

# Coupling atomic and nuclear physics degrees of freedom in highly charged ions

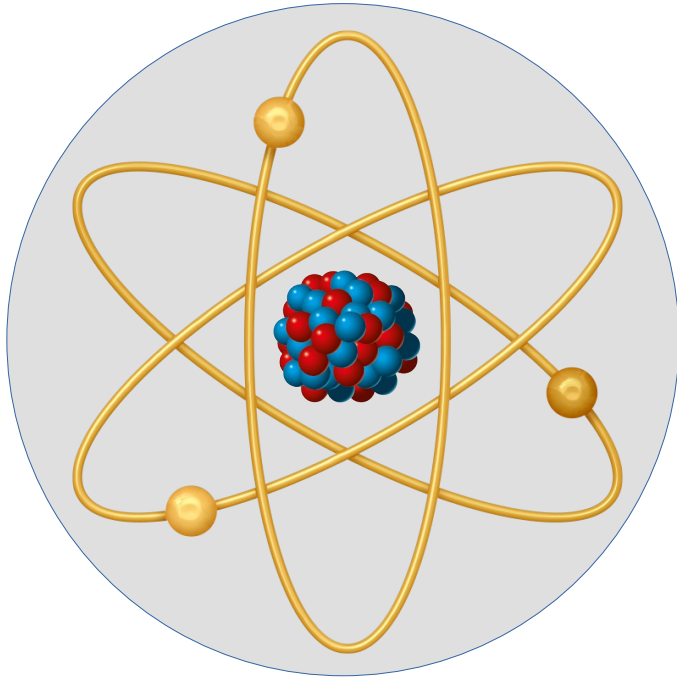
Adriana Pálffy  
University of Würzburg, Germany



Nuclear Pole Seminar, Orsay

12 June 2026

# Overlap between atomic and nuclear physics



- Precision measurements of atomic electrons explore nuclear physics properties

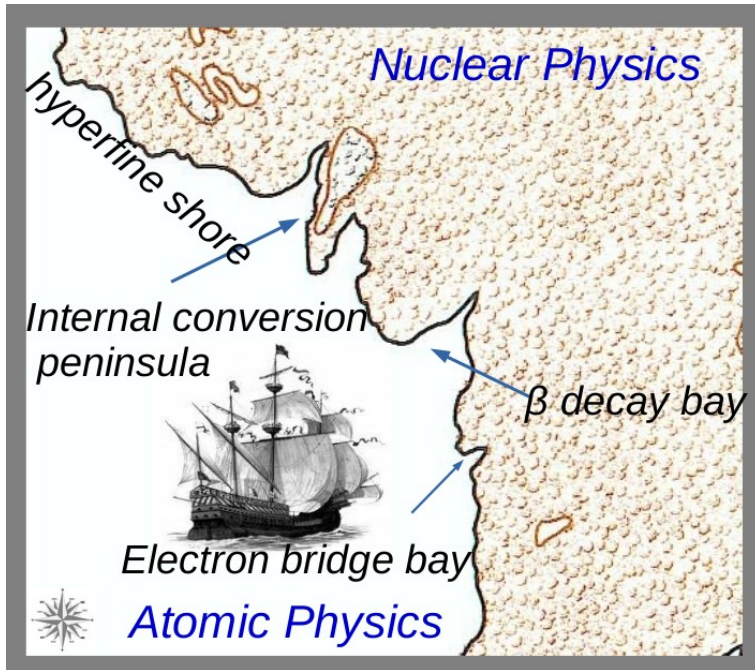
Isotope shifts  
Hyperfine splitting  
Atomic parity violation

- Nuclear processes directly involving atomic electrons

Bound beta decay & electron capture  
Internal conversion  
Nuclear excitation by electron capture  
Electronic bridge

*AP, Contemporary Physics 51, 471 (2010)*

# Overlap between atomic and nuclear physics



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# Nuclear processes involving electrons

- Electron capture (EC)  $p + e_b \rightarrow n + \nu_e$
- Bound beta decay  $n \rightarrow p + e_b + \tilde{\nu}_e$

VOLUME 77, NUMBER 26

PHYSICAL REVIEW LETTERS

23 DECEMBER 1996

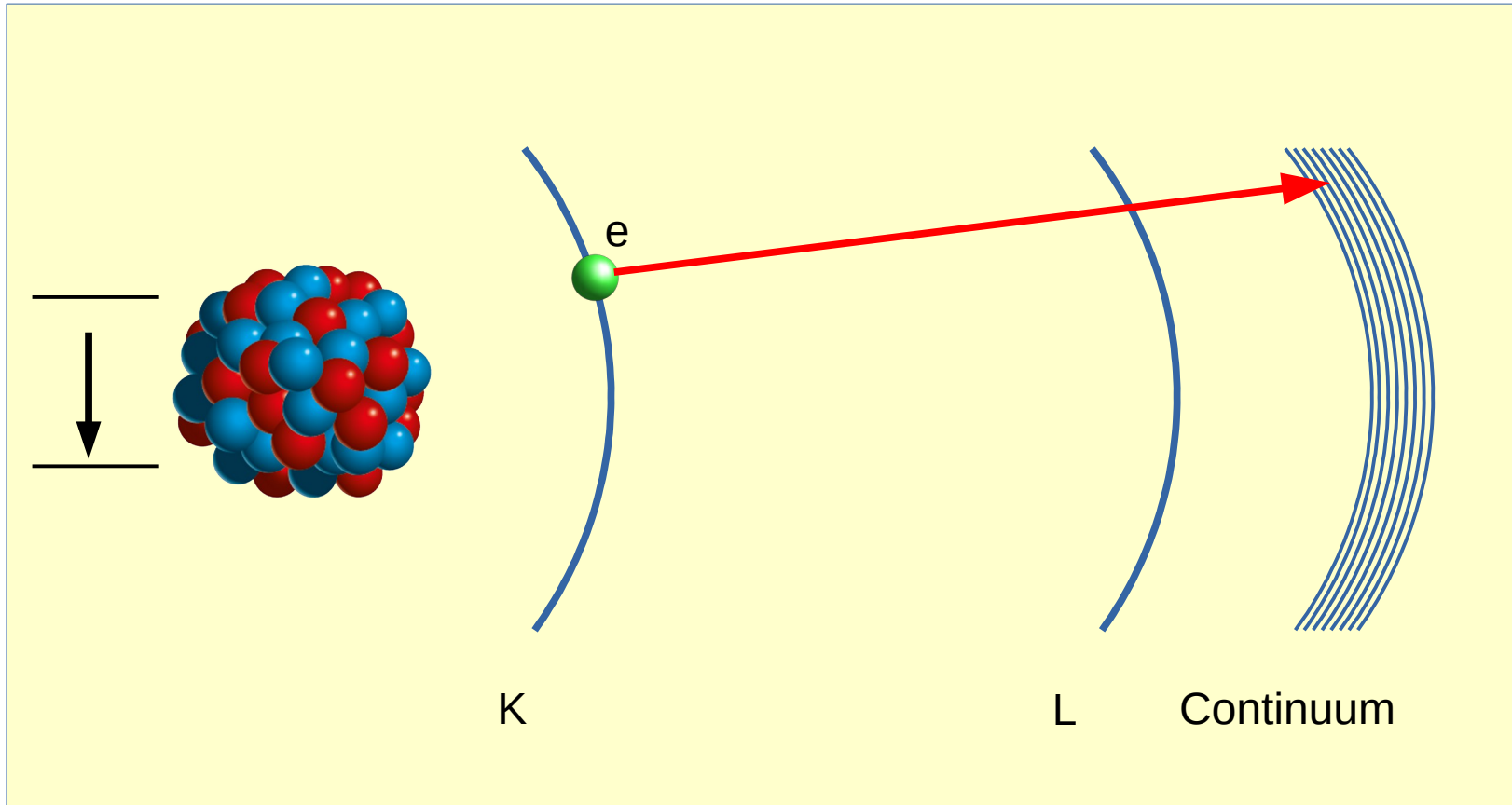
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## Observation of Bound-State $\beta^-$ Decay of Fully Ionized $^{187}\text{Re}$ : $^{187}\text{Re}$ - $^{187}\text{Os}$ Cosmochronometry

F. Bosch,<sup>1</sup> T. Faestermann,<sup>2</sup> J. Friese,<sup>2</sup> F. Heine,<sup>2</sup> P. Kienle,<sup>2</sup> E. Wefers,<sup>2</sup> K. Zeitelhack,<sup>2</sup> K. Beckert,<sup>1</sup> B. Franzke,<sup>1</sup>  
O. Klepper,<sup>1</sup> C. Kozhuharov,<sup>1</sup> G. Menzel,<sup>1</sup> R. Moshhammer,<sup>1</sup> F. Nolden,<sup>1</sup> H. Reich,<sup>1</sup> B. Schlitt,<sup>1</sup> M. Steck,<sup>1</sup>  
T. Stöhlker,<sup>1</sup> T. Winkler,<sup>1</sup> and K. Takahashi<sup>2,3</sup>

