Electronic Bridge schemes in ^{229}Th doped LiCAF

Tobias Kirschbaum[1], Martin Pimon[2], and Adriana Pálffy[1]

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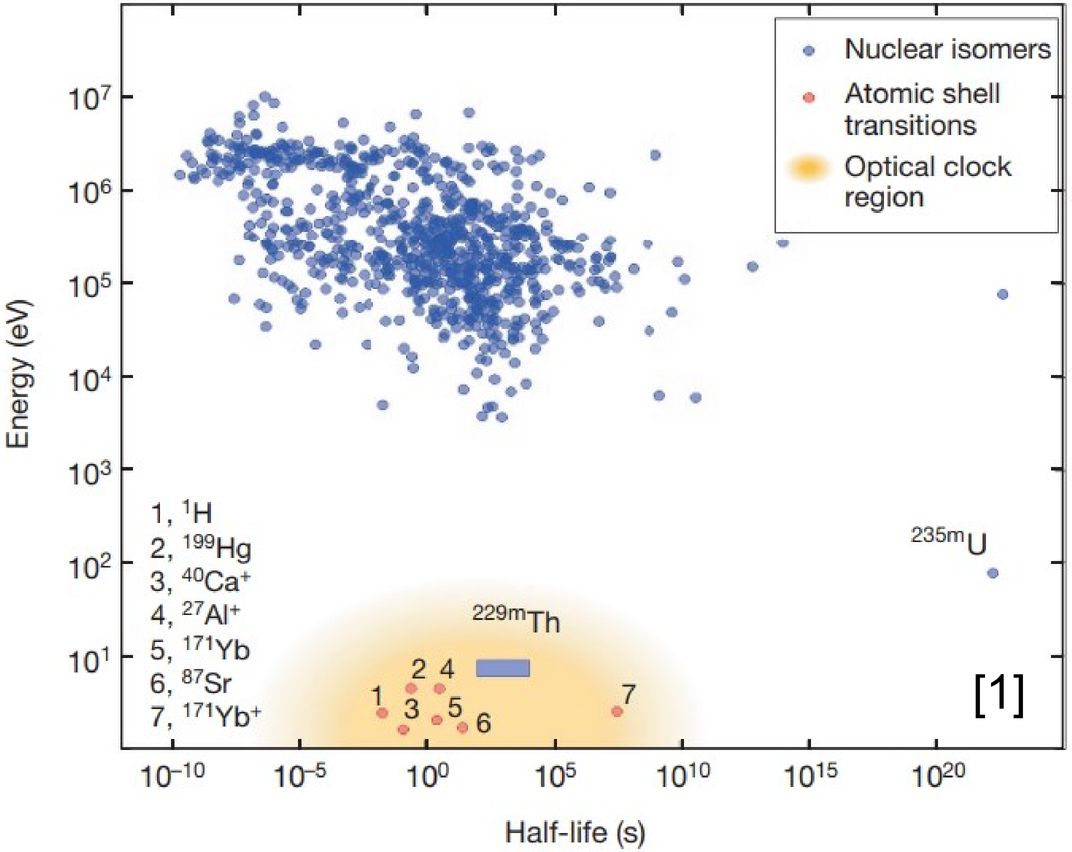
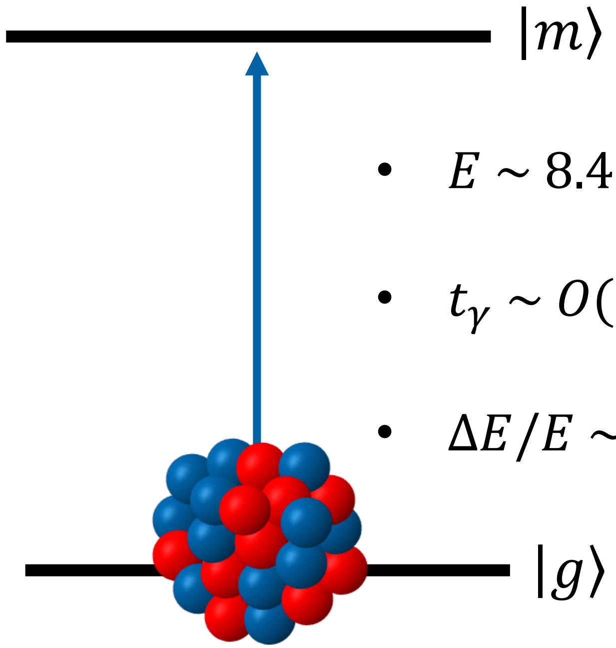
What is special about ^{229}Th (Z=90)?

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



^{229}Th: Lowest excitation energy amongst all nuclei!



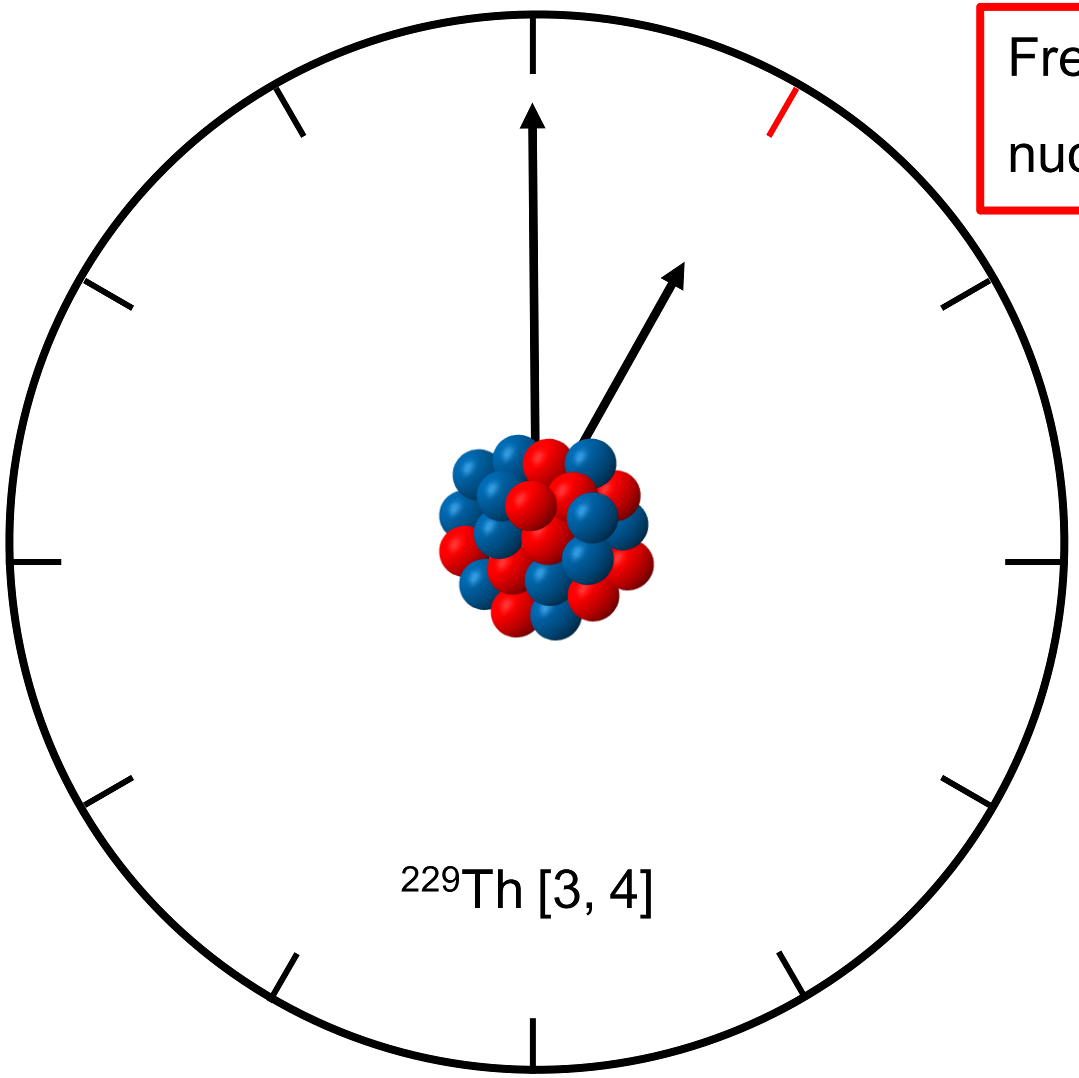
- $E \sim 8.4$ eV [2]
- $t_\gamma \sim O(10^3)$ s
- $\Delta E/E \sim 10^{-20}$

→ Ideal candidate for a nuclear clock with outstanding properties

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

[2] J. Tiedau *et al.*, Phys. Rev. Lett. **132**, 182501 (2024)

^{229}Th nuclear clock

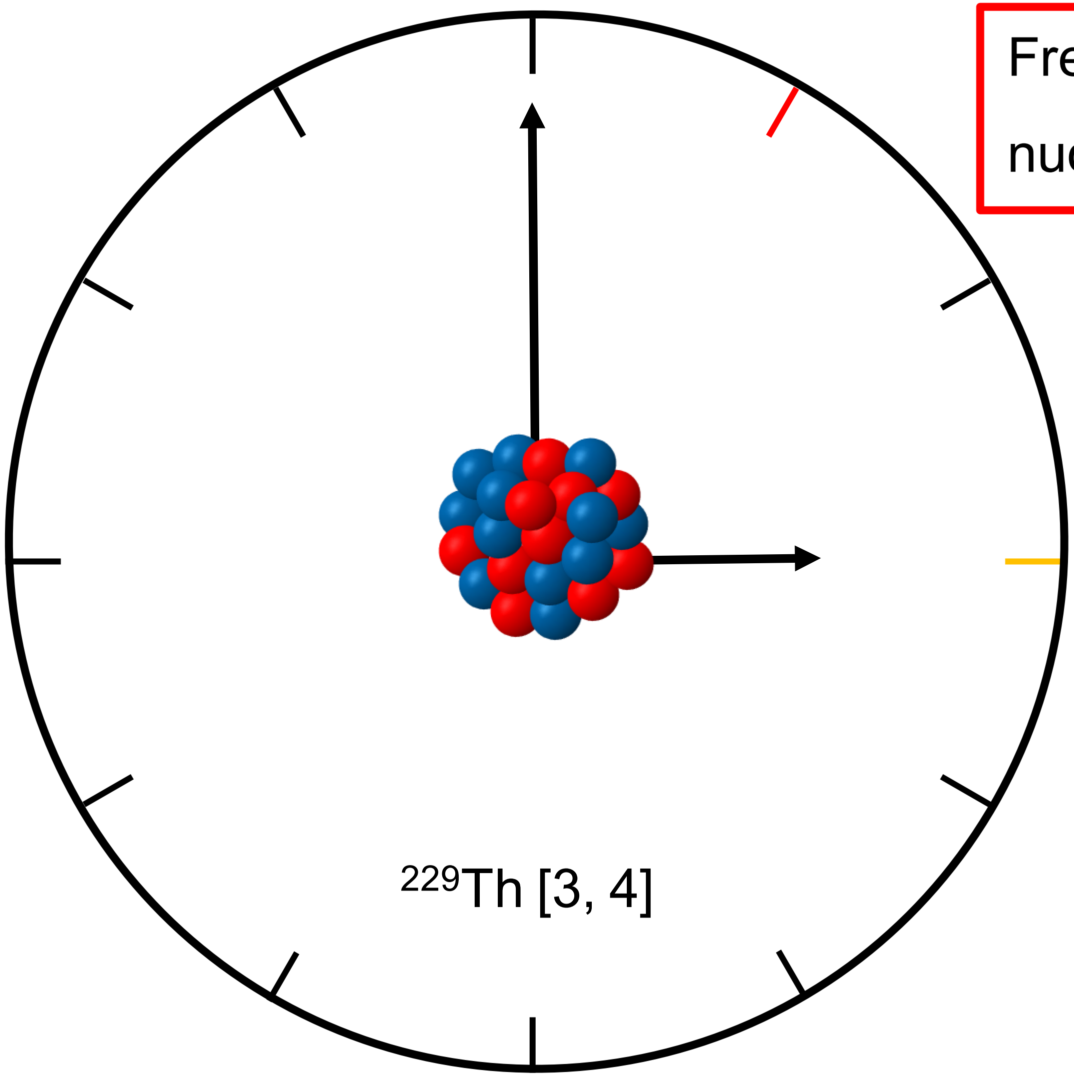


Frequency standard based on a nuclear transition

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

[4] P. Thirolf *et al.*, Eur. Phys. J. Spec. Top., 1-19 (2024)

