



# EVOLUTION OF CHANGES IN MEAN-SQUARE CHARGE RADII IN CALIFORNIUM ISOTOPES

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For the RADRIS collaboration

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|          |          |          |          |           |           |           |           |           |           |           |           |           |           |           |           |           |           |           |          |         |         |          |          |  |  |  |  |  |
|----------|----------|----------|----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|----------|---------|---------|----------|----------|--|--|--|--|--|
| 1<br>H   |          |          |          |           |           |           |           |           |           |           |           |           |           |           |           |           | 2<br>He   |           |          |         |         |          |          |  |  |  |  |  |
| 3<br>Li  | 4<br>Be  |          |          |           |           |           |           |           |           |           |           |           |           |           |           |           |           | 5<br>B    | 6<br>C   | 7<br>N  | 8<br>O  | 9<br>F   | 10<br>Ne |  |  |  |  |  |
| 11<br>Na | 12<br>Mg |          |          |           |           |           |           |           |           |           |           |           |           |           |           |           |           | 13<br>Al  | 14<br>Si | 15<br>P | 16<br>S | 17<br>Cl | 18<br>Ar |  |  |  |  |  |
| 19<br>K  | 20<br>Ca | 21<br>Sc | 22<br>Ti | 23<br>V   | 24<br>Cr  | 25<br>Mn  | 26<br>Fe  | 27<br>Co  | 28<br>Ni  | 29<br>Cu  | 30<br>Zn  | 31<br>Ga  | 32<br>Ge  | 33<br>As  | 34<br>Se  | 35<br>Br  | 36<br>Kr  |           |          |         |         |          |          |  |  |  |  |  |
| 37<br>Rb | 38<br>Sr | 39<br>Y  | 40<br>Zr | 41<br>Nb  | 42<br>Mo  | 43<br>Tc  | 44<br>Ru  | 45<br>Rh  | 46<br>Pd  | 47<br>Ag  | 48<br>Cd  | 49<br>In  | 50<br>Sn  | 51<br>Sb  | 52<br>Te  | 53<br>I   | 54<br>Xe  |           |          |         |         |          |          |  |  |  |  |  |
| 55<br>Cs | 56<br>Ba |          |          | 72<br>Hf  | 73<br>Ta  | 74<br>W   | 75<br>Re  | 76<br>Os  | 77<br>Ir  | 78<br>Pt  | 79<br>Au  | 80<br>Hg  | 81<br>Tl  | 82<br>Pb  | 83<br>Bi  | 84<br>Po  | 85<br>At  | 86<br>Rn  |          |         |         |          |          |  |  |  |  |  |
| 87<br>Fr | 88<br>Ra |          |          | 104<br>Rf | 105<br>Db | 106<br>Sg | 107<br>Bh | 108<br>Hs | 109<br>Mt | 110<br>Ds | 111<br>Rg | 112<br>Cn | 113<br>Nh | 114<br>Fl | 115<br>Mc | 116<br>Lv | 117<br>Ts | 118<br>Og |          |         |         |          |          |  |  |  |  |  |
|          |          |          |          |           |           |           |           |           |           |           |           |           |           |           |           |           |           |           | →        |         |         |          |          |  |  |  |  |  |
|          |          | 57<br>La | 58<br>Ce | 59<br>Pr  | 60<br>Nd  | 61<br>Pm  | 62<br>Sm  | 63<br>Eu  | 64<br>Gd  | 65<br>Tb  | 66<br>Dy  | 67<br>Ho  | 68<br>Er  | 69<br>Tm  | 70<br>Yb  | 71<br>Lu  |           |           |          |         |         |          |          |  |  |  |  |  |
|          |          | 89<br>Ac | 90<br>Th | 91<br>Pa  | 92<br>U   | 93<br>Np  | 94<br>Pu  | 95<br>Am  | 96<br>Cm  | 97<br>Bk  | 98<br>Cf  | 99<br>Es  | 100<br>Fm | 101<br>Md | 102<br>No | 103<br>Lr |           |           |          |         |         |          |          |  |  |  |  |  |

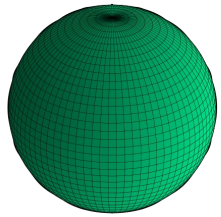
## Pairing

Studying their properties provide stringest tests of nuclear models

# PHYSICS OF HEAVY ELEMENTS

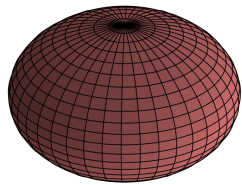
## Nuclear Structure

### Quadrupole Deformation



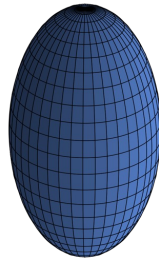
$$Q_S = 0$$

Spherical



$$Q_S < 0$$

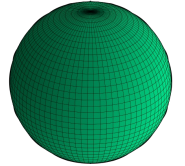
Oblate



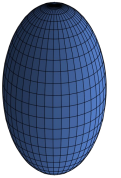
$$Q_S > 0$$

Prolate

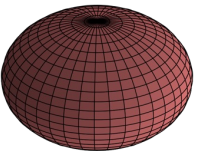
- Rare in heavier systems
- Normally found around magic N, Z



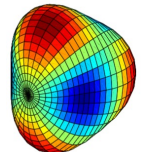
- Prevalent deformation (in heavy actinides)
- Often minimizes the nucleus' total energy



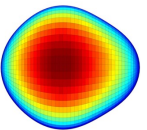
- Oblate g.s. more common in lighter nuclei
- Often coexists with prolate deformation



- Exotic triaxiality (no symmetry axis) can occur



- Higher order multipoles (pear shaped Ra-224)



# PHYSICS OF HEAVY ELEMENTS

Isotope Shift

Frequency shift of an electronic transition for different N

$$\delta\nu^{A,A'} = \frac{m^{A'} - m^A}{m^{A'} \cdot m^A} \cdot K + F \cdot \delta\langle r^2 \rangle^{A,A'}$$

Property connects electronic shell with nucleus

$$V(r = 0) \neq \infty$$

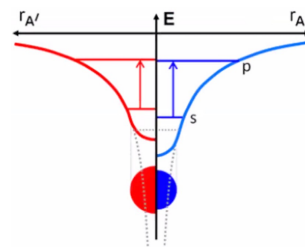
$$m_{\text{Nucl.}} \neq \infty$$

COM motion

Not to be confused with the  
(inverse) of the reduced mass

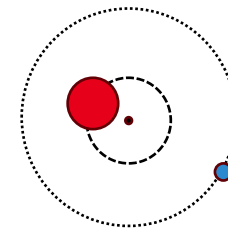
$$F = F(\langle r^2 \rangle, \langle r^4 \rangle, \langle r^6 \rangle, \dots)$$