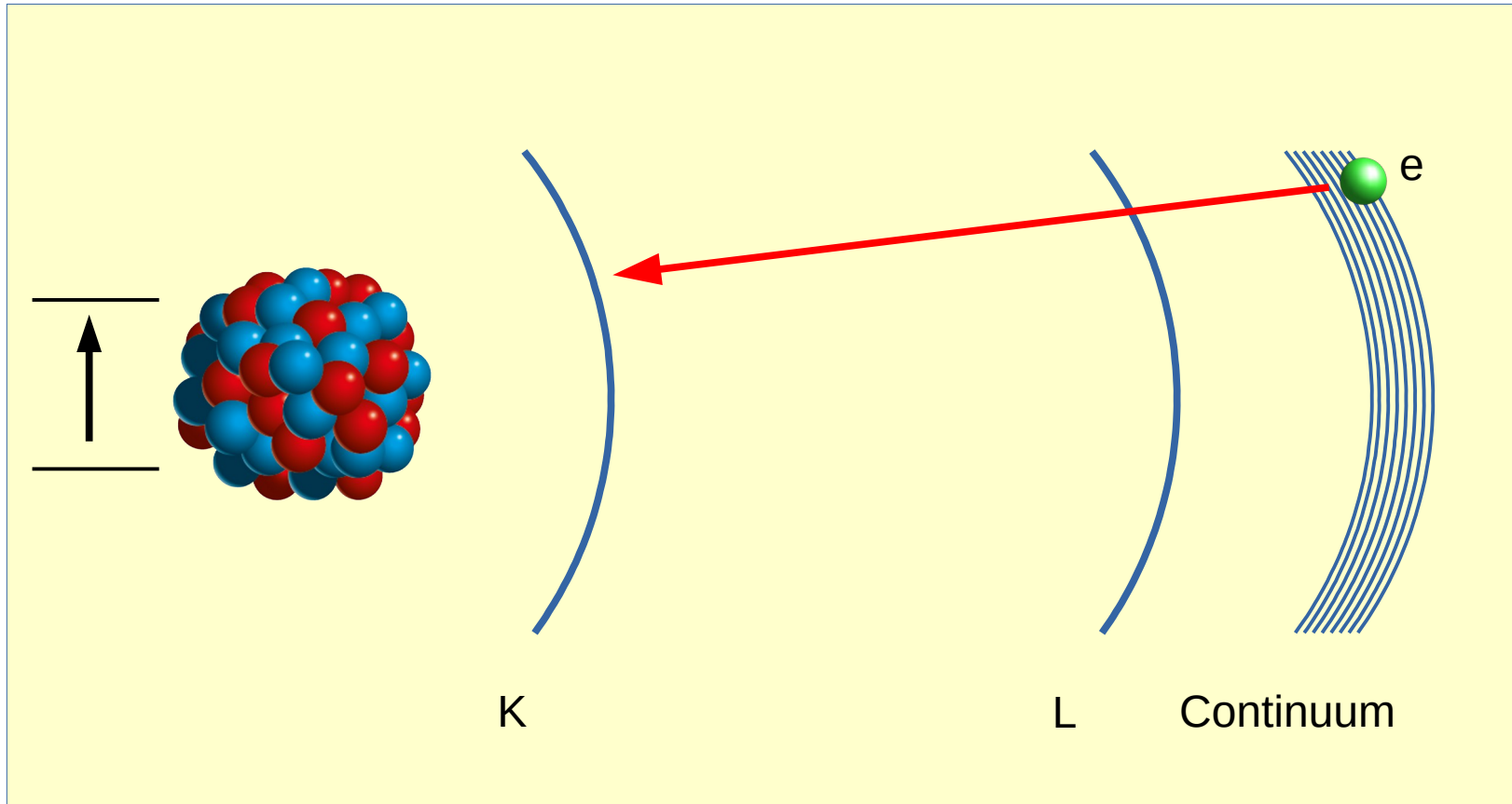
# Internal conversion and NEEC

Internal conversion



# Internal conversion and NEEC

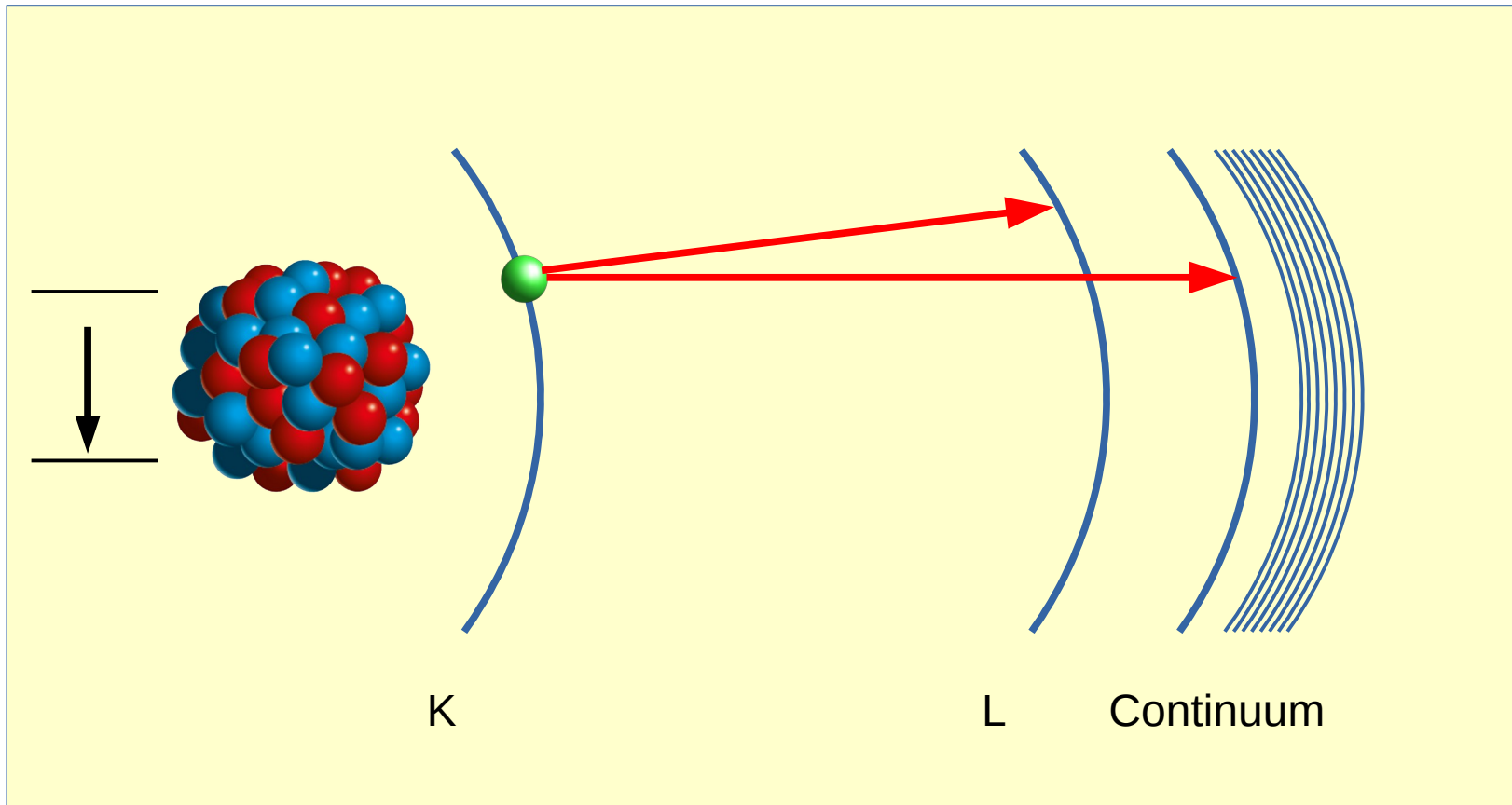
Nuclear excitation by electron capture (NEEC)



Chronologically, IC - 1924, NEEC proposed – 1976, observed (?) 2018

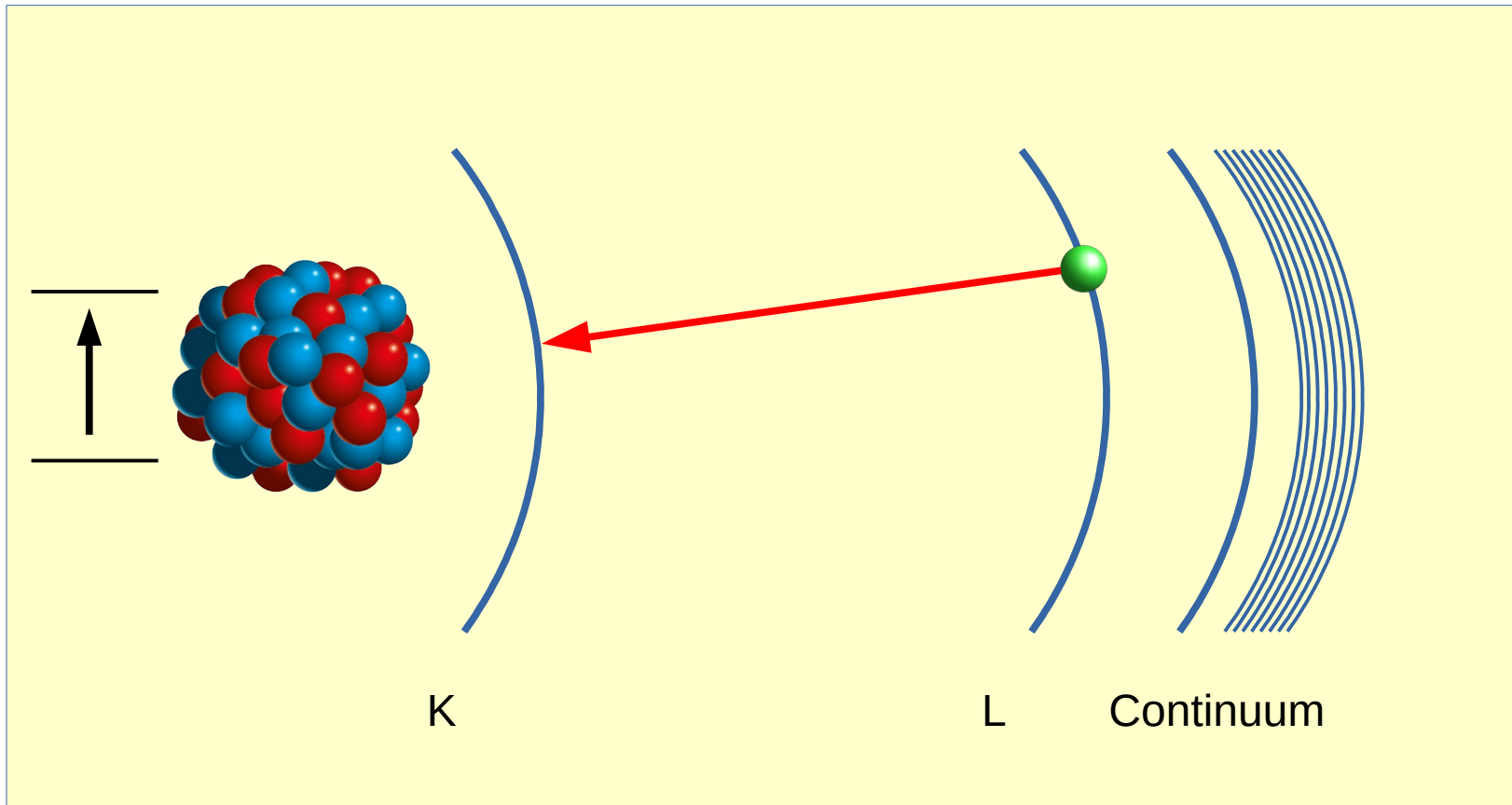
# Bound internal conversion and NEET

Bound internal conversion



# Bound internal conversion and NEET

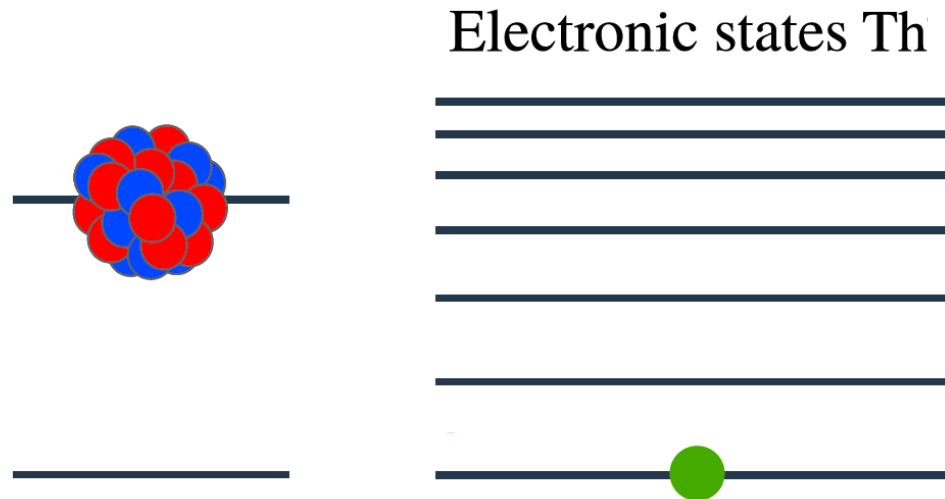
Nuclear excitation by electron transition



Difficult to find in nature such perfect matches of atomic and nuclear transition energies!

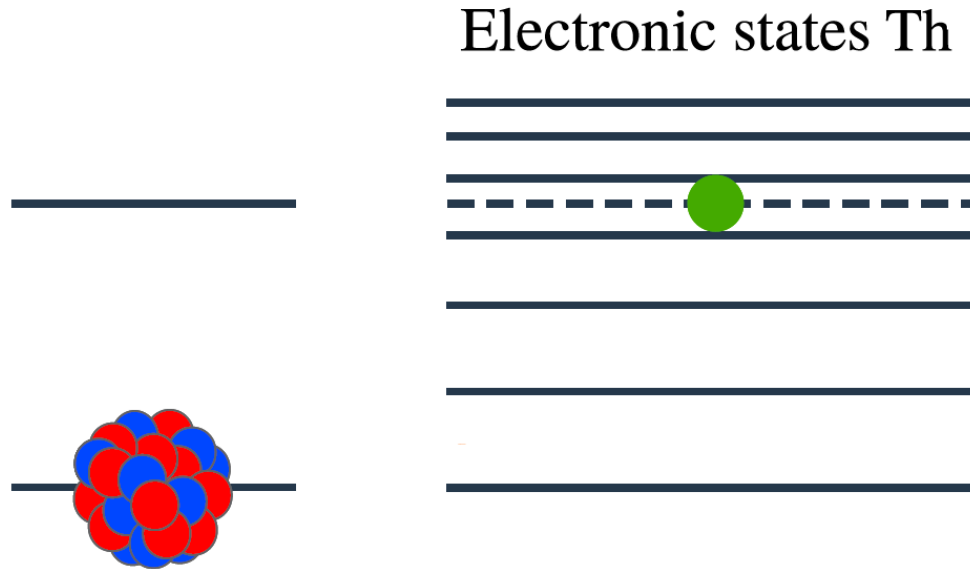
# Electronic bridge (EB)

Nuclear decay via IC not possible, energy is not sufficient to ionize a bound electron!