^{229}Th nuclear clock



Frequency standard based on a nuclear transition

High resilience against external perturbations
→ Better precision

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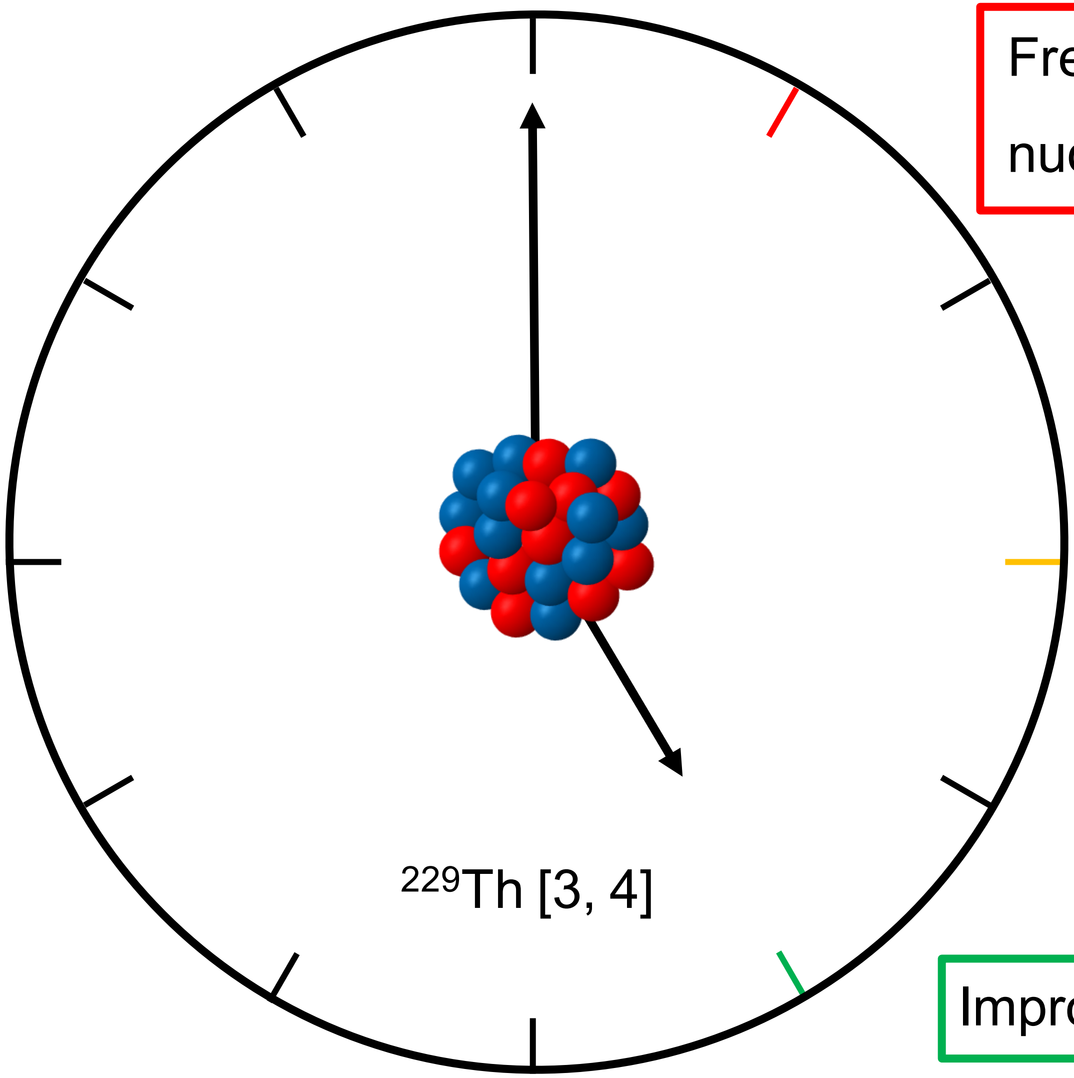

Frequency standard based on a nuclear transition

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Improving satellite navigation

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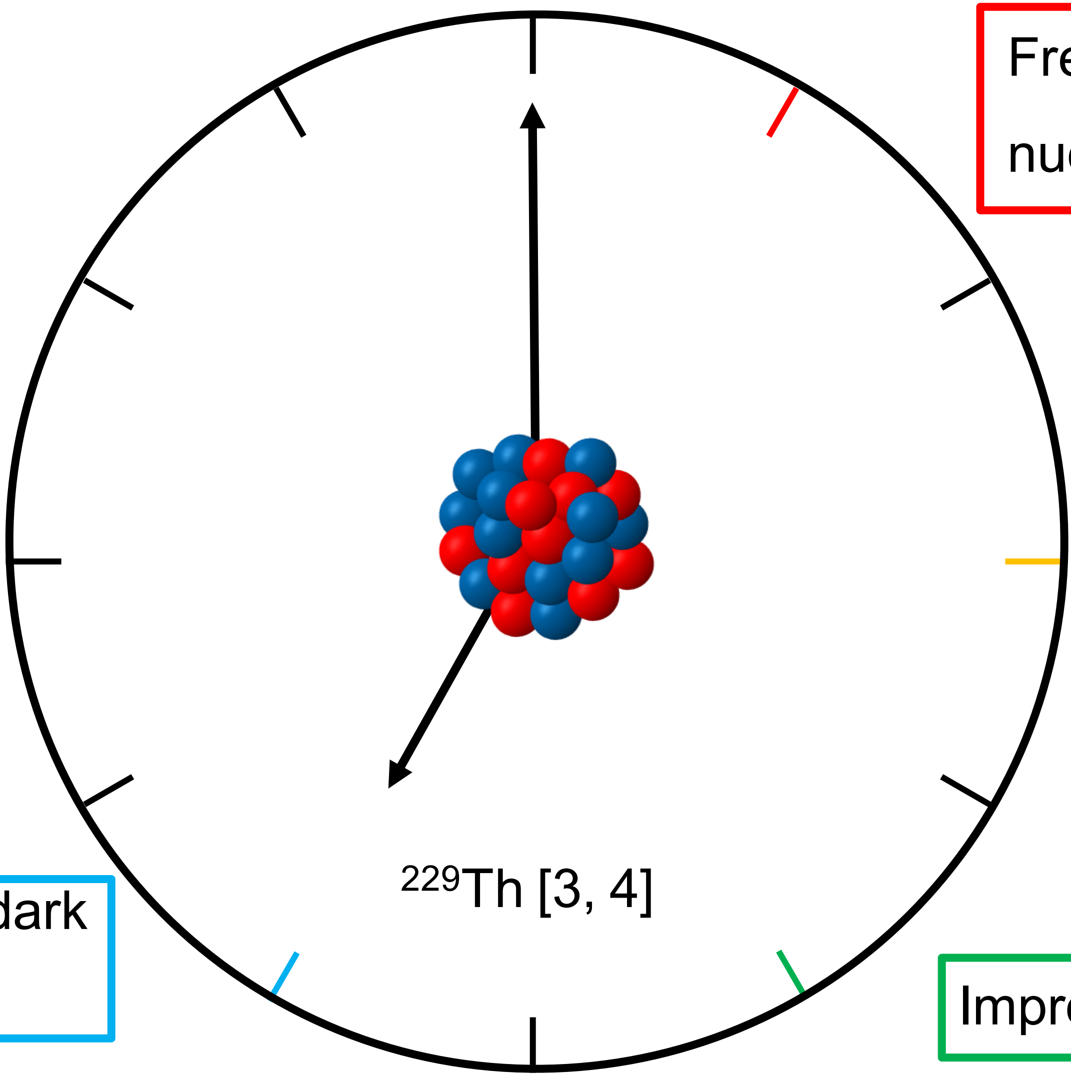

Frequency standard based on a nuclear transition

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Benchmark for searching dark matter

[3] E. Peik *et al.*, Quantum Sci. Technol. **6,** 034002 (2021)
[4] P. Thirolf *et al.*, Eur. Phys. J. Spec. Top., 1-19 (2024)

^{229}Th nuclear clock

Frequency standard based on a nuclear transition

High resilience against external perturbations
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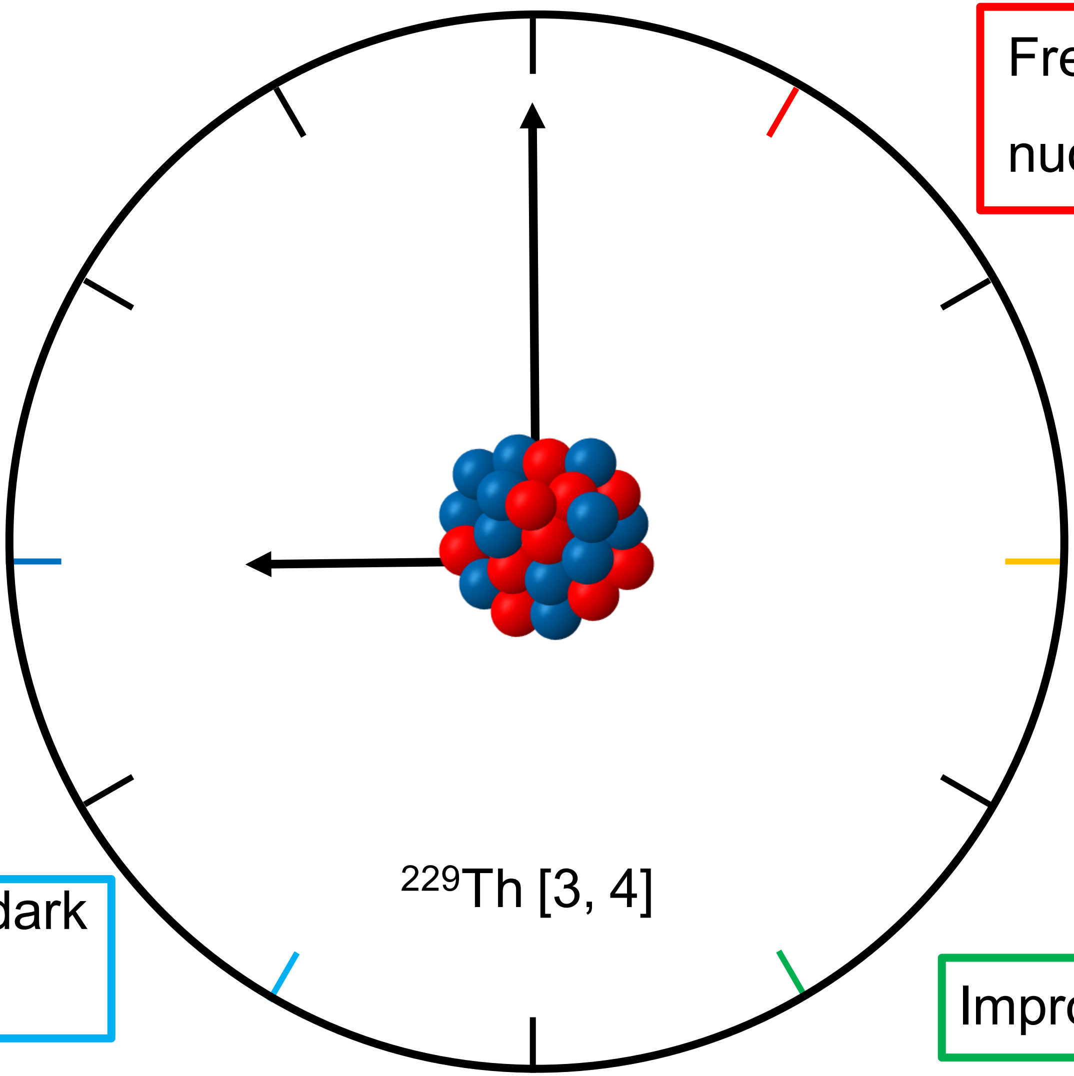
Are fundamental constants (e.g. $\alpha, \Lambda_{\mathrm{QCD}}$) really constant?



