

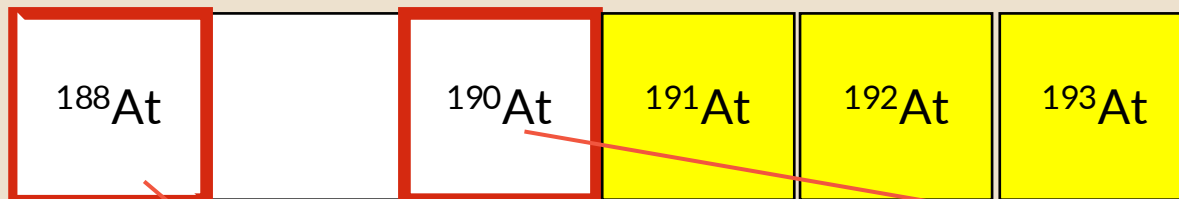


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# Decay spectroscopy studies on the two new isotopes of astatine

Henna Kokkonen

# $^{188}\text{At}$ & $^{190}\text{At}$



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## New proton emitter $^{188}\text{At}$ implies an interaction unprecedented in heavy nuclei

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We report the discovery of a new atomic nucleus  $^{188}\text{At}$ , which is the heaviest proton-emitting isotope known to date. The new activity was observed through the  $^{107}\text{Ag}(^{84}\text{Sr}, 3n)^{188}\text{At}$  fusion-evaporation reaction using the focal-plane spectrometer of the gas-filled recoil separator in the Accelerator Laboratory of the University of Jyväskylä, Finland. To fully interpret the experimental data, we have expanded the non-adiabatic quasiparticle model to treat nuclei in the beyond-lead region. The description reproduced the measured decay rate and pointed towards emission from an extremely prolate-deformed state with a dominant  $s_{1/2}$  proton component in the wave function. The Thomas-Ehrman shift can be enhanced in low angular momentum states, but such effects have not been observed in heavy nuclei. The single-proton separation energy of  $^{188}\text{At}$  deviates from that extrapolated from the systematics, which can be interpreted as the first evidence of this effect in heavy nuclei.

PHYSICAL REVIEW C **107**, 064312 (2023)

### Properties of the new $\alpha$ -decaying isotope $^{190}\text{At}$

H. Kokkonen<sup>1,\*</sup>, K. Auranen<sup>1</sup>, J. Uusitalo<sup>1</sup>, S. Eeckhaud<sup>1</sup>, T. Grahn<sup>1</sup>, P. T. Greenlees<sup>1</sup>, P. Jones<sup>1,†</sup>, R. Julin<sup>1</sup>, S. Juutinen<sup>1</sup>, M. Leino<sup>1</sup>, A.-P. Leppänen<sup>1,‡</sup>, M. Nyman<sup>1,§</sup>, J. Pakarinen<sup>1</sup>, P. Rahkila<sup>1</sup>, J. Sarén<sup>1</sup>, C. Scholey<sup>1,¶</sup>, J. Sorri<sup>1,||</sup> and M. Venhart<sup>1,2</sup>

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The  $\alpha$  decay of a new isotope  $^{190}\text{At}$  has been studied via the  $^{109}\text{Ag}(^{84}\text{Sr}, 3n)^{190}\text{At}$  fusion-evaporation reaction by employing a gas-filled recoil separator. An  $\alpha$ -particle energy of 7750(20) keV and a half-life of  $1.0^{+1.4}_{-0.4}$  ms were measured. The measured decay properties correspond to an unhindered  $\alpha$  decay, suggesting the same spin and parity of ( $10^-$ ) as those of the final state of the decay. The systematics of the nearby nuclei and the predictions of selected atomic mass models were compared with the measured decay properties.

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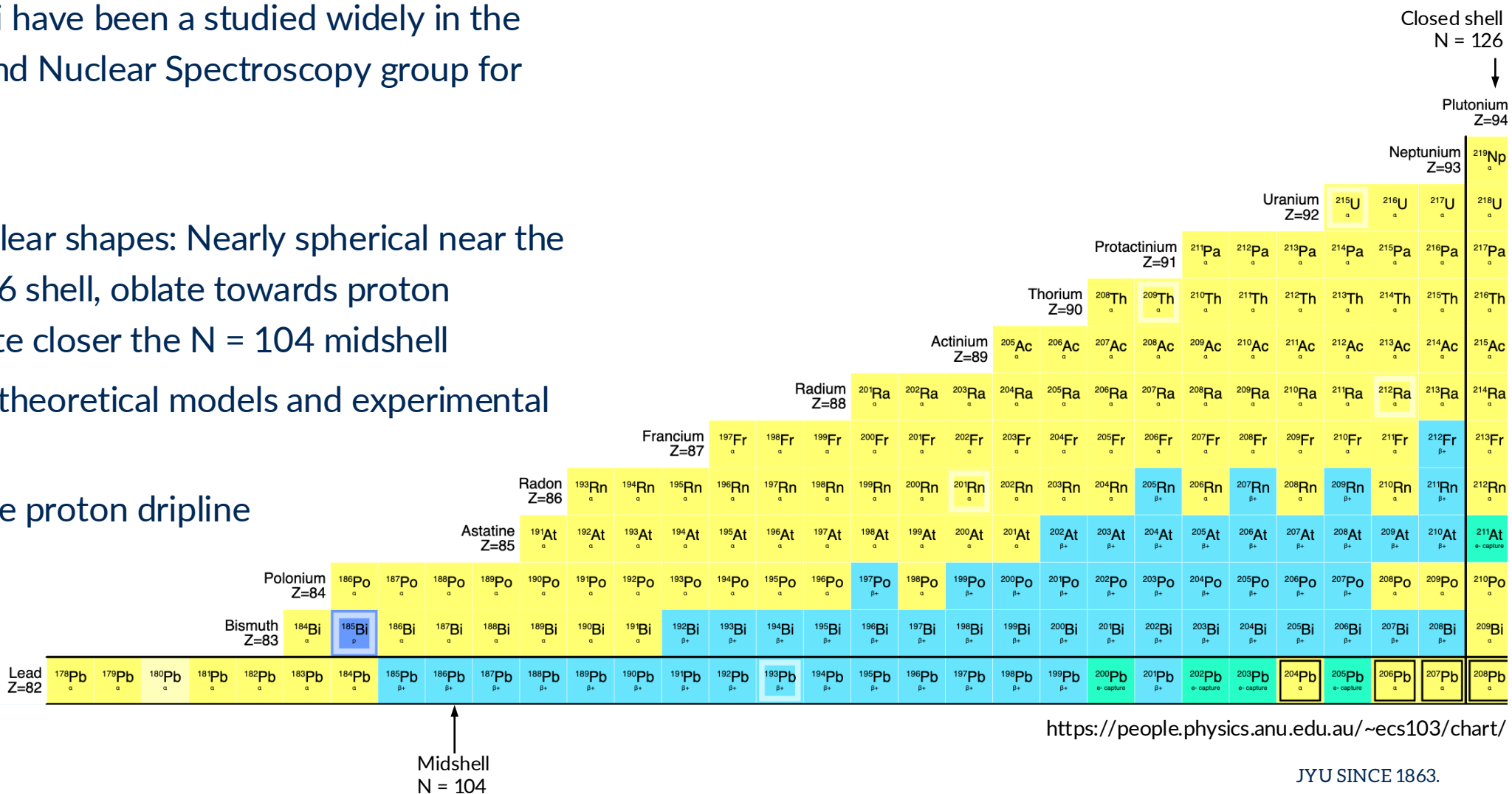
# Introduction



Trans-lead nuclei have been studied widely in the JYFL-ACCLAB and Nuclear Spectroscopy group for several decades

## Why?

- Multitude nuclear shapes: Nearly spherical near the closed  $N = 126$  shell, oblate towards proton dripline, prolate closer the  $N = 104$  midshell
- Challenge for theoretical models and experimental methods
- Location of the proton dripline



# Introduction



## Neutron-deficient astatine nuclei in general:

Odd-mass astatine nuclei have been studied via  $\gamma$ -ray spectroscopy

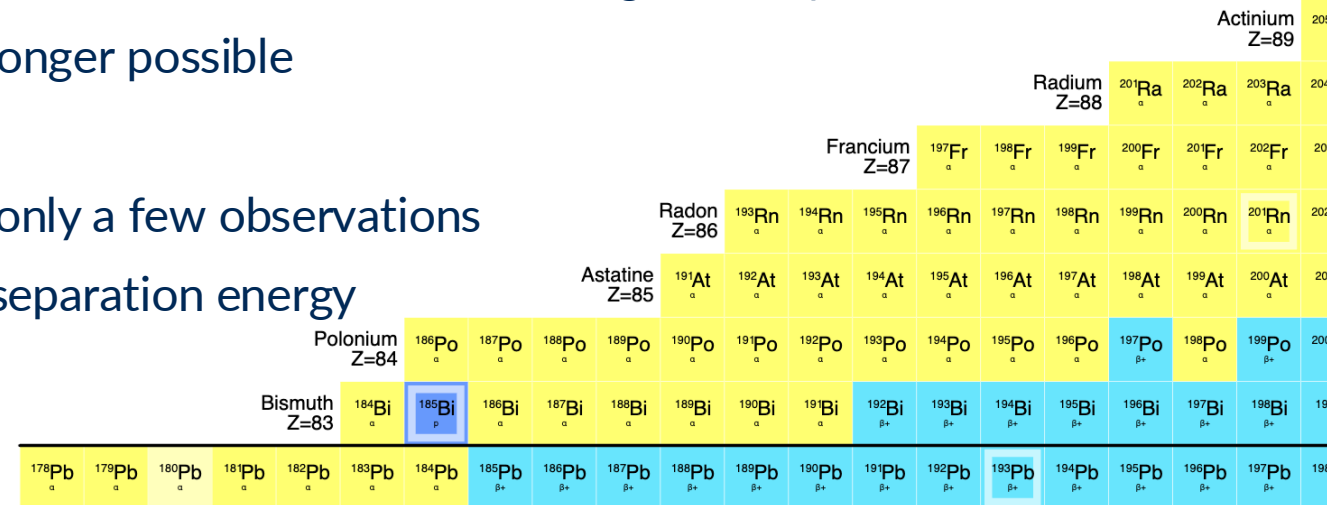
- Spherical/weakly oblate shape down to  $^{197}\text{At}$ ,  $9/2^-$  g.s., isomeric state  $1/2^+$
- The At nuclei become deformed as the mass decreases,  $1/2^+$  becomes ground state at  $^{195}\text{At}$

