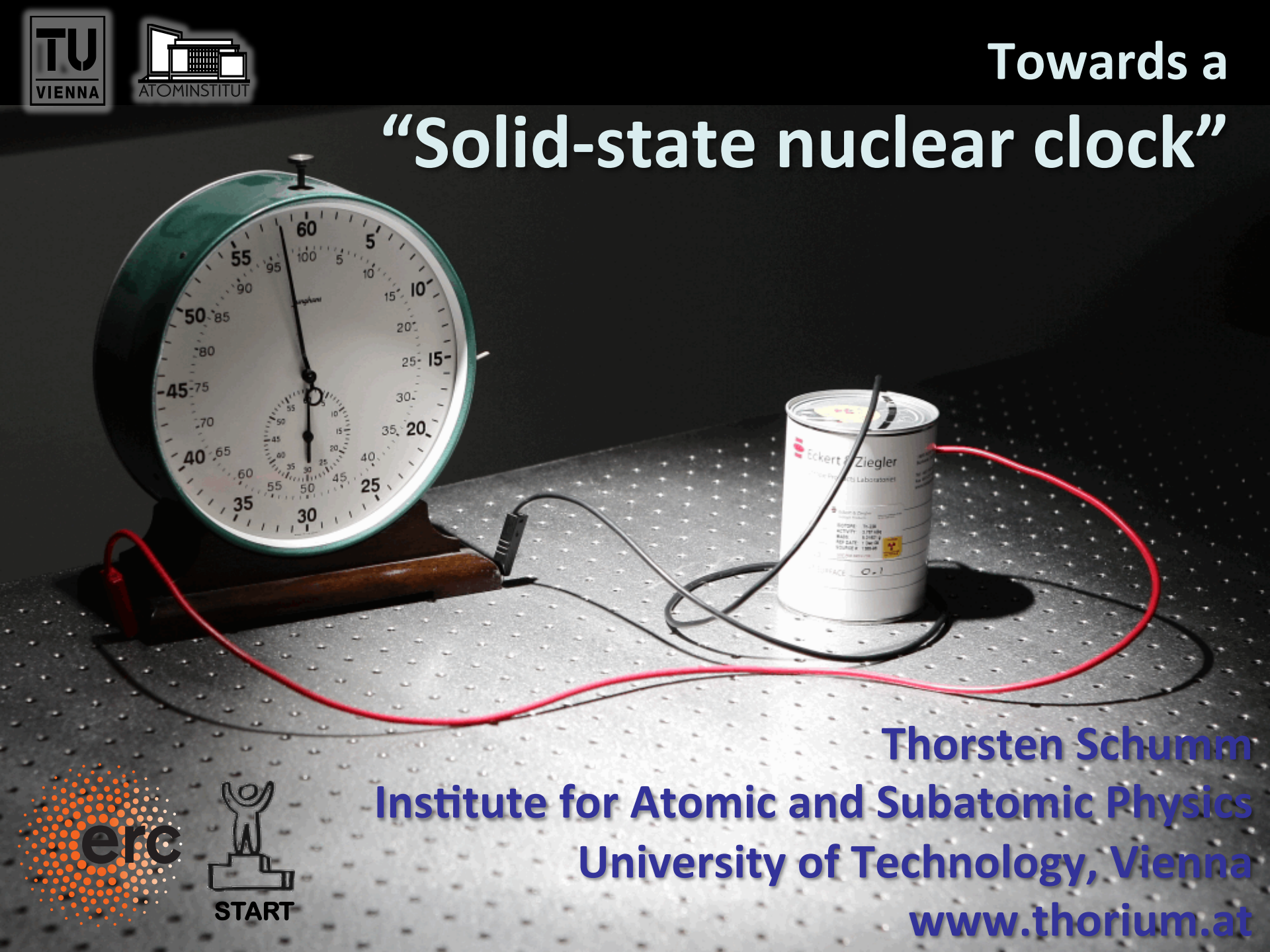
Towards a "Solid-state nuclear clock"



Thorsten Schumm
Institute for Atomic and Subatomic Physics
University of Technology, Vienna
www.thorium.at