Benchmark for searching dark matter

Improving satellite navigation

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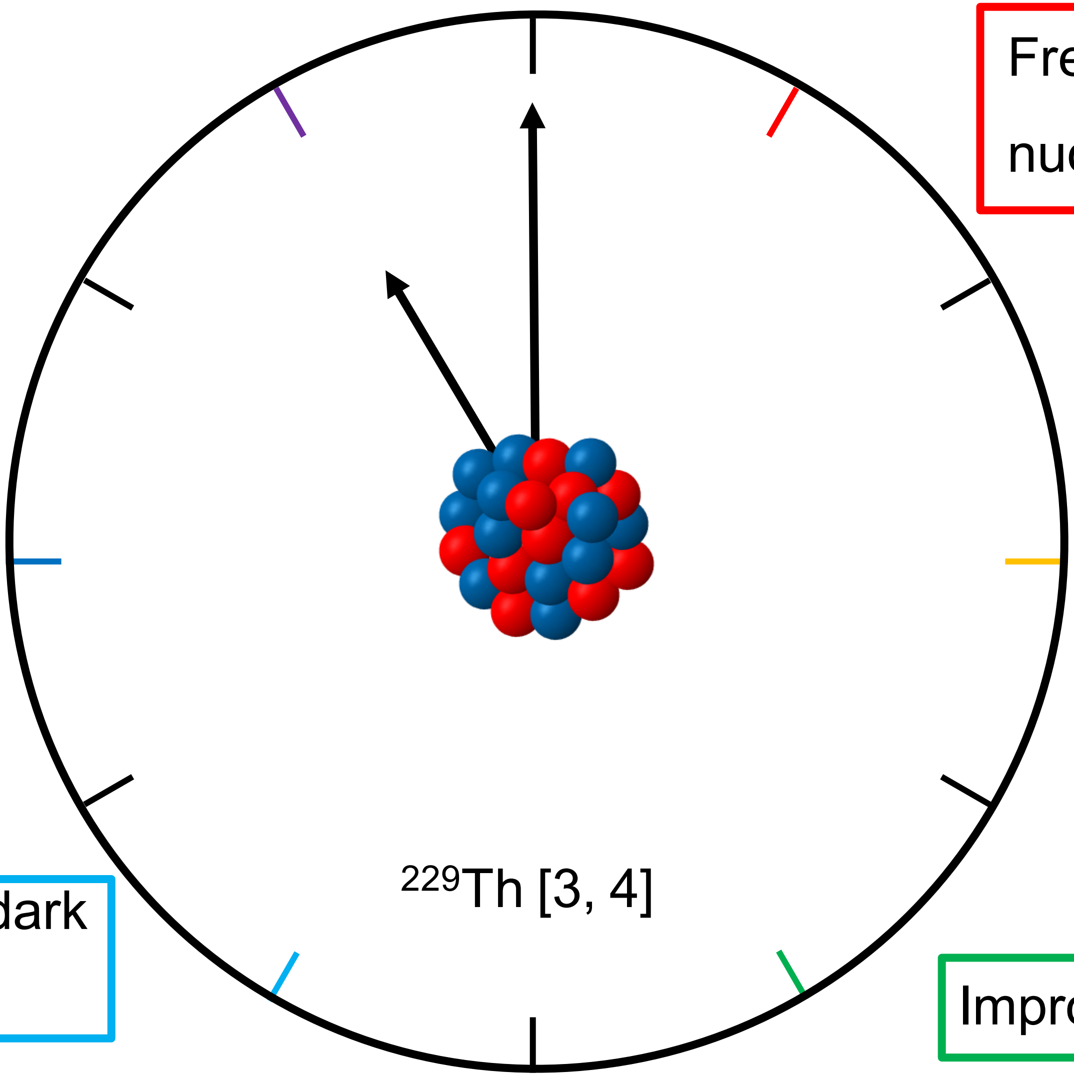
^{229}Th nuclear clock

Challenges: Uncertainty on transition energy, **lack of narrowband VUV lasers**

Are fundamental constants (e.g. $\alpha, \Lambda_{\mathrm{QCD}}$) really constant?

Benchmark for searching dark matter



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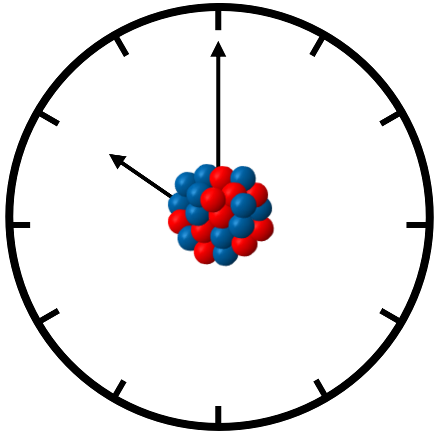
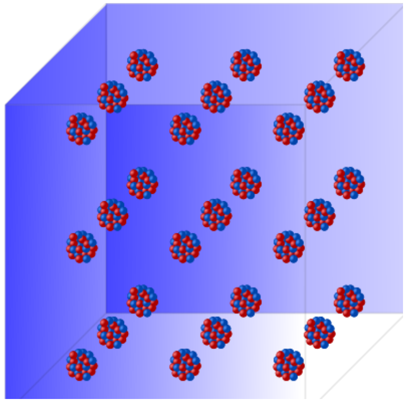
Outline

1) Nuclear clock approaches

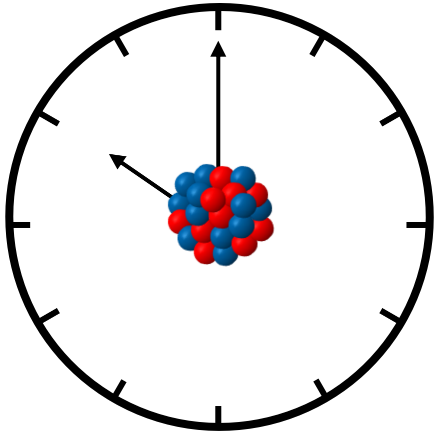


2) ^{229}Th in large band gap crystals



3) Electronic Bridge schemes in ^{229}Th:LiCAF

Outline



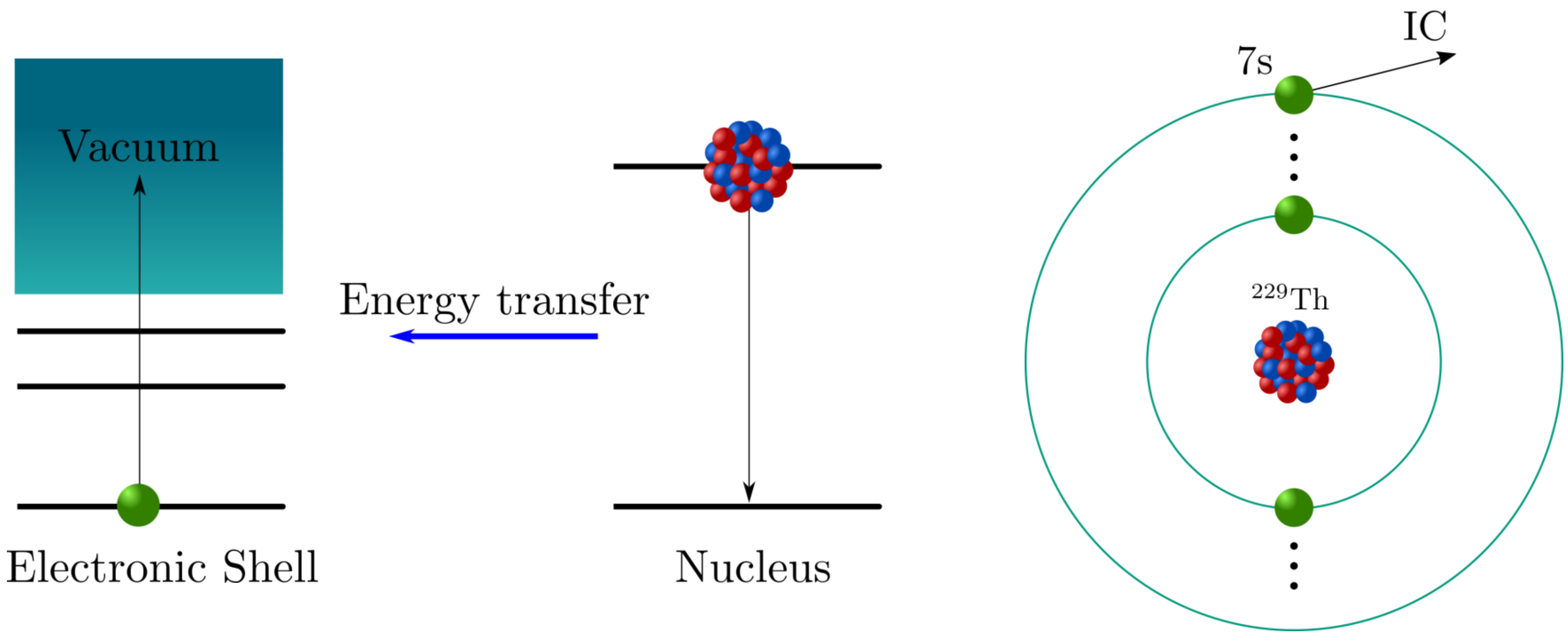
1) Nuclear clock approaches

2) ^{229}Th in large band gap crystals

3) Electronic Bridge schemes in ^{229}Th:LiCAF

Coupling to atomic shells

In neutral ^{229}Th **internal conversion (IC)** is the preferred decay channel: $\frac{\Gamma_{IC}}{\Gamma_{\gamma}} \sim O(10^8)$