# Electronic bridge (EB)

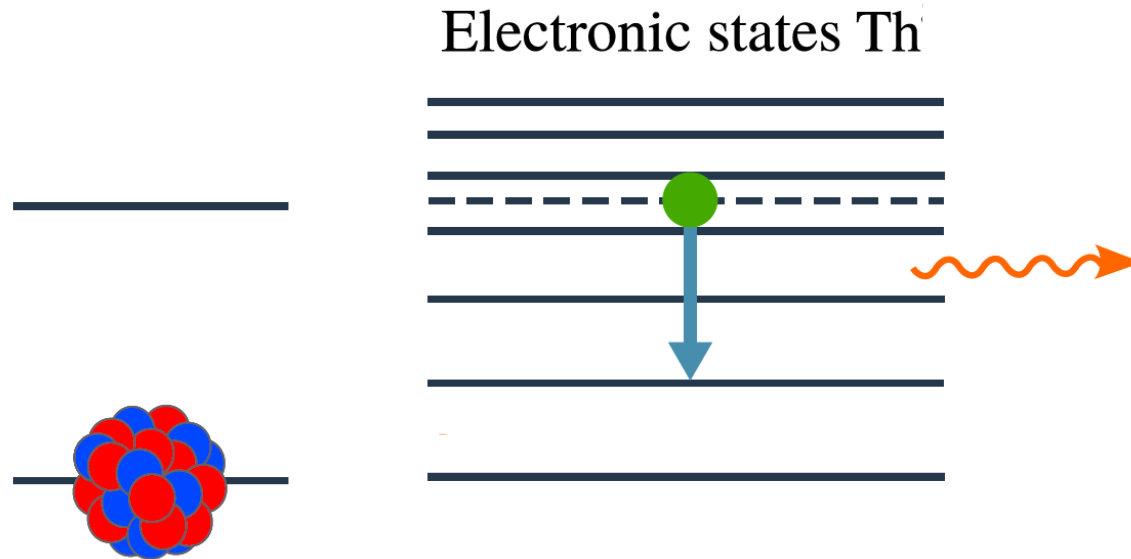
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There is no electronic state at the right energy! **Virtual state!**

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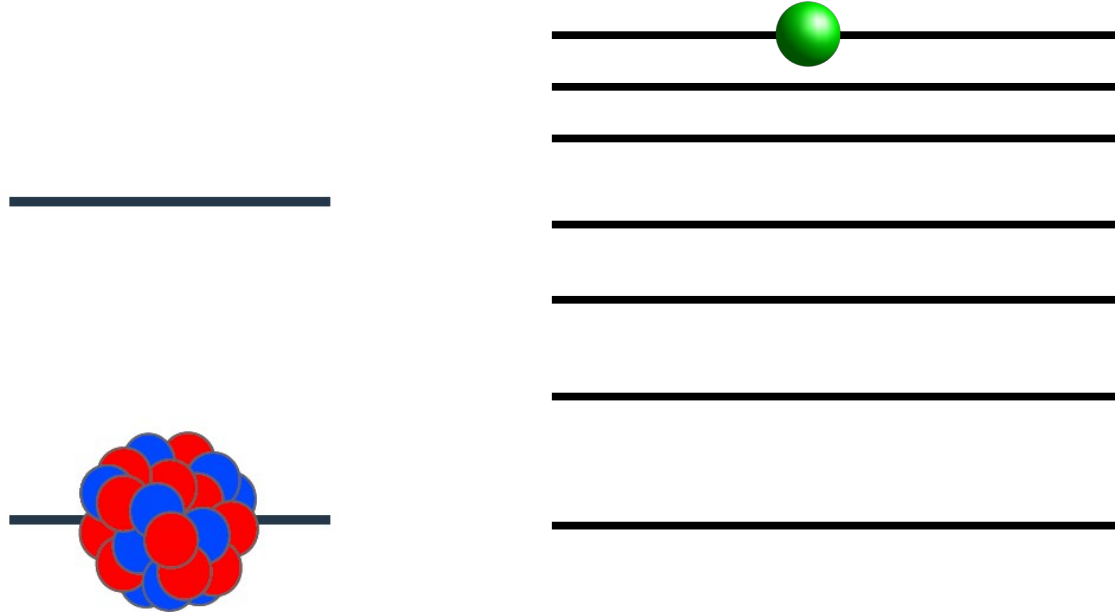


*S. G. Porsev et al, PRL 105, 182501 (2010)*

There is no electronic state at the right energy! **Virtual state!** Decays by emitting a photon!

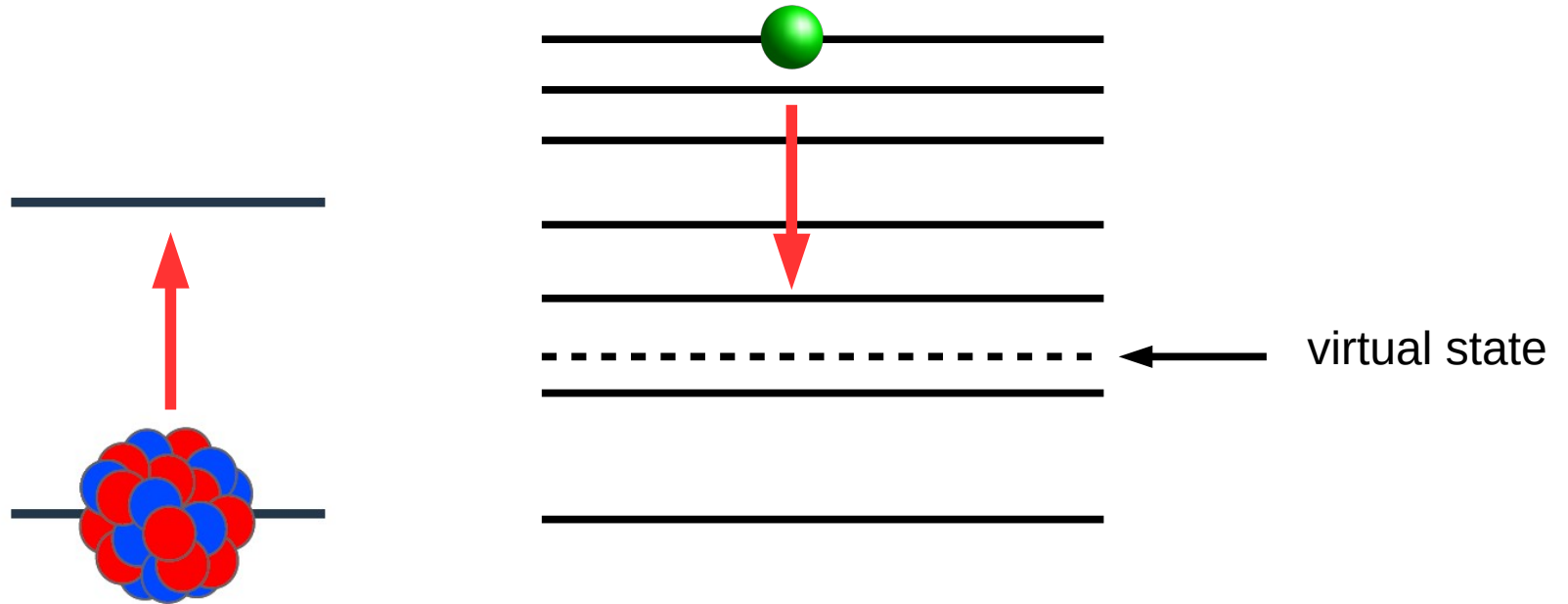
# Imagine EB with nuclear excitation

Excitation energy from the electronic shell does not match nuclear transition energy



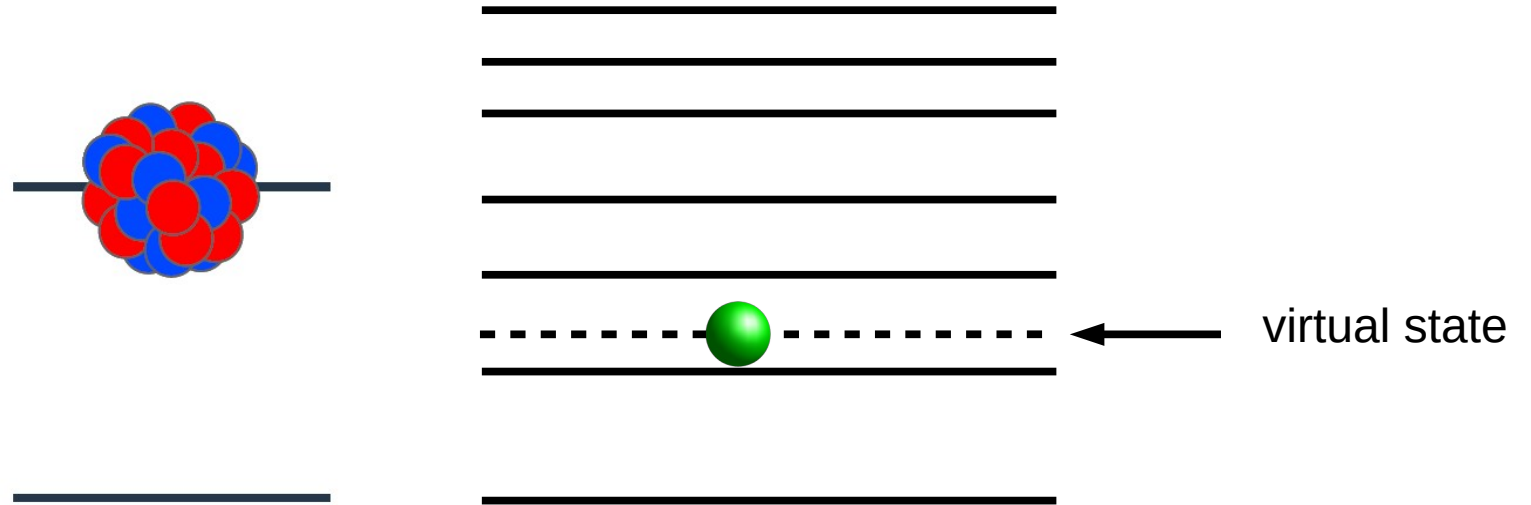
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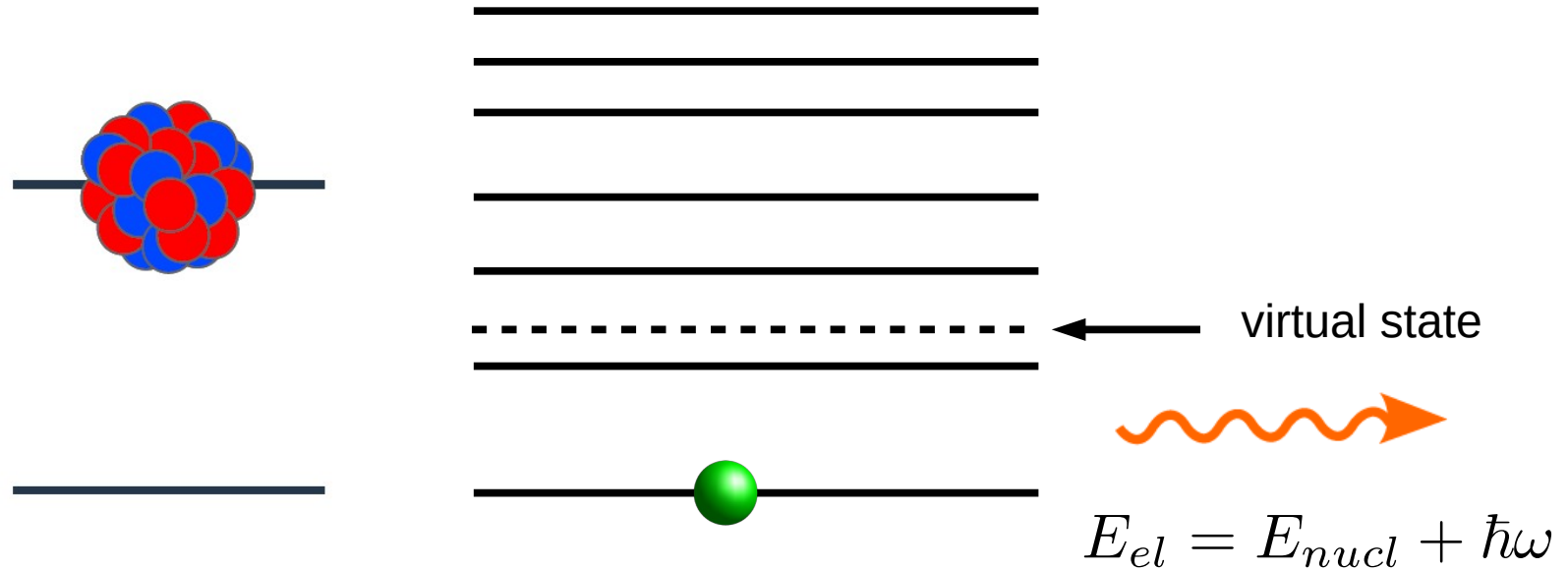
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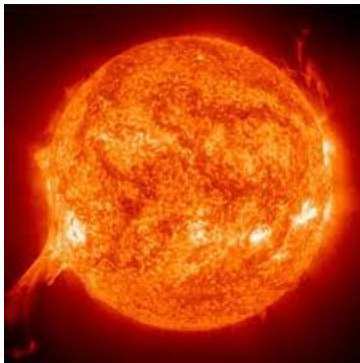
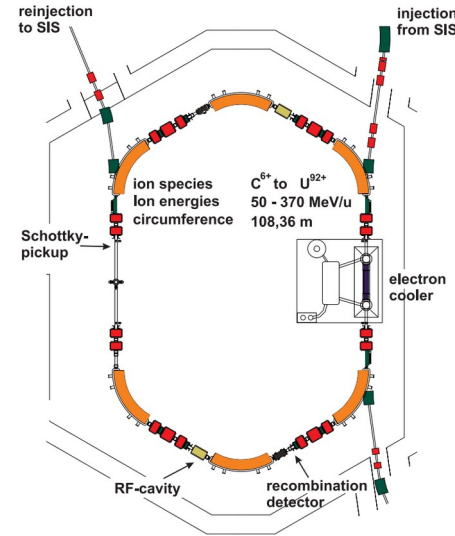
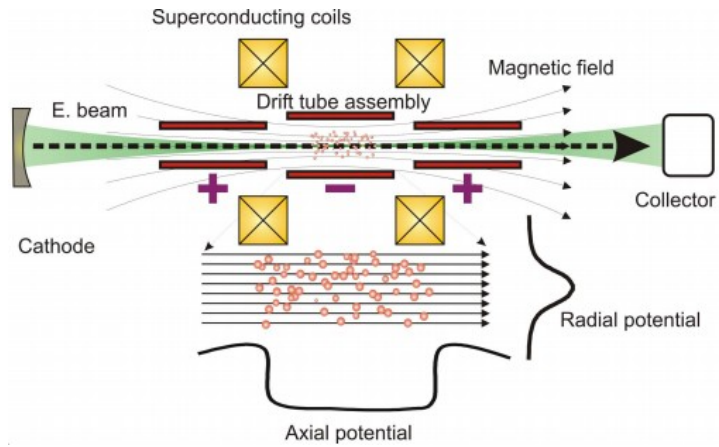
# Imagine EB with nuclear excitation