Towards lighter astatine nuclei, production cross sections and half-lives decreases significantly

- $\gamma$ -ray spectroscopy and laser spectroscopy are no longer possible

**Decay spectroscopy** efficient technique to study even only a few observations

- Particle energy, half-life, mass-excess, one-proton separation energy



<https://people.physics.anu.edu.au/~ecs103/chart/>

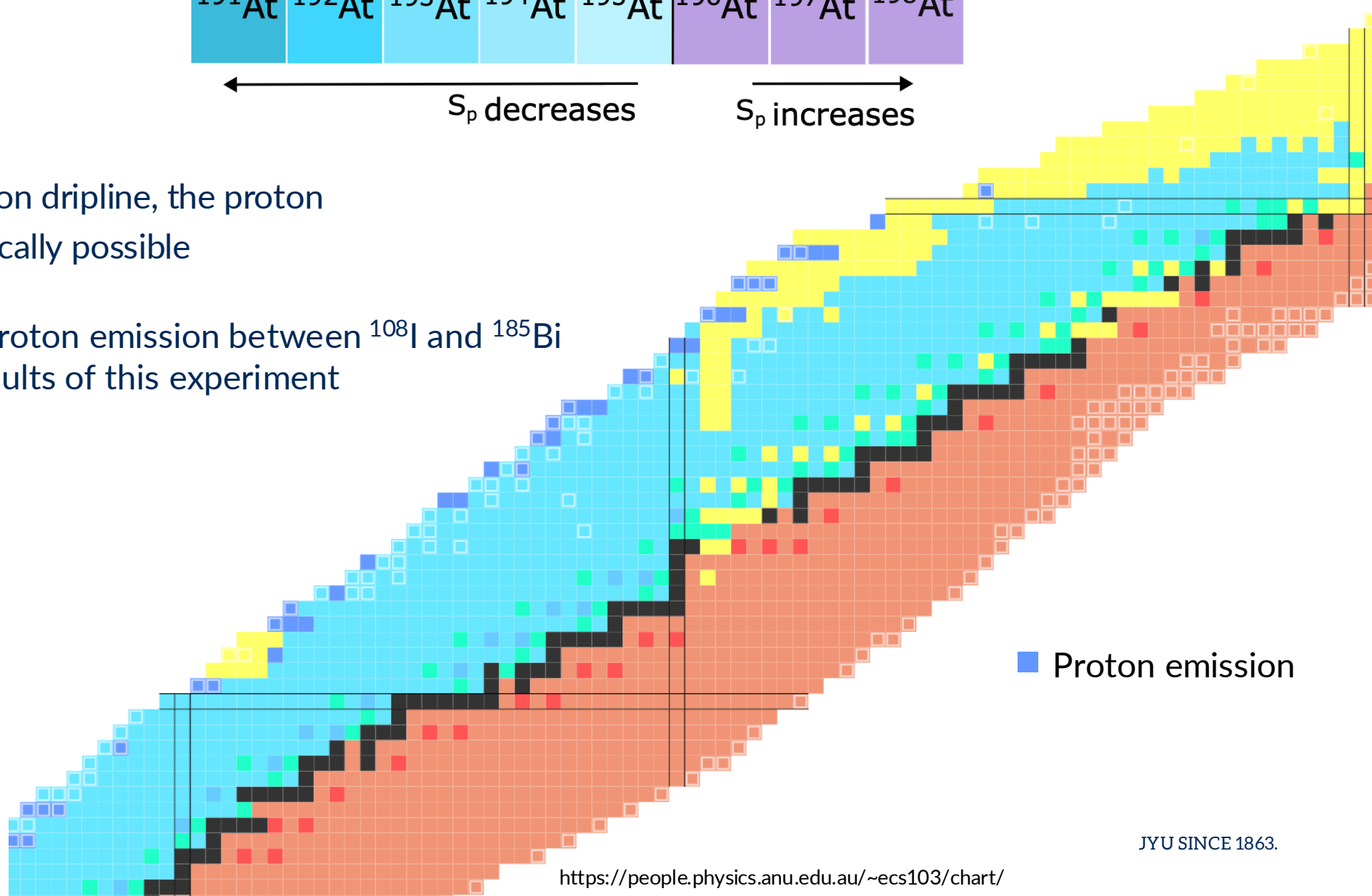
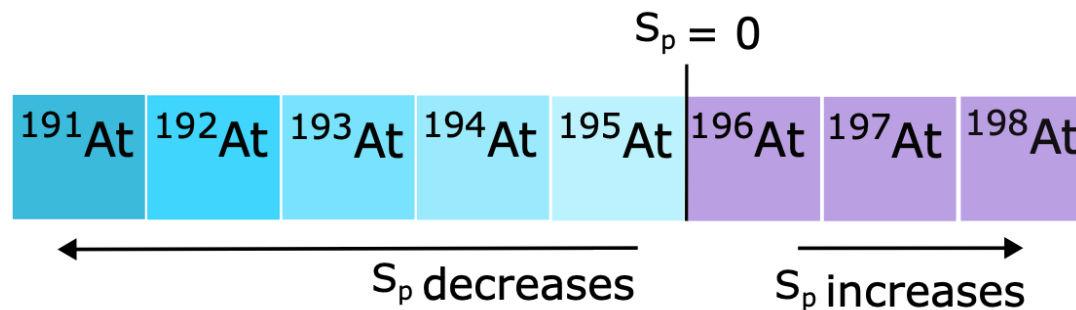
K. Auranen, et al. Phys. Rev. C 90, 024310 (2014), K. Auranen, et al. Phys. Rev. C 95, 044311 (2017)  
 M. Nyman, et al. Phys. Rev. C 88, 054320 (2013), H. Kettunen, et al. Eur Phys J A 16, 457–467 (2003)

# Introduction

## Proton emission

Moving beyond the proton dripline, the proton emission comes energetically possible

- Approx 50 cases of proton emission between  $^{108}\text{I}$  and  $^{185}\text{Bi}$  known before the results of this experiment

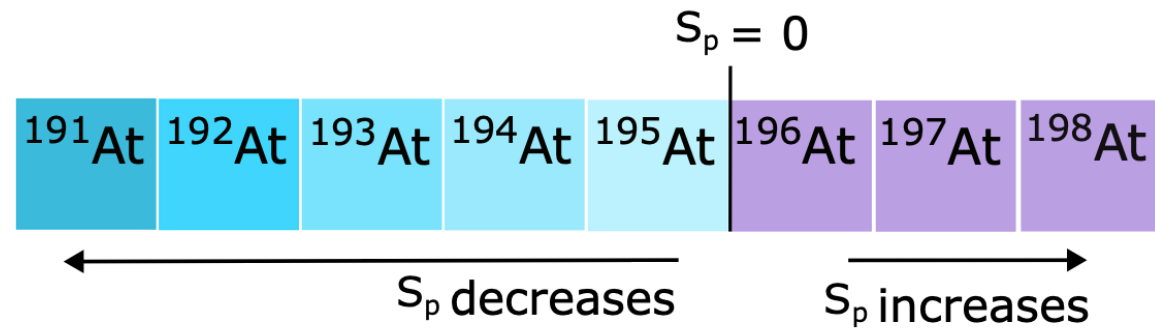


# Introduction



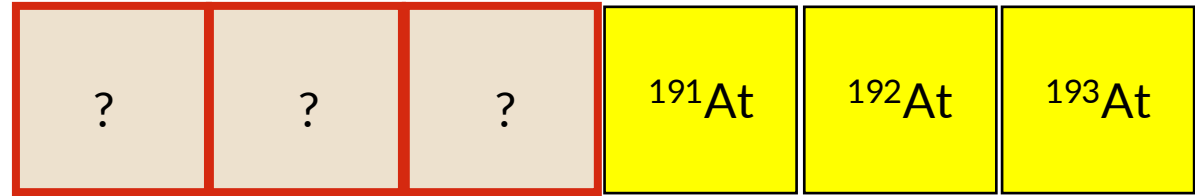
## Proton emission

Moving beyond the proton dripline, the proton emission comes energetically possible