However: IC decay of ^{229}Th has led to the first direct evidence of the isomer in 2016 [1]

Recently: Observation of radiative decay [5], direct laser excitation followed soon after [2]

[1] L. v. d. Wense *et al.*, Nature **533,** 47-53 (2016)
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[5] S. Kraemer *et al.*, Nature **617**, 706-710 (2023)

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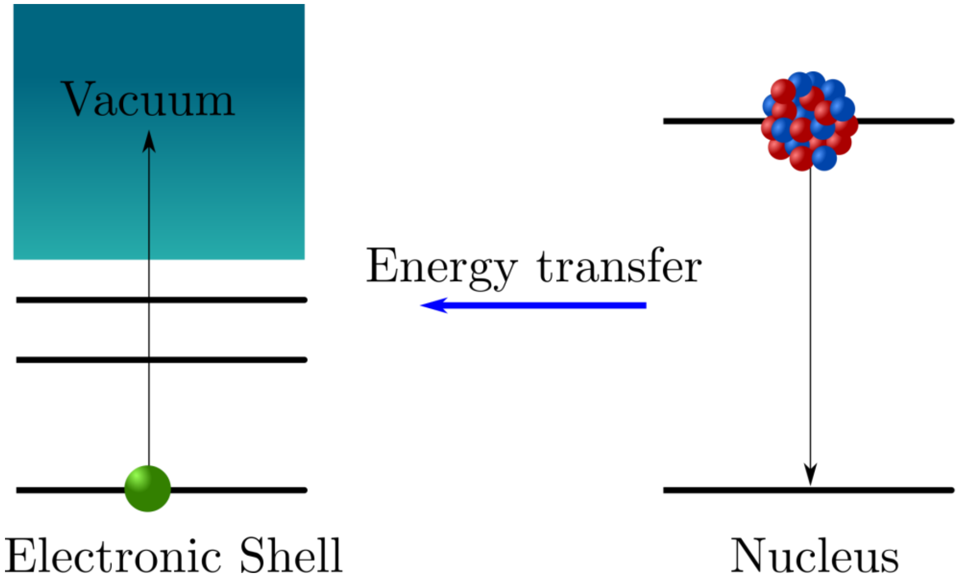

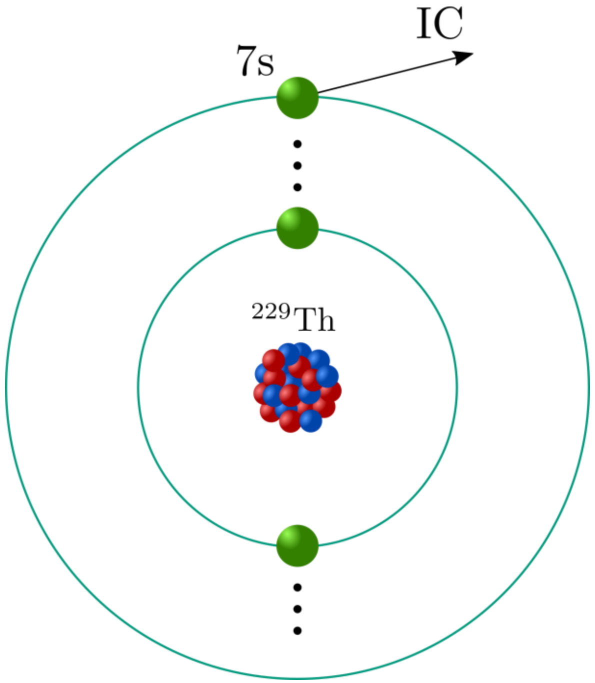

IC energetically forbidden for $q \geq 1$

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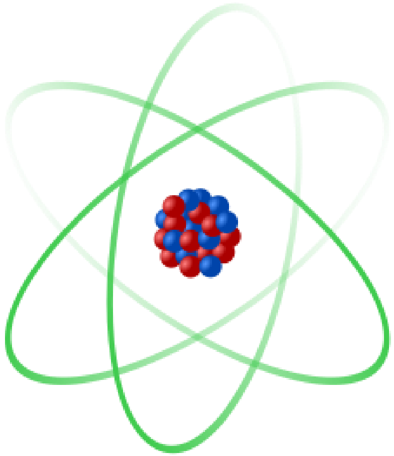
Main approaches for a nuclear clock

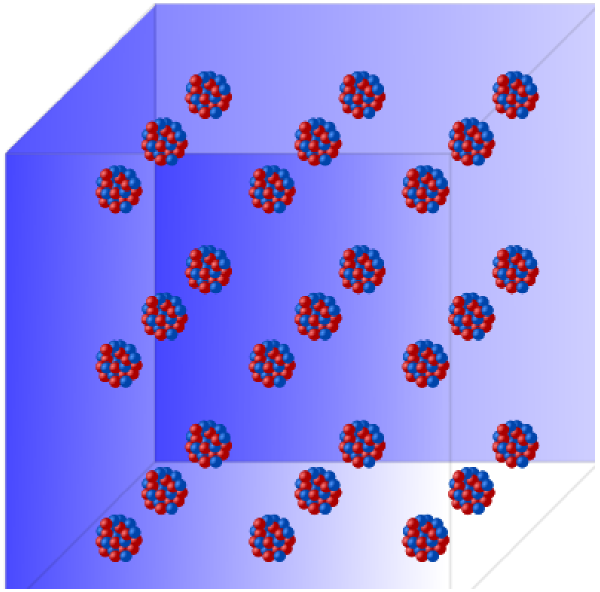
1.) Single ion nuclear clock [6]

- Trapped ion
- Similar to atomic clocks



2.) Solid state nuclear clock [7]

- Novel approach
- ^{229}Th nuclei doped in transparent crystal



[6] E. Peik and C. Tamm, EPL **61,** 181 (2003)

[7] G. A. Kazakov *et al.*, New J. Phys. **14**, 083019 (2012)

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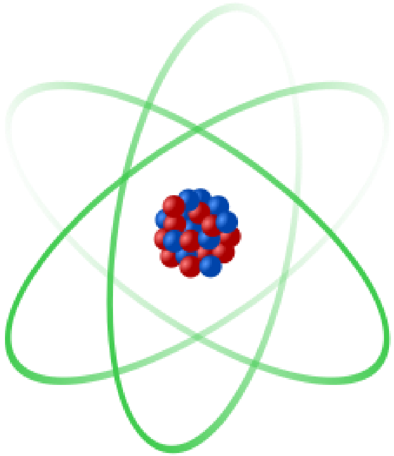

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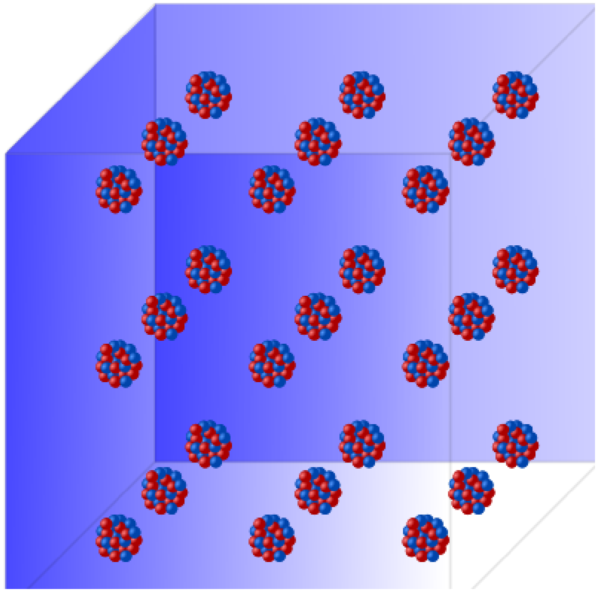


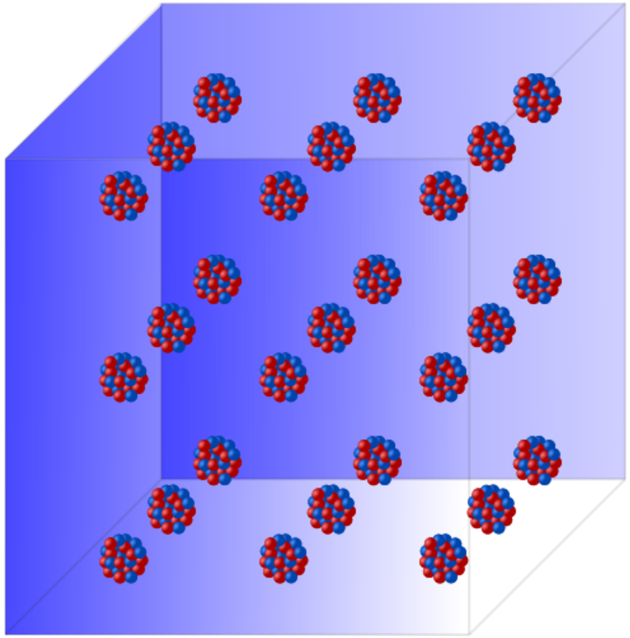
[6] E. Peik and C. Tamm, EPL **61,** 181 (2003)

[7] G. A. Kazakov *et al.*, New J. Phys. **14**, 083019 (2012)

Pros and cons of the solid state approach

Large band gap crystals ($\Delta_g > 8.4$ eV) are an ideal inert host for ^{229}Th [7]:

- Transparent + $^{229}Th^{q+}$ ($q \geq 1$)
- Logistical benefits with high doping densities up to $\approx 10^{18}$ cm^{-3}
- More nuclei can be interrogated at the same time → Better clock stability



However:

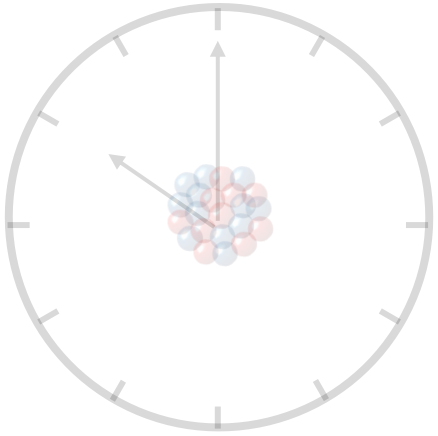
- Background radioactivity
- Systematic crystal effects
- Rabi and Ramsey interrogation schemes are **not** applicable due to crystal lattice effects

  → Clock operation via counting of nuclear fluorescence photons

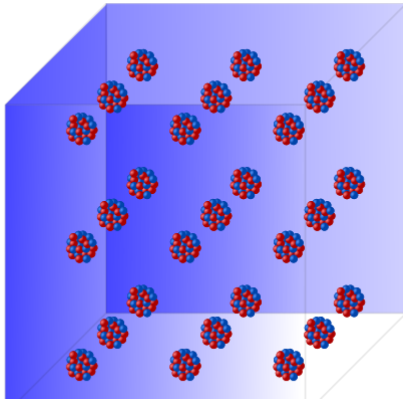
[7] G. A. Kazakov *et al.*, New J. Phys. **14**, 083019 (2012)