Excitation energy from the electronic shell does not match nuclear transition energy



# (Highly) charged ions

Nuclear excitation processes require atomic shell vacancies or even highly charged ions

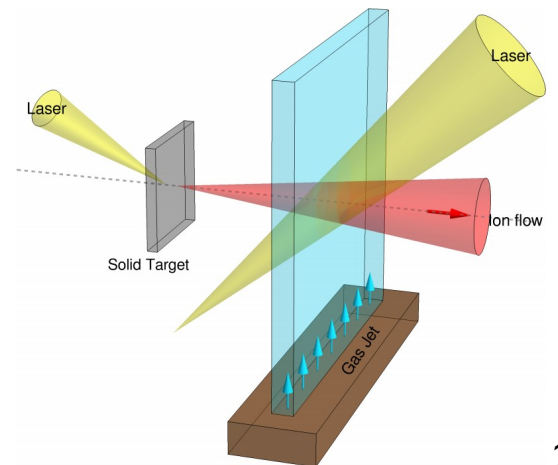


Storage rings

EBIT

Plasmas

Nuclear reactions



# Outline

## I. NEEC in highly charged ions

- ... controversy about experimental observation
- ... using electronic vortex beams
- ... signature from precision mass measurements

## II. Nuclear coupling to the atomic shell in $^{229}\text{Th}$

- ... in Th-doped crystals
- ... via nuclear hyperfine mixing

# Outline

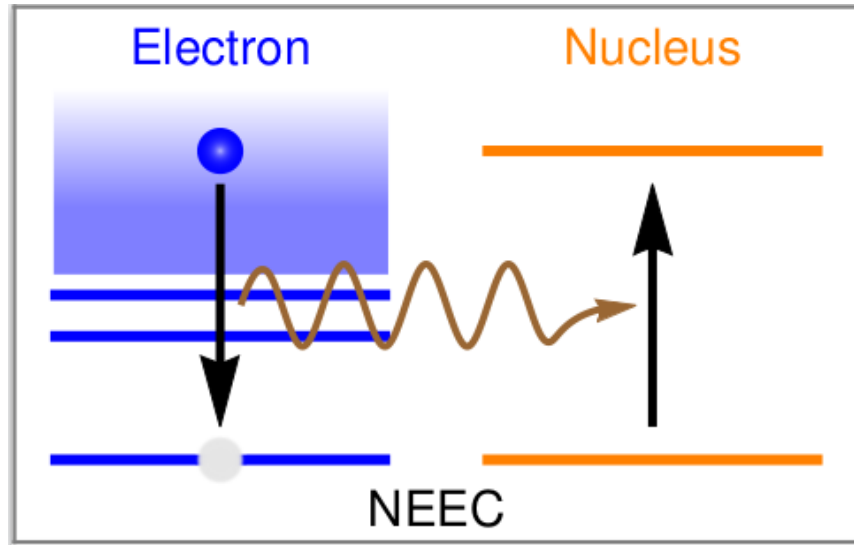
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# Interest in NEEC



- first proposed theoretically by Goldanskii & Namiot *Phys. Lett. 62B (1976)*
- First experimental observation claimed in 2018 *C. J. Chiara et al., Nature 554, 216*  
contested by others  
*S. Guo et al., Phys. Rev. Lett. 128, 242502 (2022)*

## Study of:

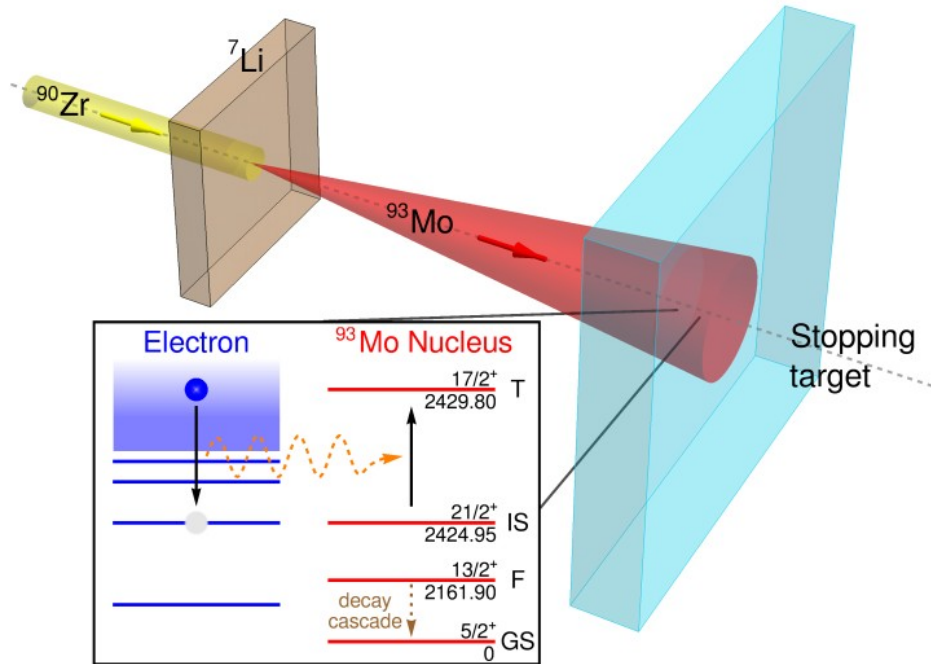
population mechanisms of excited nuclear levels  
atomic vacancy effects on nuclear lifetime

## Relevant for:

dense astrophysical plasmas  
depletion of isomers

# NEEC in beam-based scenario

- First experimental evidence of NEEC was reported in a beam-target scenario



- Depletion probability  $P_{exc} = 0.01$  per  $^{93m}\text{Mo}$  was reported

*C. J. Chiara et al., Nature, 554, 216 (2018)*

- Observed excitation probability was attributed to NEEC process

- A theoretical analysis of NEEC rates for the experimental setting reports  $P_{exc} = 10^{-11}$ !

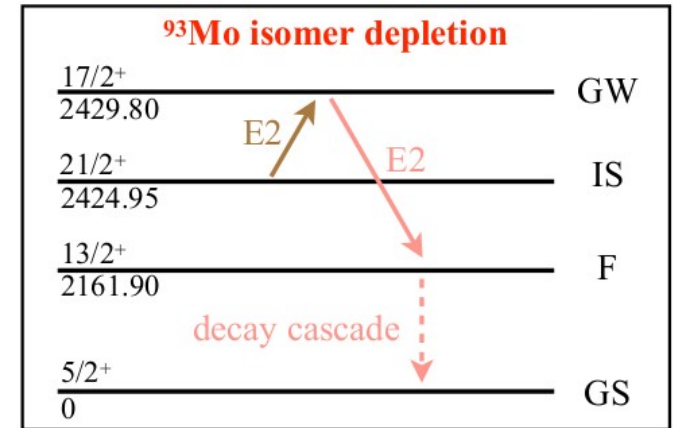
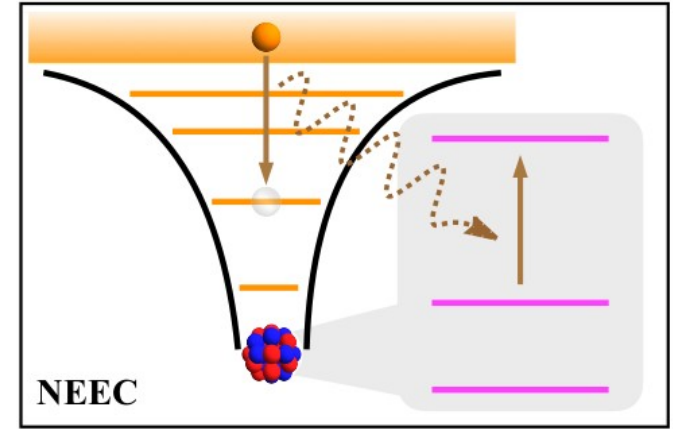
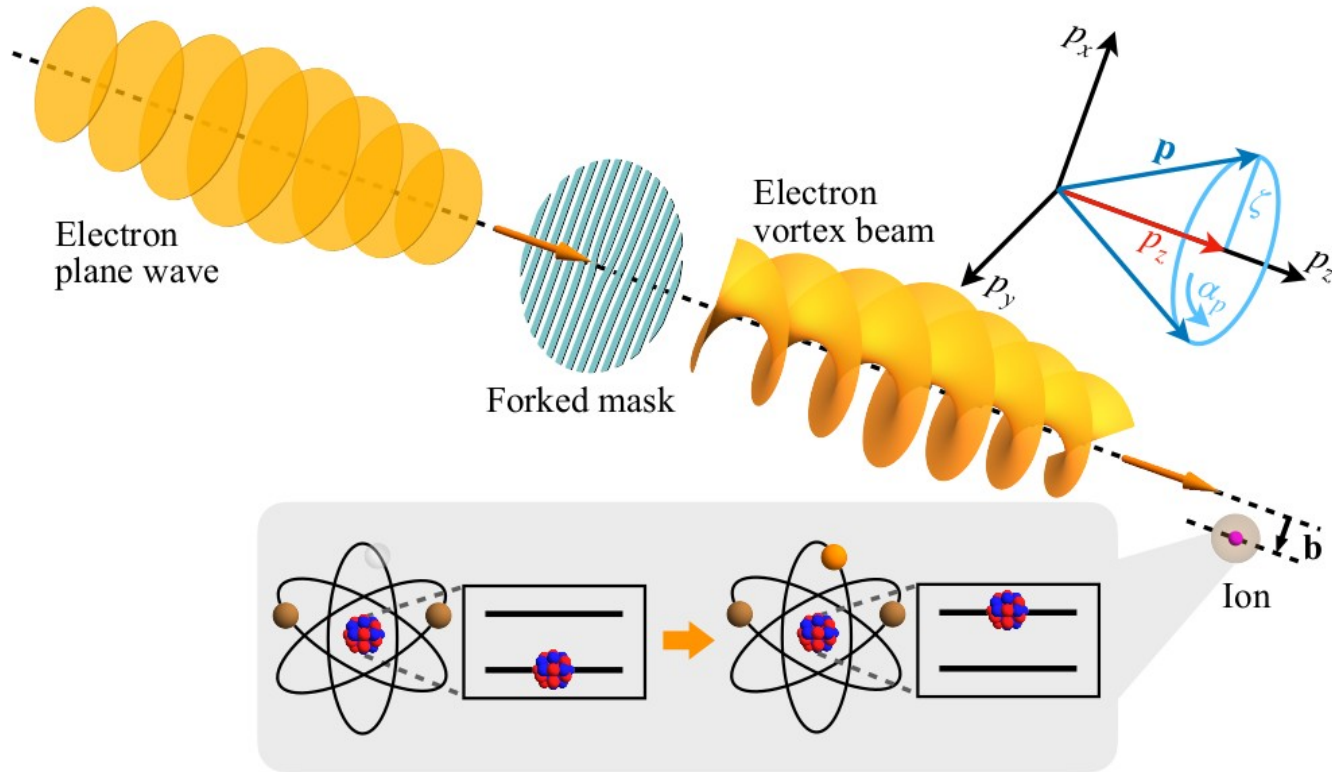
*Wu, Keitel, Pálffy, Phys. Rev. Lett. 122, 212501 (2019)*

- Debate is still in progress.