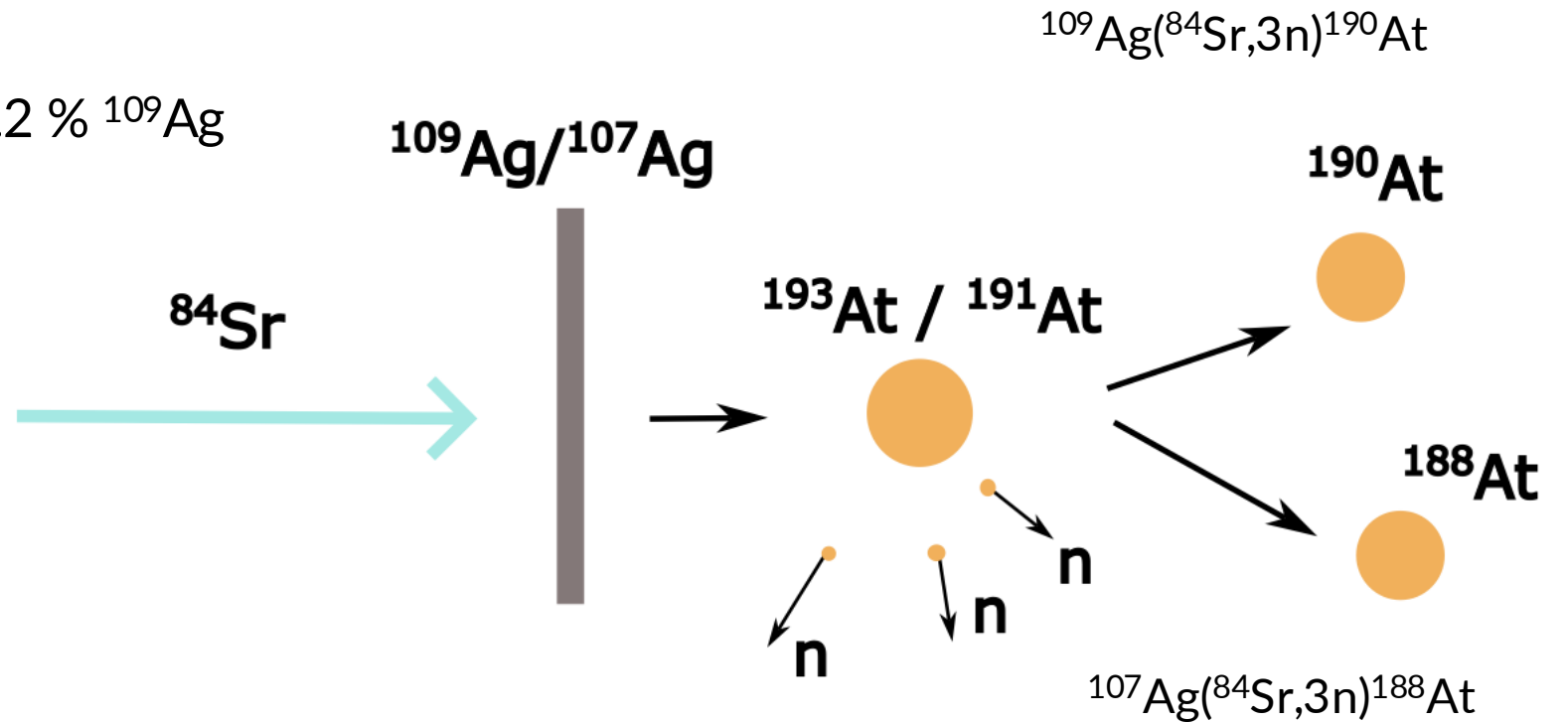


- Approx 50 cases of proton emission between  $^{108}\text{I}$  and  $^{185}\text{Bi}$  known before the results of this experiment
- Proton-emission rate depends on the particle energy, angular momentum, and the deformation
  - Spherical proton emitters can be described by simple phenomenological models, well-deformed become more complex to model
  - *Non-adiabatic quasiparticle model*

# Experimental methods

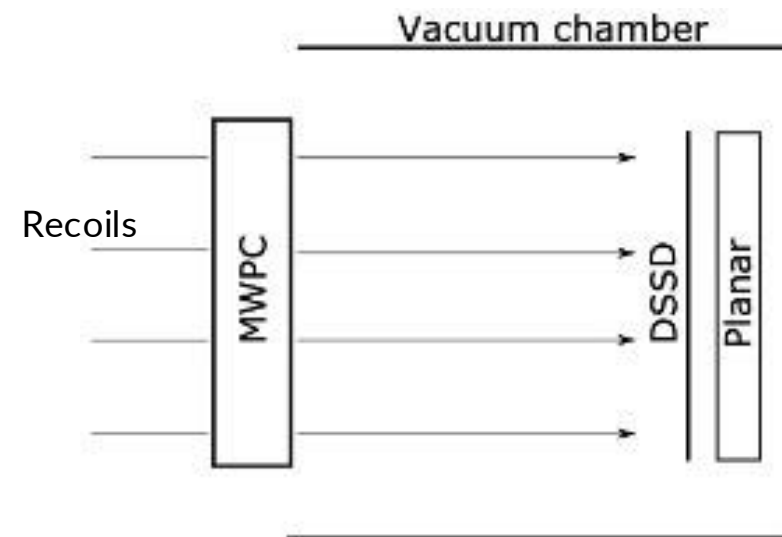
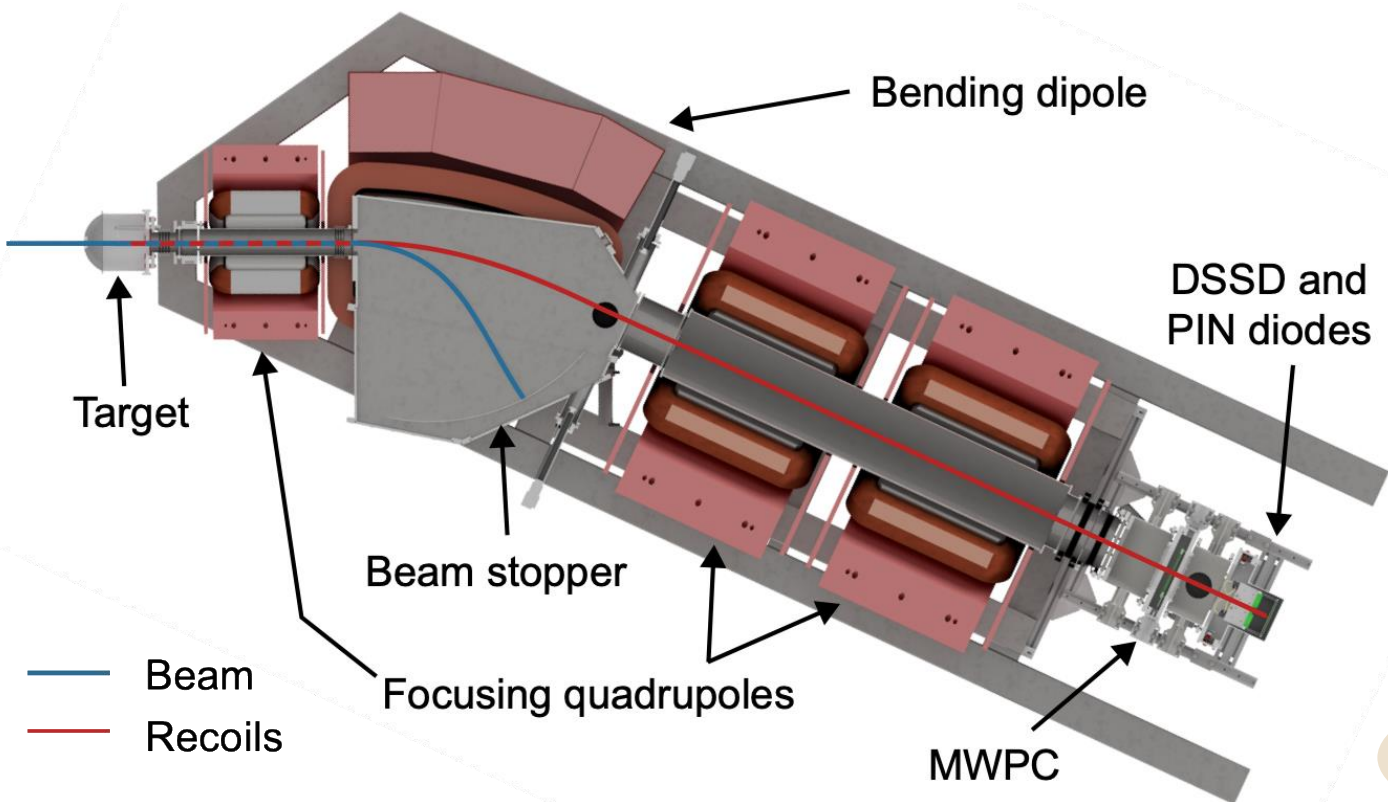


- JYFL-ACCLAB, Finland
- Fusion-evaporation reaction
- $^{nat}\text{Ag}$  target: 51.8 %  $^{107}\text{Ag}$ , 48.2 %  $^{109}\text{Ag}$
- Beamtime 181 h = 7.5 days



# Experimental methods

- K130 cyclotron
- RITU (Recoil Ion Transport Unit):



Detector chamber including MWPC, DSSD, planar

To count an event as a decay, it was required to be measured only in the DSSD and in specific time window.

Gas-filled separator: High transmission. Gas interacts with recoils changing the charge state.