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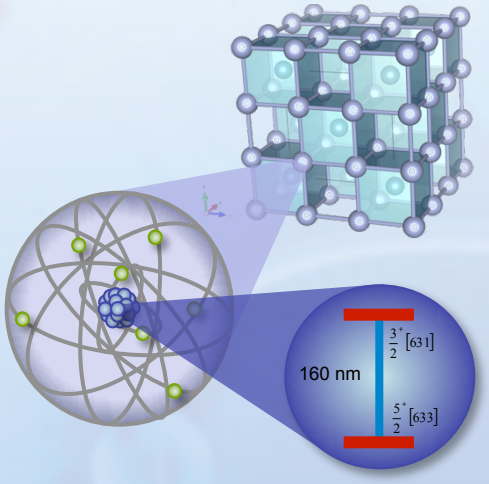
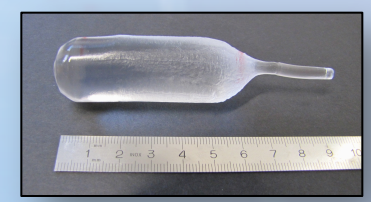
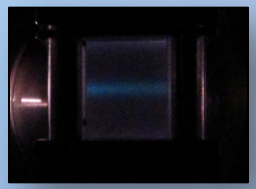
START

Towards a “Solid-state optical nuclear clock” Outline







Part 1 - A nuclear transition in a solid-state environment

- doping Thorium: microscopic configuration
- electronic structure calculations for Th-doped CaF_2
- quantum physics of the nucleus (in the crystal)

Part 2 - Consequences for a clock

- fluorescence interrogation scheme
- projected clock performance

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Part 3 - Experimental efforts

- ^{232}Th-doped CaF_2 crystals:
  - bulk characterization
  - transmission + luminescence

Part 4 - Conclusions and outlook

- Towards homegrown crystals
- Towards a UV frequency comb

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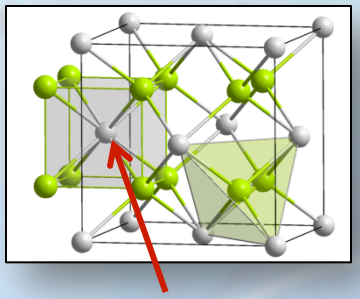
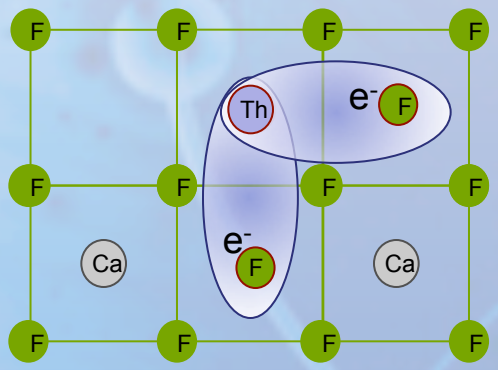
The basic Idea:
…doping Thorium into CaF_2



put ^{229}Th here!

^{229}Thorium replaces an Ca ion in a CaF_2 crystal

- a macroscopic (up to 10^{18}) number of nuclei for spectroscopy (clock)
- a UV transparent host matrix to hold them (line shifts/broadenings)



Pure CaF_2 properties:

- highest bandgap insulator (11.6 eV, VUV transparent)
- simple cubic lattice structure (can be calculated)
- „easy“ to grow (Czochralski, Bridgeman, MicroPulling)
- high damage threshold, well characterized...

Thorium doping:

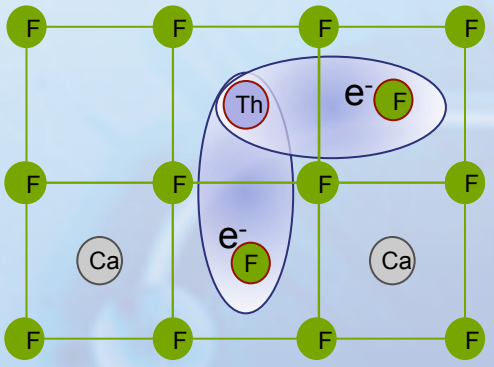
- Thorium chemically prefers 4+ state (Radon-like)
- 4+ ion instead of 2+ ion: needs **charge compensation**
- ...will break the symmetry + modify electronic structure

2 main questions (...in this talk):

- Will Thorium-doping change the crystal properties (e.g. transparency)?
- What will be the effects of the crystal on the nuclear transition (clock performance)?