Field Shift F



Dominant  
contribution  
in heavy  
systems

Mass Shift K



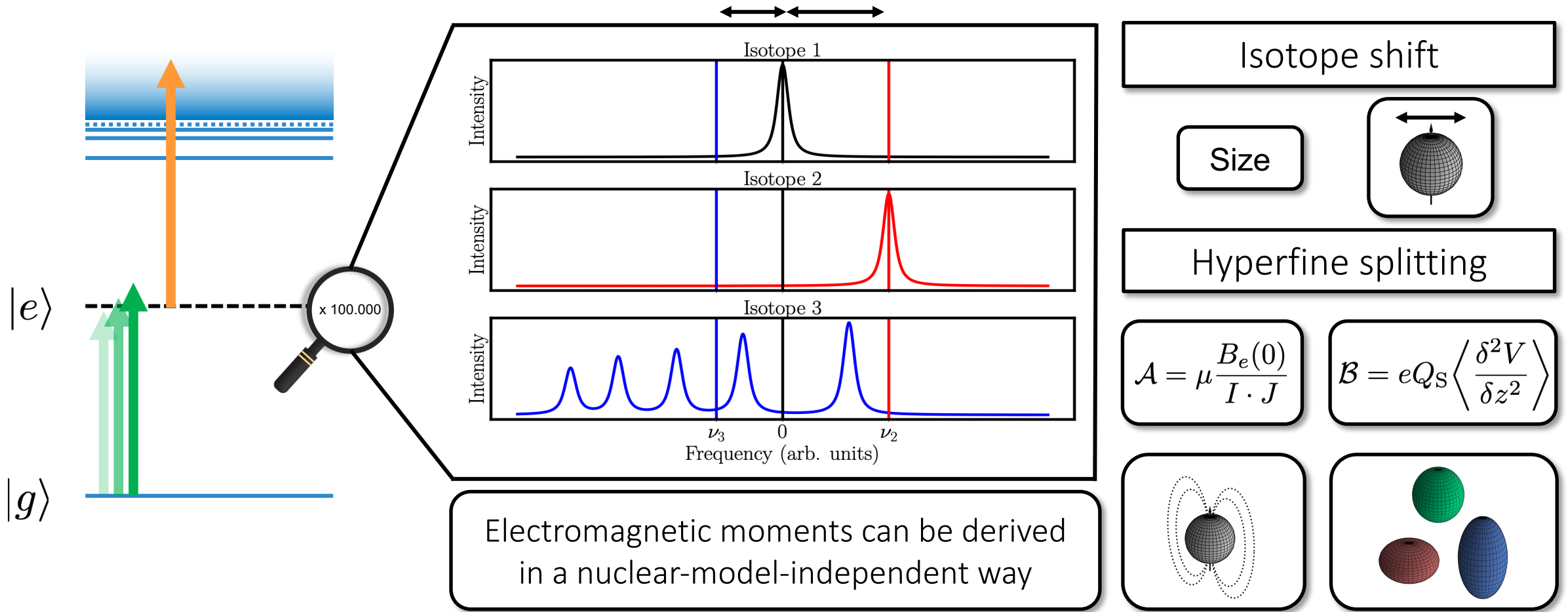
NMS

+

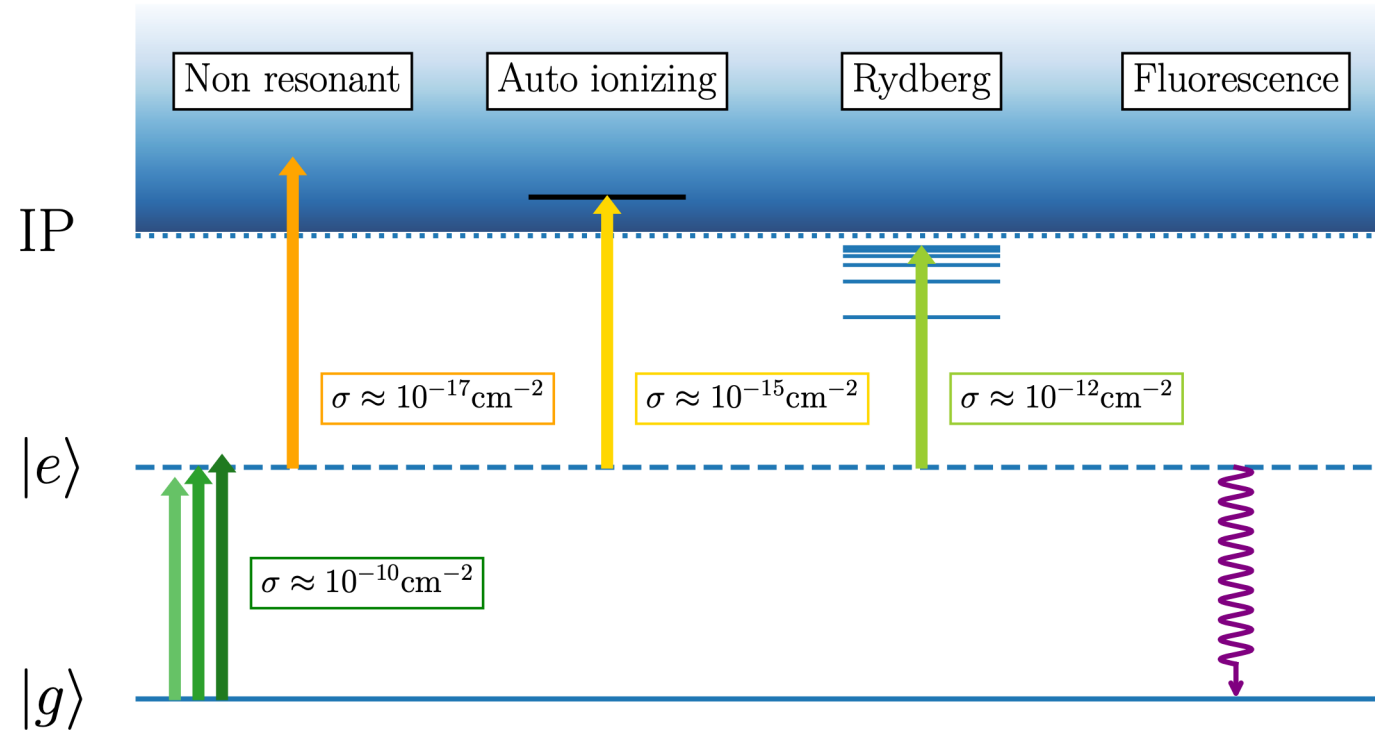
SMS



# DERIVING NUCLEAR PROPERTIES FROM LASER SPECTROSCOPY



# RESONANCE IONIZATION SPECTROSCOPY



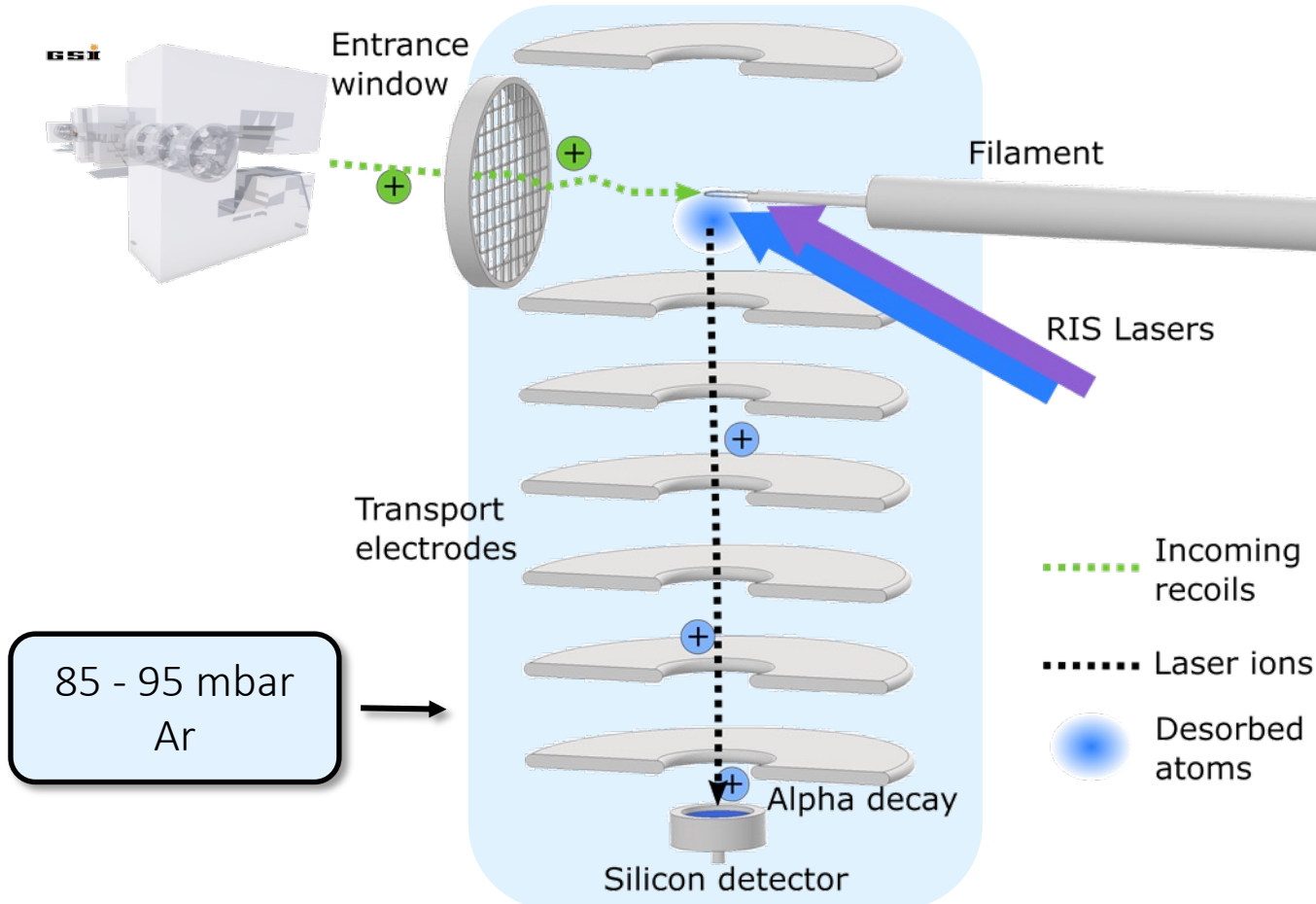
Non resonant: no knowledge about level scheme needed

AI: higher cross section than non-resonant excitation

Rydberg: can be used to determine ionization potential

Laser spectroscopy is a highly selective and efficient technique

# RADIATION DETECTED RESONANCE IONIZATION SPECTROSCOPY (RADRIS)



## Procedure:

1. Production and separation
2. Thermalizing incoming fusion products
3. Collection and neutralization on filament
4. Evaporation of neutral atoms
5. Two-step resonance laser ionization
6. Subsequent detection by  $\alpha$ -decay

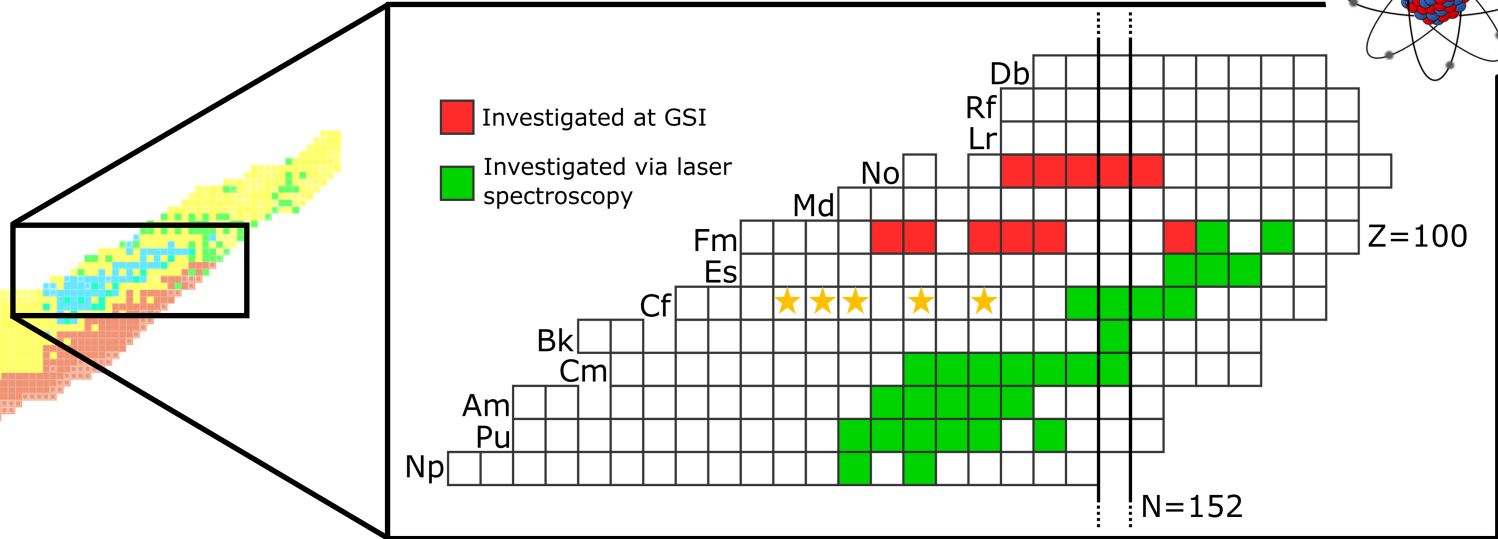
J. Warbinek et al., Atoms 10, 41 (2022)

## ISOTOPIC CHAIN IN CALIFORNIUM

Z

## Primary decay mode

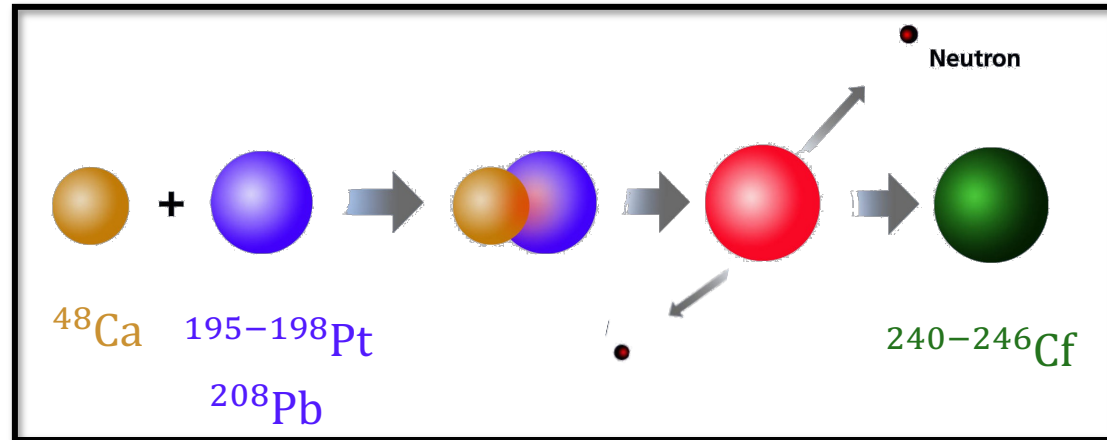
- |             |              |
|-------------|--------------|
| ■ Stable    |              |
| ■ $\beta^-$ | ■ $2\beta^-$ |
| ■ $\beta^+$ | ■ $2\beta^+$ |
| ■ $\alpha$  | ■ $p/2p$     |
| ■ Fission   | ■ $n/2n$     |


 $^{240}\text{Cf}, ^{241}\text{Cf}, ^{242}\text{Cf}, ^{244}\text{Cf}, ^{246}\text{Cf}$ 
<https://people.physics.anu.edu.au/~ecs103/chart/>