Outline

1) Nuclear clock approaches

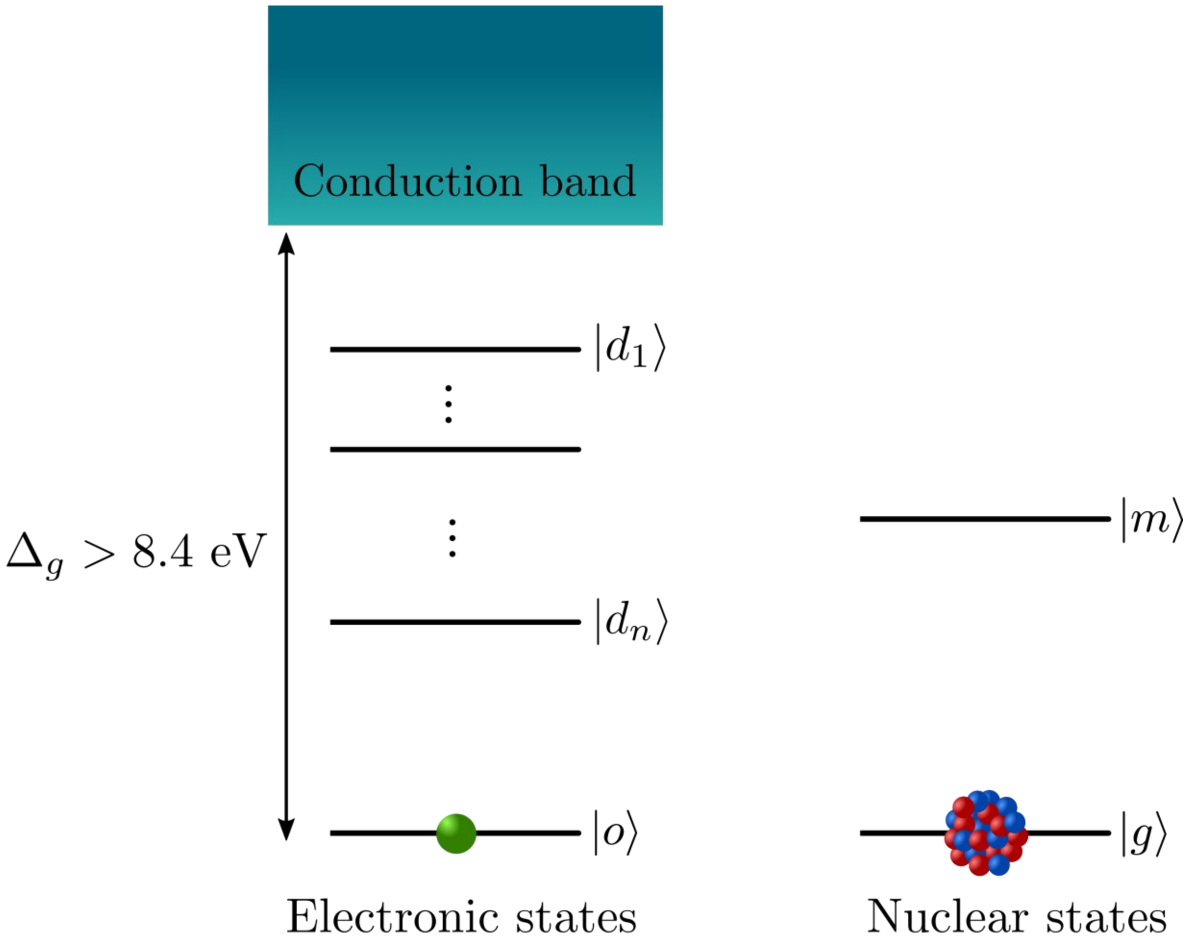


2) ^{229}Th in large band gap crystals



3) Electronic Bridge schemes in ^{229}Th:LiCAF

Doping ^{229}Th nuclei in large band gap crystals



- Possible host materials: LiCAF ($LiCaAlF_6$) or CaF_2
- Ab initio Density Functional Theory (DFT) simulations of such systems predict the formation of **defect states** localized around the ^{229}Th nucleus close to 8 eV [8, 9]

→ Above or below?

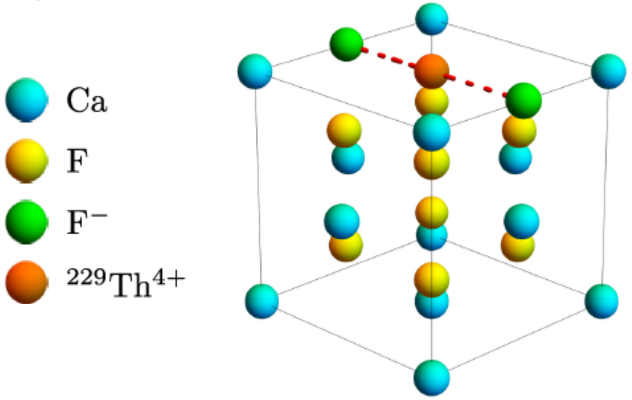
- $\Gamma_d \gg \Gamma_\gamma$

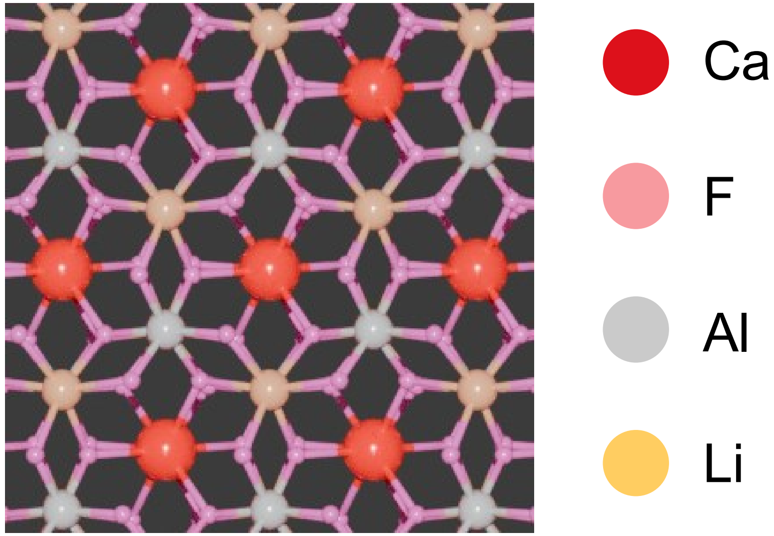
Nuisance?

No, these states can be used for isomer excitation via Electronic Bridge (EB)

[8] P. Dessovic *et al.*, J. Phys. Condens. Matter **26**, 105402 (2014) [9] M. Pimon, PhD Thesis, TU Wien (2021)

$^{229}Th:CaF_2$ versus $^{229}Th:LiCAF$



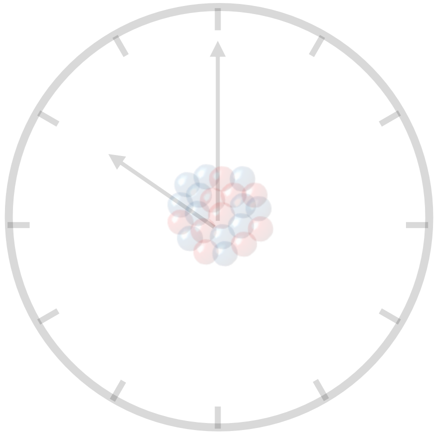


- $\Delta_g \sim 12$ eV bare CaF_2
- ^{229}Th replaces Ca ion → 2 Fluorine interstitials for charge compensation
- Formation of 8 defect states

- $\Delta_g \sim 12.7$ eV bare LiCAF
- Several preferred doping orientations + charge compensation mechanisms [10]
- Here: ^{229}Th replaces Al ion with neighboring Li vacancy
- Formation of 12 defect states

[10] M. Pimon *et al.*, Adv. Theory Simul. **5**, 2200185 (2022)

Outline



1) Nuclear clock approaches

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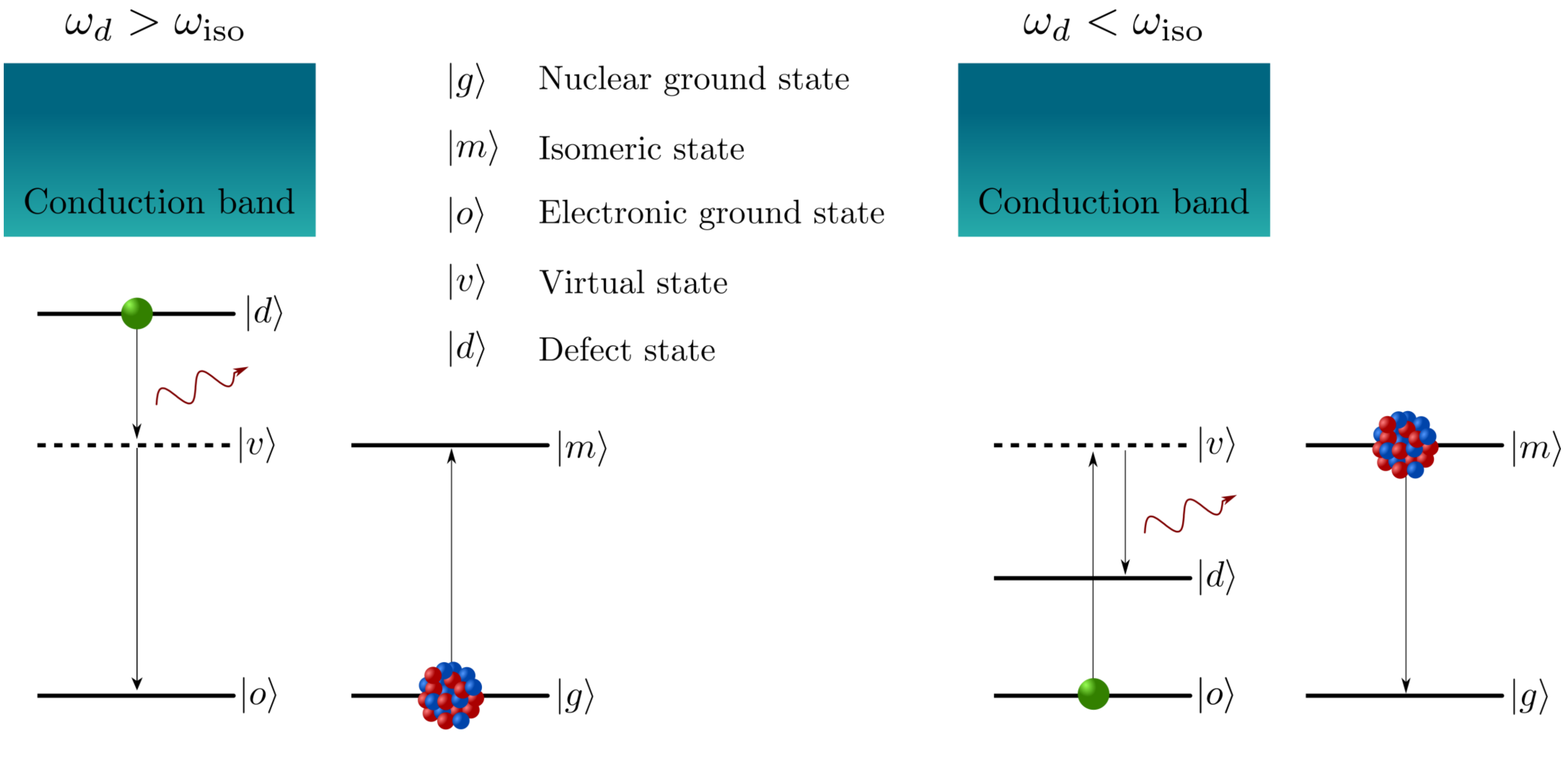
3) Electronic Bridge schemes in ^{229}Th:LiCAF