# Decay spectroscopy on $^{190}\text{At}$

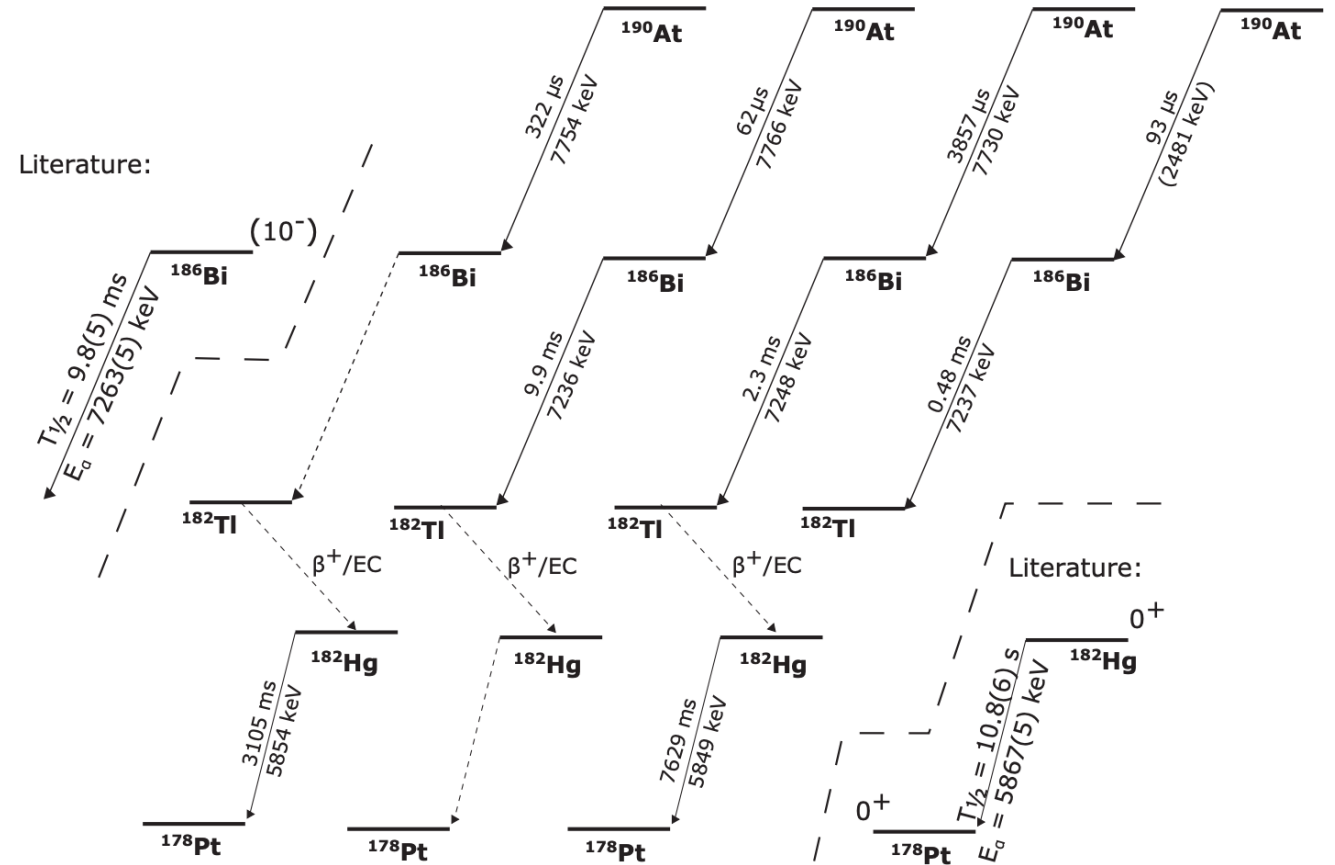
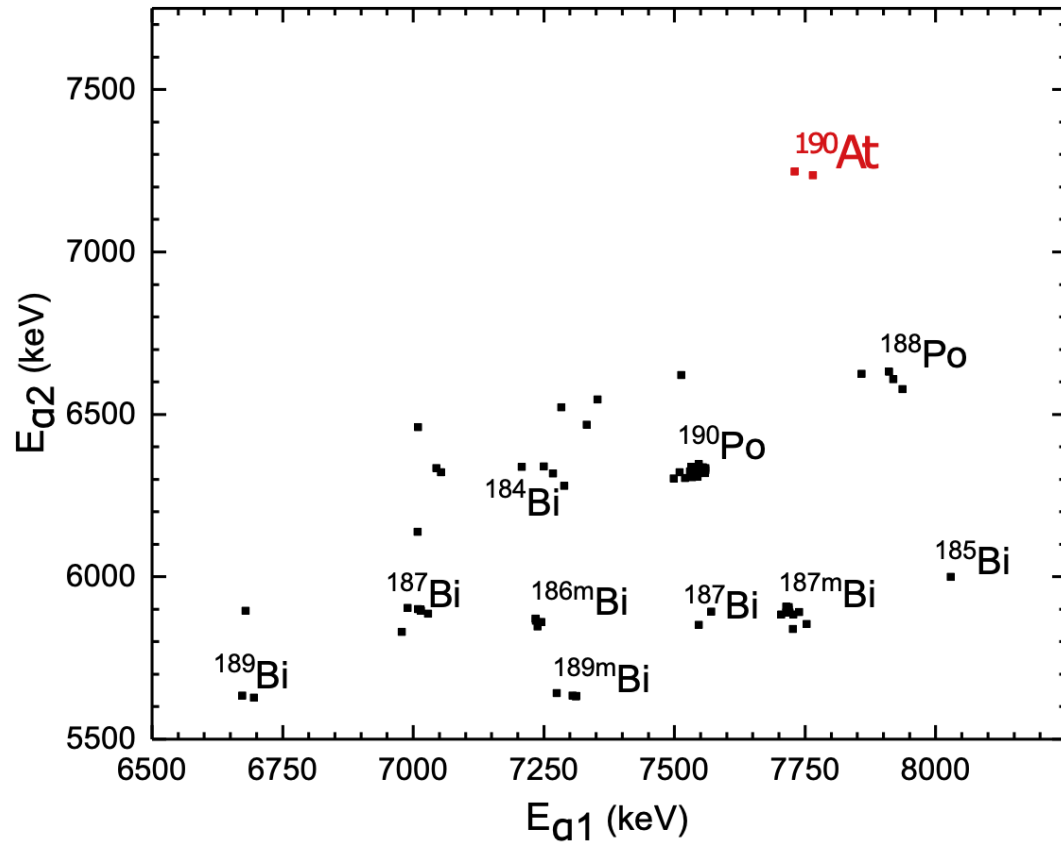
# Decay spectroscopy on $^{190}\text{At}$

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- Four  $\alpha$  - decay events measured:

$$E_\alpha = 7750(20) \text{ keV}$$

- $T_{1/2} = 1_{-0.4}^{+1.4} \text{ ms}$



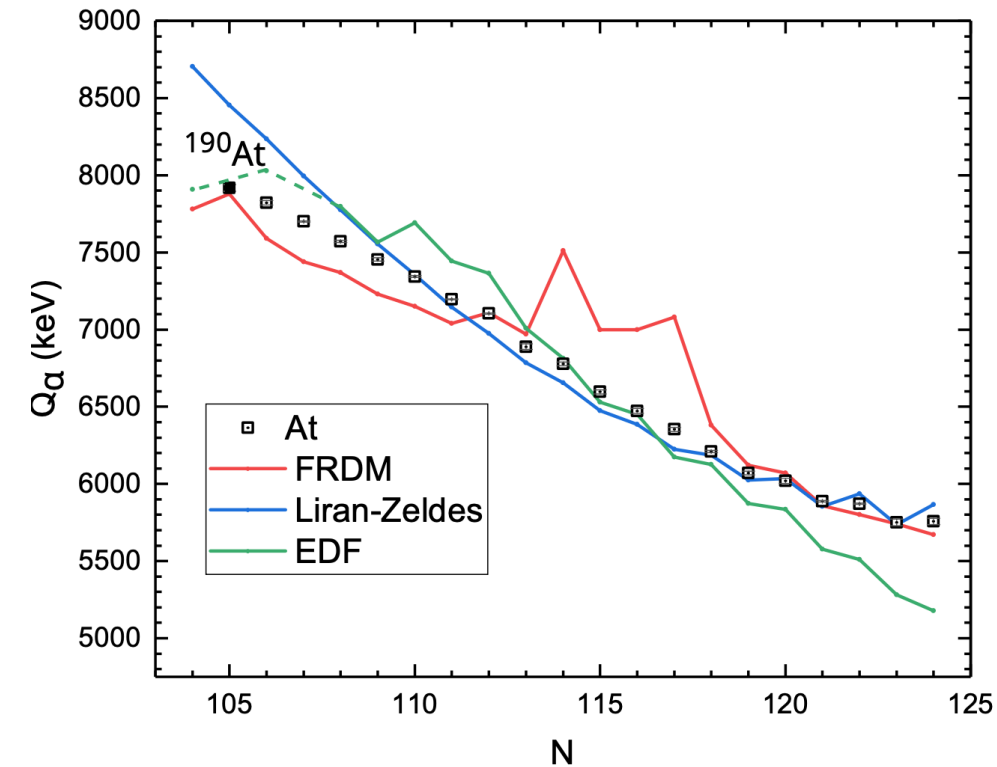
# Discussion: $^{190}\text{At}$

Results were compared to the systematics and different mass model predictions.

Possibility for proton emission was considered by estimating the partial half-life:

- *Proton emission Q value* extracted from the measured results: 1330(40) keV
- Suggested ( $10^-$ ) state for the observed  $\alpha$  decay
  - $^{186}\text{Bi}$  ( $10^-$ )
  - Calculated hindrance factor close to one
  - Fusion-evaporation favours high-spin states

- ➔ Partial half-life for proton emission was approximated to be 30 s ( $h_{9/2}$  state), 90 ms ( $f_{7/2}$  state), 2.5 ms ( $s_{1/2}$ ). However, it is not possible to couple  $s_{1/2}$  proton to any expected neutron state and obtain spin and parity of  $10^-$ 
  - Cannot compete with the observed  $\alpha$  decay



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# Decay spectroscopy on $^{190}\text{At}$



- ✓ Results confirmed later by an independent experiment performed in ANL by A. N. Andreyev, D. Seweryniak, et al.

PHYSICAL REVIEW C **108**, 034303 (2023)

## $\alpha$ decay of the neutron-deficient isotope $^{190}\text{At}$

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The  $\alpha$  decay of the neutron-deficient  $^{190}\text{At}$  isotope was observed following the  $^{103}\text{Rh}(^{90}\text{Zr}, 3n)^{190}\text{At}$  reaction at Argonne National Laboratory. The reaction products were separated from the beam using the Argonne Gas-Filled Analyzer and implanted into a double-sided Si strip detector. The spatial and temporal correlations between implanted nuclei and subsequent  $\alpha$  decays towards the known daughter isotope  $^{186}\text{Bi}$  were used to identify and characterize  $^{190}\text{At}$  nuclei. Two possible decay scenarios are proposed for the  $^{190}\text{At} \rightarrow ^{186}\text{Bi}$  decay.

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### I. INTRODUCTION

Neutron-deficient astatine ( $Z = 85$ ) nuclei demonstrate a rich variety of shape coexistence and shape evolution phenomena. The ground states of the odd- $A$  isotopes with  $A \geq 197$  are usually associated with the occupation of the proton  $\pi h_{9/2}$  orbital, leading to  $J_{\pi}^{\pi} = 9/2^{-}$  [1,2]. Moving away from the  $N = 126$  shell closure towards the neutron-deficient side, the excitation energies of  $1/2^{+}$  and  $7/2^{-}$  states, where known, decrease as a function of the neutron number, resulting in low-lying  $1/2^{+}$  isomers in  $^{197,199,201,203}\text{At}$ . In the lighter isotopes  $^{191,193,195}\text{At}$ , the  $1/2^{+}$  state becomes the ground state, with the closely lying  $7/2^{-}$  isomeric states at excitation energies as low as 50(30) keV in  $^{191}\text{At}$  and 5(10) keV in  $^{193}\text{At}$  [1]. These  $1/2^{+}$  states have been interpreted as spherical-shell model intruder states, created by  $\pi(4p-1h)$  proton excitations across the  $Z = 82$  shell gap, resulting in a proton hole in the  $s_{1/2}$  orbital. Alternatively, in the deformed shell-model approach, such  $1/2^{+}$  states are associated with the population of oblate-deformed Nilsson orbitals.