The Th:CaF_2 system
…a 3-step calculation…

Solid-state system



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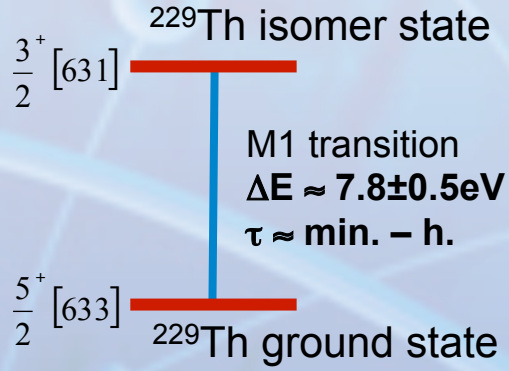
Nuclear system



= ?

Step 1: Determine charge compensation mechanism

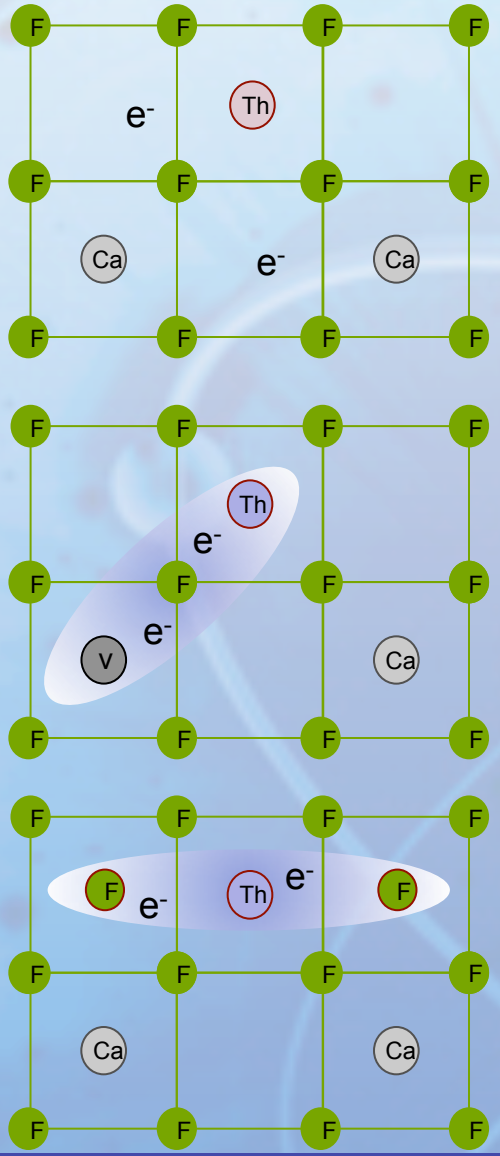
„Atomistic“ simulations based on GULP code (R. Jackson, Keele Univ.)

Step 2: Determine electronic structure of doping complex

DFT calculations based on VASP code (P. Mohn, CMS, Vienna)

Step 3: Quantum physics of the nuclear states ➔ **clock performance?**

The Th:CaF_2 system
Step 1: Charge compensation



Charge compensation mechanisms

Possible candidates:

- 2 x F-interstitial (many spatial combinations)
- Ca vacancy (nearest Ca)
- (impurity compensation, e.g. O_2)

