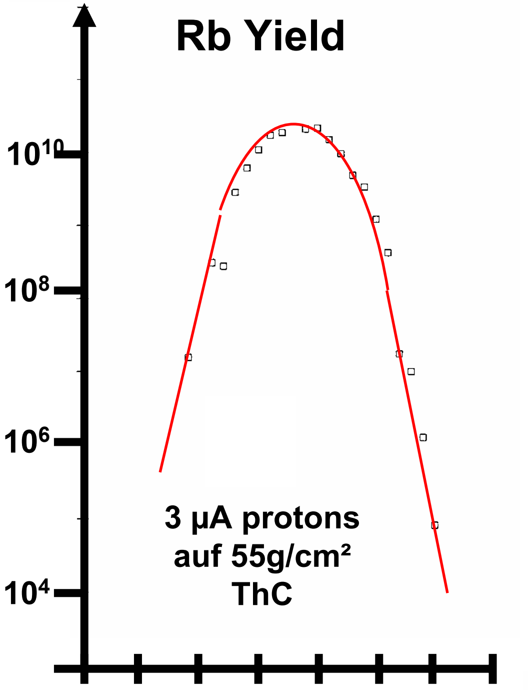
1 GeV/u U + H

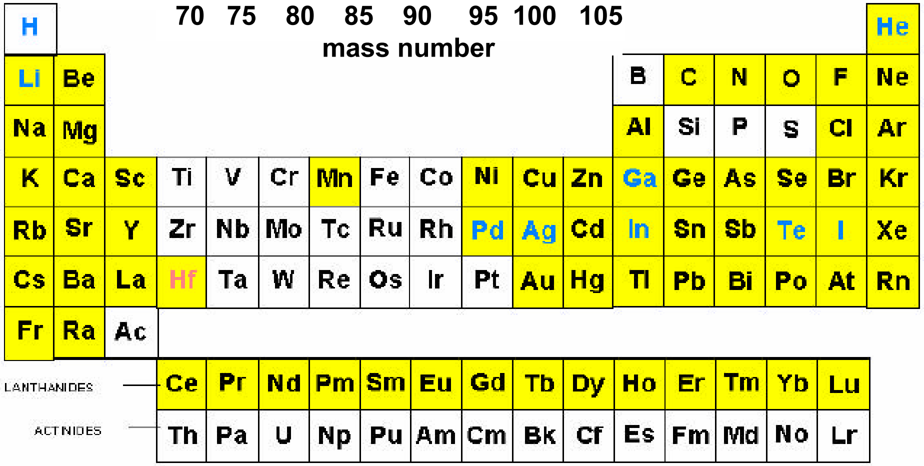


about 1000 nuclear residues identified

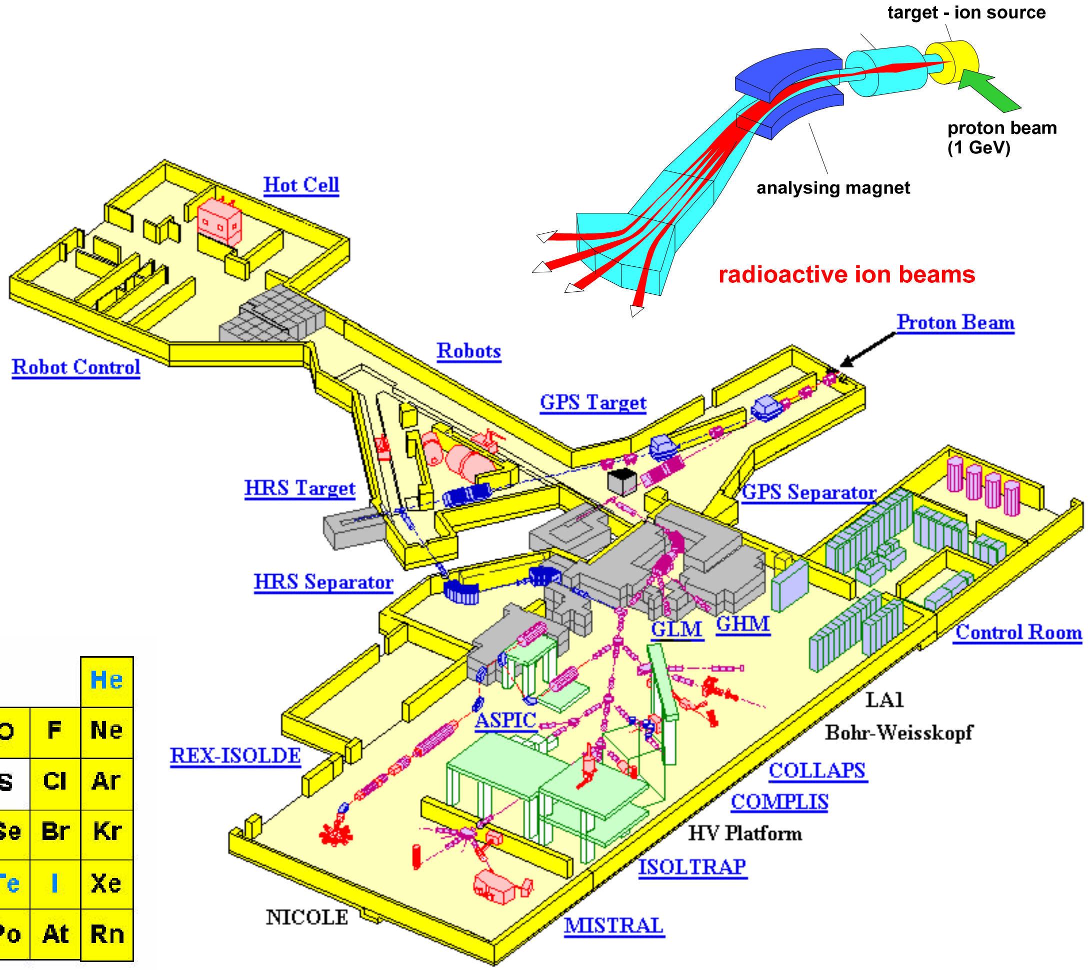


The ISOLDE Facility at CERN





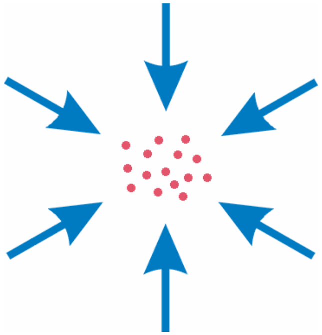
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac															
LANTHANIDES			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
ACTINIDES			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	