In the lightest known odd-odd  $^{192,194,196}\text{At}$  isotopes, complex  $\alpha$ -decay schemes were observed [3–5]. The odd proton is expected to occupy a  $\pi h_{9/2}$ , a  $\pi f_{7/2}$ , or a  $\pi s_{1/2}$  state, coupled to the odd neutron in one of the low-lying  $\nu p_{1/2}$ ,  $\nu p_{3/2}$ ,  $\nu f_{5/2}$ ,

or  $\nu i_{13/2}$  orbitals, resulting in a multitude of closely spaced states. Similarly complex proton-neutron configurations also exist in the odd-odd Bi  $\alpha$ -decay daughter isotopes. Details can be found in Table II in Ref. [4] for the  $^{194}\text{At}/^{190}\text{Bi}$  pair and in Refs. [6,7] for  $^{184,186,188,190}\text{Bi}$  isotopes. In particular, two isomeric states were identified in  $^{192}\text{At}$  [3]. The decay pattern of one of the isomers suggests that it is based on the oblate-deformed  $\pi f_{7/2} \otimes \nu i_{13/2}$  configuration.

The changes in the mean-square charge radii of the At nuclei have been extracted from measured hyperfine-structure constants and isotope shifts [2]. The ground-state mean-square charge radii of the  $^{194-198}\text{At}$  isotopes strongly deviate from the trend of the spherical lead isotopes, which was interpreted as the result of an onset of oblate deformation when approaching the neutron midshell at  $N = 104$ , as also suggested by the macroscopic-microscopic calculations [8] shown in Fig. 1. Also, cases for shape coexistence have been identified in  $^{197,199}\text{At}$ , for which a significant difference in the charge radii for near-spherical ( $9/2^{-}$ ) ground states and isomeric ( $1/2^{+}$ ) states has been observed. The latter confirms the deformed nature of the  $1/2^{+}$  states. Interestingly, while the heavier isotopes  $^{191,192}\text{At}$  are calculated to be oblate with  $\beta_2 = -0.21$ ,  $^{189,190}\text{At}$  are predicted to be prolate deformed with  $\beta_2 = 0.30$  [8] (see Fig. 1).

As seen in the heavier At isotopes, several states decaying via  $\alpha$  emission could exist in  $^{190}\text{At}$ . The aim of this study was to search for the  $^{190}\text{At}$  isotope and establish its decay properties.

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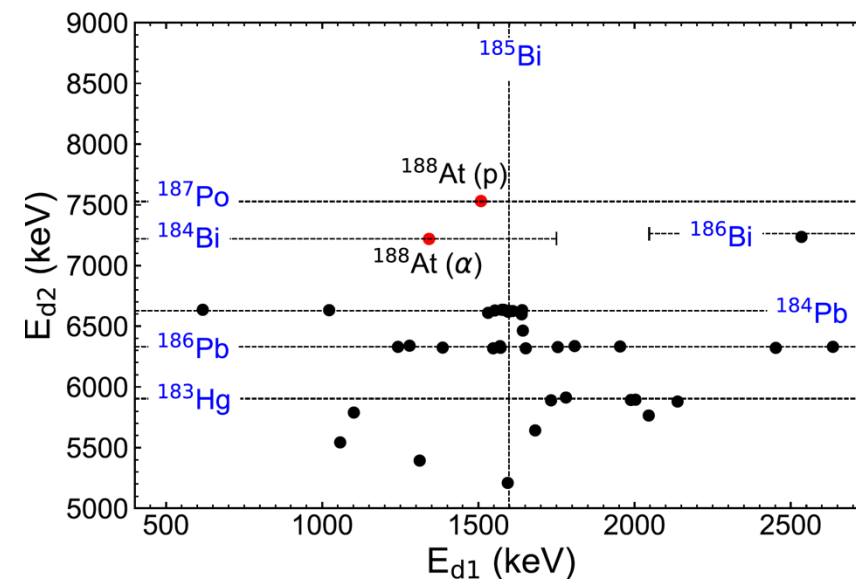
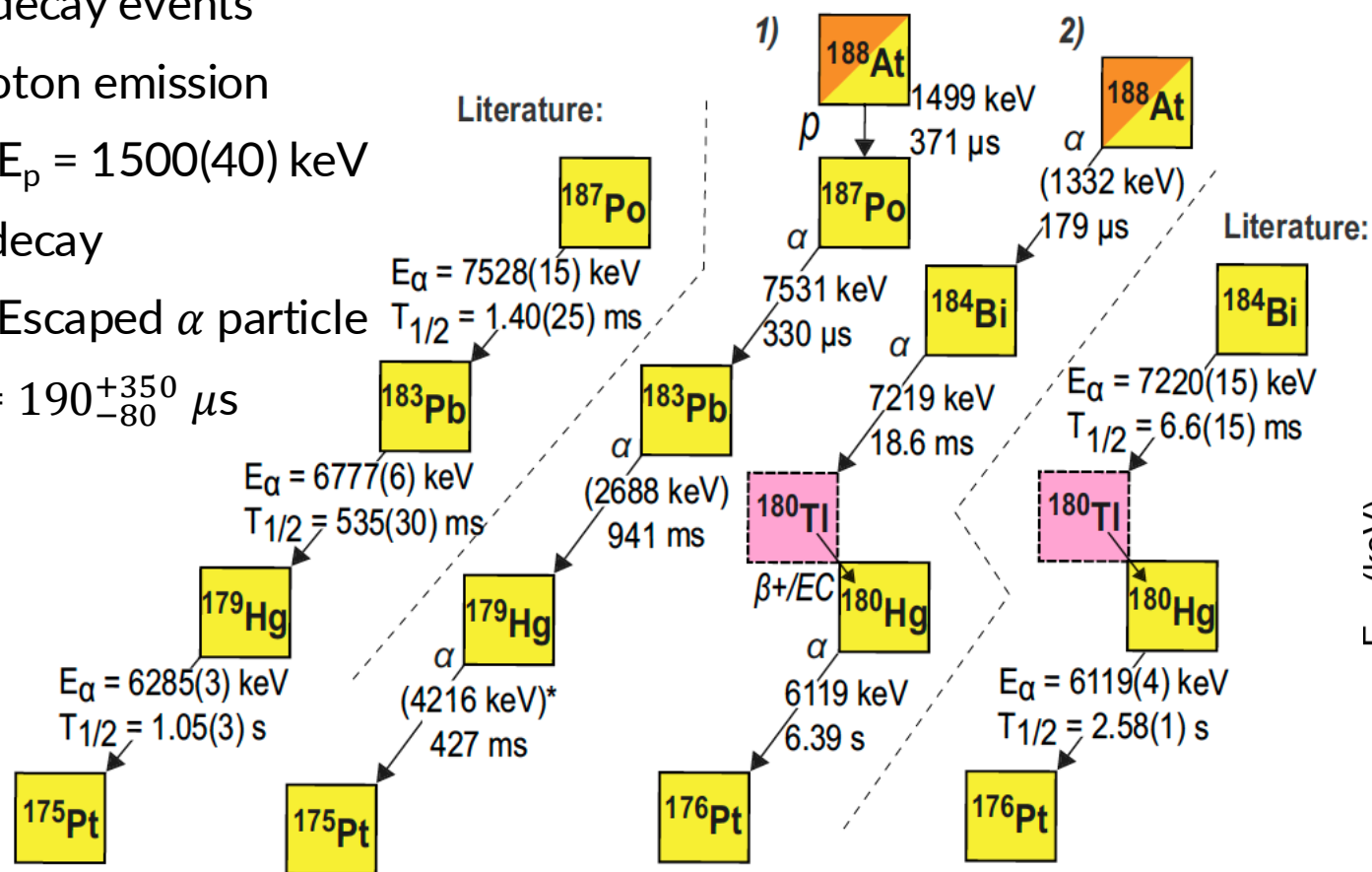


# Decay spectroscopy on $^{188}\text{At}$

# Decay spectroscopy on $^{188}\text{At}$



- Two decay events
  - Proton emission
    - $E_p = 1500(40)$  keV
  - $\alpha$  decay
    - Escaped  $\alpha$  particle
- $T_{1/2} = 190_{-80}^{+350} \mu\text{s}$



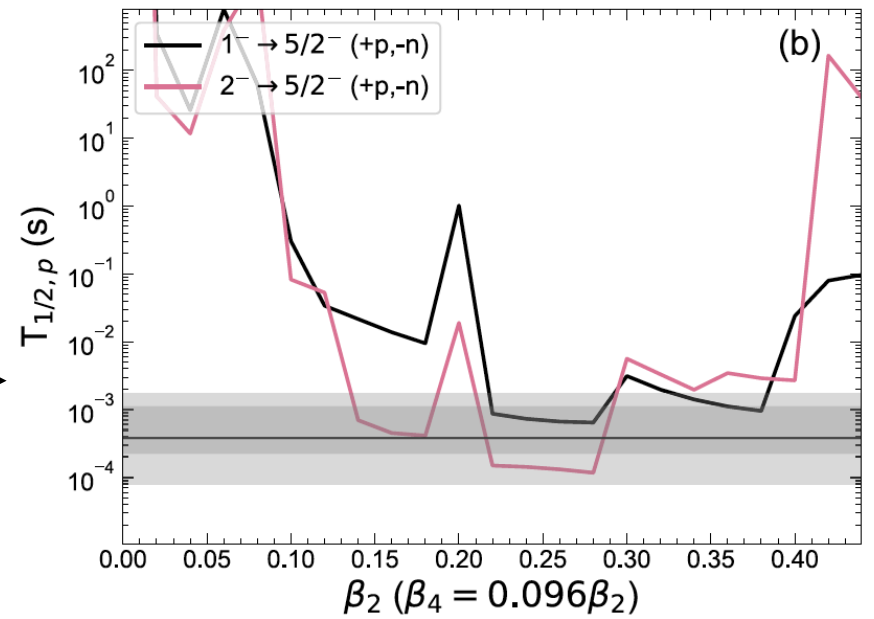
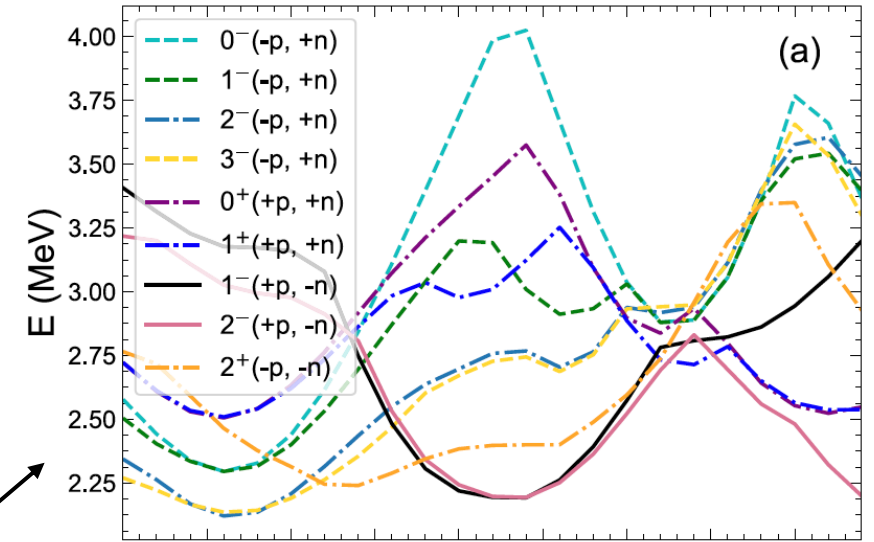
# Discussion: $^{188}\text{At}$

