

Recent advances in mass spectrometry of radioactive nuclei

Tommi Eronen

Outline

- Mass measurement techniques with "stopped" ions
 - Penning trap mass spectrometry (PTMS)
 - Multi-reflection time-of-flight mass spectrometry (MR-TOF-MS)
- Some insights to used ion manipulation methods
 - In-trap decay
 - Retrapping, stacking
- Glimpse to potential new techniques



Atomic mass spectrometry at low energy facilities



- Radioactive beam production at high voltage 5...100 kV platform
 - This is the transfer potential
- and then slowed down with near-same potential to be trapped
- Transfer inside with
 - Low E (1-5 kV)
 - Pulsed drift tubes
- Atomic mass spectrometry with stored ions
 - Penning traps (100 V)
 - MR-TOFS (few kV)

IGISOL facility



(I)

(1,2,4,5,6)

(3)

(c) Ion Injection / Ejection

Storage











- Separation of mass in time-of-flight
- Same E_{kin} for all ions, masses separate in time-of-flight (TOF)

Injection

Pulsed Reflectors

Pulsed Drift Tube

 $- t = a\sqrt{m/q} + b$

- Mass resolving power $R = \frac{M}{\Delta M} = \frac{t}{2\Delta t}$ (= 0.5 ... 10 x 10⁵)
- Trapping
 - Switch mirrors or in-trap lifting

Time-focus considerations

- Aim: time focus at detector
- From RFQ buncher: push-pull creates too early time focus
- Tune spectrometer to focus
 - lons with higher *E* have increased *t*
- Turn-number dependent
 - $\frac{\partial \delta_T}{\partial \delta_E}$ tunable by changing ions *E* inside MRTOF



R. N. Wolf et al., IJMS 313, 8 (2012)

Time-focus tuning for certain turn number



8 4.4.2025

MR-TOF spectrum

- $R = \frac{M}{\Delta M} = \frac{t}{2\Delta t}$
- Non-scanning





Jyväskylä Buncher + MRTOF





Mass measurement of state of ⁹⁴Ag

- Hot-cavity ion source "ISOL with heavy-ion fusion evaporation reaction"
 - Mikael Reponen et al.



JYU SINCE 1863.

Inductively Heated Hot Cavity Cather Laser Ion Source

IGISOL typically: Gas cell. This time no buffer gas in chamber: reaction products caught in graphite catcher.





⁹⁴Ag

- N=Z nucleus, N=Z=47, has 21⁺, 7⁺ and 0⁺ gs
- 21⁺ state is p- and 2p emitter
- 21⁺ state decay:





I. Mukha et al. Nature 439 (2006) 298.

(Non-existence?) of two-proton radioactivity in ⁹⁴Ag 21+ state



- Subsequent study by Cerny et al. confirmed the 0.79 MeV proton, but saw no evidence for two-proton decay or the other 1.01 MeV single proton
- J. Cerny PRL 103, 152502 (2009)

A. Kankainen et al. PRL 101, 142503 (2008)

Mass measurement of 21⁺ and 7⁺ ⁹⁴Ag states

Laser 1st step used to select the state to be ionized



How about 0⁺ ground state?



Laser frequency scan



⁹⁴Ag measurements conclusion

- Mukha measured: $E_{2p} = ~1.9$ MeV
- Our measurement:
 - 94Ag(21+) to ⁹²Rh ground state Q_{2p} (94Ag (21 +) → 92Rh(6+)) < 1.9 MeV
- 2p decay not energetically possible

M. Reponen and V. Virtanen et al. (to be published) 2025

(At this moment, MR-TOF assisted laser-spectroscopy measurement running!)

N=Z hot cavity + MR-TOF measurement outlook



• ⁹³Pd:

- p-decay daughter of ⁹⁴Ag
- Measured also at FRS ion catcher @ GSI



• ⁹²Pd (N=Z=46)



JYU SINCE 1863.

Progress so far... and possible future mass adventures



• Hot-cavity + MRTOF is a powerful combination

MRTOF in-trap contamination removal



RIKEN zero-degree spectrometer M. Rosenbusch et al. NIMA 1047, 167824 (2023)



FRS ion catcher MRTOF S. Ayet et al. PRC 99, 064313 (2019)

MRTOF is the method of choice for $T_{1/2}$ < 50 ms

- For N=Z nuclei below ¹⁰⁰Sn
- ISOLTRAP Sn isotopes
 - Down to ¹⁰³Sn
- Smoothening of the mass surface





JYU SINCE 1863.