*J. Rzadkiewicz et al., PRL 127, 042501 (2021)*

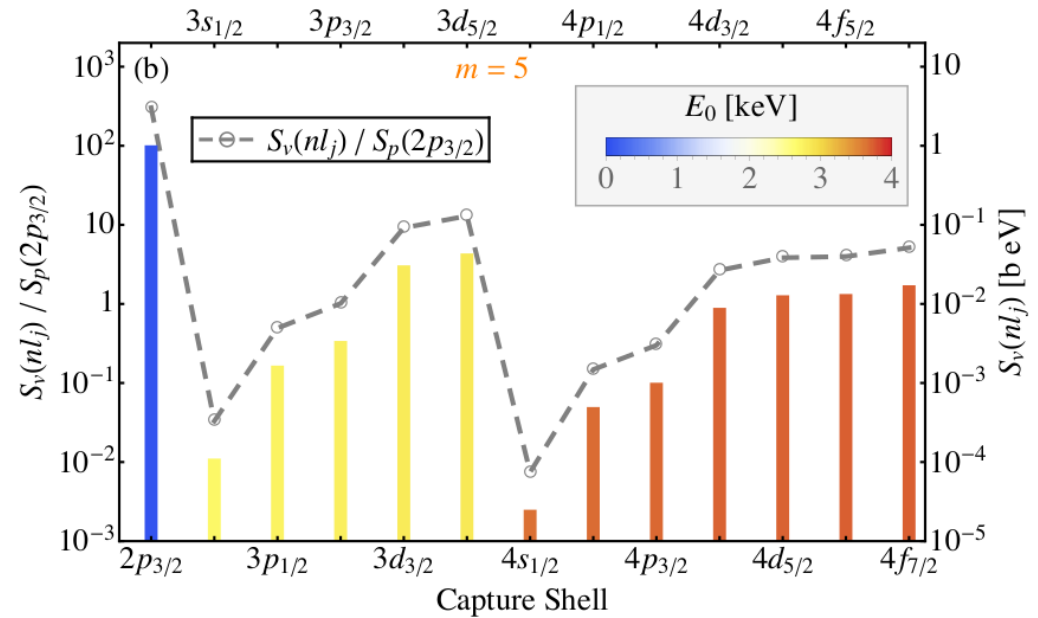
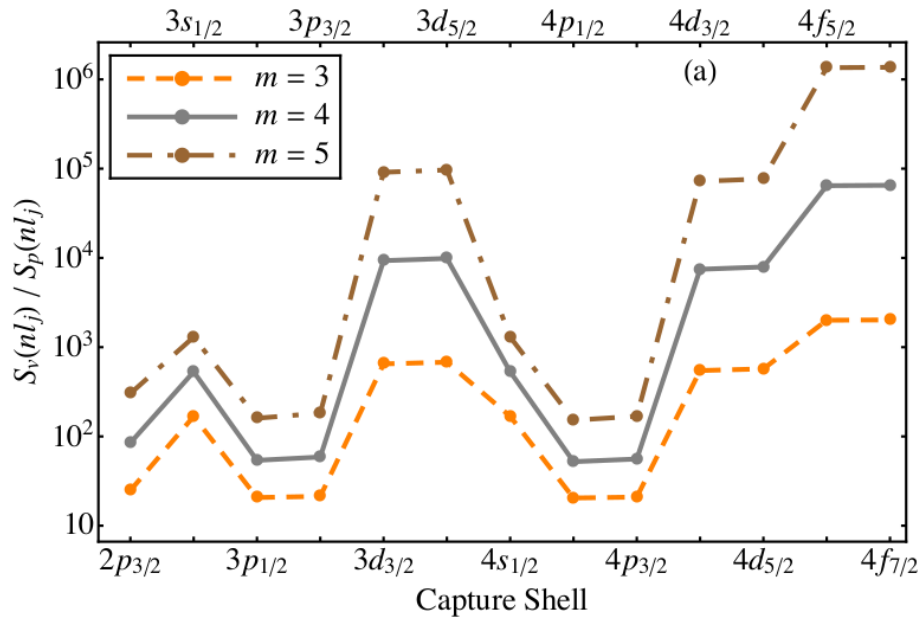
*S. Guo et al., PRL 128, 242502 (2022)*

# Spatially shaped electron beam



Electron vortex beam carry orbital angular momentum

# Vortex beam NEEC for $^{93m}\text{Mo}$

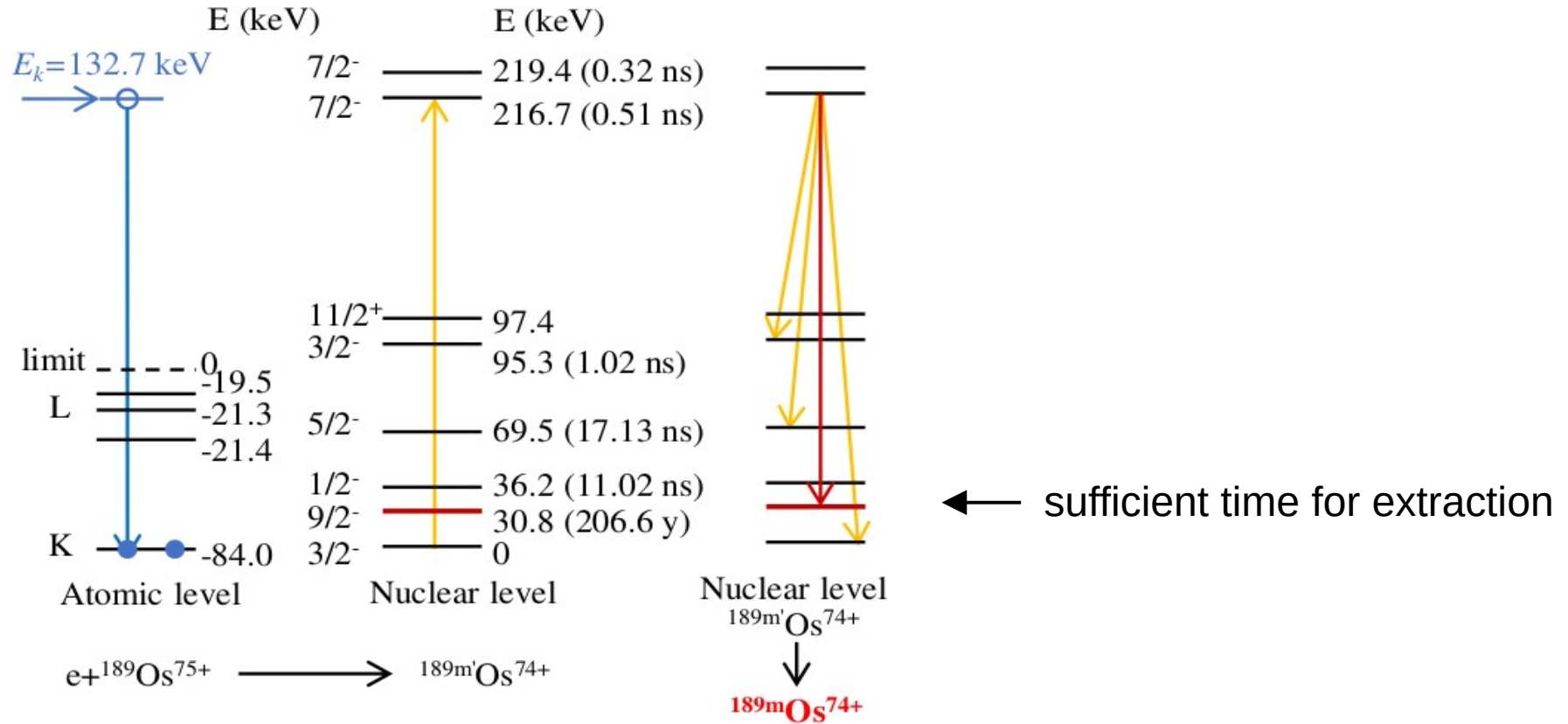


$$\zeta = p_z; \zeta b = 1$$

- The choice of  $b$  is crucial for the enhancement
- An average over arbitrary impact parameters up to infinity  
 $\Rightarrow$  enhancement factor  $p/p_z$

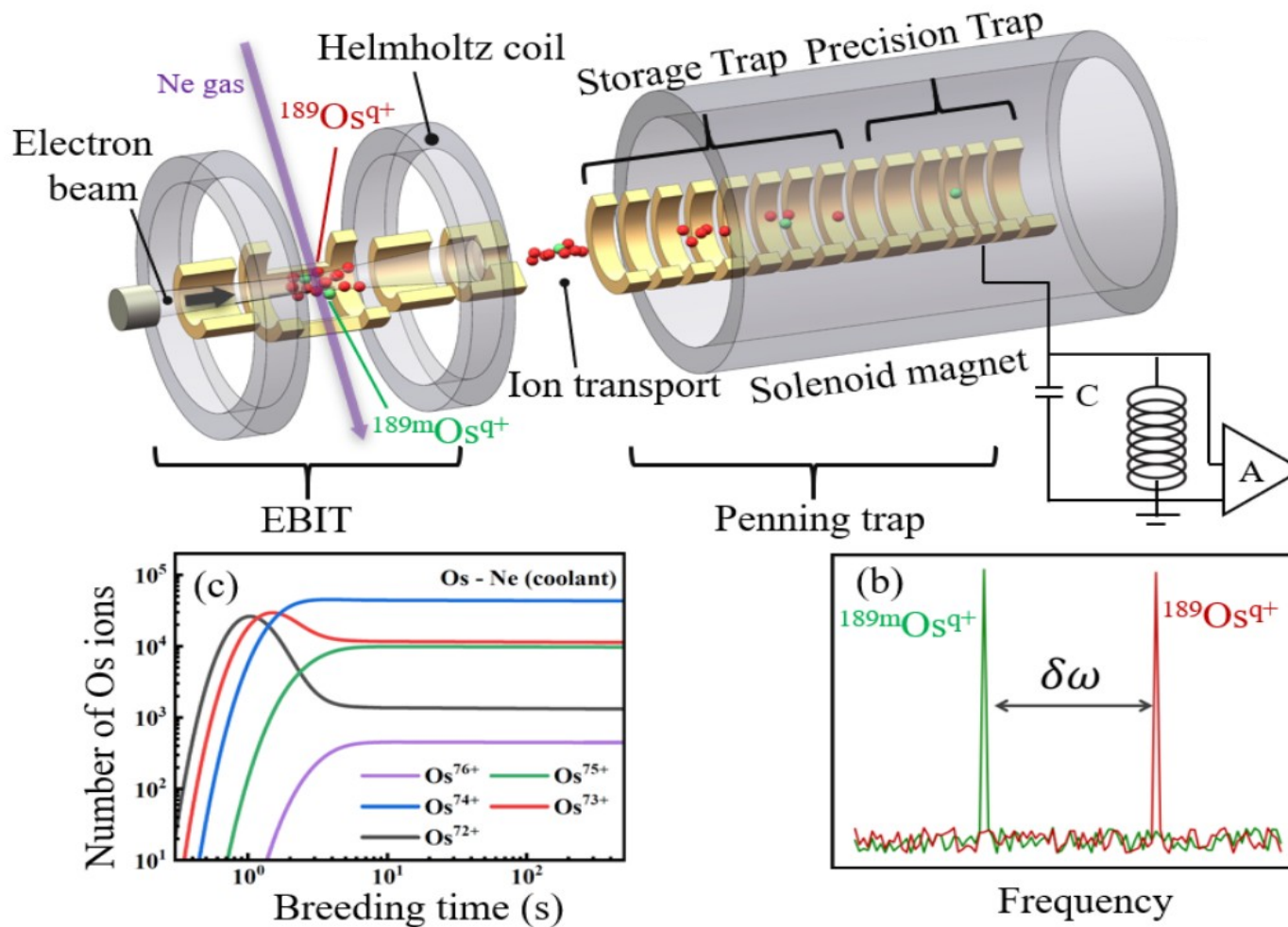
Y. Wu, S. Gargiulo, F. Carbone, C. H. Keitel, AP, PRL 128, 162501 (2022)

# NEEC in $^{189}\text{Os}$

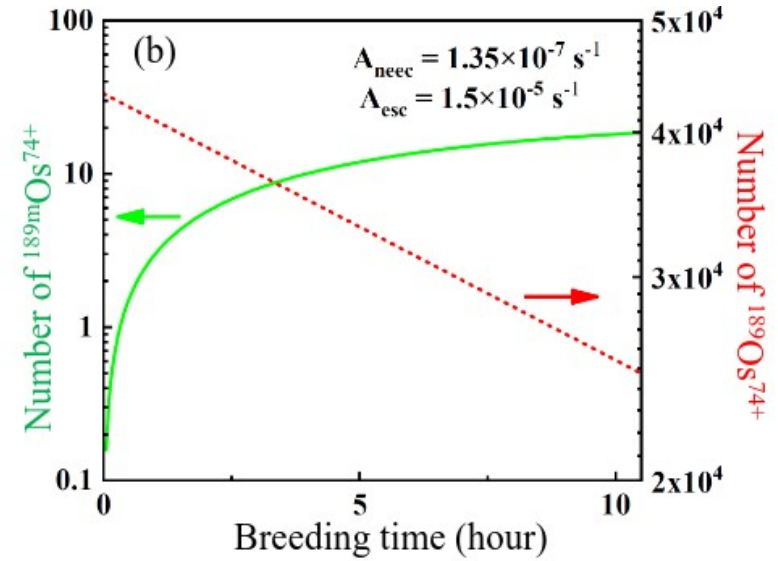
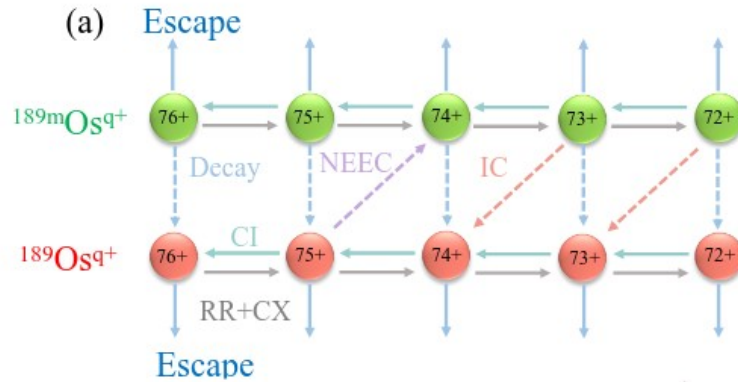