N



# PRODUCTION OF CALIFORNIUM AT GSI



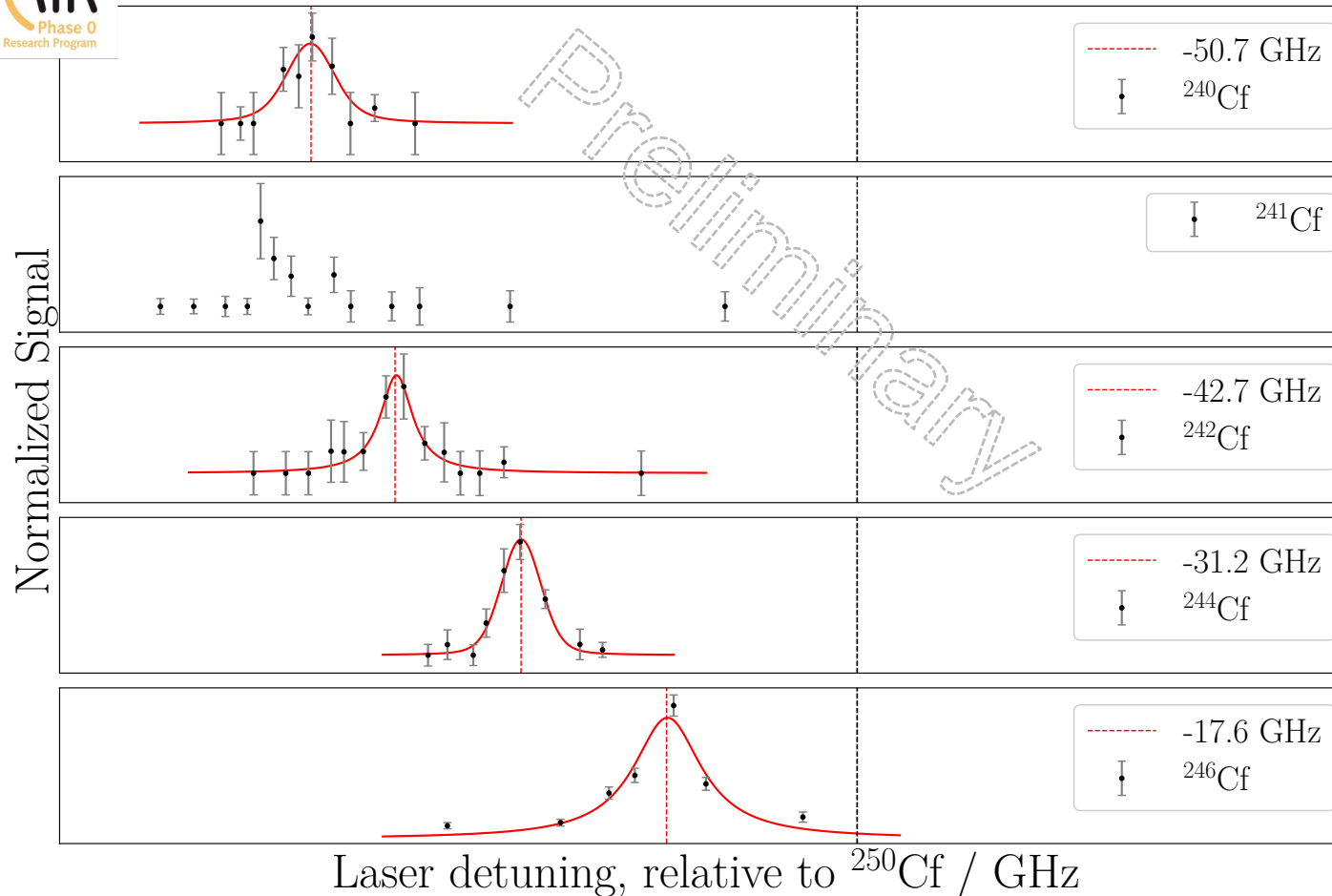
- Direct production of  $^{240-244}\text{Cf}$  with Pt
- Indirect production via  $^{254}\text{No}$  via subsequent  $\alpha$  decays
- Long half-life challenging for RADRIS method

| Isotope           | Half-life    | Reaction                                                                                                | Ions per hour |
|-------------------|--------------|---------------------------------------------------------------------------------------------------------|---------------|
| $^{240}\text{Cf}$ | 0.96(15) min | $^{195}\text{Pt}(^{48}\text{Ca}, 3n)$                                                                   | $\approx 1$   |
| $^{241}\text{Cf}$ | 3.78(70) min | $^{196}\text{Pt}(^{48}\text{Ca}, 3n)$                                                                   | 1 - 1.5       |
| $^{242}\text{Cf}$ | 3.5(2) min   | $^{196}\text{Pt}(^{48}\text{Ca}, 2n)$                                                                   | 5 - 6         |
| $^{244}\text{Cf}$ | 19.4(6) min  | $^{198}\text{Pt}(^{48}\text{Ca}, 2n)$                                                                   | 9 - 10        |
| $^{246}\text{Cf}$ | 35.7(5) h    | $^{208}\text{Pb}(^{48}\text{Ca}, 2n)$<br>$^{254}\text{No} \xrightarrow{\alpha, \alpha} ^{246}\text{Cf}$ | 18 - 22       |

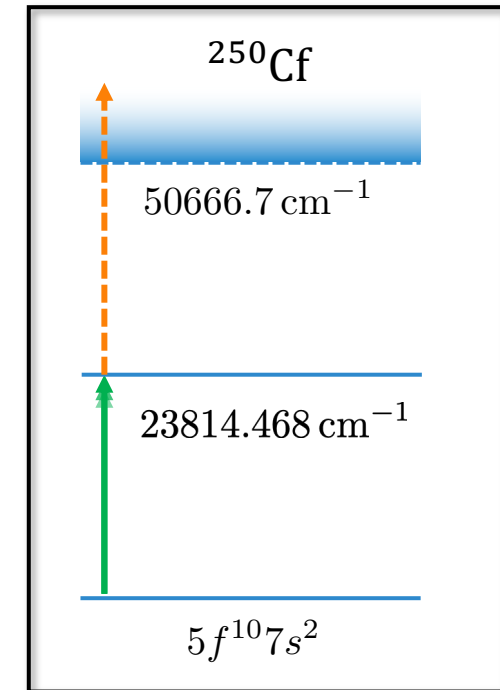
# ISOTOPE SHIFTS



Isotope shift in  $^{240}\text{Cf}$ ,  $^{242}\text{Cf}$ ,  $^{244}\text{Cf}$  and  $^{246}\text{Cf}$



Reference Isotope



Weber et al. *Atoms* **2022**, 10(2), 51

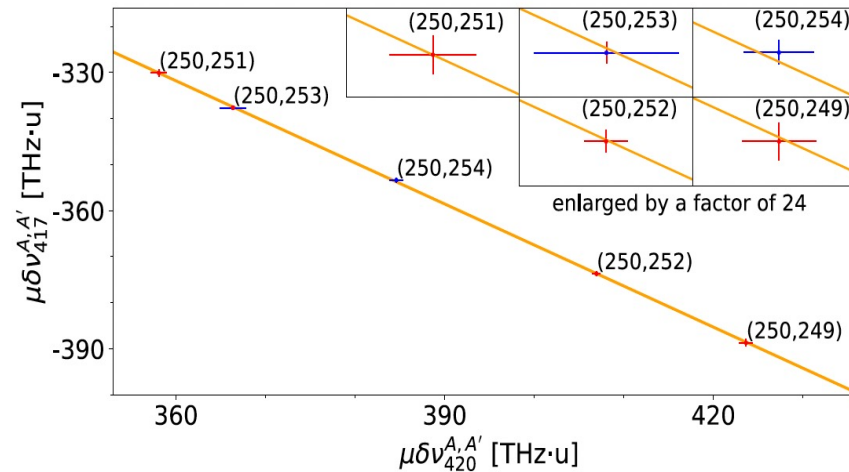
# FROM ISOTOPE SHIFT TO CHANGES IN MEAN-SQUARE CHARGE RADII

Weber et al. *Atoms* **2022**, 10(2), 51

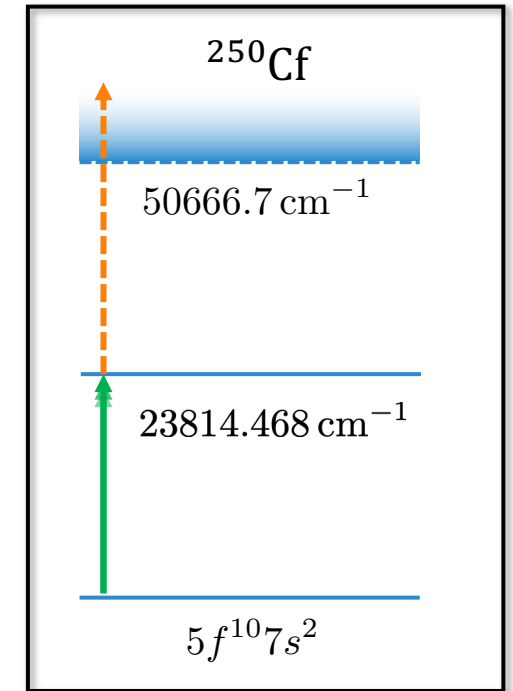
$$\delta\langle r^2 \rangle^{A,A'} = \left( \delta\nu^{A,A'} - \frac{K}{\mu} \right) \cdot \frac{1}{F}$$

Input from atomic theory  
(CI, MCDHF,...)

High uncertainty due to  
large number of open  
f-shells in the actinides



$$\mu \cdot \delta\nu_i^{A,A'} = \underbrace{\mu \cdot \frac{F_i}{F_j}}_{\text{slope}} \cdot \delta\nu_j^{A,A'} - \underbrace{\frac{F_i}{F_j} \cdot K_j + K_i}_{\text{intercept}}$$



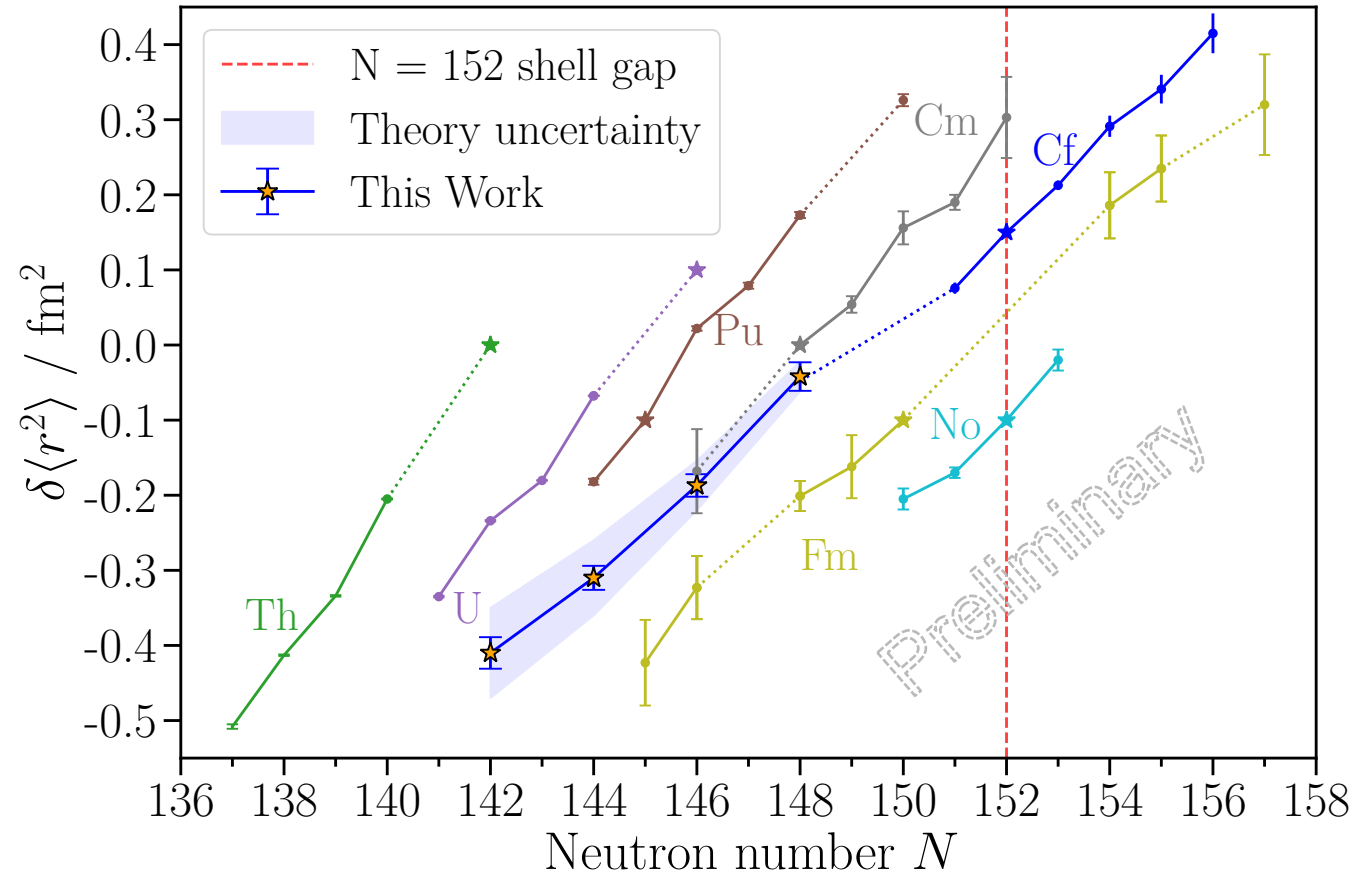
$$F_{420} = 91(27) \text{ GHz/fm}^2$$

$$\frac{F_{417}}{F_{420}} = -0.891(6)$$

$$F_{420} = 91(9) \text{ GHz/fm}^2$$

# FROM ISOTOPE SHIFT TO CHANGES IN MEAN-SQUARE CHARGE RADII

- Mostly smooth isotope-shift evolution, with no large universal kink established at  $N = 152$
- Bulk size is less sensitive to the subshell closure
- Similar trend as recently established in an isotopic chain on fermium