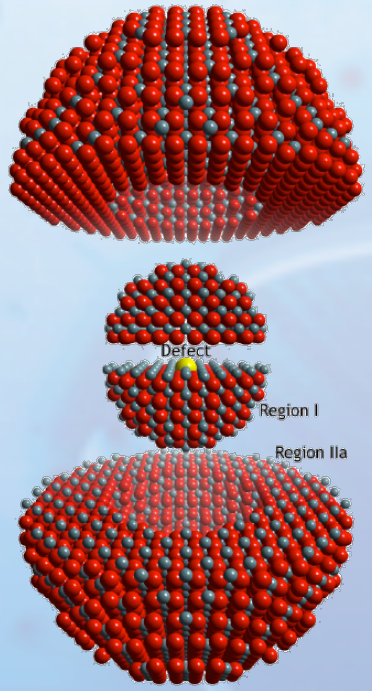
Calculate solution Energies using GULP code

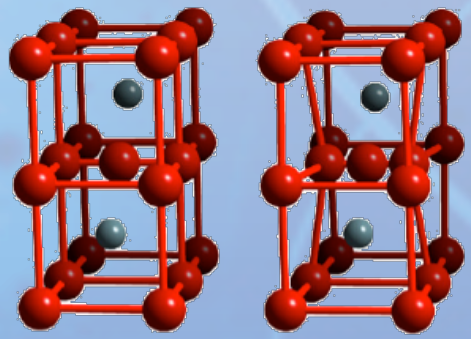
Example: Ca vacancy (only one possibility):

$$E_{sol} = -E_{latt}(ThF_4) + E(Th_{Ca}^{**}) + E(V_{Ca}^{''}) + 2E_{latt}(CaF_2)$$

- energy to „melt“ ThF_4
- energy to replace Ca with Th
- energy to remove one Ca
- binding energy of CaF_2

The Th:CaF_2 system
Step 1: Charge compensation





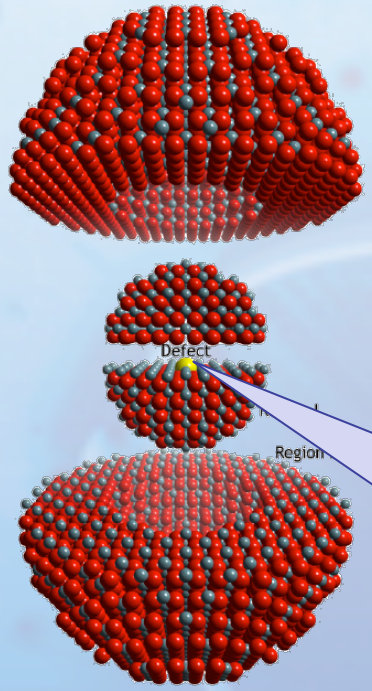
How it is done (Rob Jackson, Keele University)

- Model ion interactions with (classical) **Buckingham potential**

$$V_{Buck}(r_{i,j}) = \underbrace{A_{i,j}\exp\left(-\frac{r_{i,j}}{\rho_{i,j}}\right)}_{\text{repulsive}} \underbrace{-\frac{C_{i,j}}{r_{i,j}^6}}_{\text{attractive}}$$

- Determine potential parameters by fitting the undoped structures (lattice spacing, dielectric properties, stress tensors...)
- Start with a given geometry
- Relax all ion positions in region 1 (scaling region 2)
- Determine solution energy for relaxed system
- search for configuration of the lowest solution energy...

The Th:CaF_2 system
Step 1: Charge compensation



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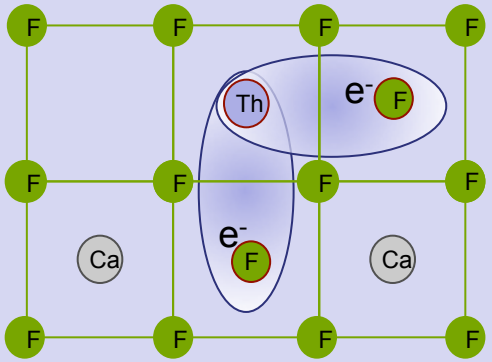
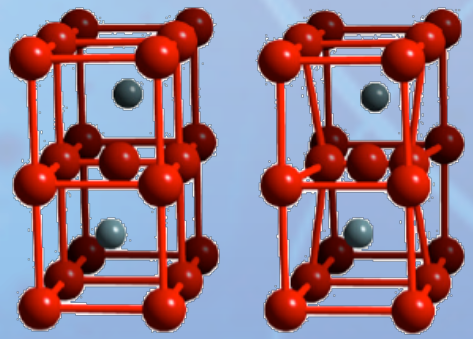
Result: System will go to a 90° F-interstitial compensation