B. Tu, ..., AP, ..., X. Kong, Y. Ma, PRC 112, 054307 (2025)

# EBIT scenario



# Estimates



- 0.07% isomer probability, 500 ions stored, isomer detection in 2 days
- Scan for energy with 30 eV electron beam for approx. 60 days
- Unambiguous NEEC signature
- Challenging but not impossible

*B. Tu, ..., AP, ..., X. Kong, Y. Ma, PRC 112, 054307 (2025)*

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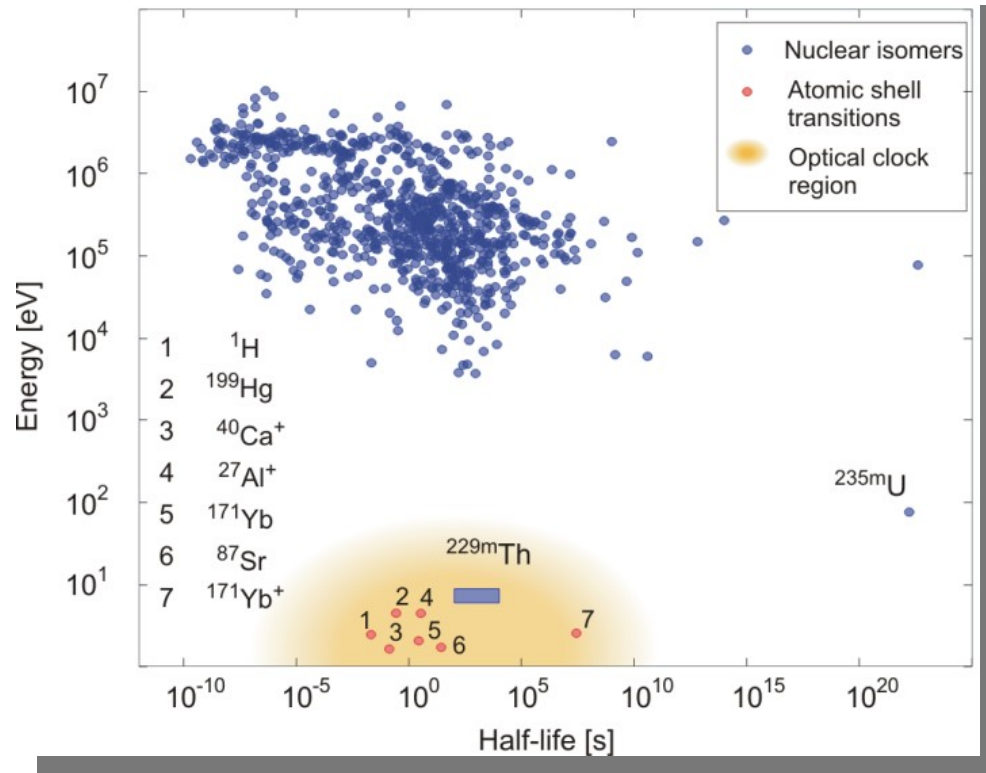
## II. Nuclear coupling to the atomic shell in $^{229}\text{Th}$

- ... in Th-doped crystals
- ... via nuclear hyperfine mixing

# What's special about $^{229}\text{Th}$ ( $Z=90$ )



Thor, the Scandinavian god of thunder inspired the name of Thorium (1828)



lowest excitation energy of all presently known excited nuclear states

$$E \simeq 8.4 \text{ eV}$$
$$\Delta E/E \simeq 10^{-20}$$

- insensitivity to external perturbations
- largely immune to systematic frequency shifts

*L. v.d. Wense et al., Nature 533, 47-53 (2016)*

*E. Peik et al., Quantum Sci. Technol. 6, 034002 (2021)*

## <sup>229</sup>Th nuclear clock

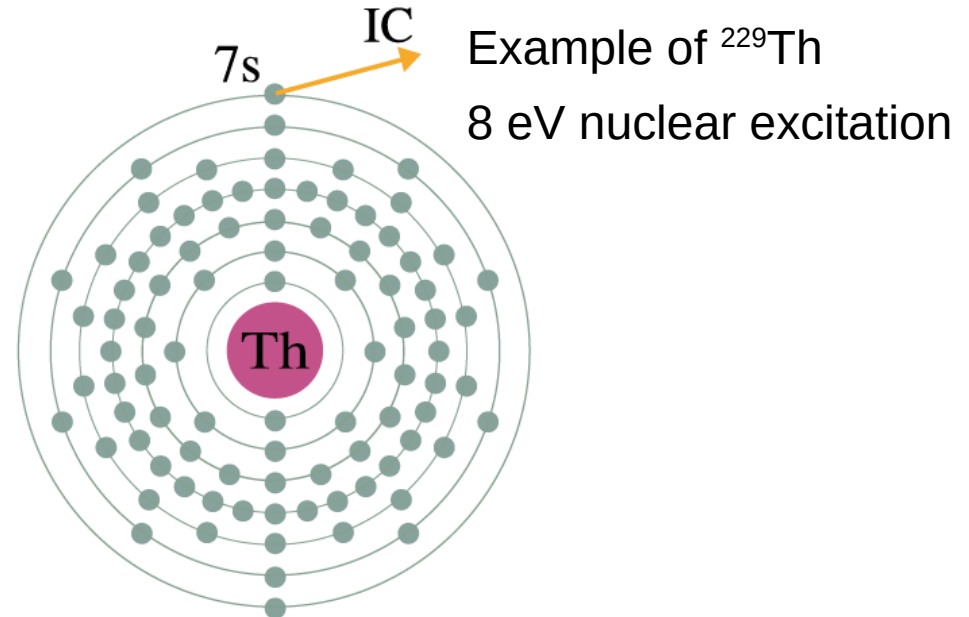
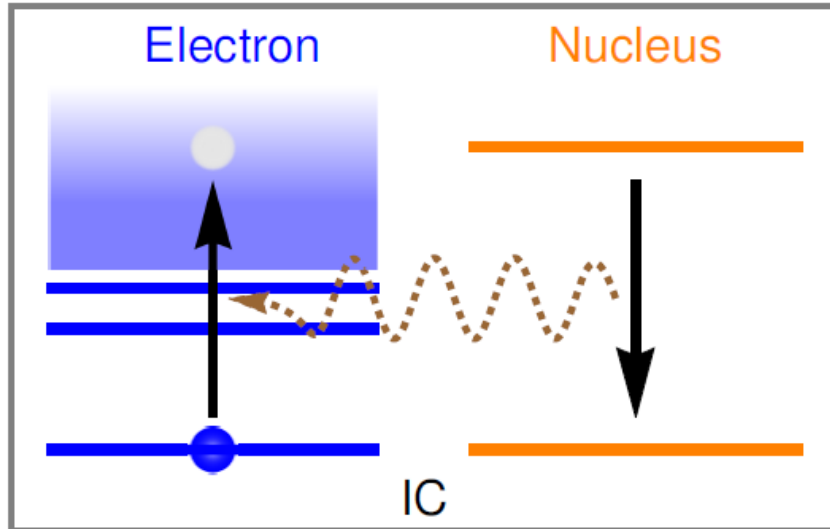


Art: Daria Bilous

- Promises better precision than present record
- Useful for independent secondary standard
- Improve GPS accuracy
- Search for dark matter
- Are fundamental constants constant in time?  $\alpha$ ,  $\Lambda_{QCD}$

# Nuclear processes involving electrons

For low-energy transition, **internal conversion** is the preferred decay channel!



9 orders of magnitude stronger than radiative decay

First direct evidence of isomer in 2016

Determination of transition energy via IC

*L. v. d. Wense et al., Nature 533, 47 (2016)*

*B. Seiferle et al., Nature 573, 243 (2019)*

# Nuclear structure for $^{229}\text{Th}$ ( $Z=90$ , $A=229$ )



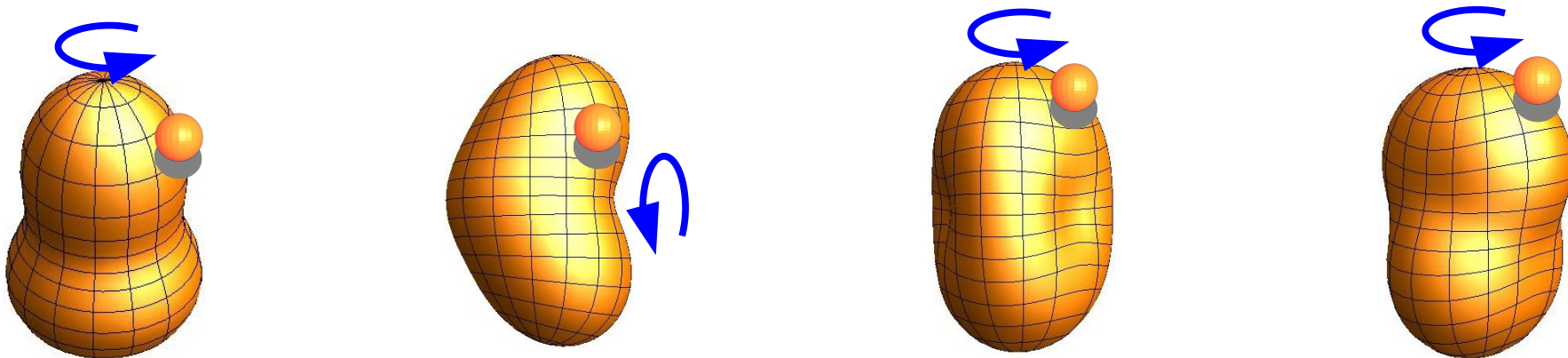
Nikolay Minkov

*Minkov and AP,  
Phys. Rev. Lett. 118, 212501 (2017)  
Phys. Rev. Lett. 122, 162502 (2019)  
Phys. Rev. C 103, 014313 (2021)*

## Macroscopic model

- 90 protons + 138 neutrons form an even-even core
- Quadrupole and octupole deformations
- 1 single-particle neutron on top
- Coriolis interaction between them

Low-lying states may appear also in other actinides, but 8 eV remains a “miracle”!



# Microscopic model

Skyrme Hartree-Fock plus BCS approach to  $^{229}\text{Th}$  and neighboring nuclei

- Model describes single-particle degrees of freedom and pairing
- It correctly reproduces the low-lying spin order and predicts small isomer energies for  $^{229}\text{Th}$  and neighboring nuclei  $^{227}\text{Ra}$  and  $^{231}\text{U}$
- Phenomenon is not typical for  $^{229}\text{Th}$ , but eV energy might remain unique
- Provides values for gyromagnetic factors – new calculation for magnetic moment

Quenching

$$q_s = 0.765$$

$$q_R = 0.9$$

Old values

$$q_s = 0.6$$

$$q_R = 0.6$$

Magnetic moment

$$\mu_G = 0.74\mu_N$$

$$\mu_I = -0.31\mu_N$$

*N. Minkov and AP  
PRL 122, 162502 (2019)*

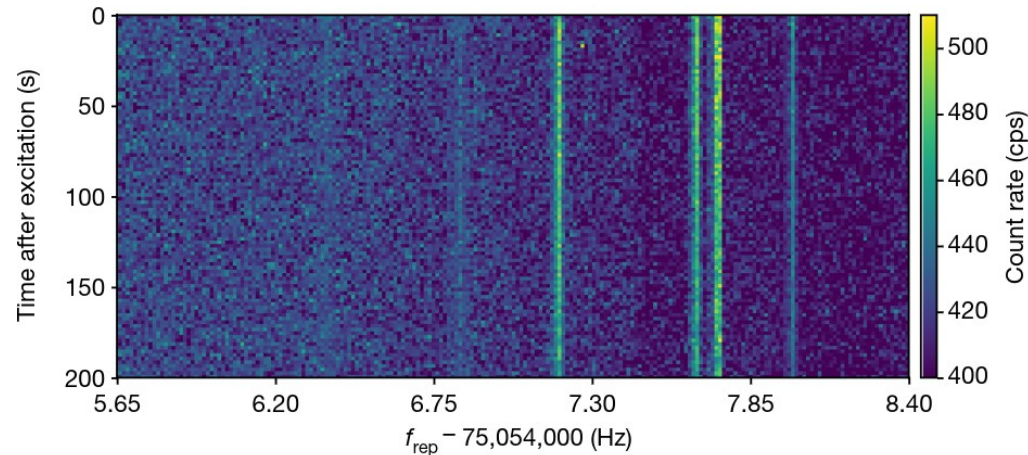
*N. Minkov, AP, L. Bonneau, P. Quentin  
Phys. Rev. C 110, 034327 (2024)*