> 70 elements, > 600 isotopes

Principle of Storing and Cooling

Radial force



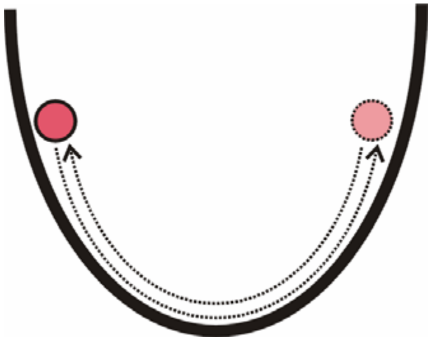
electric fields

magnetic fields

light fields

“infinite” storage time

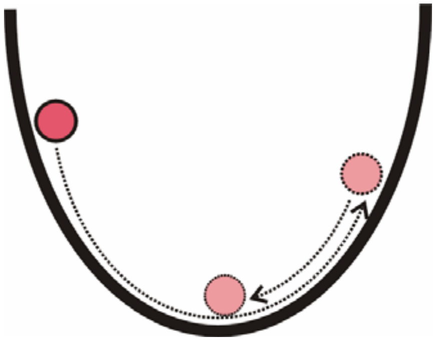
Harmonic potential



harmonic oscillation

2 or 3 independent eigen frequencies

Cooling

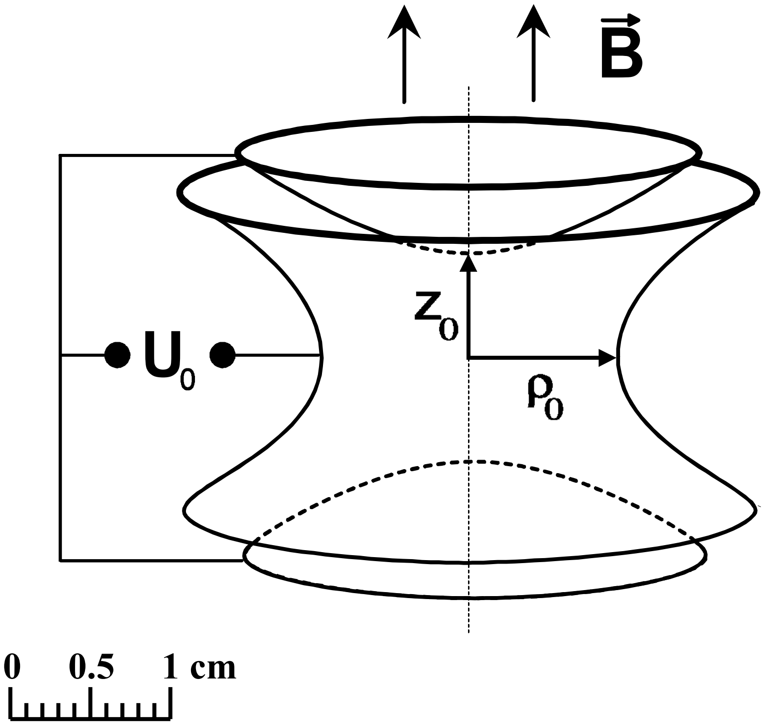


decrease of oscillation amplitude

reduction of trap imperfections

Storage Devices

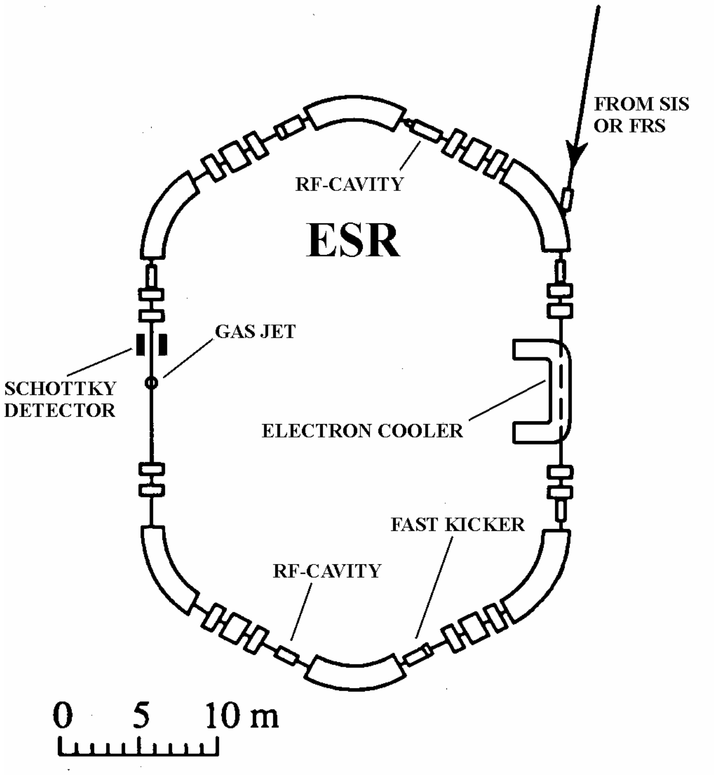
PENNING and PAUL TRAP



particles at nearly rest in space

many cooling techniques

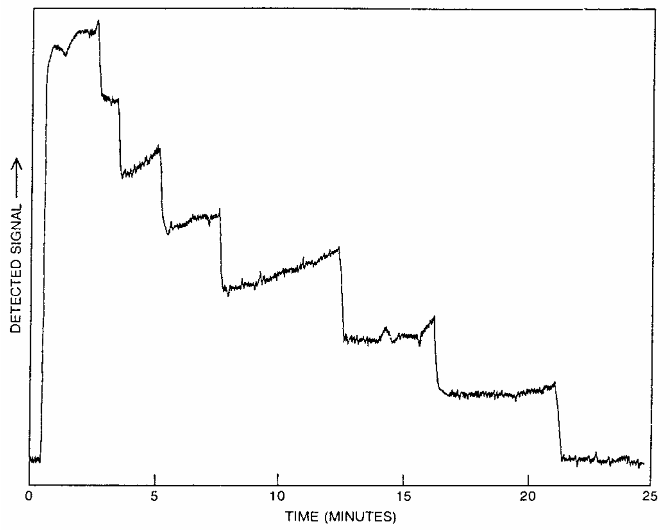
STORAGE RING



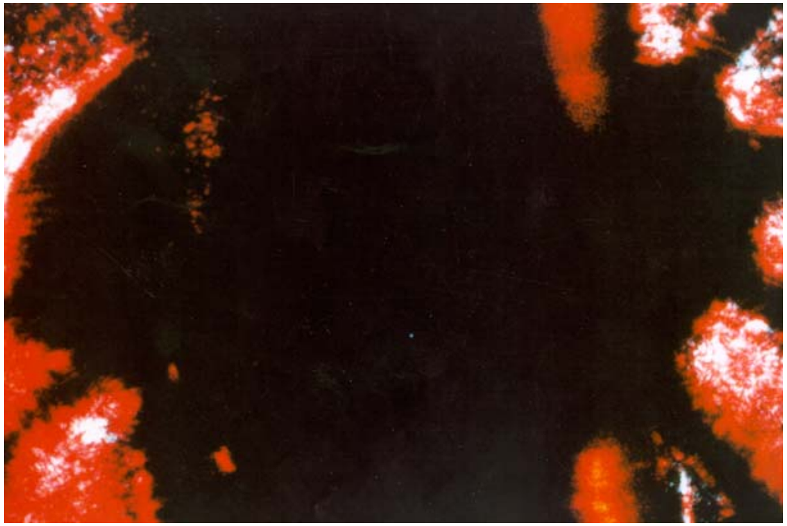
relativistic particles

electron, stochastic, laser cooling

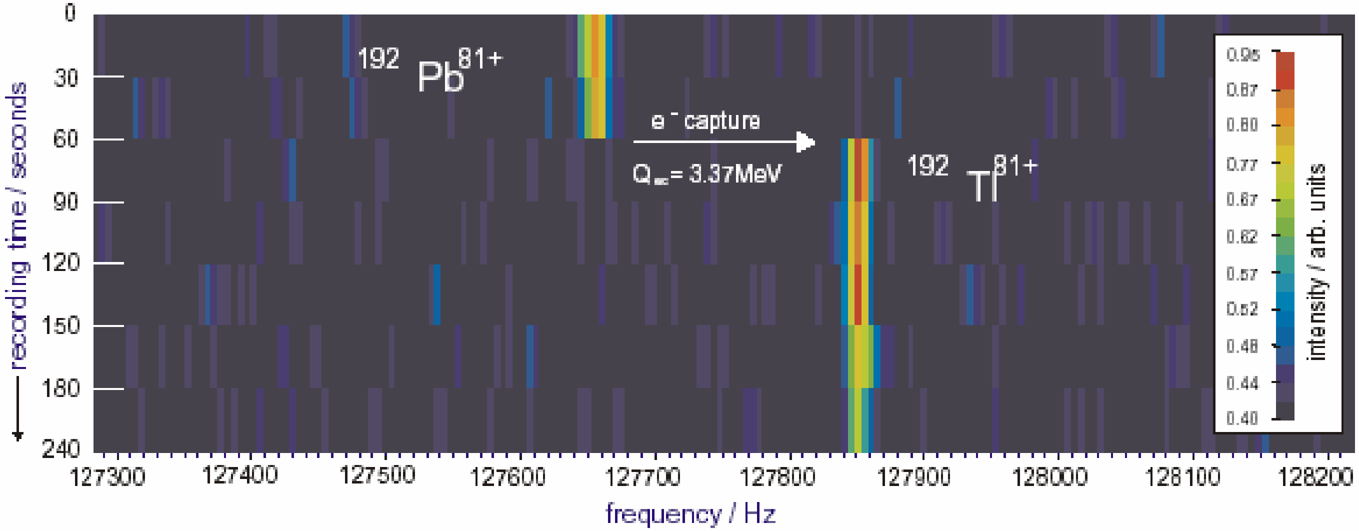
Single-Ion Sensitivity



Electronic Detection of 1–7 Electrons in a Penning Trap Dehmelt et al.