# FROM ISOTOPE SHIFT TO CHANGES IN MEAN-SQUARE CHARGE RADII

- Mostly smooth isotope-shift evolution, with no large universal kink established at  $N = 152$
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## Article

# Smooth trends in fermium charge radii and the impact of shell effects

<https://doi.org/10.1038/s41586-024-08062-z>

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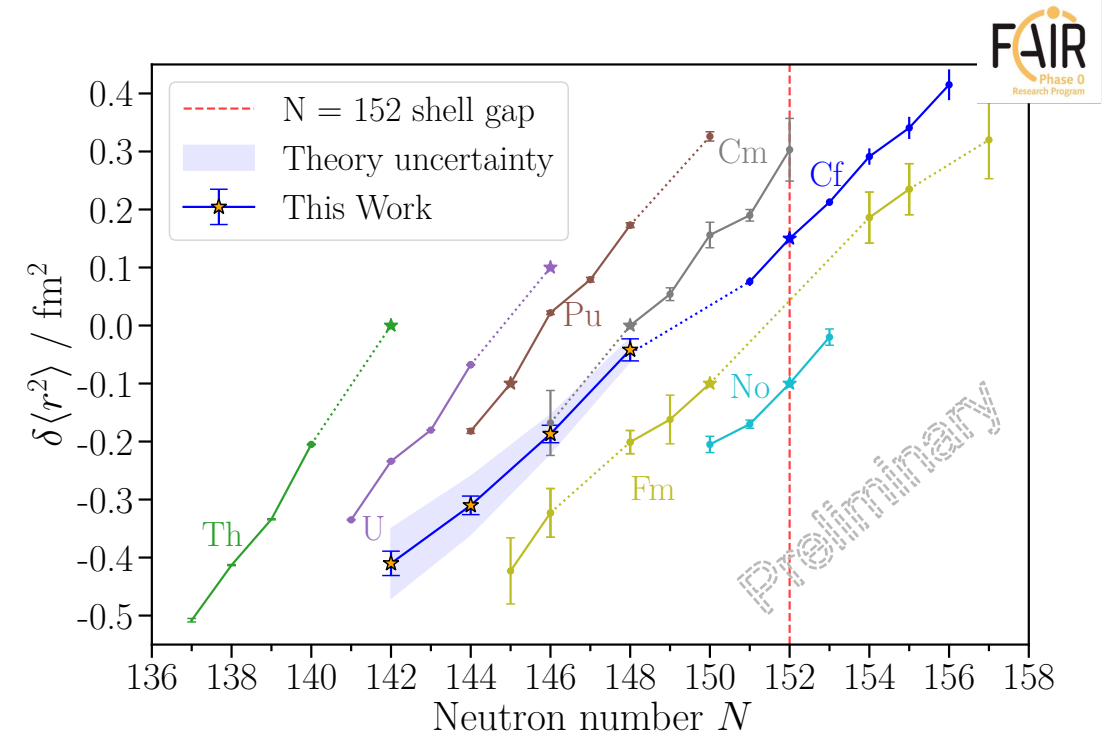
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# SUMMARY AND OUTLOOK

- Isotopic shifts and changes in rms charge radii were determined for even-even Cf isotopes:  $^{240}\text{Cf}$ ,  $^{242}\text{Cf}$ ,  $^{244}\text{Cf}$  and  $^{246}\text{Cf}$
- Smooth trend for charge radii continue to show up in the heavy actinide region
  - Collective behavior masks single particle effects that impact the charge radius
- Long-lived  $^{252}\text{Fm}$  (25.4h) on the deformed subshell closures  $N = 152$  and  $Z = 100$  now in range for the RADRIS technique







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und Raumfahrt



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**GANIL**

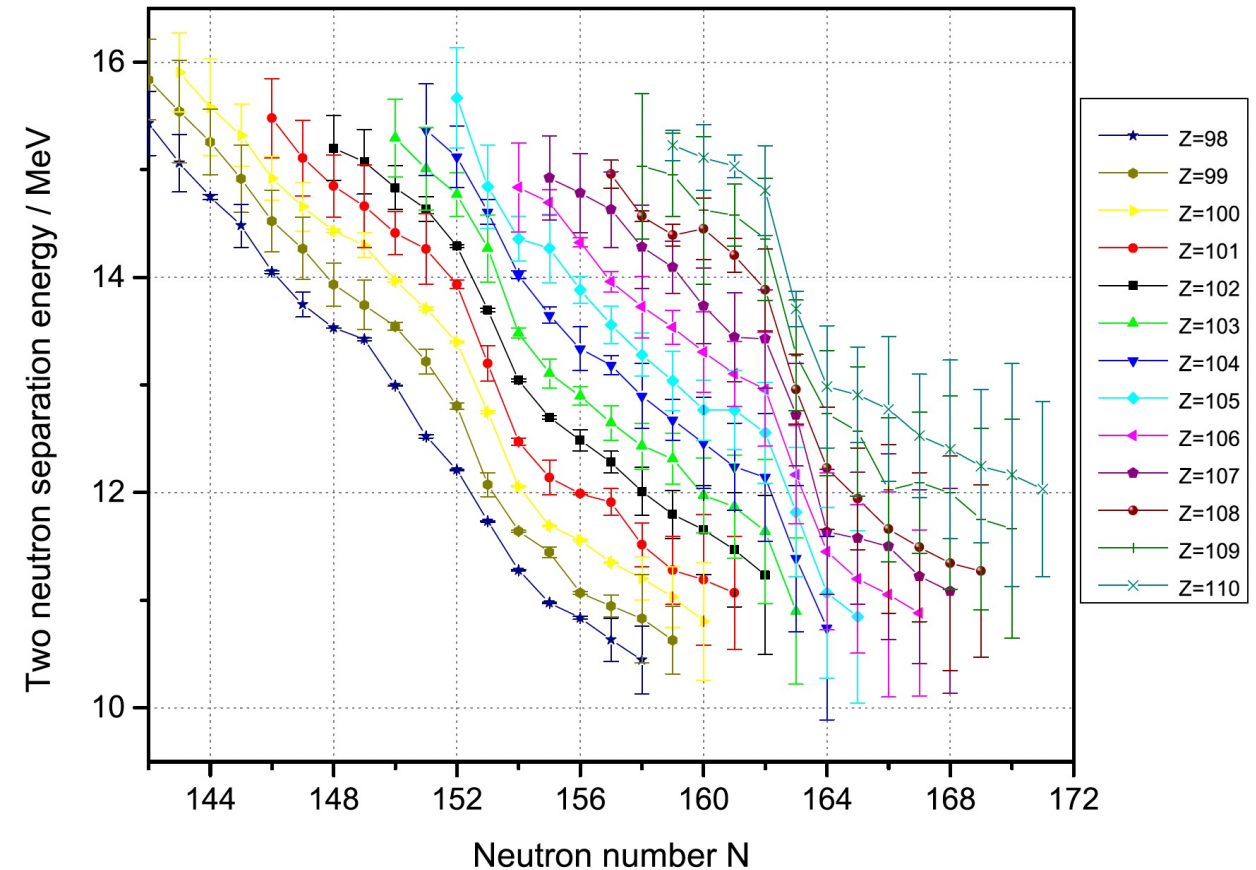
This Marie Skłodowska-Curie Action (MSCA) Innovative Training Network (ITN) receives funding from the European Union H2020 Framework Programme under grant agreement no. 861198.



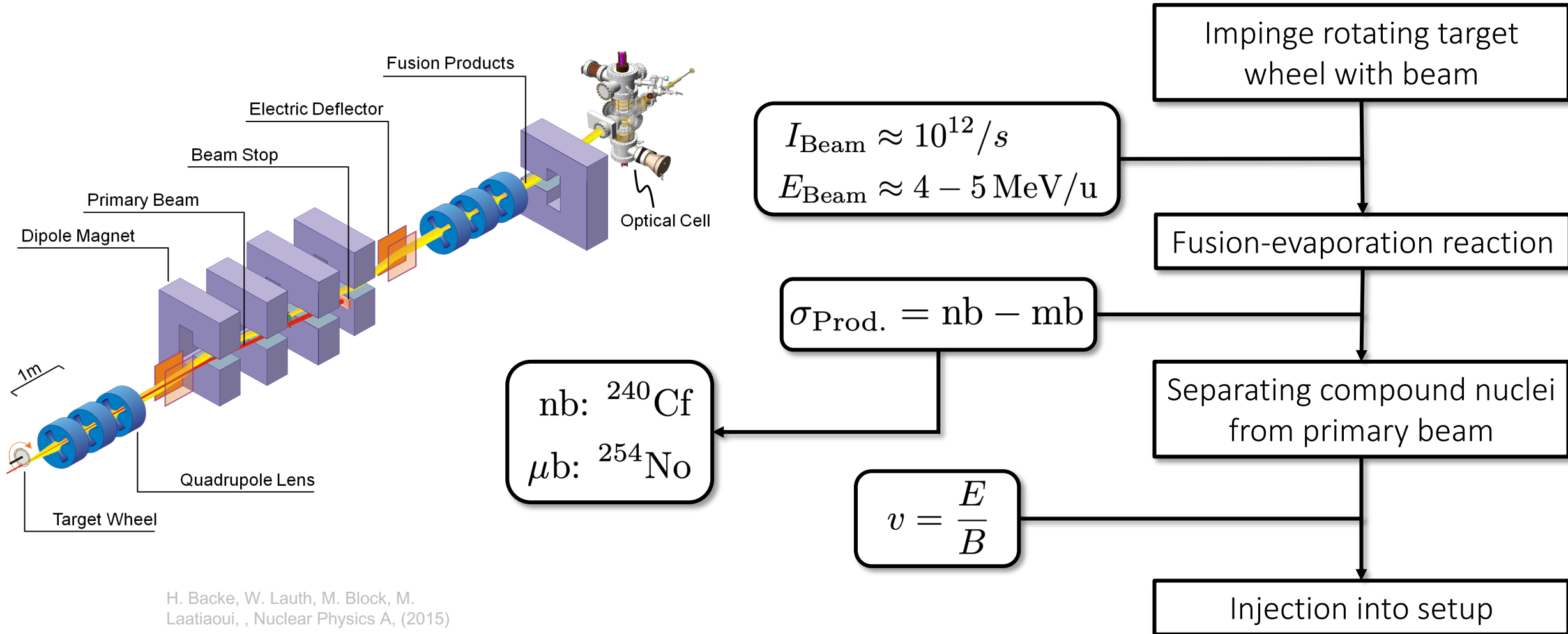
# SHELL EFFECT SENSITIVE QUANTITIES

*M. Block / Nuclear Physics A 944 (2015) 471–491*

- $B(E2)$  values
- $E(2_1^+)$
- Spacing in rotational bands
- $Q_\alpha$
- Half-lives
- Hindrance factors
- ....