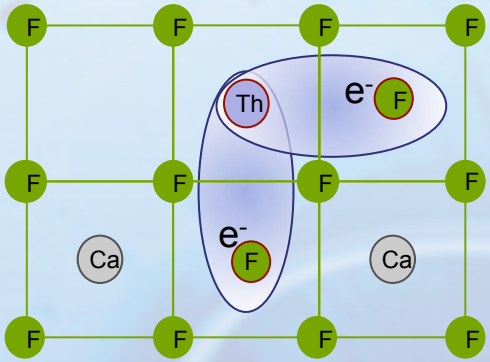


(also confirmed by VASP calculations)

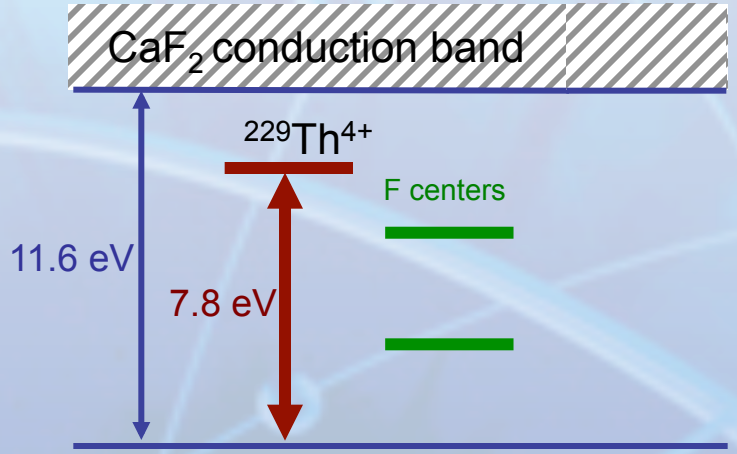


unrelaxed relaxed

The Th:CaF_2 system
Step 2: electronic structure calculations



What about the electronic structure?

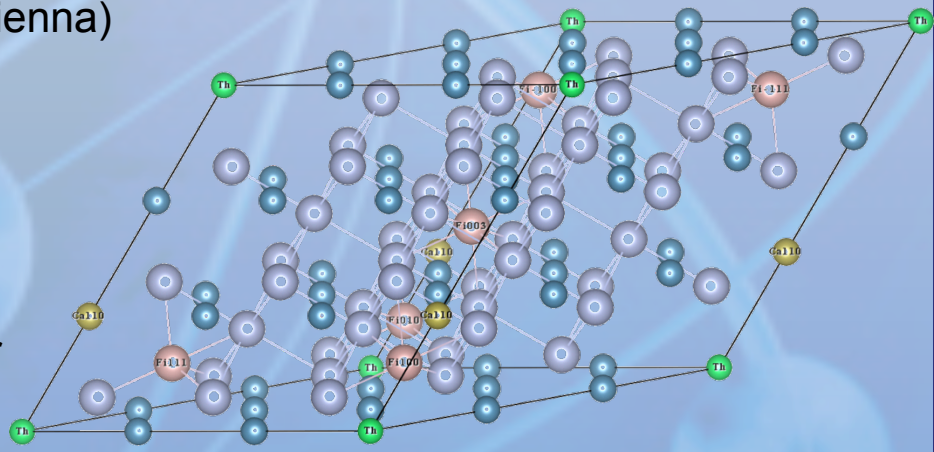


- new absorption levels?
- band gap shifts?
- effects of impurities?

Full electronic structure calculations using VASP code...