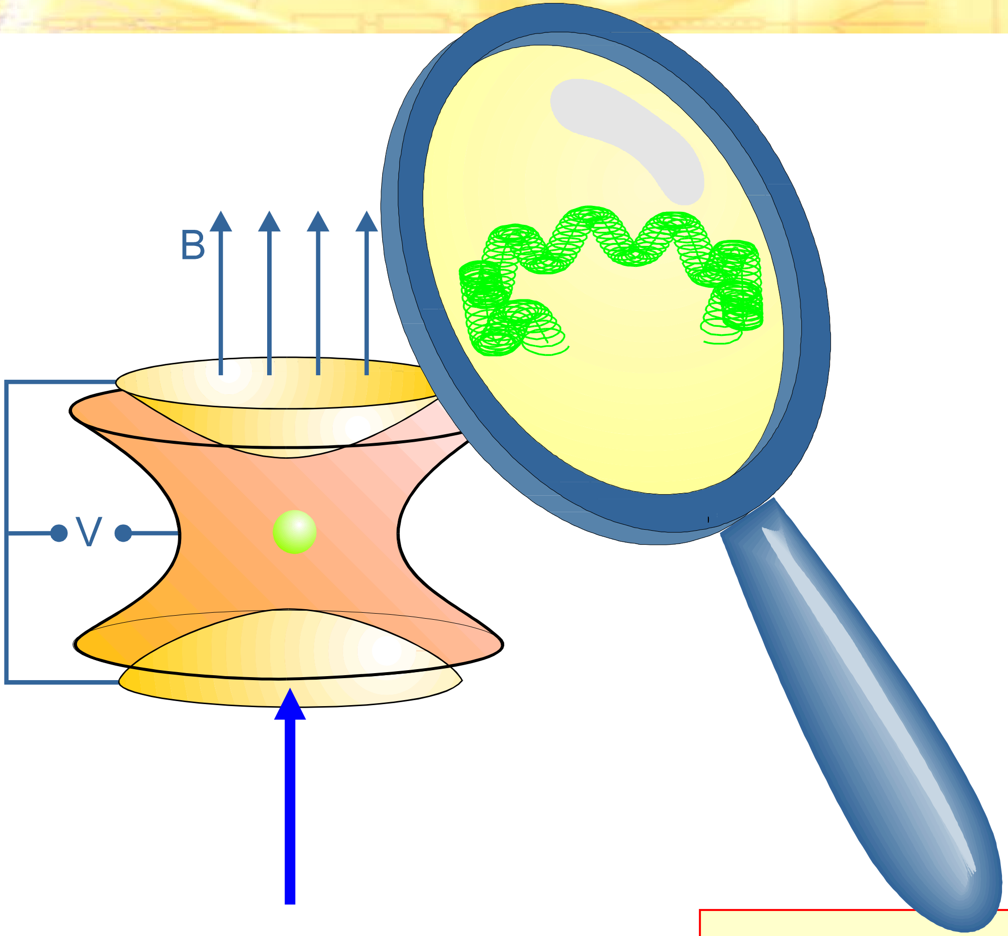


Optical Detection of a Single Barium Ion in a Paul Trap
Dehmelt, Toscheck et al.



Electron Capture of a Single Tungsten Ion in the Storage Ring ESR
Bosch et al.

Principle of Mass Measurements in Penning Traps



ION SOURCE:
stable isotopes
radioactive isotopes
highly charged ions
electrons
antiprotons