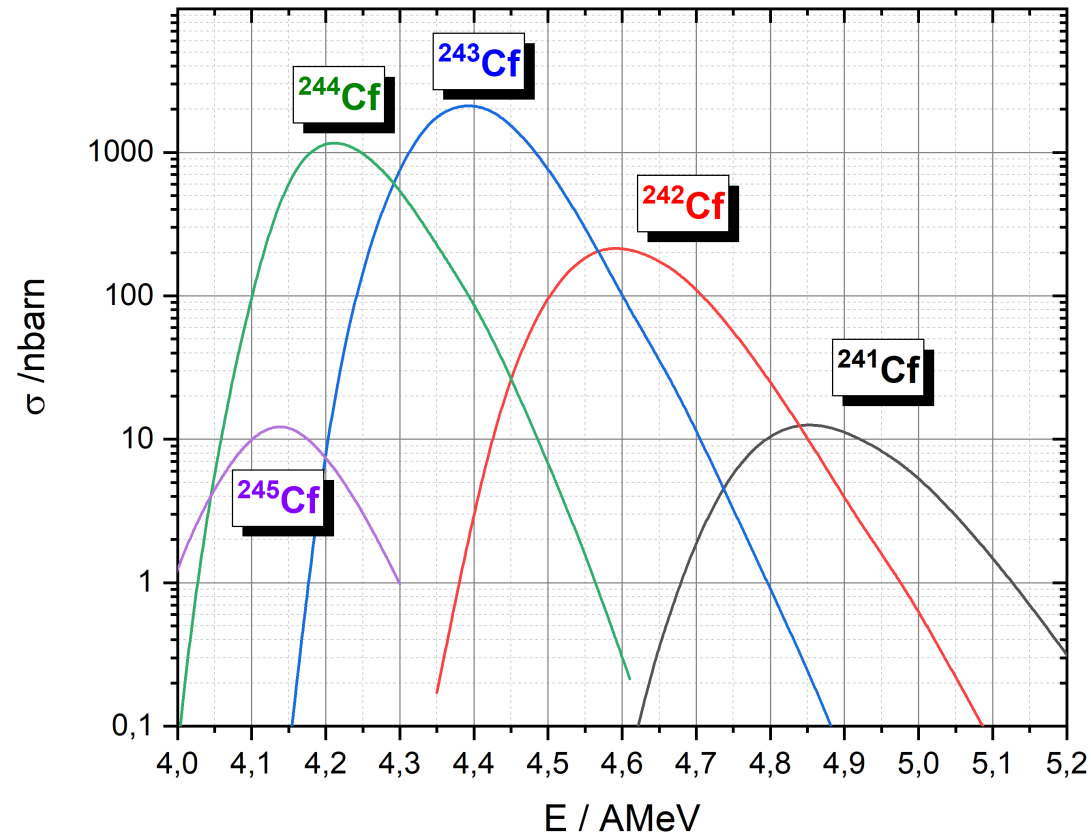


# PRODUCING HEAVY ELEMENTS

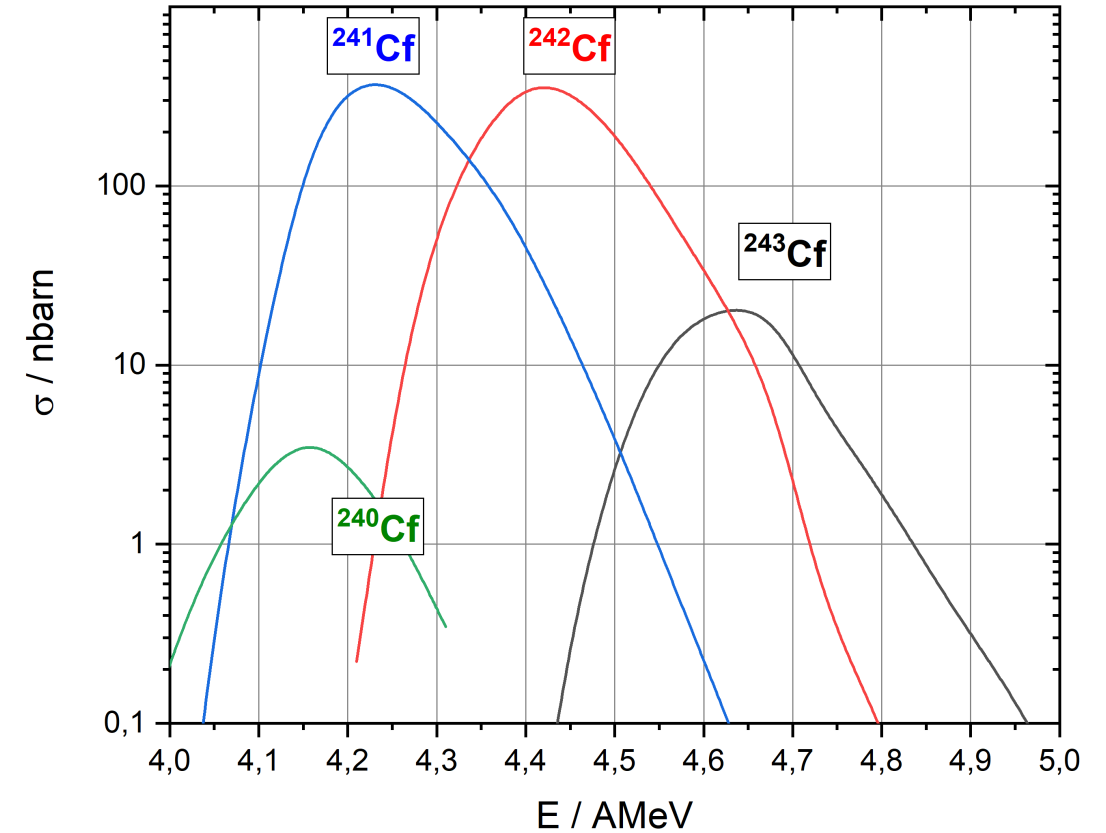


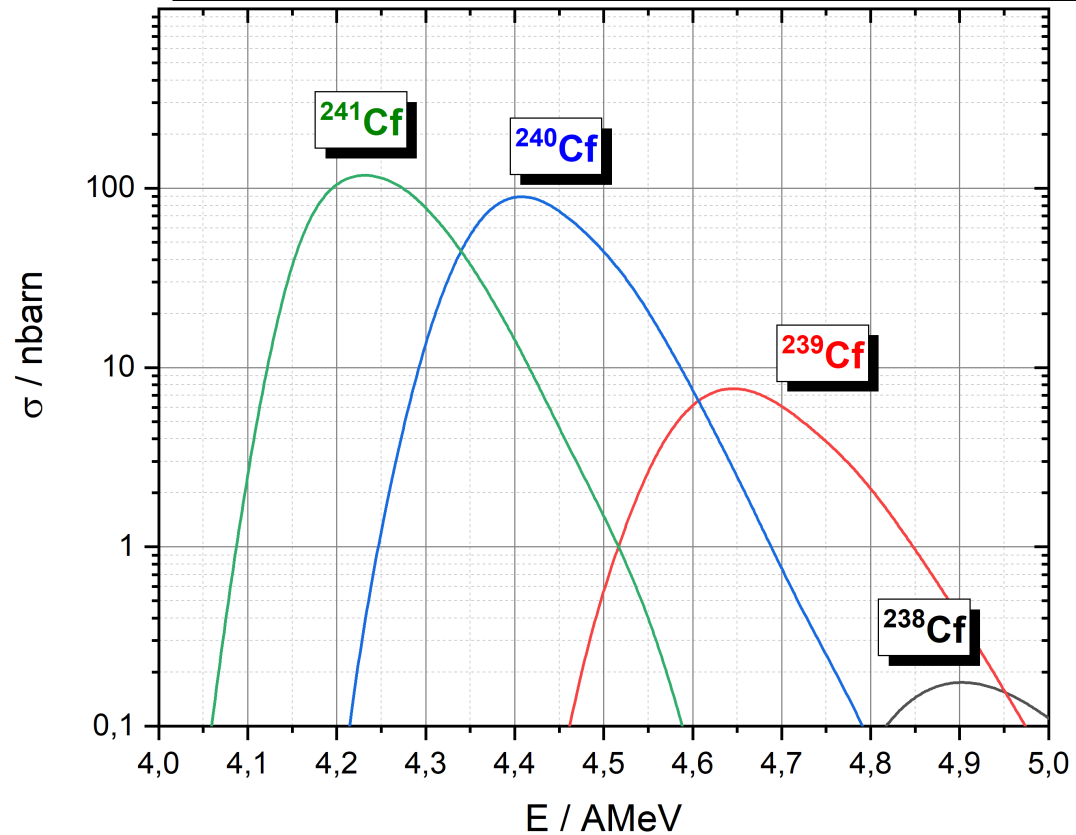
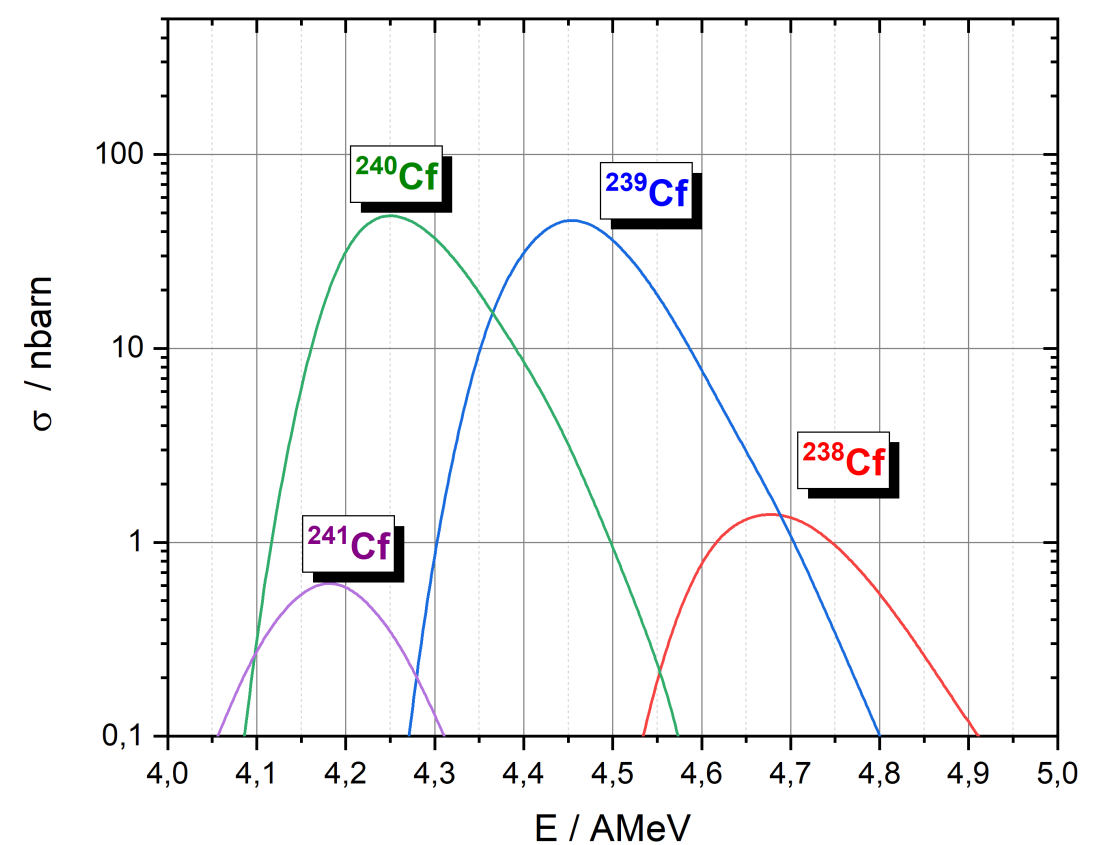
H. Backe, W. Lauth, M. Block, M. Laatiaoui, , Nuclear Physics A, (2015)

Anregungsfkt.  $^{48}\text{Ca} + ^{198}\text{Pt} \rightarrow ^{246}\text{Cf}^*$ , HIVAP - Rechnung



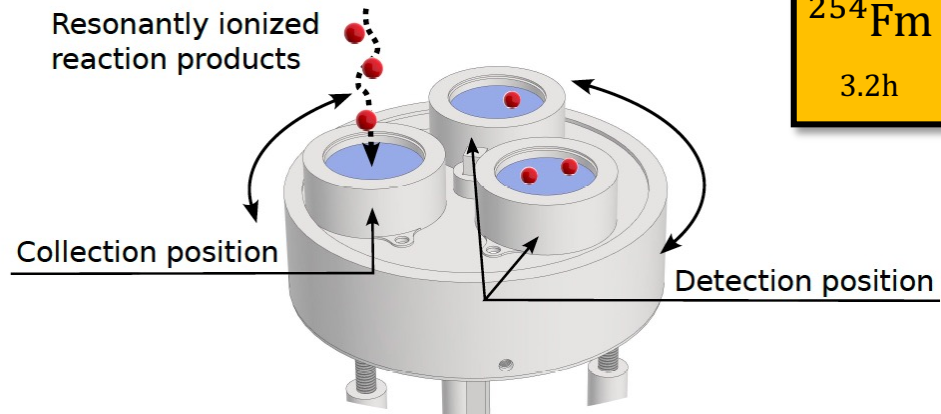
Anregungsfkt.  $^{48}\text{Ca} + ^{196}\text{Pt} \rightarrow ^{244}\text{Cf}^*$ , HIVAP - Rechnung