Penning trap mass spectrometry at IGISOL



Penning trap mass spectrometry

- Strong magnetic field B + electrostatic quadrupole potential
- Three in-trap motions:
 - Axial (z), frequency $v_z = \frac{1}{2\pi} \sqrt{\frac{qV_0}{md^2}}$
 - Radial (+,-) frequencies $v_{\pm} = \frac{1}{2} \left(v_c \pm \sqrt{v_c^2 2v_z^2} \right)$
- To measure: free-space cyclotron frequency $v_c = \frac{1}{2\pi}\omega_c = \frac{1}{2\pi}\frac{q}{m}B$
 - Through side-band coupling frequency

$$v_- + v_+ = v_c$$





Phase detection method with 2D ion detection



- For exact set duration of *t_{acc}*, let ions
- 1. revolve in v_+ motion (mass dependent)
- 2. revolve in ν_{-} motion (mass independent)
- Both accumulate phase $2\pi n + \phi$
- Detect ϕ_+ and ϕ_- , figure out $\mathrm{n_-}$ and $\mathrm{n_+}$

It is best to set t_{acc} to be exact multiples of $\frac{1}{v_c}$,

- $\phi_- - \phi_+$ will vanish (or becomes small)

$$\nu_c = \frac{(\phi_+ - \phi_-) + 2\pi n_c}{2\pi t_{acc}}$$

$$R = \frac{M}{\Delta M} = \frac{\nu_+}{\Delta \nu_+} > 10^6$$
, beyond 10⁷







-max

Mass measurements of isomers of n-rich nuclei

- PI-ICR technique $R = \frac{M}{\Delta M} > 10^7$
- Allows separation down to ~10 keV level
- Even A n-rich Rh isomers
 - Hukkanen et al. PRC 107, 014306 (2023).





Comparison to TOF-ICR



Also measured at Canadian Penning trap: https://arxiv.org/abs/2410.00389

¹¹⁴Rh states revisited





Stryjczyk PLB 862, 139359 (2025)

N-rich Ag isomers

- A. Jaries et al. PRC 110, 034326 (2024)
- J. Ruotsalainen et al. Arxiv 2408.14181 (2024)
- ¹²²Ag: NUBASE20, ENSDF: 3 states

- JYFLTRAP: only 2 seen, m1 is actually gs!





S_{2n} values of n-rich Ag

Smooth all the way



32 4.4.2025

Combatting contaminants and gaps in yield

In-trap β^- decay assisted PTMS

- Let singly-charged parent ion to decay in Penning trap
- Daughter is 2+ charged, recoil remains trapped
- Simply: Capture bunch to gas-filled preparation trap
- Wait (ideally) few half-lives
- Use sideband buffer gas cooling technique to clean rest away
- Do PI-ICR
 - Also gained x2 precision due to q=2e!
- ¹¹⁴Rh case: Learned also that ¹¹⁴Ru beta decay does not feed ¹¹⁴Rh gs
- HELPS to fill gaps in primary production (here p+U)

$\Delta E = 22.4(9) \text{ keV }!$



Combination of different mass filters

Purification Penning trap + PI-ICR



35 4.4.2025

Independent fission yields

- Mass spectroscopy means
 - Penning trap purification trap (scanning)
 - MR-TOF (non-scanning)





36 4.4.2025

Rami Korkiamäki MSc thesis Thorium-232 proton induced fission yield distribution http://urn.fi/URN:NBN:fi:jyu-202406124567

New developments

- MRTOF+JYFLTRAP coupling
 - Demonstrated but not yet efficient
- Many others..



PI-ICR with ultracold ions

KU LEUVEN

Collaboration being formed



KU LEUVEN State-selective mass spectrometry with RAPTOR 30 kV 4-pole Buncher 2-5 kV bender 2 kV Neutralization + reionization with lasers MRTOF - State selection RAPTOR - "Low energy CRIS" Unambigious spin+mass info! 2 kV 500 V 30 kV 4-pole **JYFLTRAP** 2 kV Penning trap bender

Mass-separation assisted laser spectroscopy



Stacking trap for accumulation of pure ion sample



JYU SINCE 1863.

IGISOL is space-limited after buncher.



Exotic front vs precision front



Precision measurements, "close" to stability

- Expect high yields, particular impurities
- Standard Model testing
- Neutrino physics
- PI-ICR PTMS, with ultracold ions
- Expand high-precision work from stable to radioactive nuclei
 - E.g. King plot linearity tests

Exotic nuclei, very far from stability

- Expect low yields and lots of contamination
- High contamination ratios
 - Pre-separation essential
- Masses with MR-TOF
 - Still, e.g. ¹⁰¹Sn was measured at FRIB with PI-ICR at LEBIT trap

Thank you for listening!



JYU SINCE 1863.

JYFLTRAP magnet quench and re-energization

- Energized ourselves (3rd time was a charm), shimmed too
- Thanks MLLtrap (Enrique) ffinallyor NMR Tesla-probe
- Thanks SHIPTRAP for shimming tools
- And we at JYFLTRAP have the main current lead stick