Confinement of ions in a strong magnetic field of known strength B

Mass measurement via determination of cyclotron frequency

$$\nu_c = (q/m)\cdot(B/2\pi)$$

from characteristic motion of stored ions

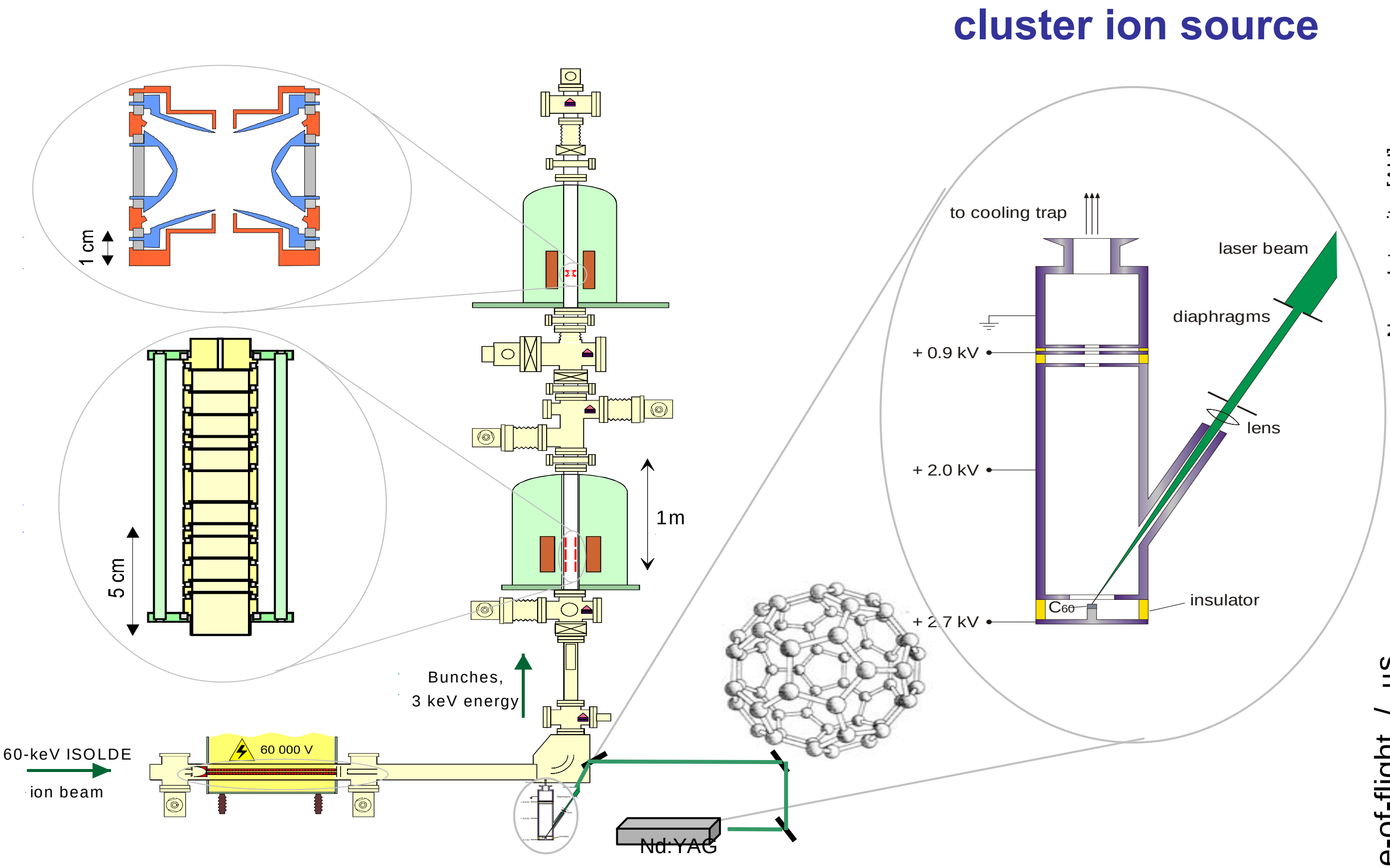
Example

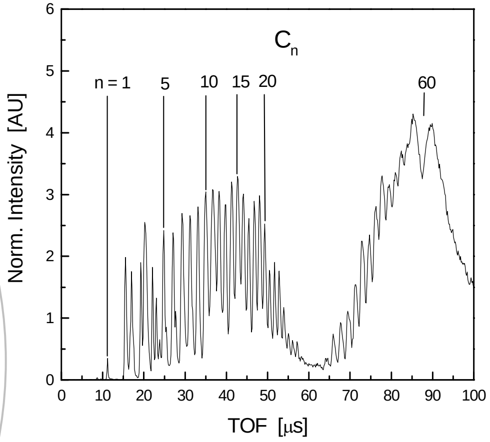
B = 6 T, q = 1, m = 100 u → ν_c = 1 MHz

T_{obs} = 1 s: $\Delta\nu_c$ = 1 Hz

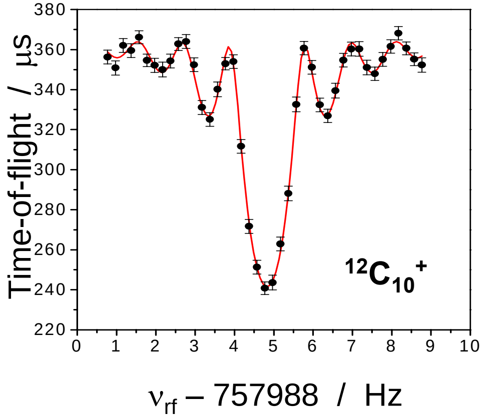
→ R = 10^6 and $\delta m/m = 10^{-8}$

Absolute Mass Measurements of Radionuclides by Use of Carbon Cluster





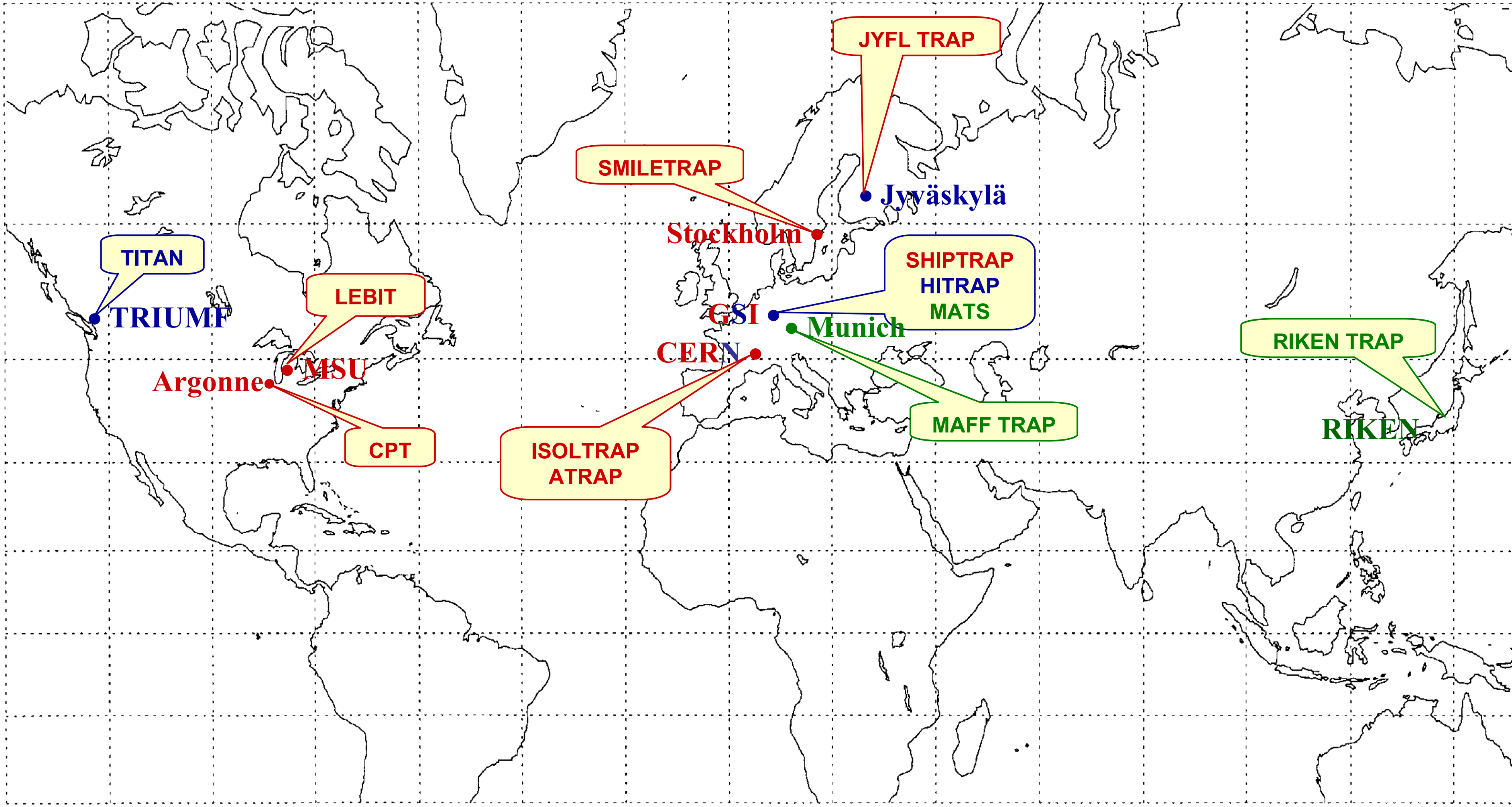
TOF resonance spectrum



Relative mass accuracy limit: $\delta m/m \geq 8\cdot10^{-9}$

K. Blaum *et al.*, Eur. Phys. J. A 15, 245 (2002)

Penning Traps at Accelerators for Mass Spectrometry



- operating facilities
- facilities under construction or test
- planned facilities

Complementarities of Penning Traps for Mass Spectrometry

Type of reaction or facility	ISOL-TRAP CERN	CPT Argo.	SHIP-TRAP GSI	JYFL-TRAP Jyväskylä	LEBIT MSU	MAFF-TRAP Munich	TITAN TRIUMF	HITRAP GSI	MATS GSI
ISOL	x						x		
fusion		x	x						
IGISOL				x					
fragmentation					x			x	x
fission by neutrons						x			
highly-charged ions						x	x	x	x
$<10^{-10}$ accuracy								x	x

- operating facilities
- facilities under construction or test
- planned facilities

the masses of about 700 isotopes were measured up to now by use of Penning traps

How to Reach Highest Accuracy?

Use
- **single ion stored in the Penning trap**
- **a highly charged ion**
- **a high-field superconducting magnet**
- **self-shielded superconducting magnet**
- **storage at low temperature**
- **long observation time**

Avoid
- **fluctuations of external magnetic fields**
- **changes of pressure**
- **changes of temperature**

Presently reached: 10^{-11} for singly charged ion

10^{-10}

MIT Group

M.P. Bradley, F. Palmer, D. Garrison, L. Ilich, S. Rusinkiewics, D.E. Pritchard, Hyperfine Interactions 108 (1997) 144
M.P. Bradley, J.V. Porto, S. Rainville, J.K. Thompson, D.E. Pritchard, Phys. Rev. Lett. 83 (1999) 4510