Non-adiabatic quasiparticle model: Theoretical description of deformed proton emitters [1]

To fully construe the experimental observations, the model was expanded to treat nuclei beyond lead by P. Siwach, L. Ferreira, E. Maglione and P. Arumugam.

- Calculated rotational energy of the states which at some deformation reproduce measured partial proton-emission half-life of  $^{188}\text{At}$
- The calculated partial proton-emission rate from the two candidate states are compared to the measured one

[1] Fiorin, G., Maglione, E. & Ferreira, L. S. *Phys. Rev. C* **67**, 054302 (2003)



H. Kokkonen, K. Auranen et al. *Nat Commun* **16**, 4985 (2025)

# Discussion: $^{188}\text{At}$

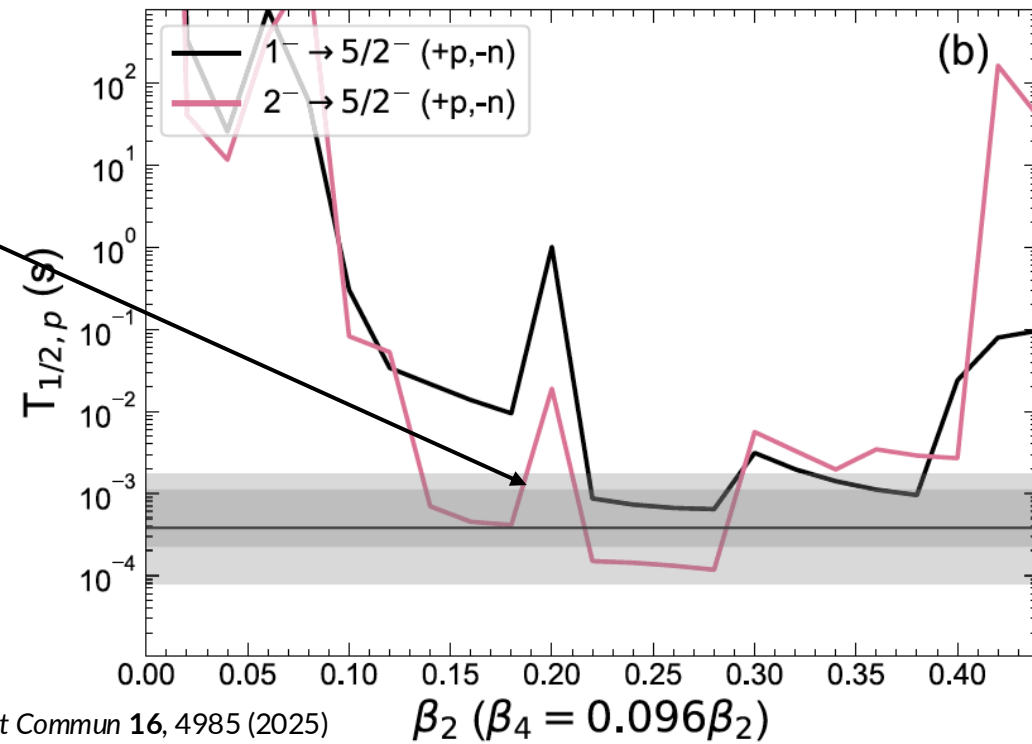
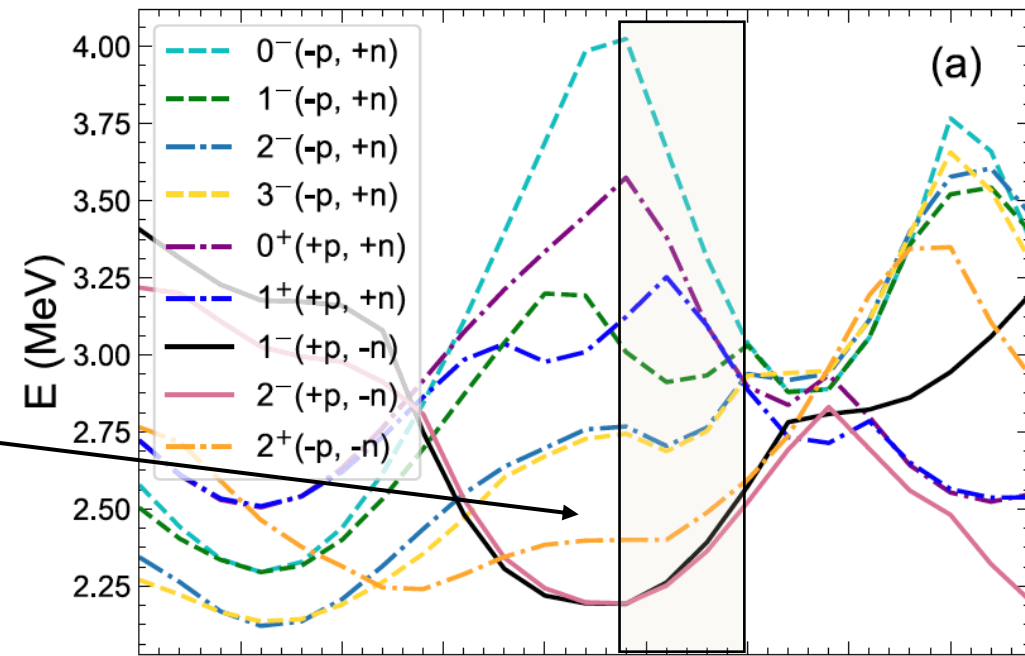
Expected deformation:  $\beta_2 = 0.24 - 0.30$

→  $1^-$  and  $2^-$  most likely candidates for the proton emitting state

Calculated proton emission half-life compared to the measured

- Candidate states' proton component:  $d_{3/2}$  in  $1^- (+p, -n)$  and  $s_{1/2}$  in  $2^- (+p, -n)$
- g.s. configurations for  $^{191,193,195}\text{At}$  are  $\pi s_{1/2}$ , as well as for the  $^{185}\text{Bi}$  proton emitting orbital is  $s_{1/2}$