Electronic Bridge schemes

[11, 12]



Spontaneous excitation

Spontaneous decay

[11] B. S. Nickerson *et al.*, Phys. Rev. Lett. **125**, 032501 (2020)

[12] B. S. Nickerson *et al.*, Phys. Rev. A **103**, 053120 (2021)

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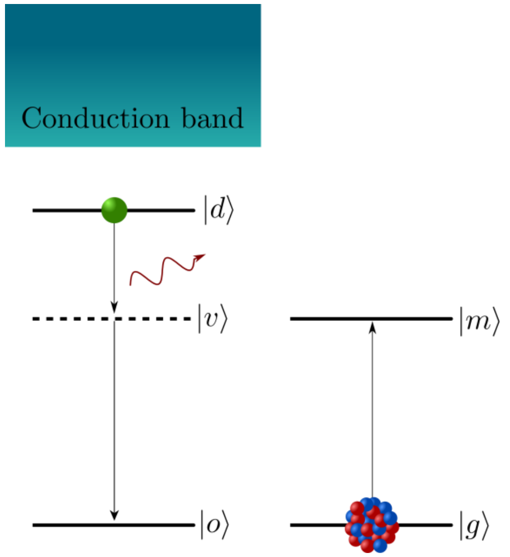
Some technical details



Using the example of spontaneous excitation [11,12]:

$$\Gamma_{\mathrm{eb}} \propto \sum_{m,g} \omega_p^3 \cdot \left|\langle m, o|\widetilde{\boldsymbol{D}}_{E1}|g, d\rangle\right|^2$$

Bridge photon of $E1$ multipolarity

Matrix element (3rd order perturbation theory):

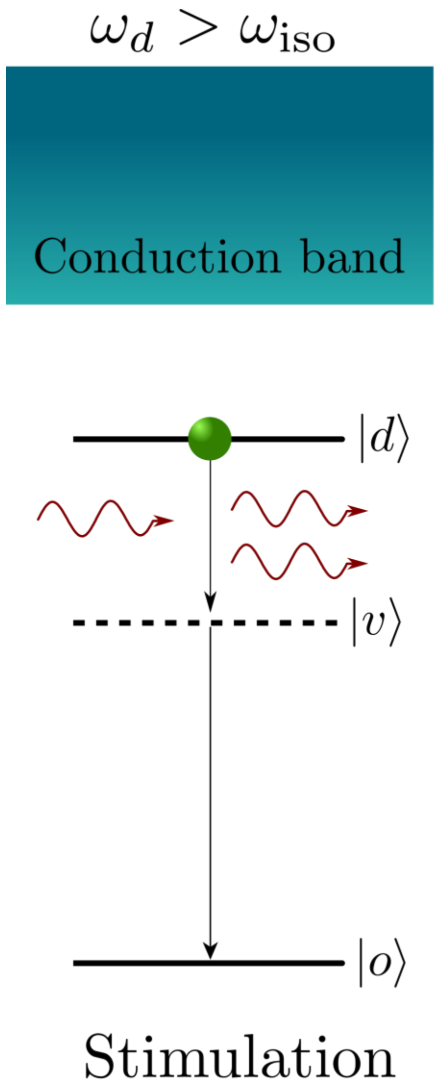
$$\langle m, o|\widetilde{\boldsymbol{D}}_{E1}|g, d\rangle = \sum_{\lambda K,q} (-1)^q \left[\sum_n \frac{\langle o|\boldsymbol{D}_{\boldsymbol{E1}}|n\rangle\langle n|T_{\lambda K,q}|d\rangle}{\omega_{dn} - \omega_{mg}} + \sum_k \frac{\langle o|T_{\lambda K,q}|k\rangle\langle k|\boldsymbol{D}_{\boldsymbol{E1}}|d\rangle}{\omega_{ok} + \omega_{mg}}\right] \langle m|M_{\lambda K,-q}|g\rangle$$

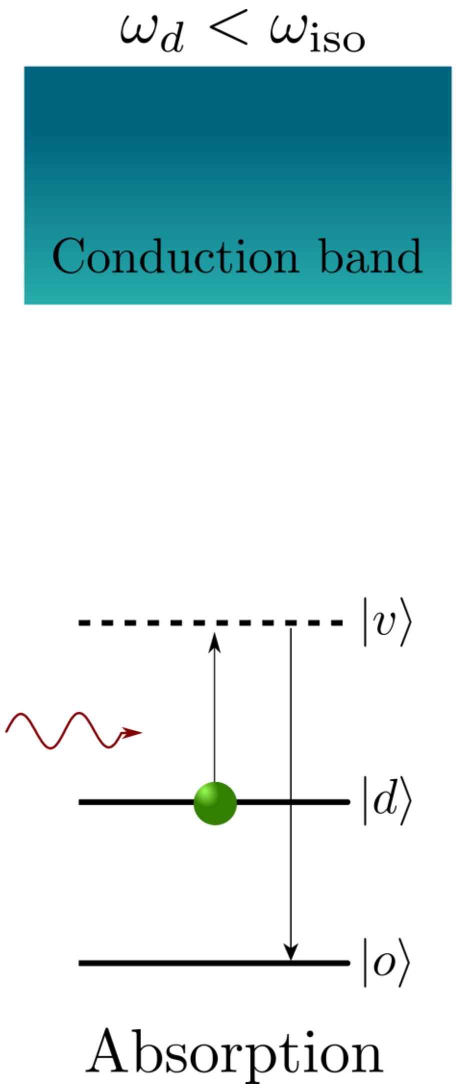
- $\boldsymbol{D}_{\boldsymbol{E1}}$: Photon emission via electronic transition
- $T_{\lambda K,q}$: Coulomb ($\lambda K = E2$)/current-current ($\lambda K = M1$) interaction between electron and nucleus
- $M_{\lambda K,-q}$: Nuclear transition via Coulomb or current-current interaction
- **Intermediate electronic states** forming the virtual states

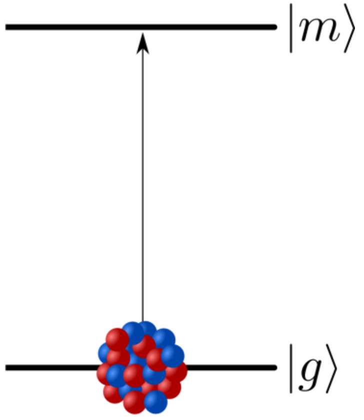
[11] B. S. Nickerson *et al.*, Phys. Rev. Lett. **125**, 032501 (2020)
[12] B. S. Nickerson *et al.*, Phys. Rev. A **103**, 053120 (2021)

Driven schemes

[11, 12]







$$\Gamma_{\rm driven} \propto \Gamma_{\rm eb} \frac{I_\omega}{E_p^3}$$

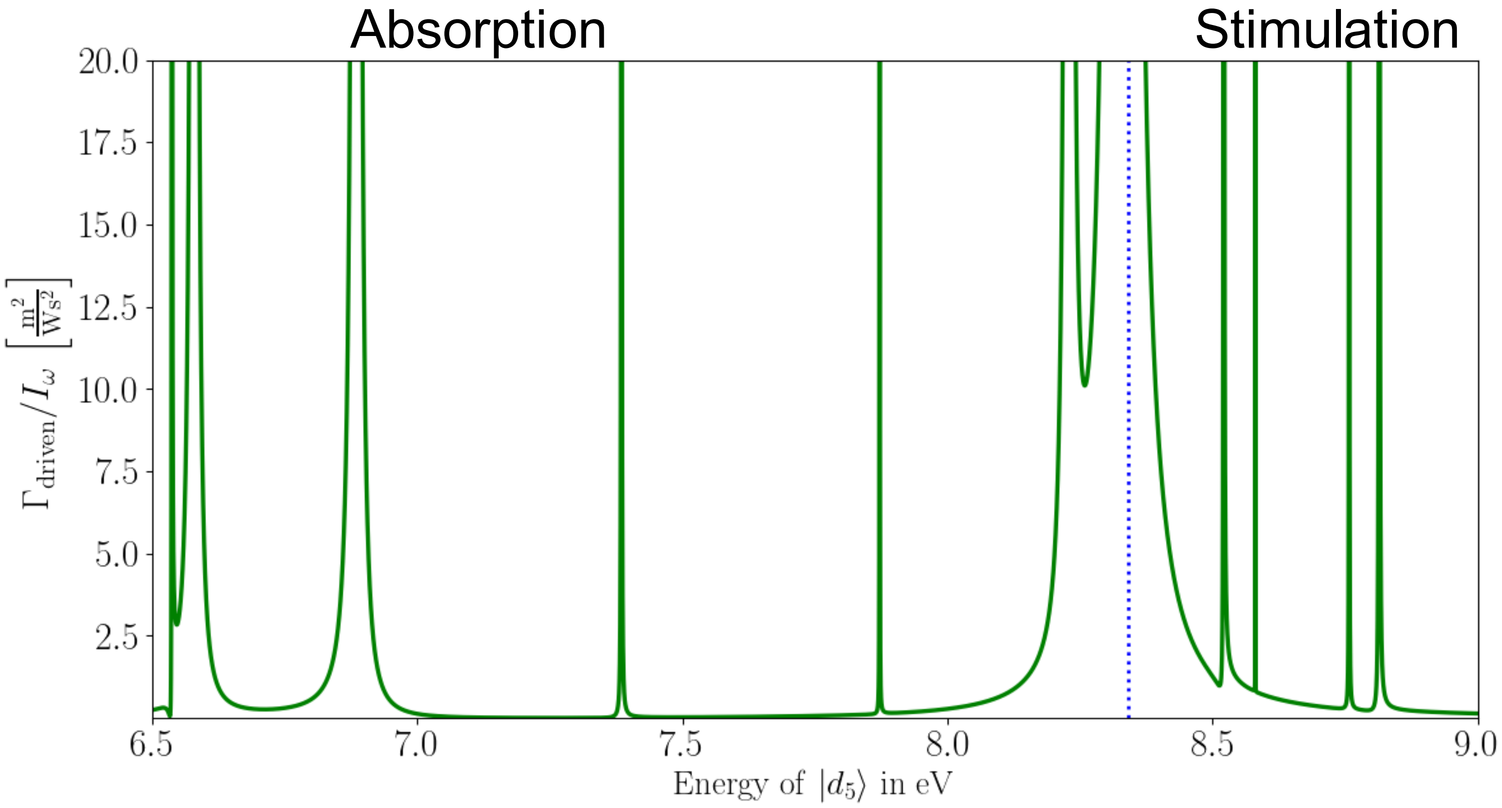
[11] B. S. Nickerson *et al.*, Phys. Rev. Lett. **125**, 032501 (2020)

[12] B. S. Nickerson *et al.*, Phys. Rev. A **103**, 053120 (2021)

Electronic Bridge spectrum

Defect state involved for EB schemes: $|d_5\rangle$ with predicted energy $E_d = 11.54$ eV ($E_1 < \cdots < E_{12}$)

→ Vary energy of $|d_5\rangle$ and other $|d_i\rangle$ around isomer energy by subtracting same constant from each state energy