Anregungsfkt.  $^{48}\text{Ca} + ^{195}\text{Pt} \rightarrow ^{243}\text{Cf}^*$ , HIVAP - RechnungAnregungsfkt.  $^{48}\text{Ca} + ^{194}\text{Pt} \rightarrow ^{242}\text{Cf}^*$ , HIVAP - Rechnung

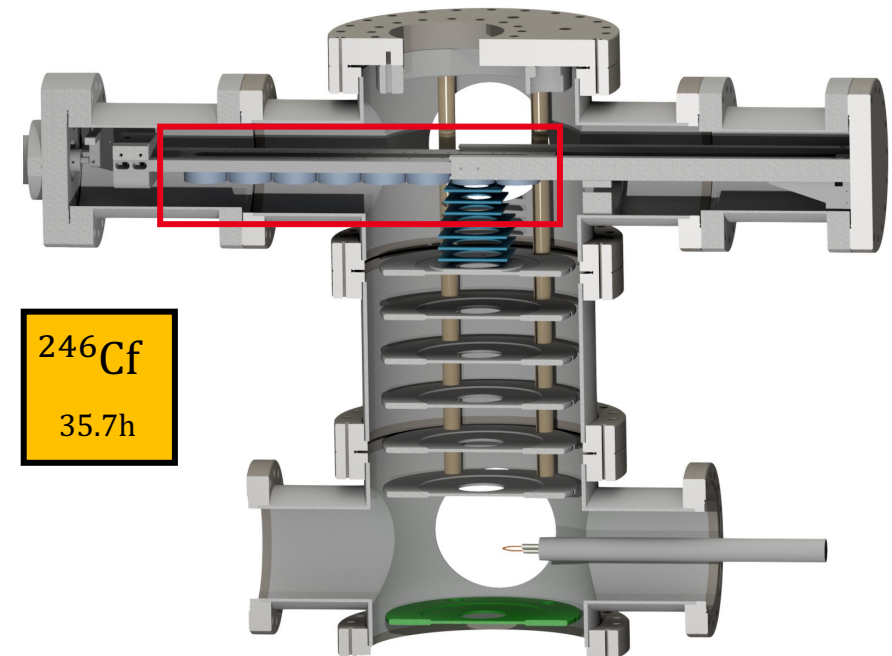
# LONG HALF-LIVES: A CHALLENGE FOR RADRIS

Three rotatable detectors



Access to longer-lived nuclei

Array of eight detectors





# PHYSICS OF HEAVY ELEMENTS

## Nilsson model

### Mean-field potential

$$V(\rho, z) = \frac{1}{2}m(\omega^2\rho^2 + \omega_z^2 z^2)$$

### Quantum Numbers

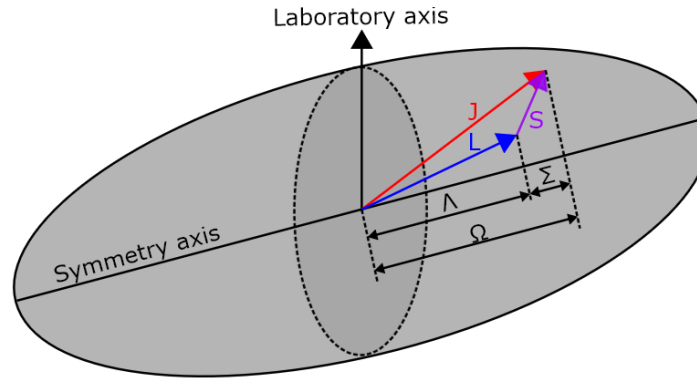
$$\Lambda, \Sigma, \Omega, K$$

### States

$$\Omega^\pi [N, n_z, \Lambda]$$

### Deformation

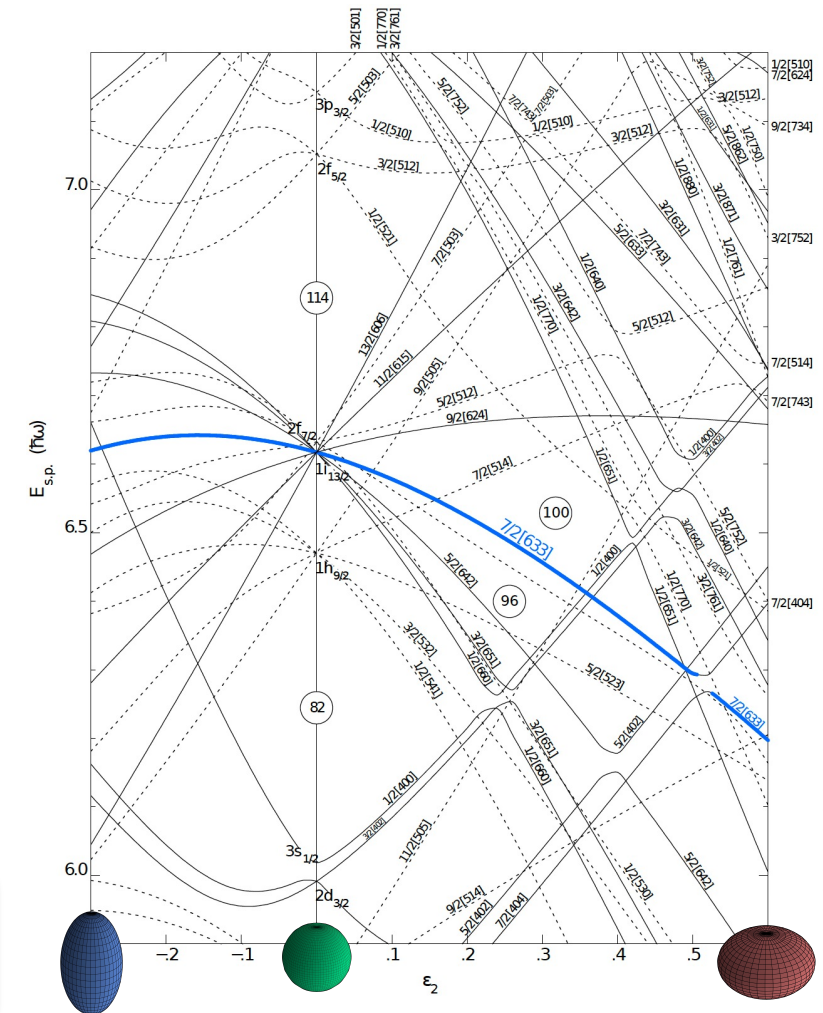
$$\delta = \frac{\Delta r}{\sqrt{\langle r^2 \rangle}} \approx \frac{3}{2} \sqrt{\frac{5}{4\pi}} \beta_2$$



## Nilsson label

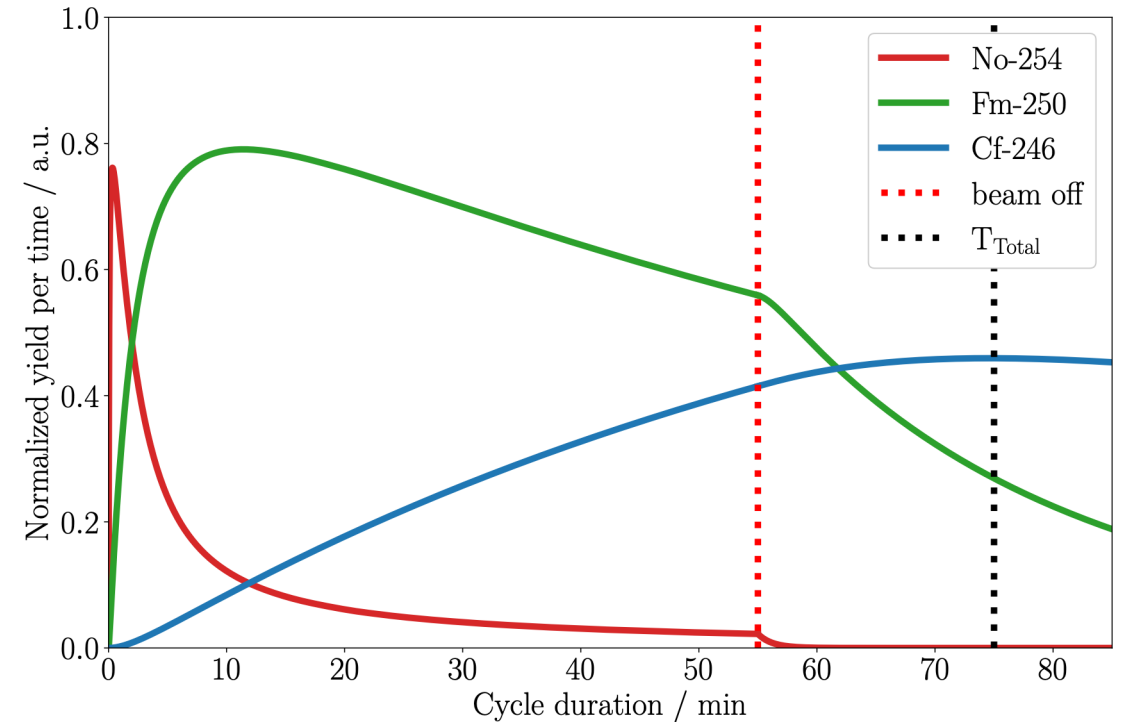
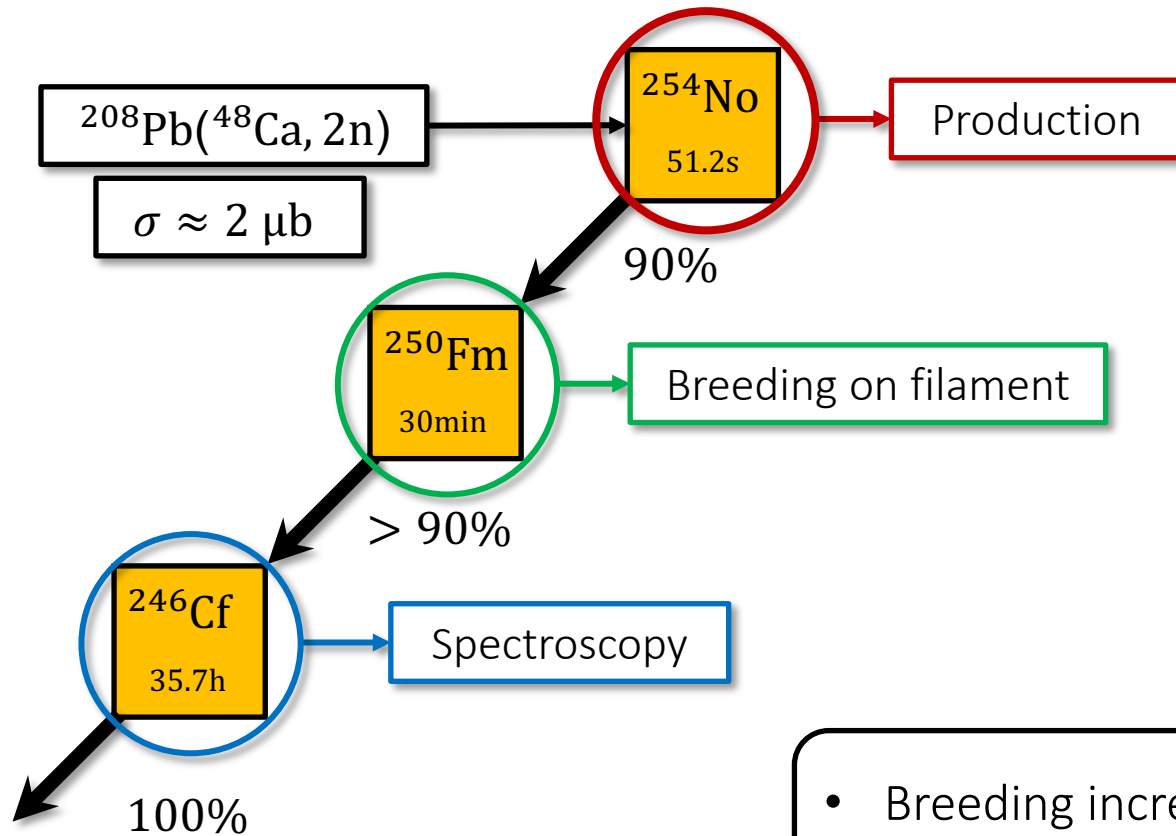
$$7/2^- [514]$$

How do orbitals evolve with deformation?