Harvard Group

G. Gabrielse, A. Khabbaz, D.S. Hall, C. Heimann, H. Kalinowsky, W. Jhe, Phys., Rev. Lett. 82 (1999) 3198

Seattle Group

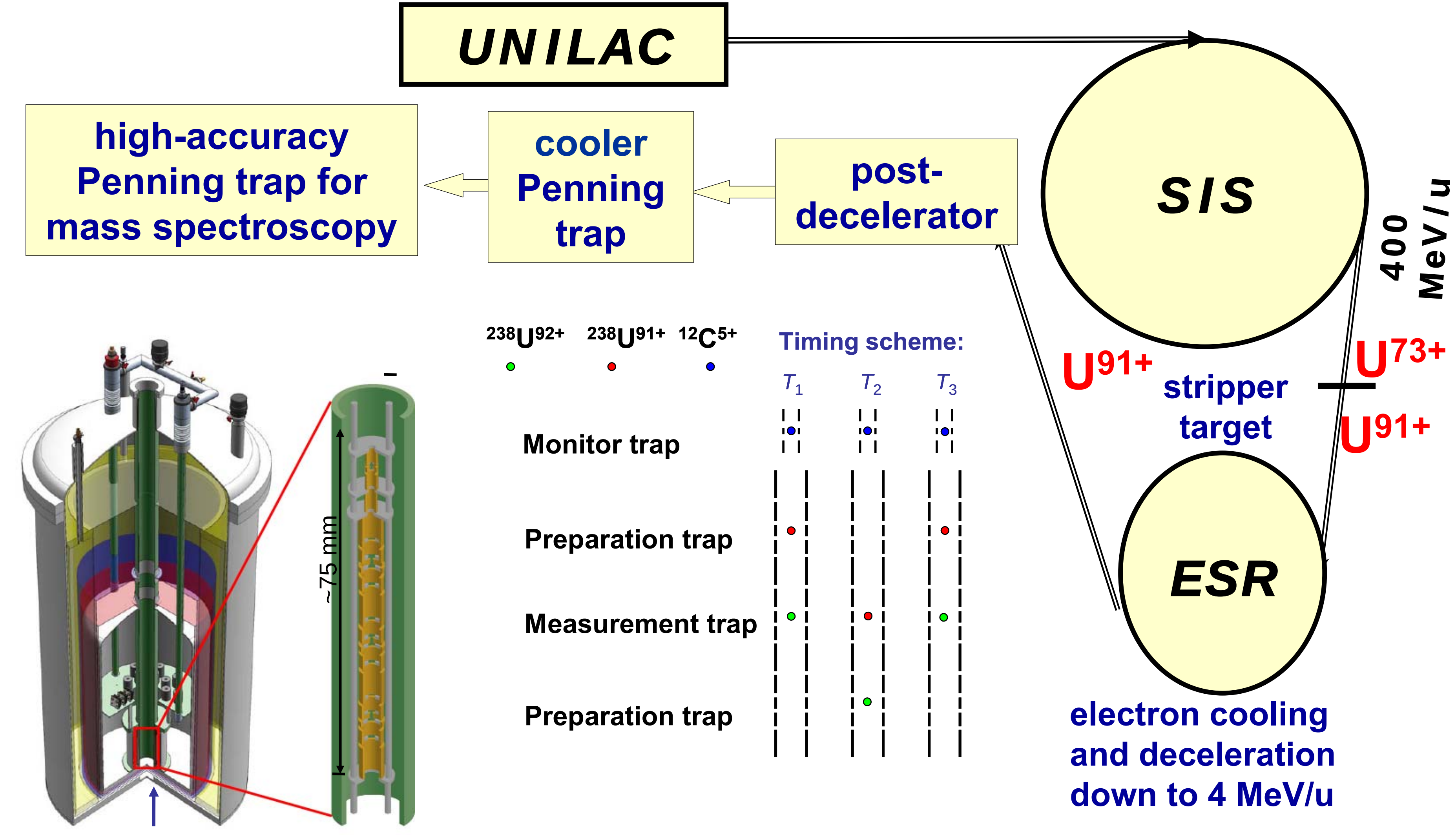
R.S. Van Dyck, Jr., D.L. Farnham, S.L. Zafonte, P.B. Schwinberg, Rev. Scientific Instr. 70 (1999) 1665.
R.S. Van Dyck, Jr., S.L. Zafonte, P.B. Schwinberg, Hyperfine Interactions 132 (2001) 163

10^{-11}

MIT Group

S. Rainville, J.K. Thompson, D.E. Pritchard, Science 303 (2004) 334

HITRAP at the Experimental Storage Ring ESR



UNILAC

SIS

400 MeV/u

U^{73+}

stripper target

U^{91+}

U^{91+}

ESR

electron cooling and deceleration down to 4 MeV/u

post-decelerator

cooler Penning trap

high-accuracy Penning trap for mass spectroscopy

$^{238}U^{92+}$ ● $^{238}U^{91+}$ ● $^{12}C^{5+}$ ●

Timing scheme:

T_1 T_2 T_3

Monitor trap

Preparation trap

Measurement trap

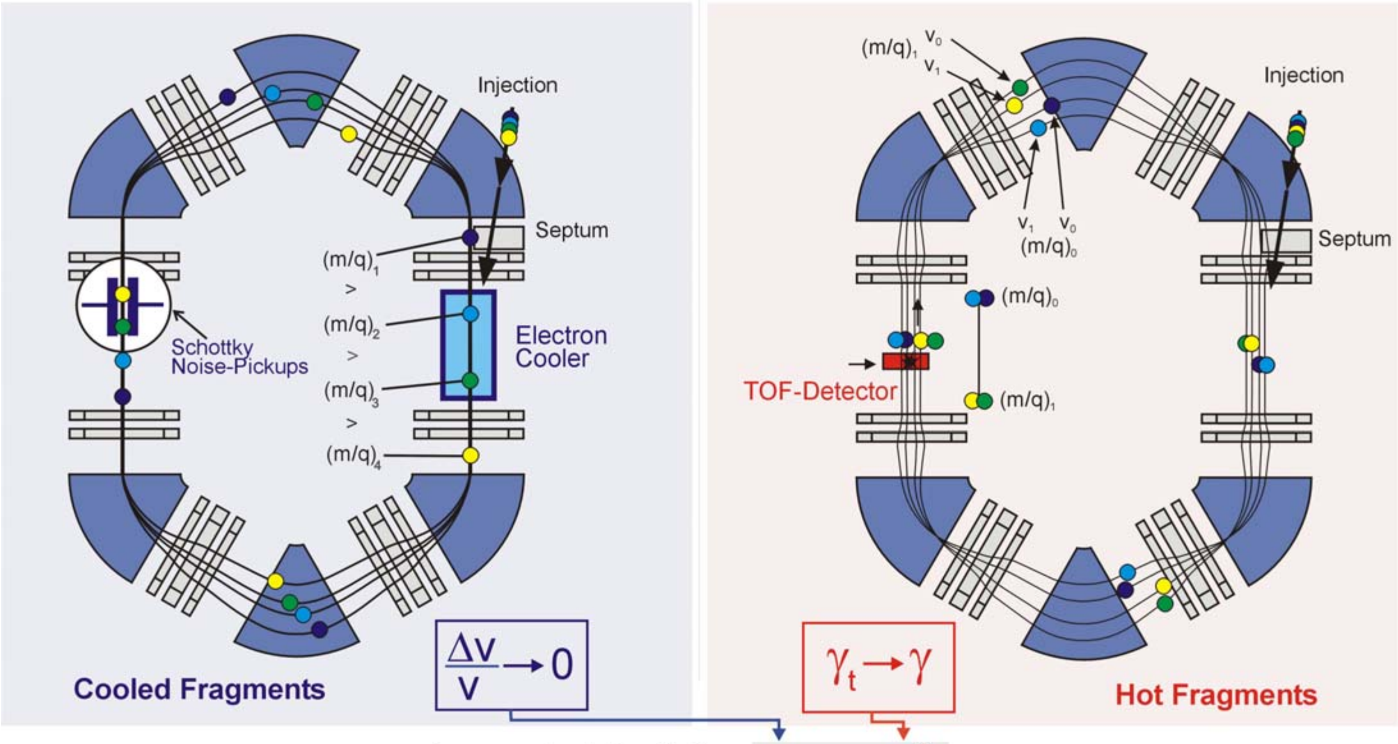
Preparation trap

~75 mm

Ions from HITRAP

K. Blaum et al.

Principle of Mass Spectrometry by Use of a Storage Ring

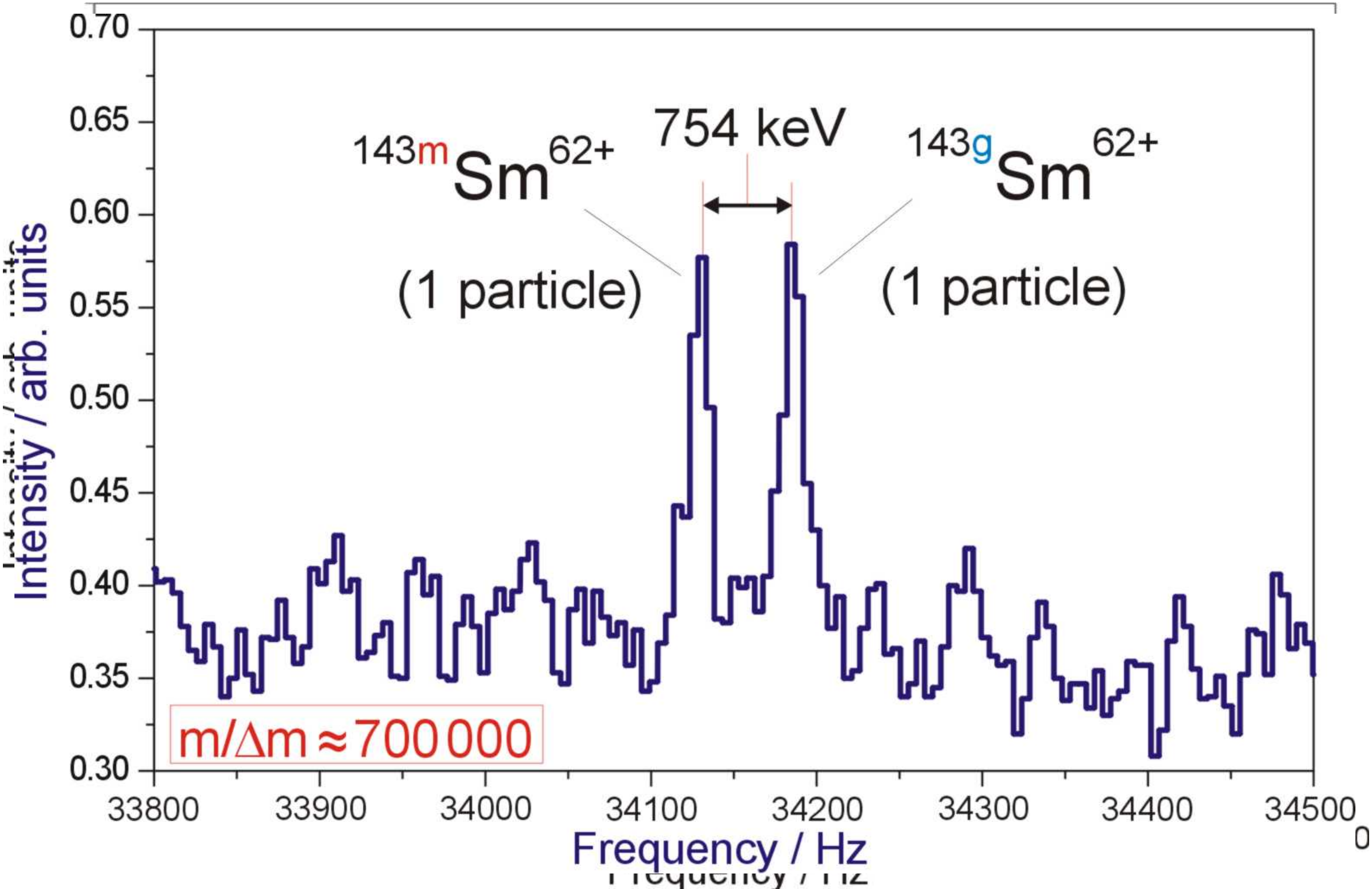


$$\frac{\Delta f}{f} = -\frac{1}{\gamma_t^2}\frac{\Delta(m/q)}{m/q} + \frac{\Delta v}{v}\left(1-\frac{\gamma^2}{\gamma_t^2}\right)$$

electron cooling required
high accuracy
high resolving power
frequency measurement

no electron cooling
short half-life
time-of-flight detection
complex ion optical setting

SMS: Broad Band Frequency Spectra



Measured Mass Surface

Masses of more than 1100 Nuclides were measured

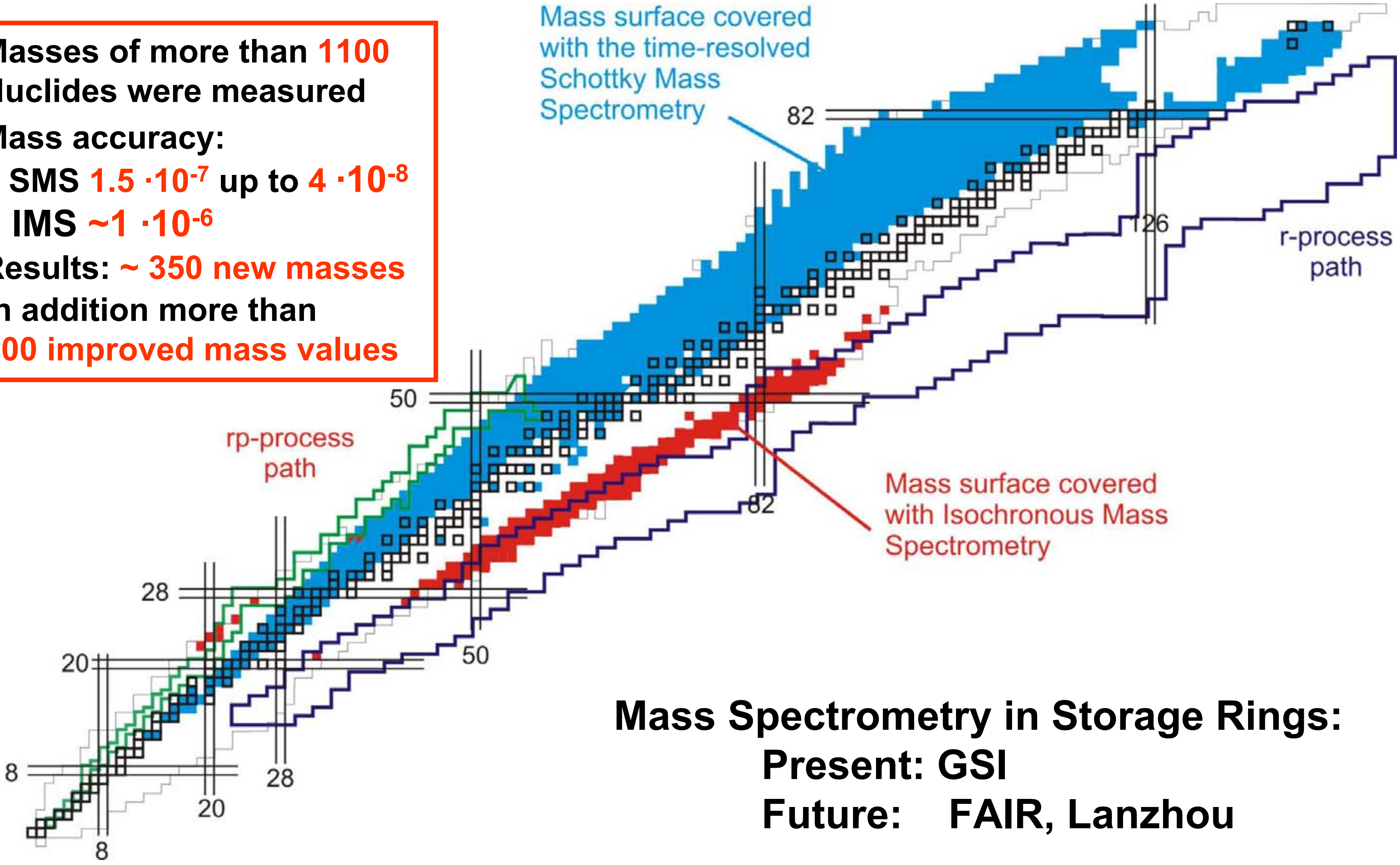
Mass accuracy:

SMS $1.5 \cdot 10^{-7}$ up to $4 \cdot 10^{-8}$

IMS $\sim 1 \cdot 10^{-6}$

Results: ~ 350 new masses

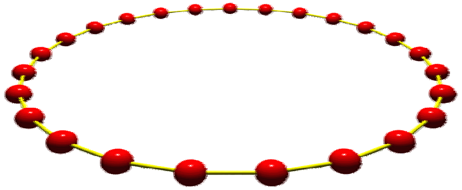
In addition more than 300 improved mass values

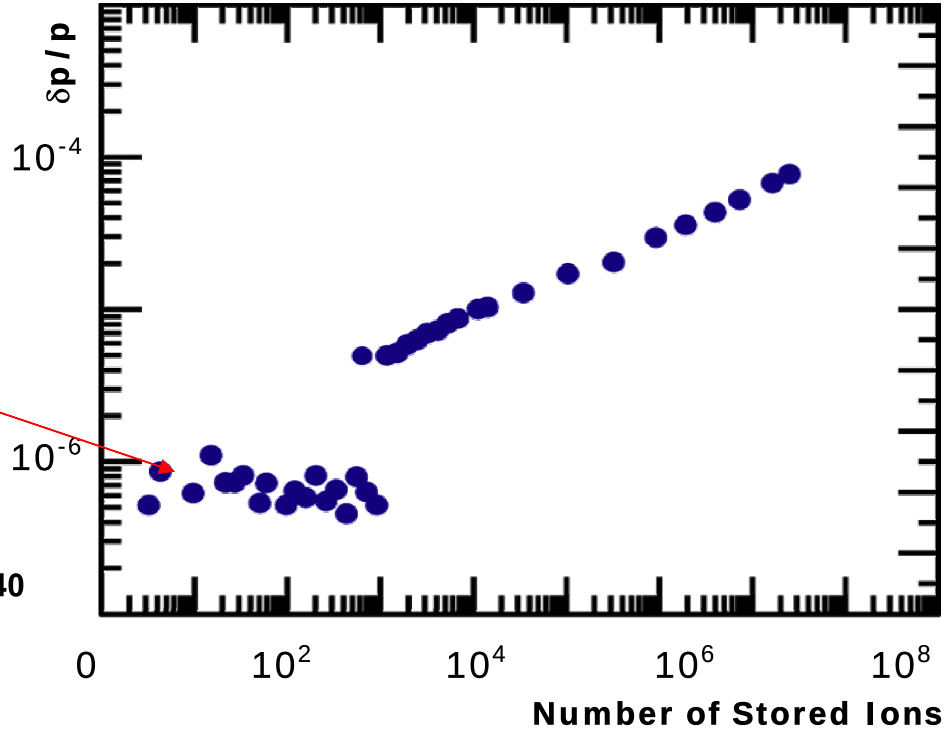


Mass Spectrometry in Storage Rings:
Present: GSI
Future: FAIR, Lanzhou

H. Geissel et al., AIP Conf. Proc. Vol. 831 (2006) 108

How to Reach Highest Accuracy?



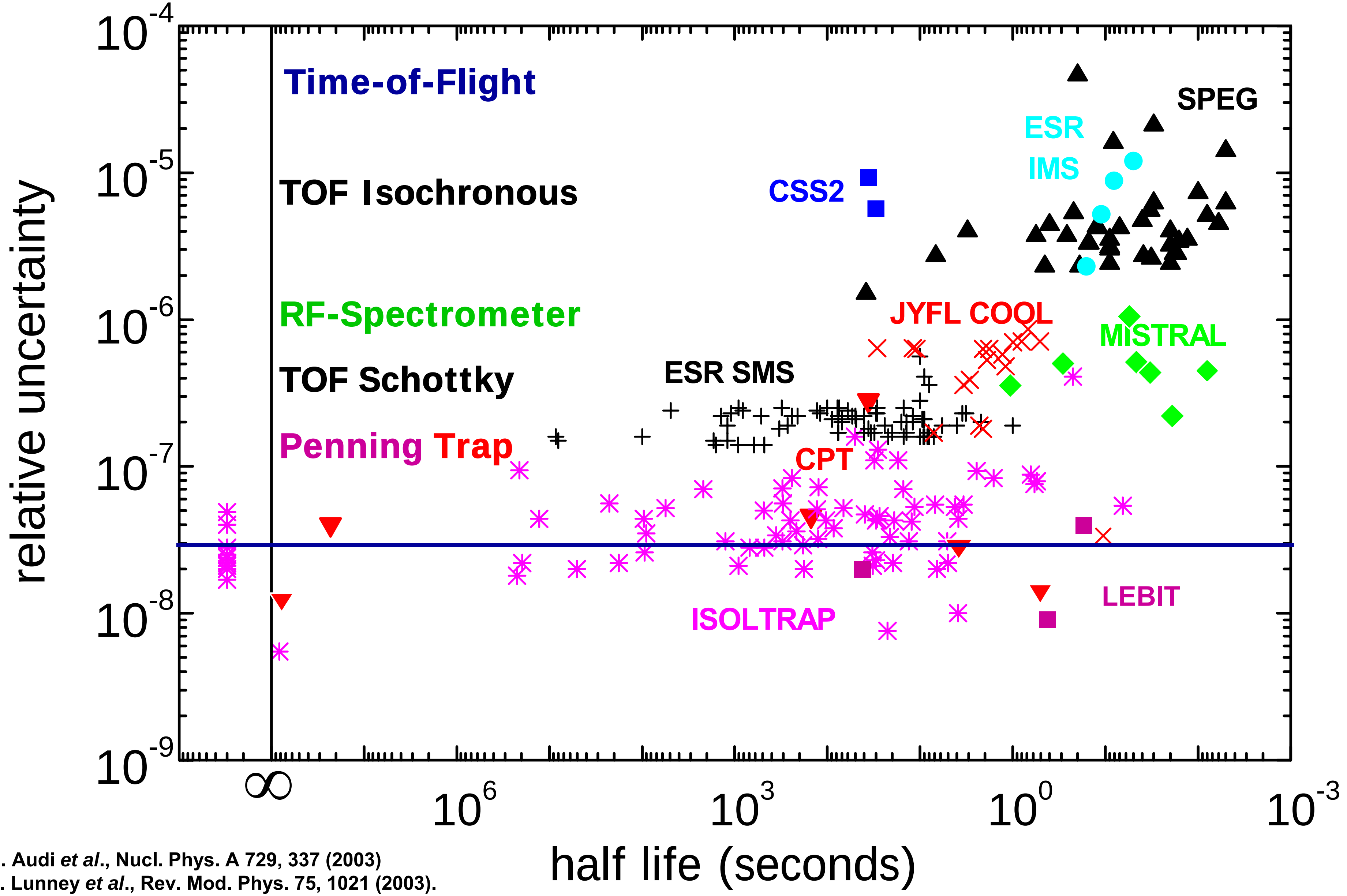


exp: B. Franzke, M. Steck et al.
theo: R.W. Hasse, Phys. Rev. Lett. 83 (1999) 340

Improve
- **stability of the voltage of the electron cooler**
- **stability of the magnetic field**