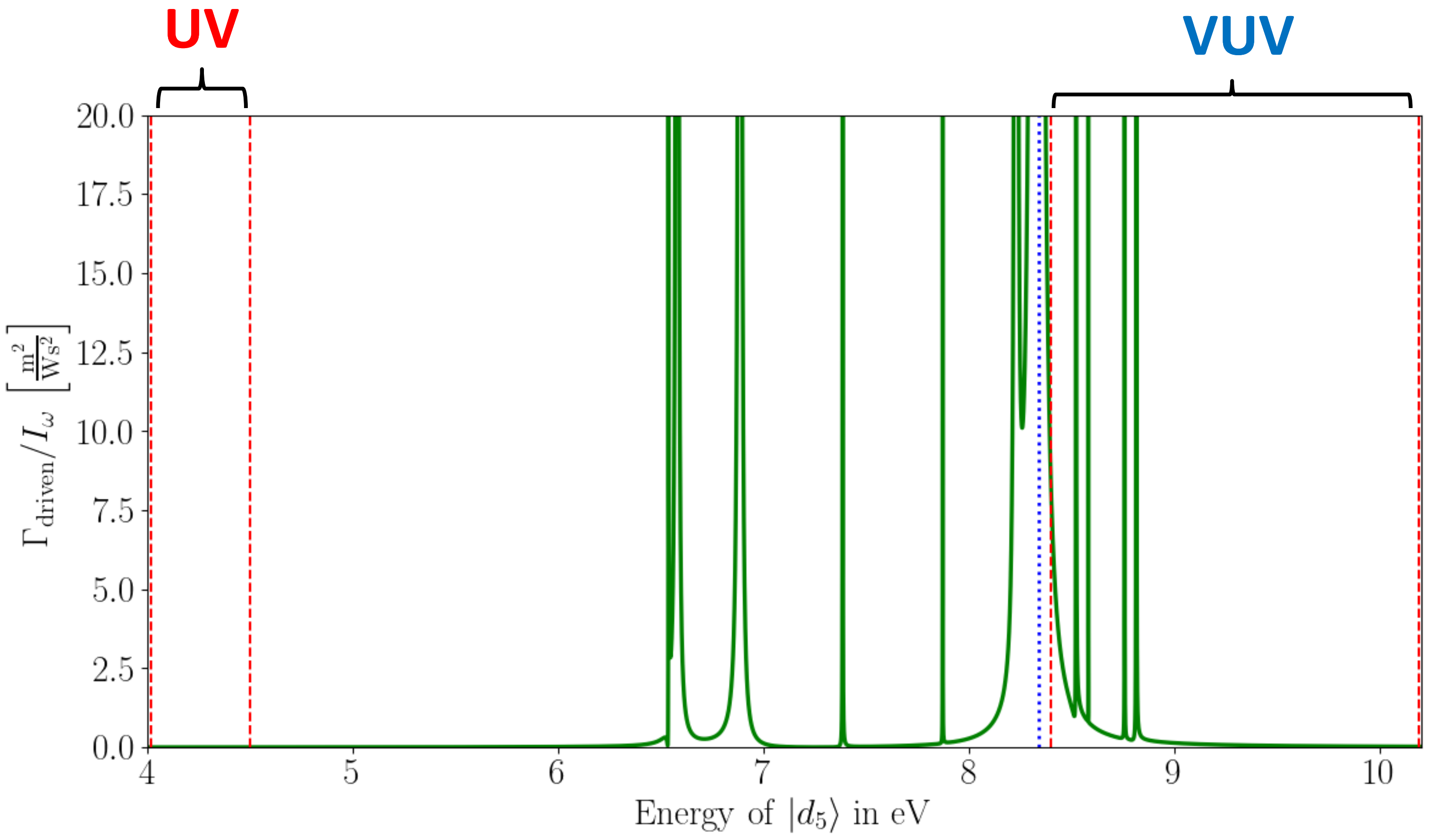


- **Resonance:** Alignment in energy of one of the defect states with the nuclear isomer
- Strong excitation when real electronic states are in the vicinity of the nuclear isomer

[13] T. Kirschbaum, M. Pimon *et al.*, in preparation (2024)

Advantage towards direct Photoexcitation?

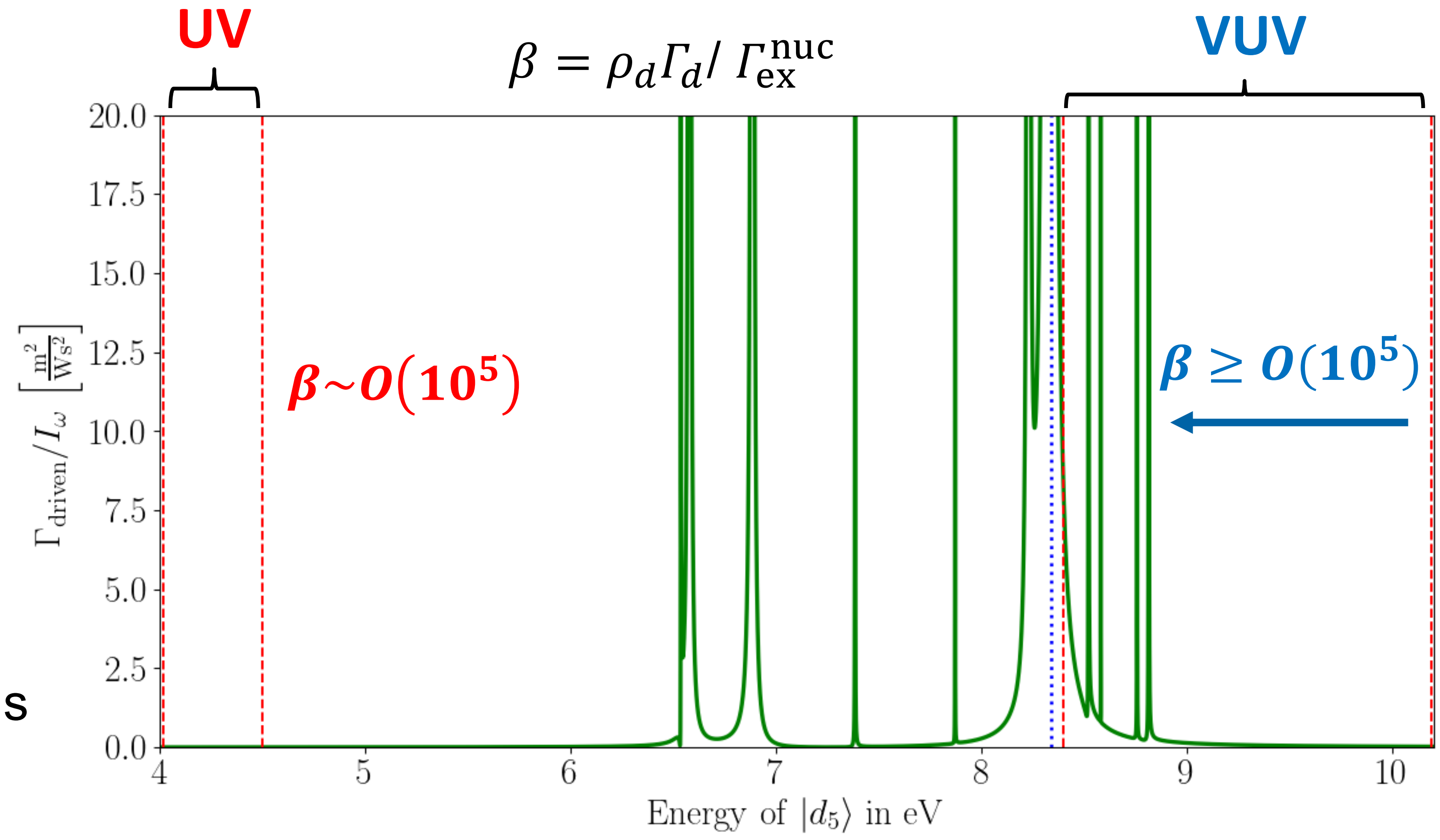
- With current laser technology [14]:
  $\boldsymbol{\Gamma_{\text{ex}}^{\text{nuc}} = 10^{-7} s^{-1}}$@ $w = 100\ \mu\text{m}$
- Enhancement: $\beta = \rho_d \Gamma_d / \Gamma_{\text{ex}}^{\text{nuc}}$
- For **demonstrative** purposes [13]:
  Enhancement of nuclear excitation via defect state in UV, VUV



[13] T. Kirschbaum, M. Pimon *et al.*, in preparation (2024)
[14] J. Thielkling *et al.*, New. J. Phys. **25**, 083026 (2023)

Number examples for nuclear excitation

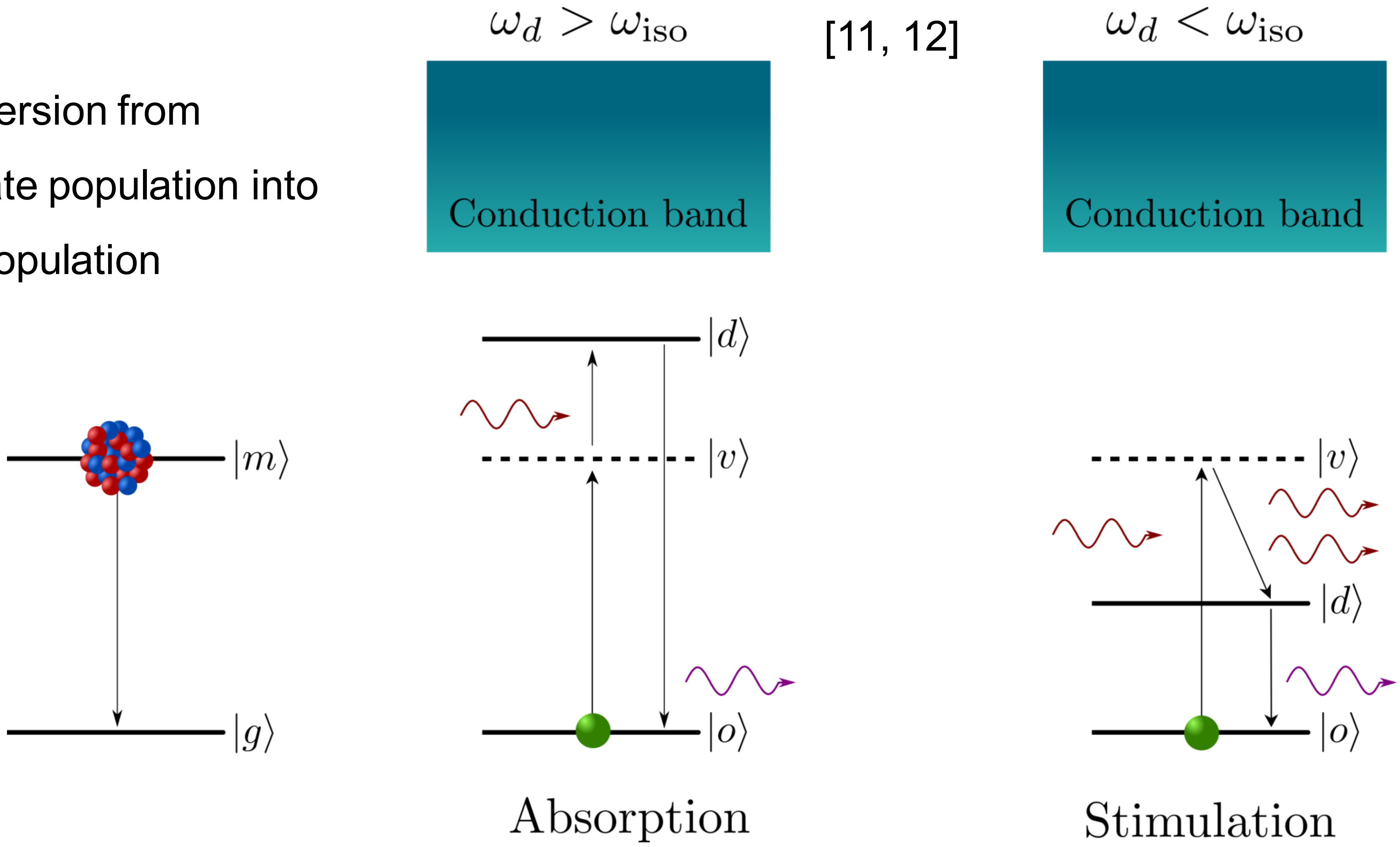
- Orders of magnitude stronger excitation compared to direct laser excitation $\Gamma_{\mathrm{ex}}^{\mathrm{nuc}}$ [13]
- **UV**: Assisting laser in UV range (e.g. tuneable UV lasers from TOPTICA)
- **VUV**: Assisting laser in optical/IR range
- However: Exact energetic position is crucial for the enhancement