# Laser excitation in Th-doped crystals

April 2024: Broadband excitation in Th:CaF<sub>2</sub>  
*J. Tiedau et al., Phys. Rev. Lett. 132, 182501 (2024)*

July 2024: Broadband excitation in Th:LiSrAlF<sub>6</sub>  
*R. Elwell et al., Phys. Rev. Lett. 133, 013201 (2024)*

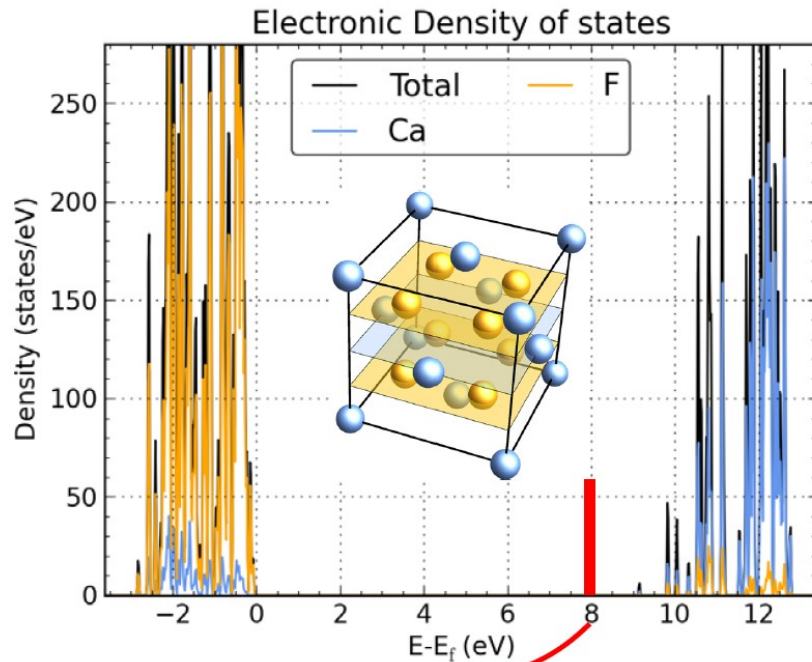
September 2024: Narrowband excitation in Th:CaF<sub>2</sub>  
*C. Zhang et al., Nature 633, 63 (2024)*



quadrupole hyperfine splitting in crystal field

# Electronic defect states

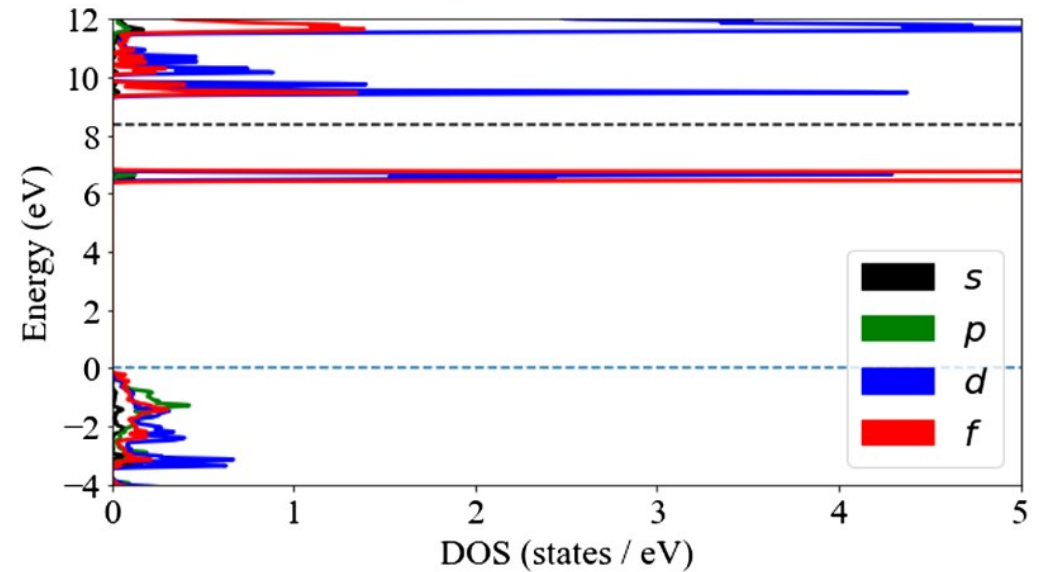
Th: CaF<sub>2</sub>



Electronic states  
localized on dopant Th ions

Th:LiSrAlF<sub>6</sub>

6

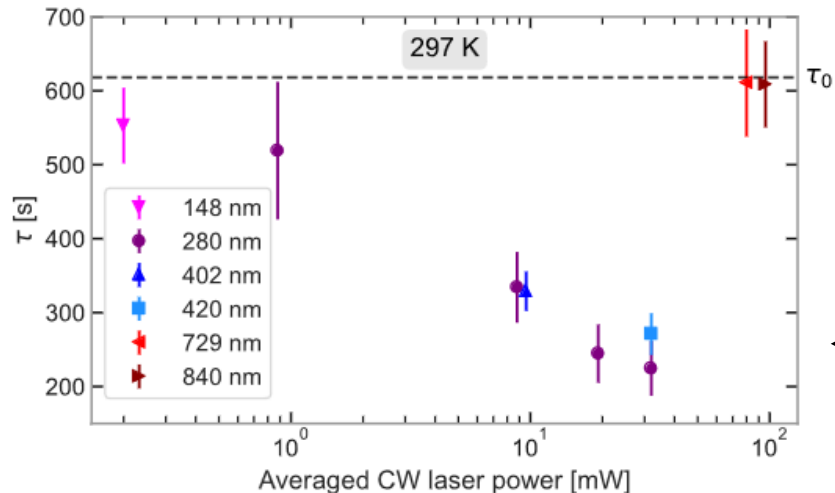


*B. S. Nickerson, ... , AP, PRL 125, 032501 (2020)*

*R. Elwell et al., Phys. Rev. Lett. 133, 013201 (2024)*

# Coupling to the atomic shell

- Only part of the nuclear decay occurs radiatively
- Quenching via coupling to atomic shell and defect states in the crystal band gap
- LiSAF: Broad redshifted fluorescence observed
- CaF<sub>2</sub> and LiSAF: Laser-assisted quenching observed
- Electronic bridge or (bound) internal conversion via defect states - theory, experiment ThO<sub>2</sub>



*H. W. T. Morgan et al., PRL 134, 253801 (2025)*

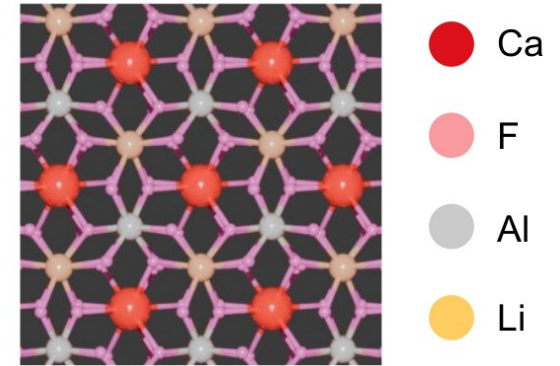
*R. Elwell et al., Nature 648, 300 (2025)*

← *F. Schaden et al., PRR 7, L022036 (2025)*

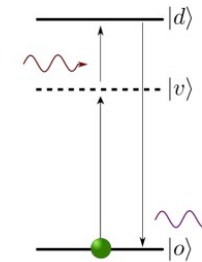
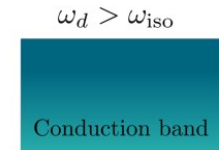
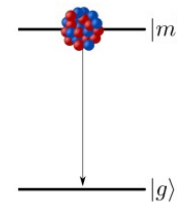
# Electronic bridge in LiSAF and LiCAF

- LiSAF has lower defect formation energies than LiCAF
- Charge compensation schemes can be controlled via fluorination
- VASP calculations

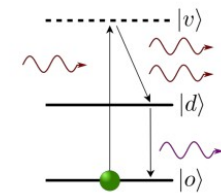
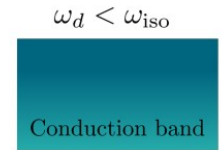
*M. Pimon, T. Kirschbaum, ..., AP, ..., PRB 113, 014107 (2026)*



- EB calculated for the relevant charge compensation schemes
- 2-3 orders of magnitude more efficient than photoexcitation



Absorption



Stimulation

*T. Kirschbaum, M. Pimon, ..., AP, PRA 113, 023108 (2026)*

# Nuclear hyperfine mixing

$$H=H_0+H_{int}$$

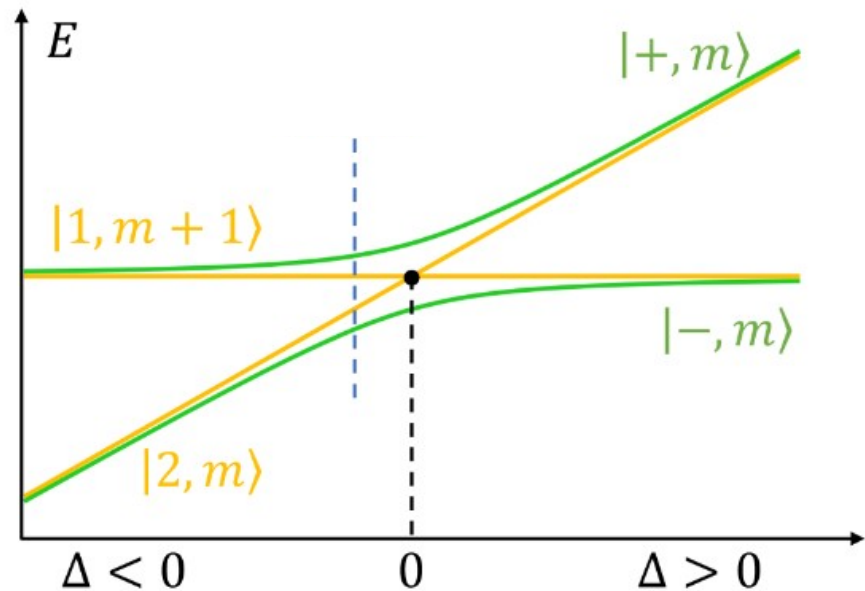
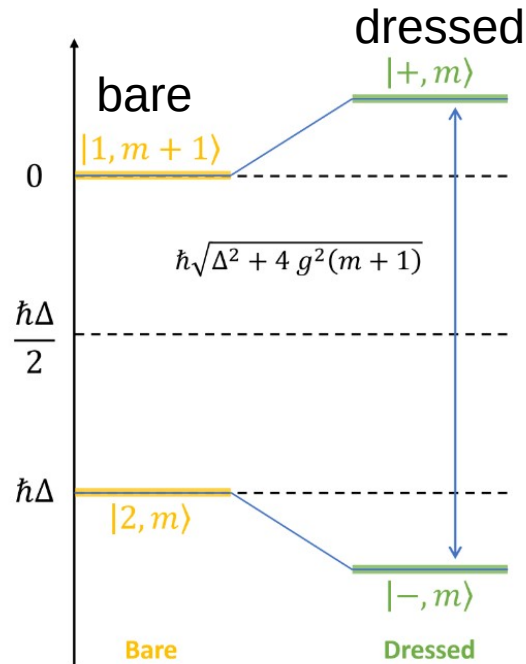
- The hyperfine interaction is a perturbation – regular hyperfine splitting
- The hyperfine interaction part strong, work in its eigenbasis – “Nuclear Hyperfine Mixing”
- NHM predicted for highly charged ions with unpaired electrons, but not observed so far