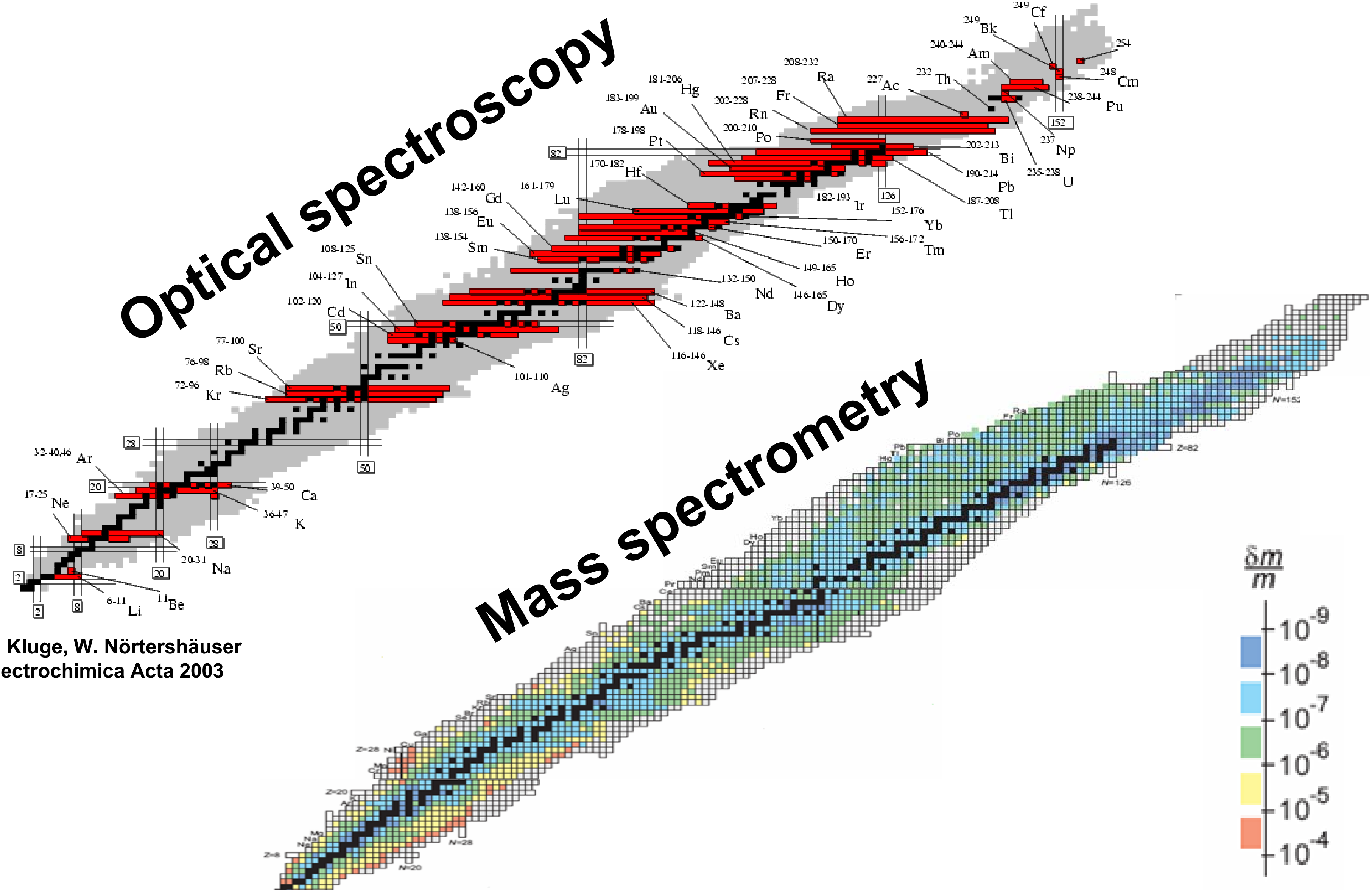
Use
- **multiple low-noise Schottky pick-up electrodes**
- **higher harmonics**

Comparison of Mass Measurements (Published since 2003)



G. Audi *et al*., Nucl. Phys. A 729, 337 (2003)
D. Lunney *et al*., Rev. Mod. Phys. 75, 1021 (2003).

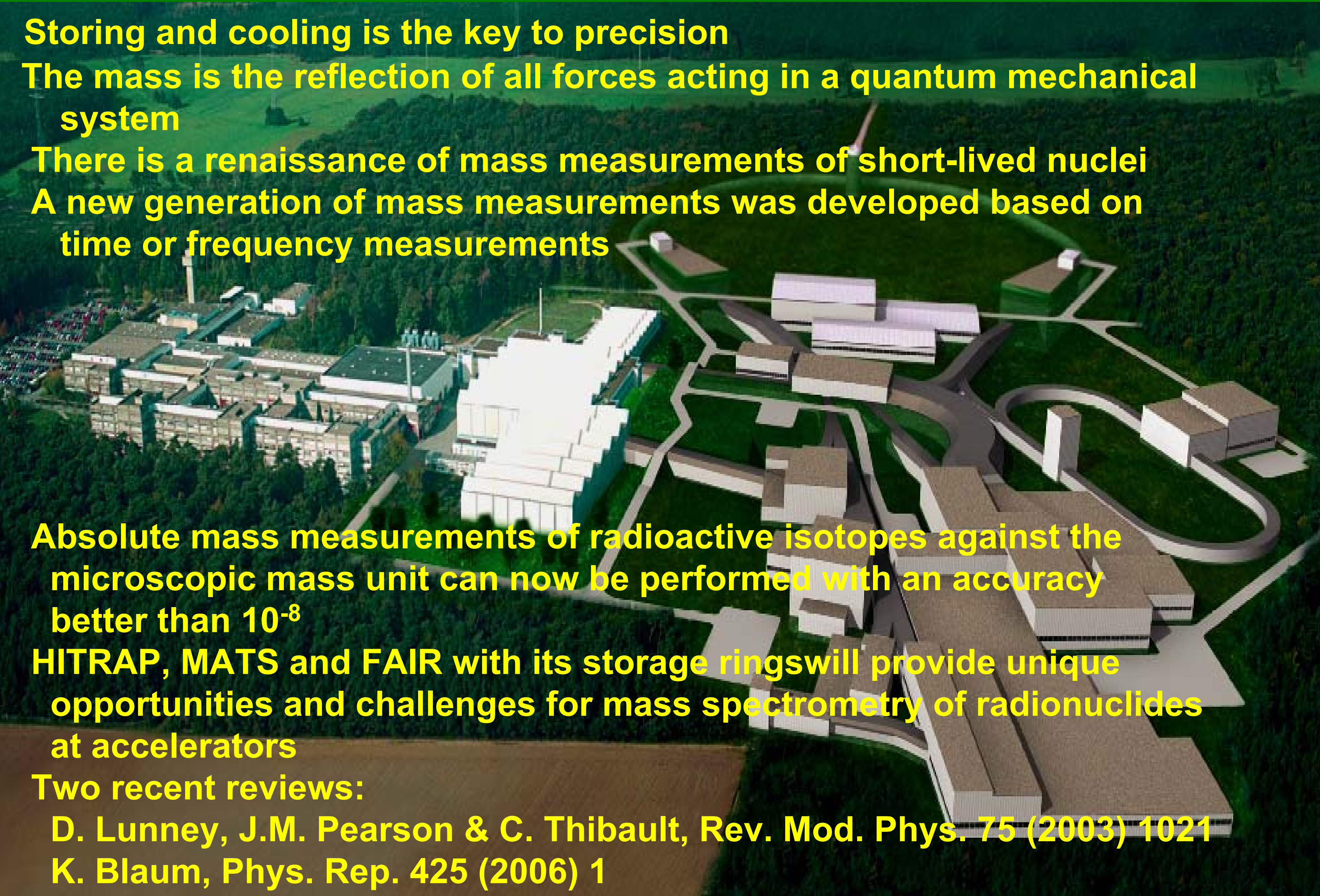
Laser Spectroscopy and Mass Spectrometry in Long Isotopic Chains



H.-J. Kluge, W. Nörtershäuser
Spectrochimica Acta 2003

K. Blaum, Phys. Rep. 425 (2006) 1

Summary and Outlook



Storing and cooling is the key to precision

The mass is the reflection of all forces acting in a quantum mechanical system

There is a renaissance of mass measurements of short-lived nuclei

A new generation of mass measurements was developed based on time or frequency measurements

Absolute mass measurements of radioactive isotopes against the microscopic mass unit can now be performed with an accuracy better than 10^{-8}

HITRAP, MATS and FAIR with its storage ringswill provide unique opportunities and challenges for mass spectrometry of radionuclides at accelerators

Two recent reviews:

D. Lunney, J.M. Pearson & C. Thibault, Rev. Mod. Phys. 75 (2003) 1021

K. Blaum, Phys. Rep. 425 (2006) 1