→  $s_{1/2}$  is expected to be at the Fermi surface

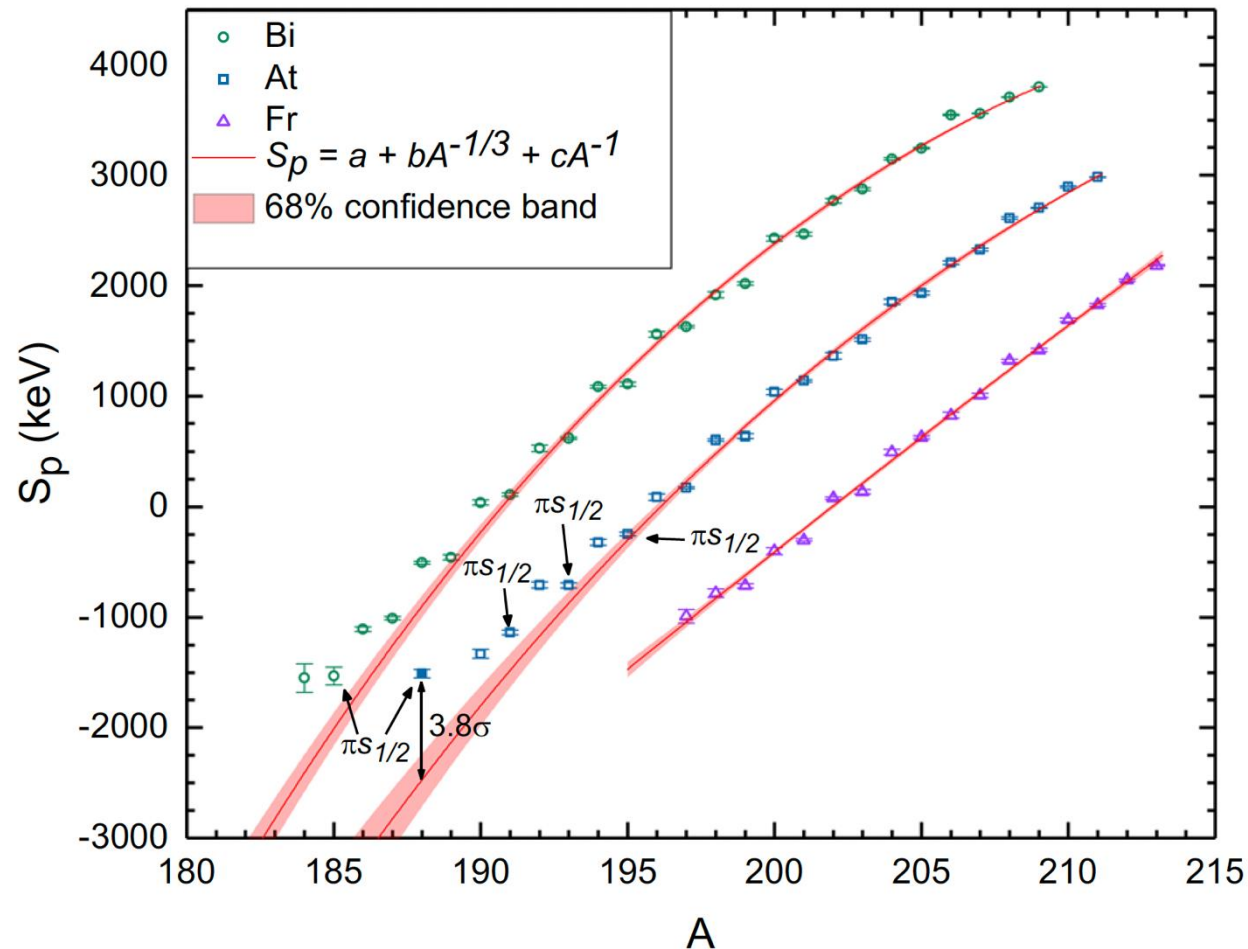


# Discussion: $^{188}\text{At}$

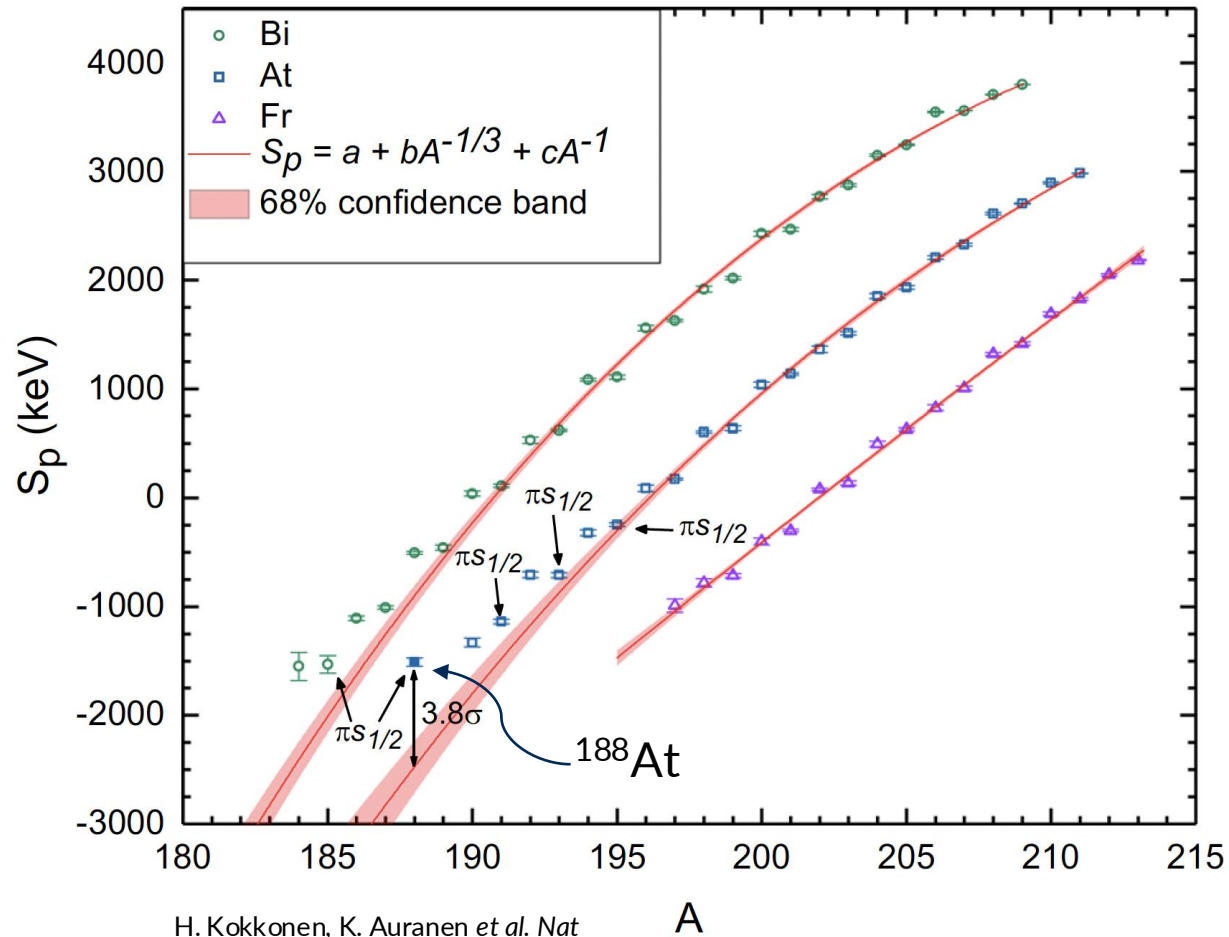


One proton separation energy,  $S_p$  was calculated from the measured data

The results were compared to the systematics of nearby isotopes:



# Discussion: $^{188}\text{At}$



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Fitting a function of  $S_p = a + bA^{-1/3} + cA^{-1}$  arising from the liquid-drop model to the proton-bound ( $S_p > 0$ ) data, extrapolated to ( $S_p < 0$ )

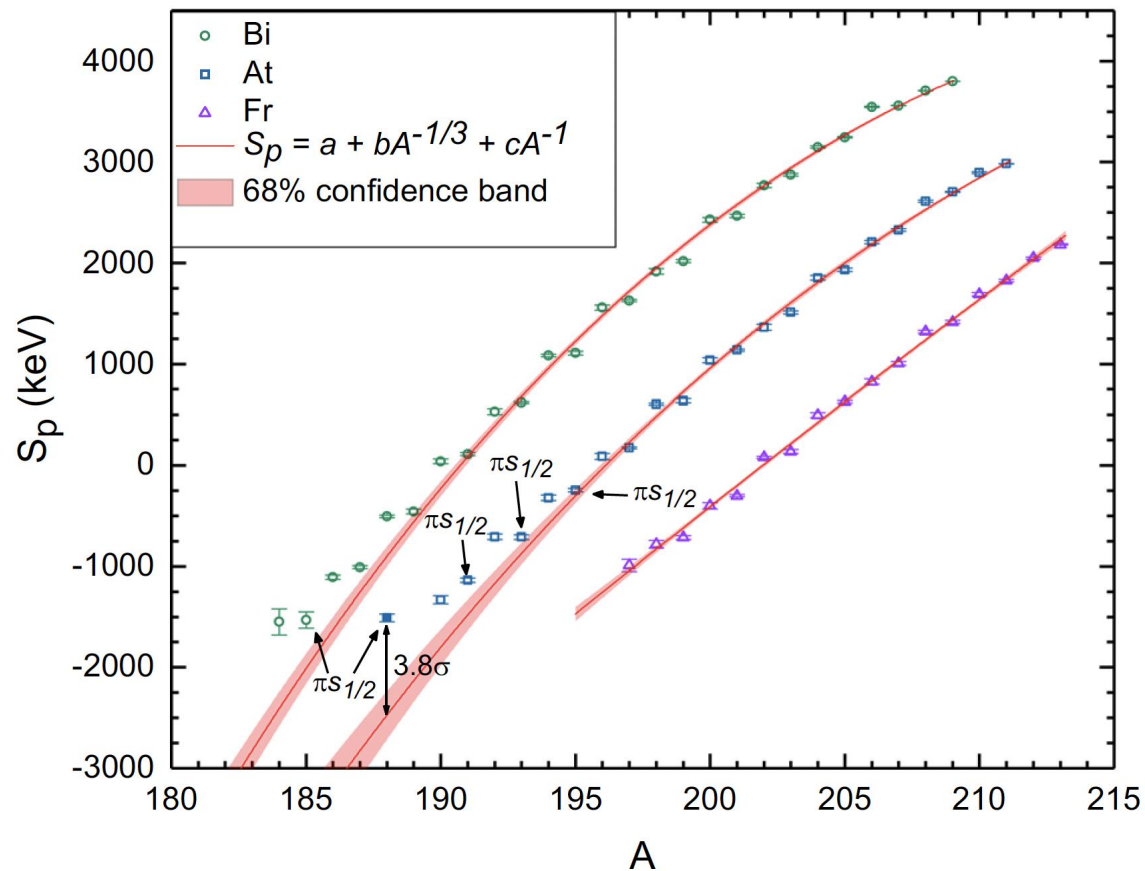
- Deviation of  $3.8\sigma$  in  $S_p(^{188}\text{At})$

Strong  $\pi s_{1/2}$  component in the wave function of the ground state

Fr isotopes have  $\pi h_{9/2}$  ground state, no such deviation in the systematics

One possible explanation for this effect is *Thomas-Ehrman effect*

# Thomas-Ehrman effect



- Phenomenon where valence protons have a wave function extending outside nuclear interior
  - The repulsive Coulomb effect is reduced
  - The valence protons appear to be more bound to the core
- Thomas-Ehrman effect is at strongest in proton unbound nuclei with low angular momentum proton orbitals [1]
- This has been characterised in many light and medium nuclei before [2,3], and never before in the heavy nuclei

[1] Shlomo, S. Reports on Progress in Physics 41(7), 957(1978)  
 [2] Comay, E., et.al. Physics Letters B 210(1), 31–34 (1988)  
 [3] Nazarewicz, et. al. Rev. C 53, 740–751 (1996)

# Summary

Two new isotopes and their decay properties were studied


Possible sign of the Thomas-Ehrman shift for the first time in heavy nuclei

## Properties of the new $\alpha$ -decaying isotope $^{190}\text{At}$

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The  $\alpha$  decay of a new isotope  $^{190}\text{At}$  has been studied via the  $^{109}\text{Ag}(^{84}\text{Sr}, 3n)^{190}\text{At}$  fusion-evaporation reaction by employing a gas-filled recoil separator. An  $\alpha$ -particle energy of 7750(20) keV and a half-life of  $1.0^{+0.4}_{-0.4}$  ms were measured. The measured decay properties correspond to an unhindered  $\alpha$  decay, suggesting the same spin and parity of ( $10^-$ ) as those of the final state of the decay. The systematics of the nearby nuclei and the predictions of selected atomic mass models were compared with the measured decay properties.