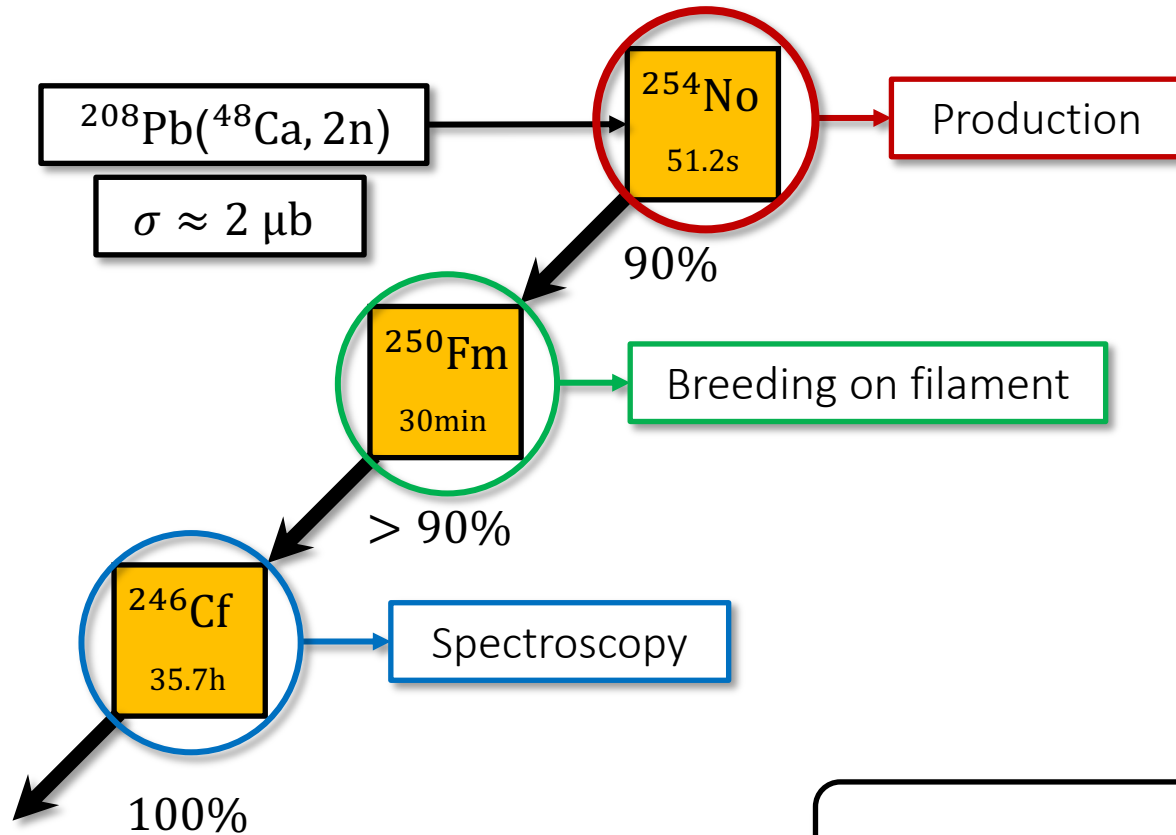
$$\epsilon_2 = \delta + \frac{1}{6}\delta^2 + \frac{5}{18}\delta^3 + \dots$$

# CYCLE FOR CALIFORNIUM 246



- Breeding increases Cf yield and decreases possible background from Fm
- Prolonging cycle past 75 min doesn't significantly increase gain

# CYCLE FOR CALIFORNIUM 246



$$\dot{N}_{\text{No}} = N_{\text{Prod}} - \lambda_{\text{No}} N_{\text{No}}$$

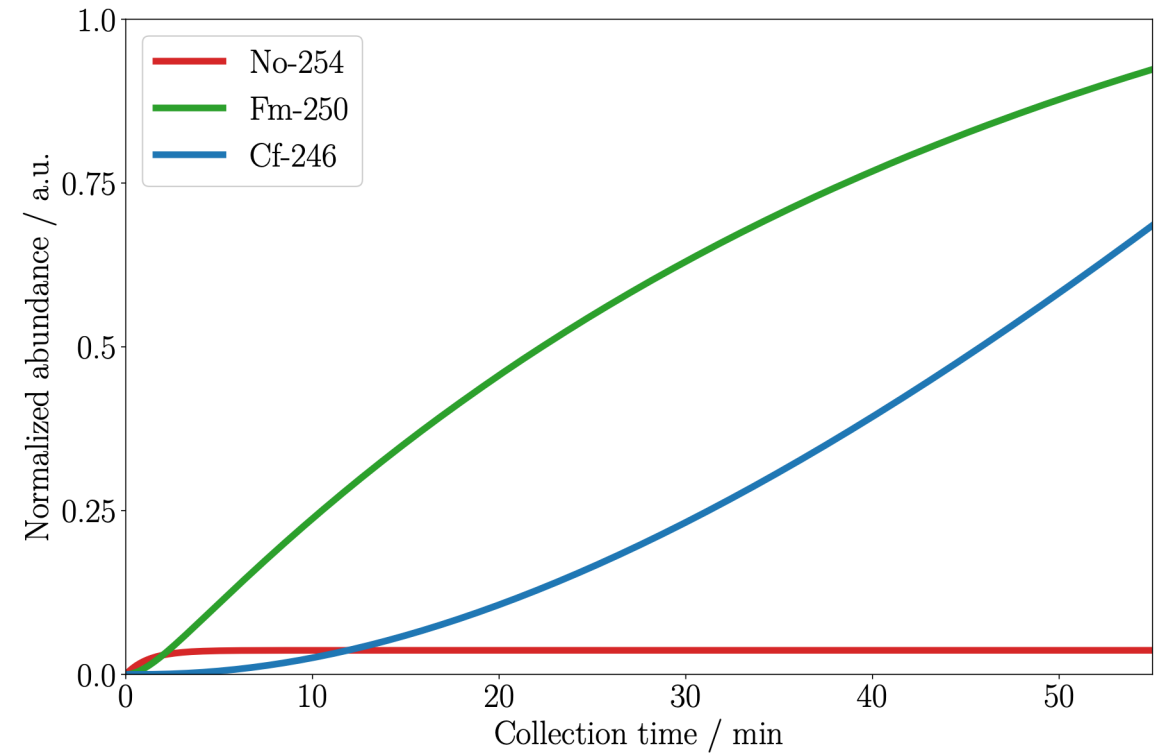
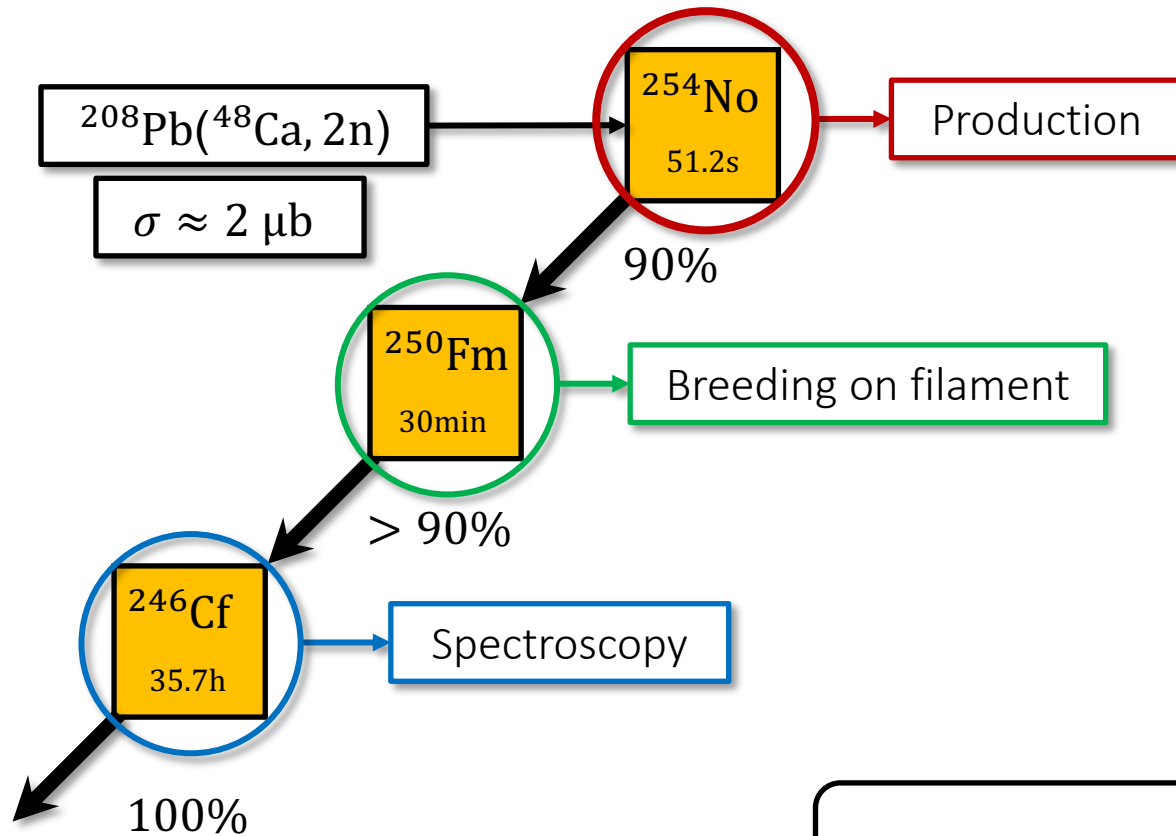
$$\dot{N}_{\text{Fm}} = \lambda_{\text{No}} N_{\text{No}} - \lambda_{\text{Fm}} N_{\text{Fm}}$$

$$\dot{N}_{\text{Cf}} = \lambda_{\text{Fm}} N_{\text{Fm}} - \lambda_{\text{Cf}} N_{\text{Cf}}$$