# Nuclear hyperfine mixing

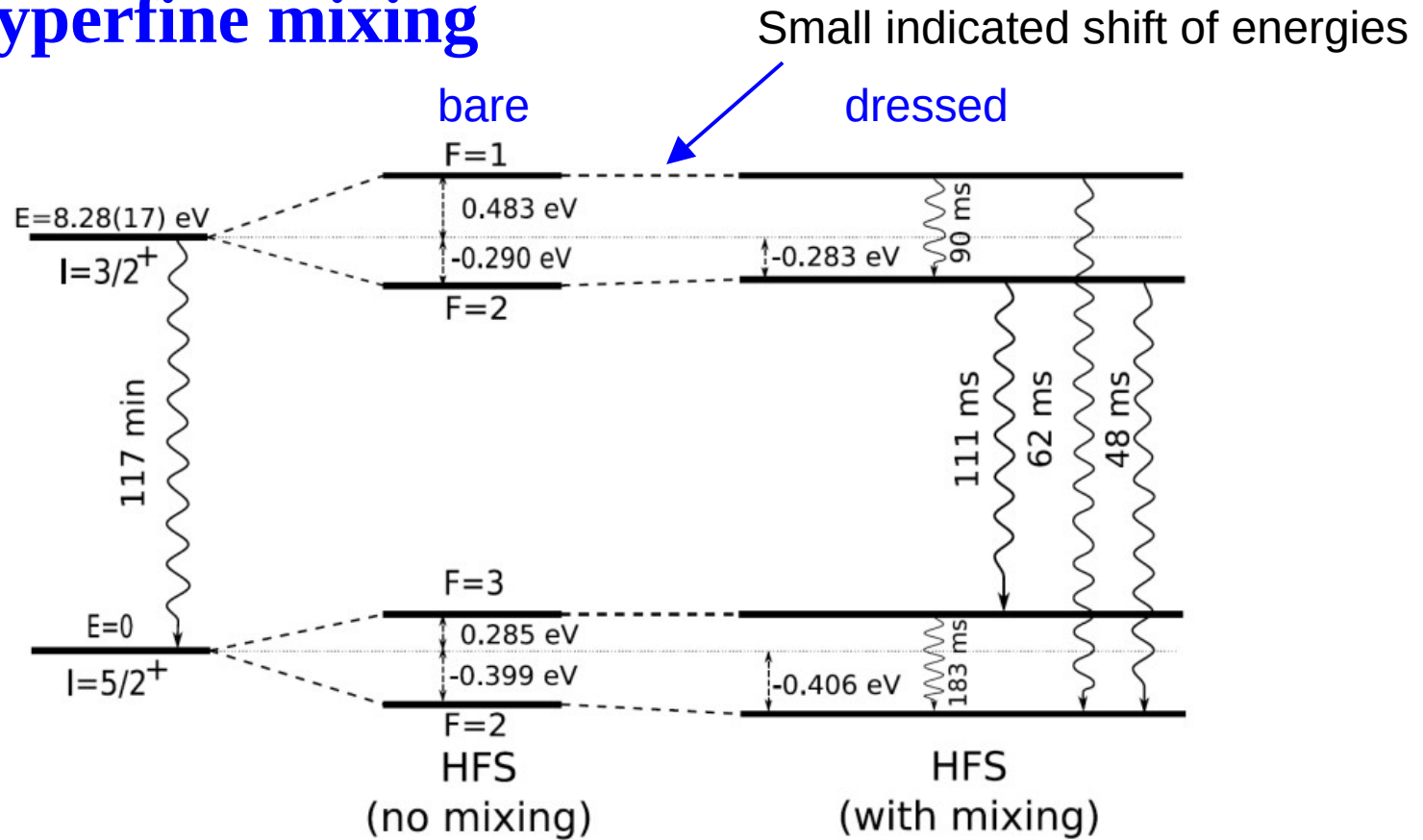
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Dressed states in quantum optics



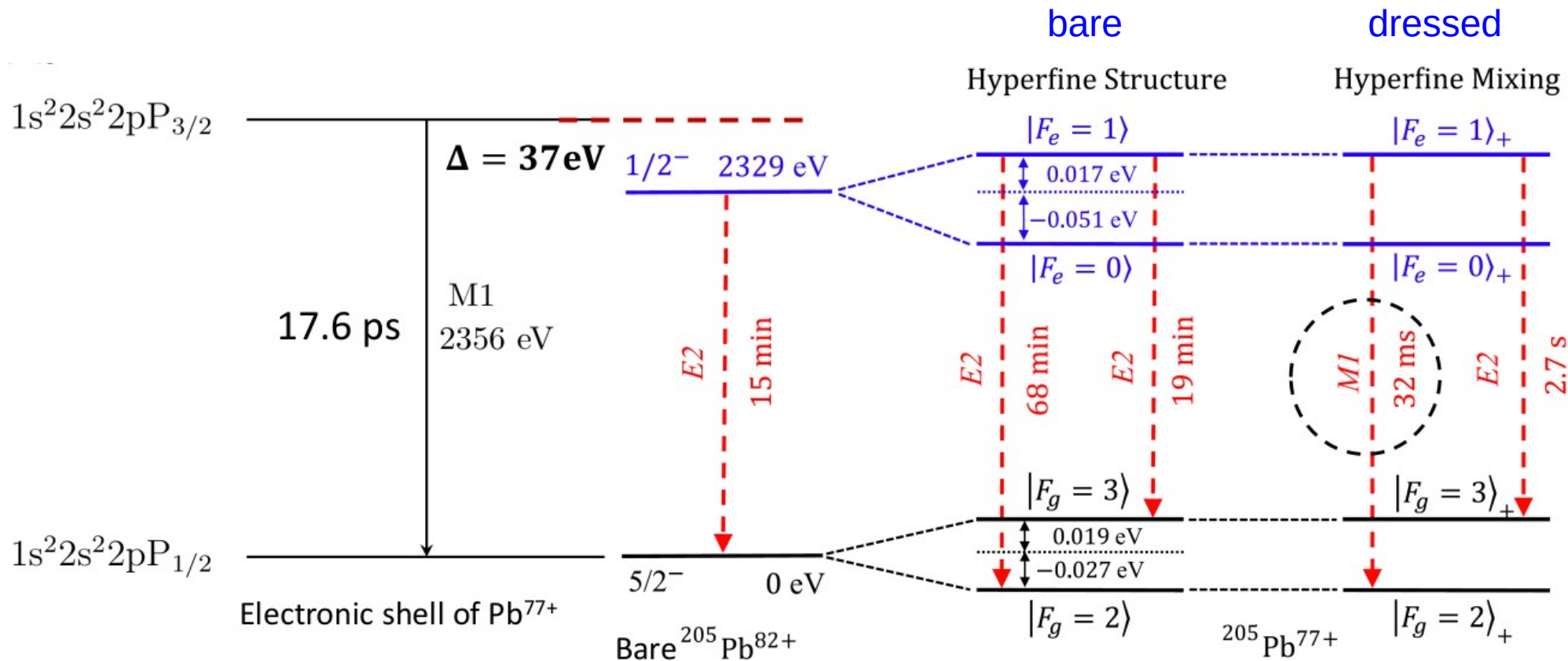
# Nuclear hyperfine mixing



In  $^{229}\text{Th}^{89+}$  the very strong 28 MT magnetic field of the unpaired electron mixes  $F = 2$  states

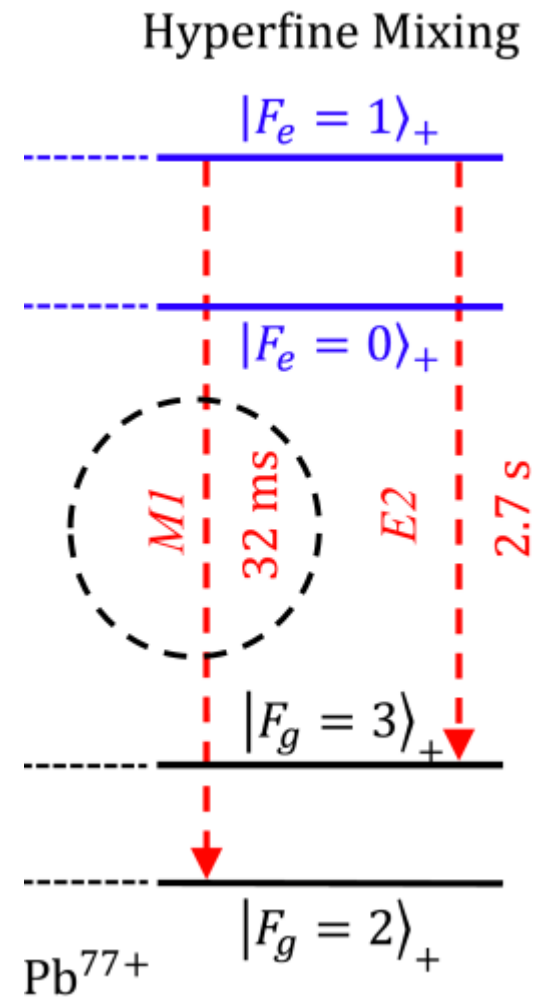
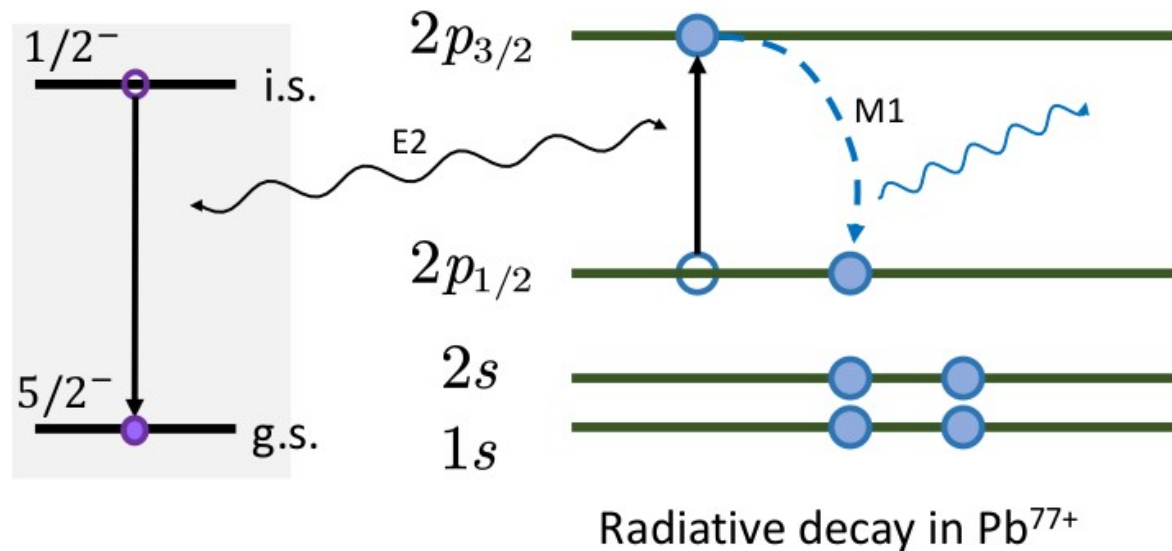
# Ongoing work at Jussieu

Common framework for NHM and BIC/IC/NEET/NEEC



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Common framework for NHM and BIC/IC/NEET/NEEC



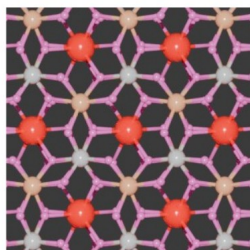
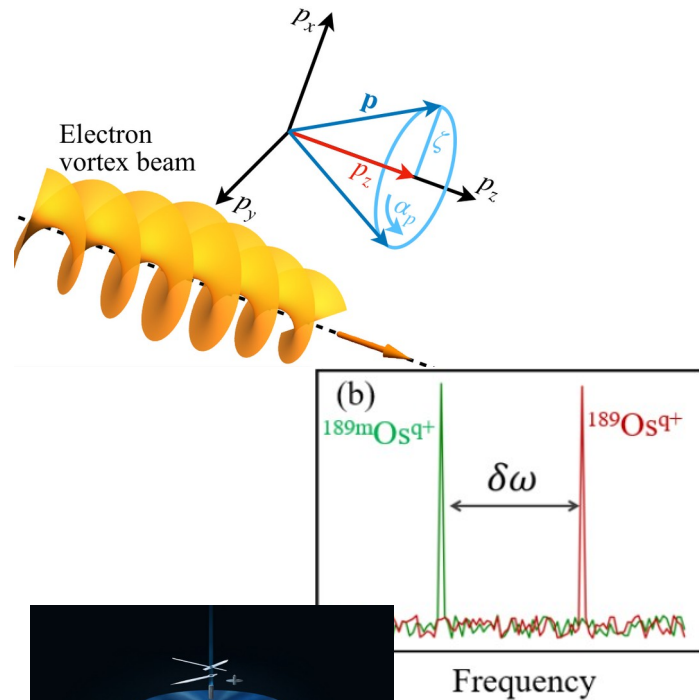
# Ongoing work at Jussieu

Common framework for NHM and BIC/IC/NEET/NEEC

	$H_{en}$	$I_g J_g$	$I_g J_e$	$I_e J_g$	$I_e J_e$
$I_g J_g$			Hyperfine quenching	NHM	×
$I_g J_e$		Hyperfine quenching		BIC/NHM	?
$I_e J_g$		NHM	BIC/NHM		Hyperfine quenching
$I_e J_e$		×	?	Hyperfine quenching	

With P. Indelicato, K. Shi. X. Ma (Lanzhou, China)

# Conclusions



- Ca
- F
- Al
- Li

- Zoo of nuclear processes that involve atomic electrons
- **NEEC experimental evidence** remains controversial
- For small impact parameters, **electron vortex beams** enhance NEEC integrated cross sections
- Precision mass measurements could provide unambiguous NEEC signature
- $^{229}\text{Th}$  provides a unique chance to develop a **nuclear clock** with increased accuracy and potentially investigate new physics
- Atomic shell brings quenching and therefore faster clock interrogation and improved performance in Th-doped crystals
- What can we expect from nuclear hyperfine mixing?

# Thanks



Paul Indelicato  
Nancy Paul



Christoph H. Keitel



Yuanbin Wu



Xiangjin Kong  
Bingsheng Tu  
Yugang Ma



Thorsten Schumm  
Martin Pimon  
Andreas Grüneis



Fabrizio Carbone  
Simone Gargiulo



***THANK YOU FOR YOUR ATTENTION!***