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## New proton emitter $^{188}\text{At}$ implies an interaction unprecedented in heavy nuclei

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We report the discovery of a new atomic nucleus  $^{188}\text{At}$ , which is the heaviest proton-emitting isotope known to date. The new activity was observed through the  $^{107}\text{Ag}(^{84}\text{Sr}, 3n)^{188}\text{At}$  fusion-evaporation reaction using the focal-plane spectrometer of the gas-filled recoil separator in the Accelerator Laboratory of the University of Jyväskylä, Finland. To fully interpret the experimental data, we have expanded the non-adiabatic quasiparticle model to treat nuclei in the beyond-lead region. The description reproduced the measured decay rate and pointed towards emission from an extremely prolate-deformed state with a dominant  $s_{1/2}$  proton component in the wave function. The Thomas-Ehrman shift can be enhanced in low angular momentum states, but such effects have not been observed in heavy nuclei. The single-proton separation energy of  $^{188}\text{At}$  deviates from that extrapolated from the systematics, which can be interpreted as the first evidence of this effect in heavy nuclei.

Properties of the new  $\alpha$ -decaying isotope  $^{190}\text{At}$ 

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The  $\alpha$  decay of a new isotope  $^{190}\text{At}$  has been studied via the  $^{109}\text{Ag}(^{84}\text{Sr}, 3n)^{190}\text{At}$  fusion-evaporation reaction

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Article

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## New proton emitter $^{188}\text{At}$ implies an interaction unprecedented in heavy nuclei

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# ILTA-SANOMAT

KOTIMAA

# Hennan, 24, gradu osoittautui täysin poikkeukselliseksi: löysi uuden atomiytimen

Suomalaisen Henna Kokkosen pro gradu -tutkielmaa on siteerattu kansainvälisessä ydinfysiikan tiedelehdessä.

yle Etusivu Venäjän hyökkäys Irar

Tutkimus

## Väitöskirjatutkija löysi Jyväskylässä uuden atomiytimen

Henna Kokkosen tutkimuksen tulokset jaettiin arvostetussa Nature Communications -lehdessä.



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Helsinki  
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**PÄIVÄN TIMANTTI:** Suomen kiinteistömarkkinat ovat kurimuksessa, ja seuraava vaihe on pakkomyynnit



**Tiede | Läpimurto ydinfysiikassa**  
Henna Kokkosen gradua siteerattiin maailman tiedejulkaisuissa

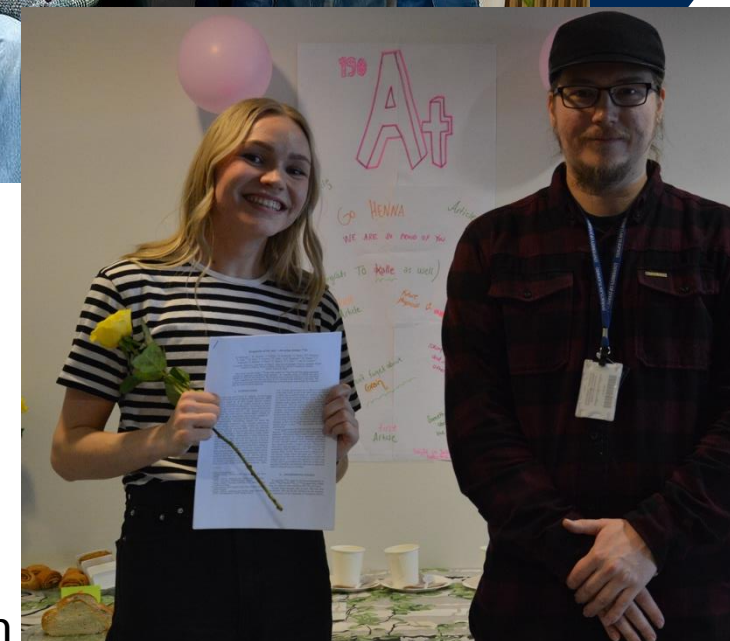
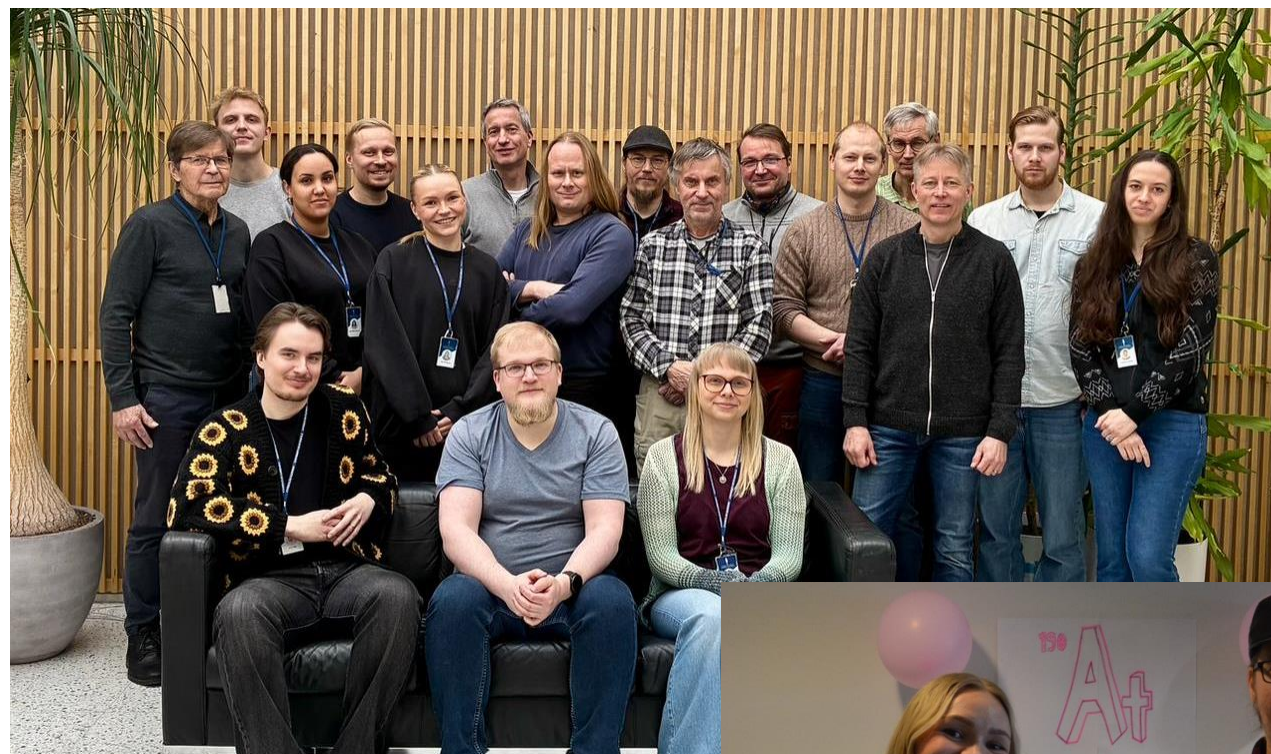
## 188At collaboration

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## Nuclear spectroscopy group in 3/2026



My phd supervisor Kalle Auranen

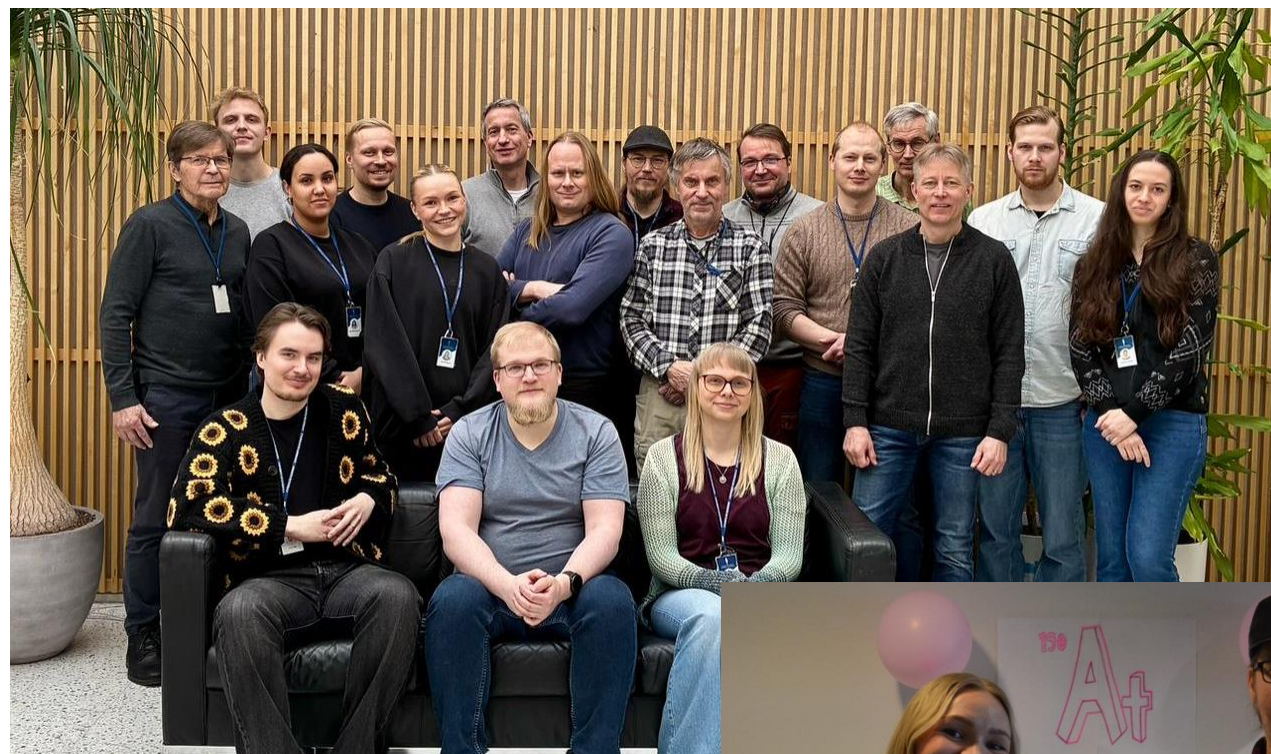
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# Thank you!