[13] T. Kirschbaum, M. Pimon *et al.*, in preparation (2024)

Quenching schemes

Quenching: Conversion from nuclear excited state population into electronic defect population

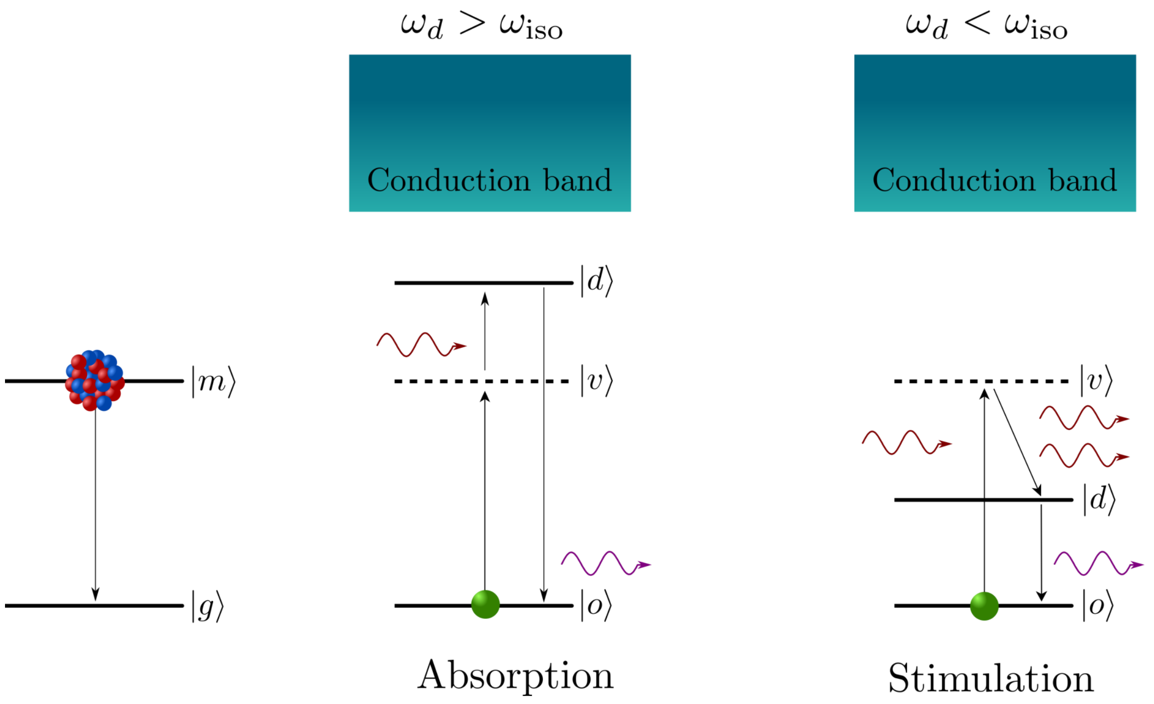


[11] B. S. Nickerson *et al.*, Phys. Rev. Lett. **125**, 032501 (2020)
[12] B. S. Nickerson *et al.*, Phys. Rev. A **103**, 053120 (2021)

Number examples for quenching

Consider quenching schemes in the **UV** spectral range and the **VUV** spectral range [13]: $\boldsymbol{\beta} = \boldsymbol{\Gamma_{qu}/\Gamma_{\gamma}}$

$$\Gamma_{\text{qu}} \propto \Gamma_{\text{eb}} \frac{I_\omega}{E_p^3}$$



UV (Stimulation): $\beta \sim O(10^2)$

VUV (Absorption): $\beta \geq O(10^3)$

Quenching scheme can lead to a controlled decay

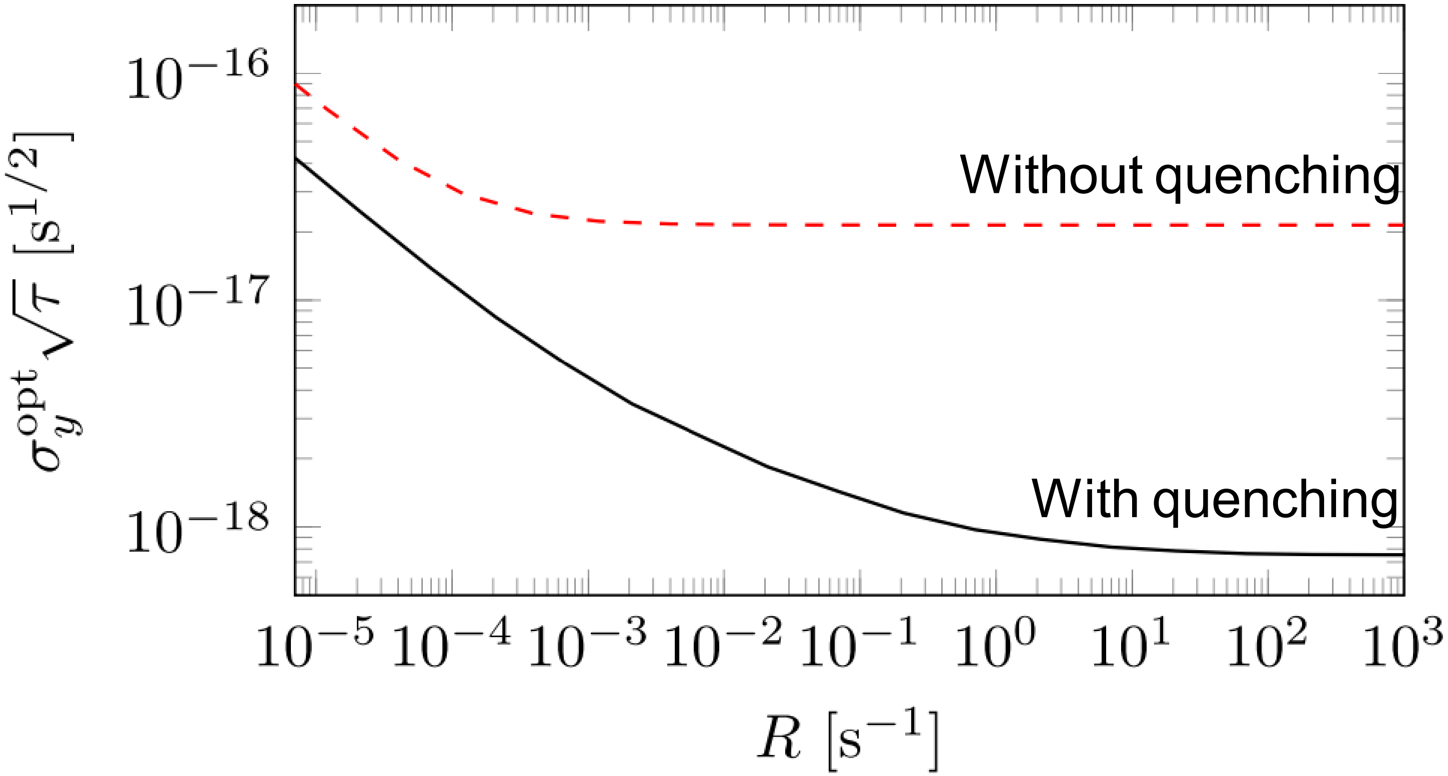
[13] T. Kirschbaum, M. Pimon *et al.*, in preparation (2024)

Quenching and influence on nuclear clock performance

- **Remember:** Within the solid state approach, flourescence photons are counted within a fixed time interval
- **Quenching**: More fluorescence photons in detection window

  → Better short term stability

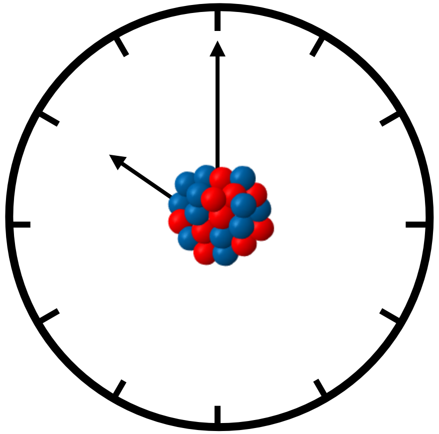
For EB in $^{229}Th:CaF_2$ [12]



Clock fractional instability as a function of excitation rate

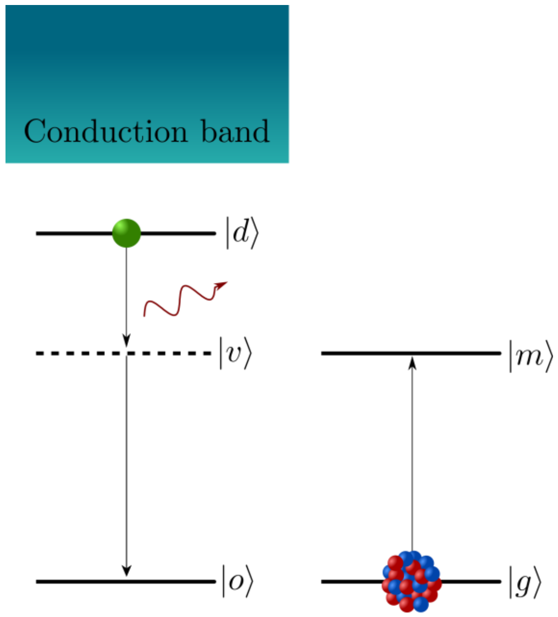
[12] B. S. Nickerson *et al.*, Phys. Rev. A **103**, 053120 (2021)

Conclusion & Outlook



- ^{229}Th provides a unique chance to develop a **nuclear clock** with increased accuracy and potentially investigate new physics

- Electronic bridge schemes via crystal defects in ^{229}Th:LiCAF
- Depending on the energetic position of the defect state, much stronger nuclear (de)excitation occurs
- **Outlook**: Experimental verification of defect states + influence of different ^{229}Th:LiCAF structures



Thanks

University of Würzburg

Technical University of Vienna

Martin Pimon

Thorsten Schumm

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Thank you for your attention!