$$N_{\text{No}}(0) = N_{\text{Cf}}(0) = N_{\text{Fm}}(0) = 0$$

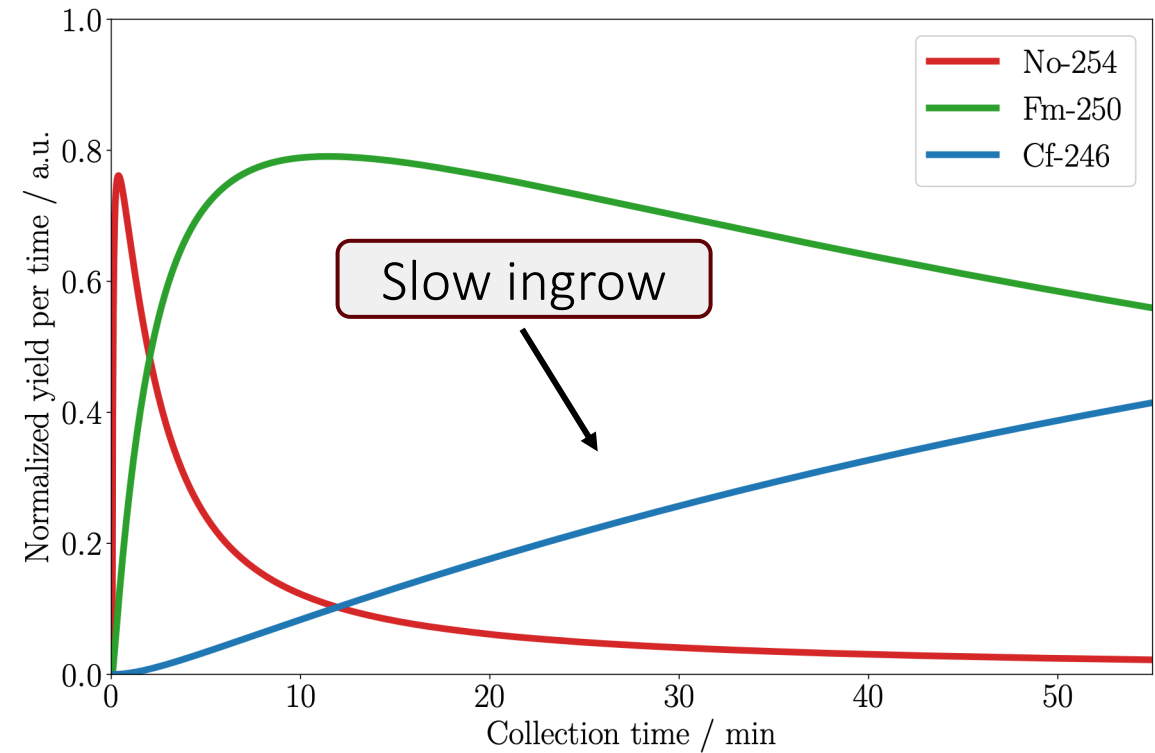
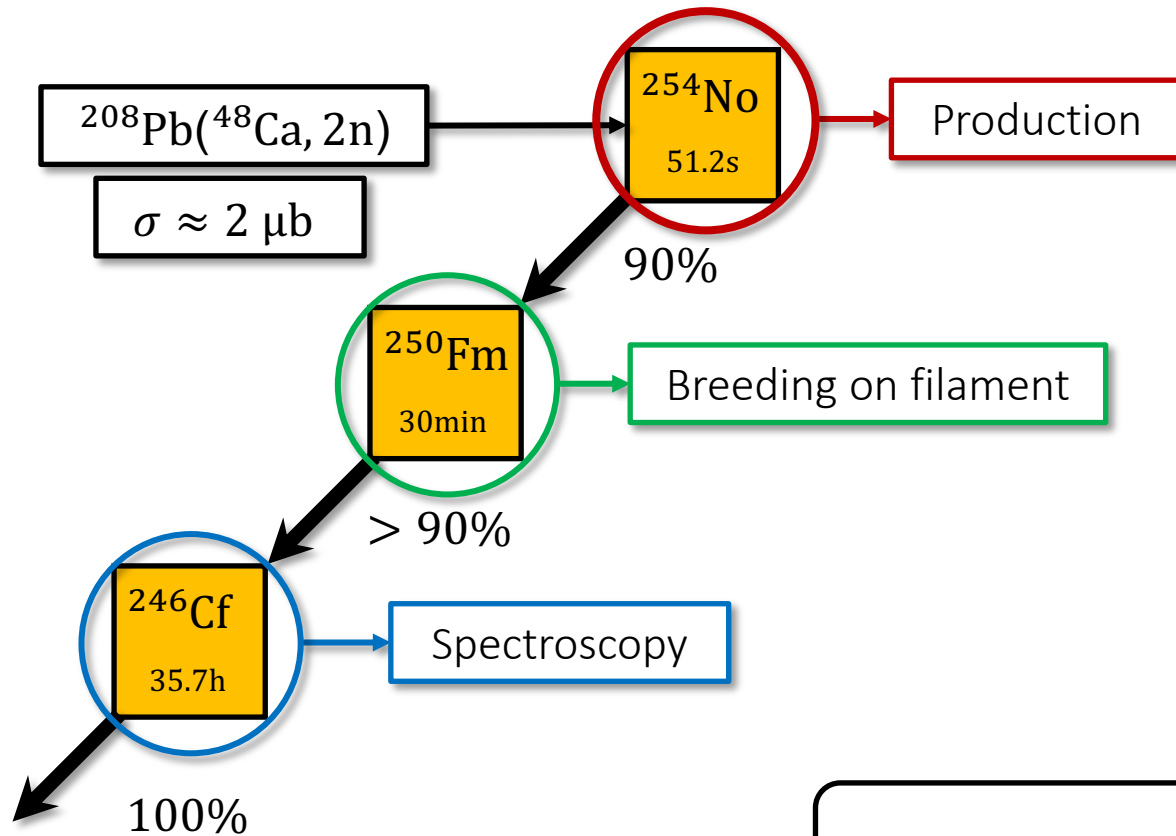
Cycle time optimized based on half-life of nuclide of interest

# CYCLE FOR CALIFORNIUM 246



Efficiency is given by normalizing abundance over time

# CYCLE FOR CALIFORNIUM 246

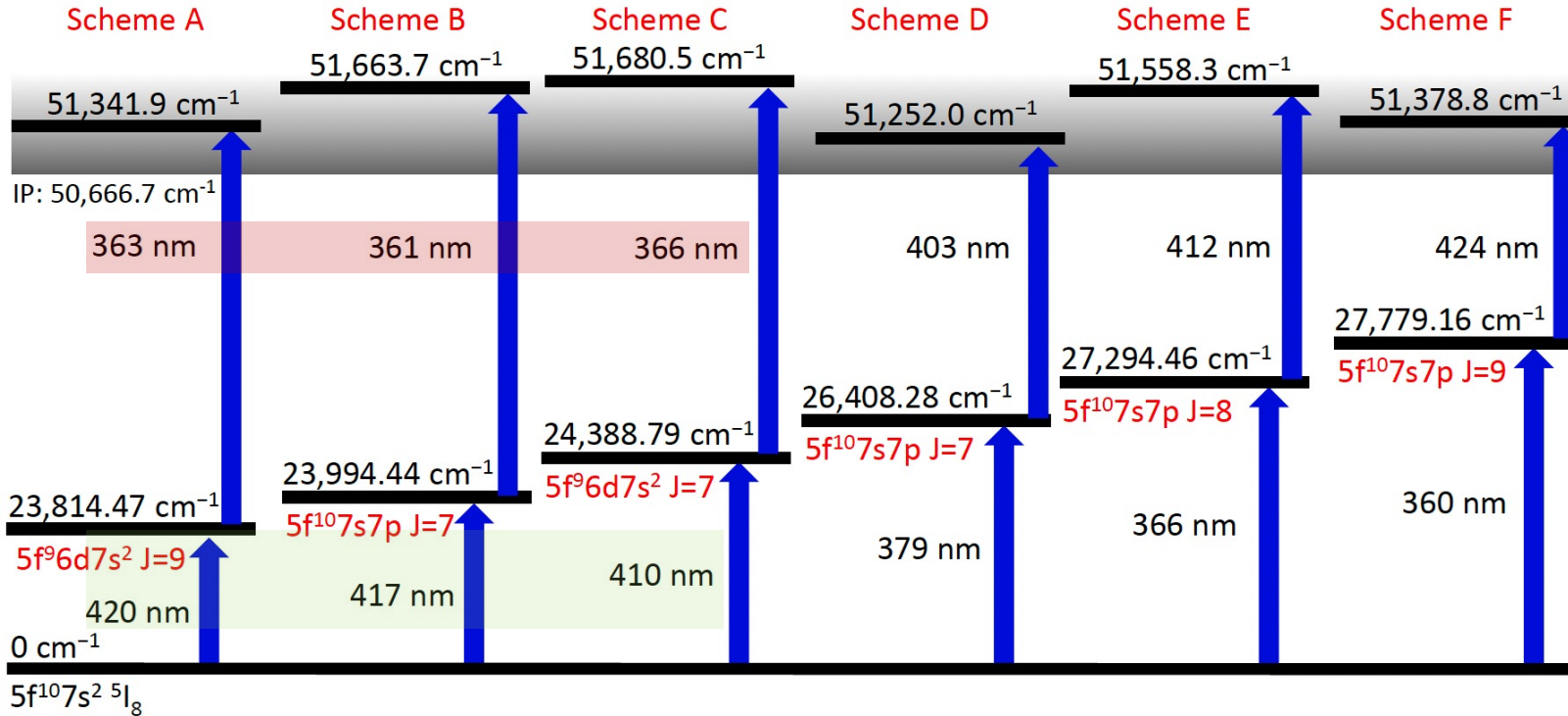


Waiting for maximized Cf yield unfeasible → compromise

# EVOLUTION OF CHANGES IN MEAN-SQUARE CHARGE RADII IN CALIFORNIUM ISOTOPES



TECHNISCHE  
UNIVERSITÄT  
DARMSTADT



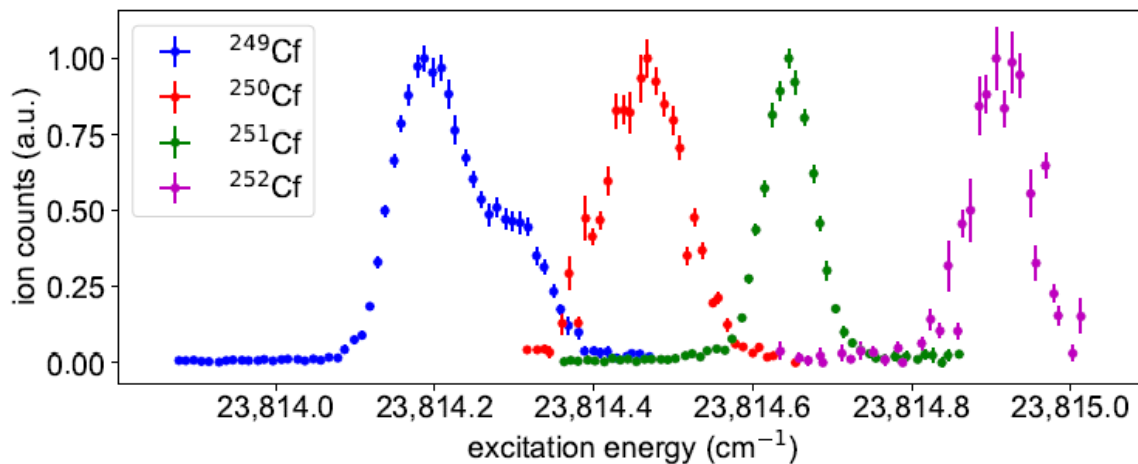
➤ Identified in  $^{250}\text{Cf}$

Non-Resonant

355 nm  
 $28169.0 \text{ cm}^{-1}$

Resonant (AI)

|     |     |         |
|-----|-----|---------|
| DMQ | 360 | 342-385 |
|-----|-----|---------|



1<sup>st</sup> Step

|                                |     |           |
|--------------------------------|-----|-----------|
| Exalite 411                    | 411 | 404 - 417 |
| Exalite 417                    | 419 | 413 - 422 |
| Stilbene 3<br>aka Stilbene 420 | 425 | 419 - 434 |



| Isotope           | Reaction                                                              | $\alpha$ -Branch (%) | $\sigma_{\text{Production}}$ (nb) | $\sigma_{\text{Effective}}$ (nb) | $\sigma_{\text{Prod.}} / \sigma_{\text{Eff.}}$ | Ions per hour |
|-------------------|-----------------------------------------------------------------------|----------------------|-----------------------------------|----------------------------------|------------------------------------------------|---------------|
| $^{240}\text{Cf}$ | $^{195}\text{Pt}(^{48}\text{Ca}, 3n)$                                 | 98                   | 9                                 | 8.8                              | 1.02                                           | $\approx 1$   |
| $^{241}\text{Cf}$ | $^{196}\text{Pt}(^{48}\text{Ca}, 3n)$                                 | 25                   | 40                                | 10                               | 4                                              | 1 - 1.5       |
| $^{242}\text{Cf}$ | $^{196}\text{Pt}(^{48}\text{Ca}, 2n)$                                 | 100                  | 40                                | 40                               | 1                                              | 5 - 6         |
| $^{243}\text{Cf}$ | $^{198}\text{Pt}(^{48}\text{Ca}, 3n)$                                 | 14                   | 200                               | 28                               | 7.14                                           | 3.5 - 4       |
| $^{244}\text{Cf}$ | $^{198}\text{Pt}(^{48}\text{Ca}, 2n)$                                 | 70(20)               | 100                               | 70                               | 1.42                                           | 9 - 10        |
| $^{245}\text{Cf}$ | $^{207}\text{Pb}(^{48}\text{Ca}, 2n)$<br>55% $\alpha$ , 15% $\alpha$  | 35.3(25)             | 1000                              | 29.12                            | 34.4                                           | 0.3 – 0.4     |
| $^{246}\text{Cf}$ | $^{208}\text{Pb}(^{48}\text{Ca}, 2n)$<br>90% $\alpha$ , >90% $\alpha$ | 100                  | 1800                              | 1458                             | 1.23                                           | 18 - 22       |

$$\sigma_{\text{Effective}} = \sigma_{\text{Production}} * \prod_i \alpha_{\text{Branch},i}$$