(collaboration with P. Mohn, P. Dessovic, CMS Vienna)

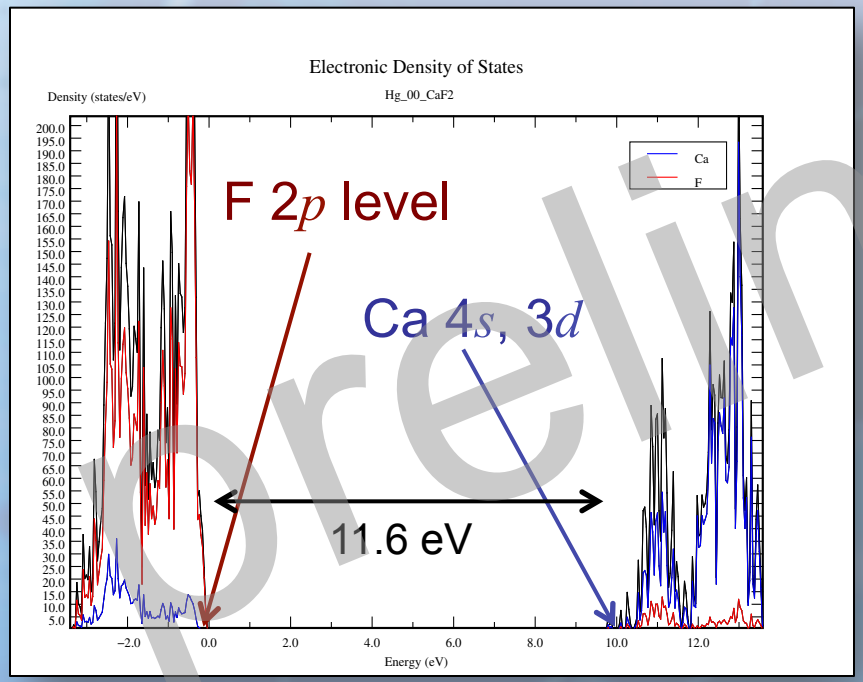
- 83 ions in „supercell“
- Full spatial relaxation
- 612 total electron orbitals
- 348 orbitals treated dynamically
- several weeks on large computer cluster



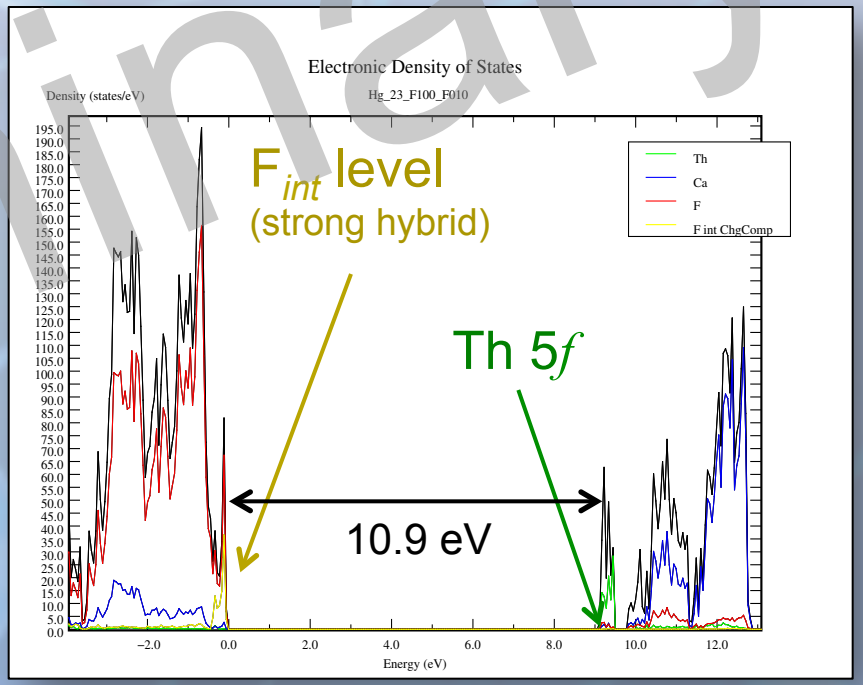
The Th:CaF_2 system
Step 2: electronic structure calculations

Results:

- Undoped CaF_2: large bandgap of 11.6 eV
- **Th-doped CaF_2: slightly reduced bandgap 10.9 eV** ☺



Undoped CaF_2

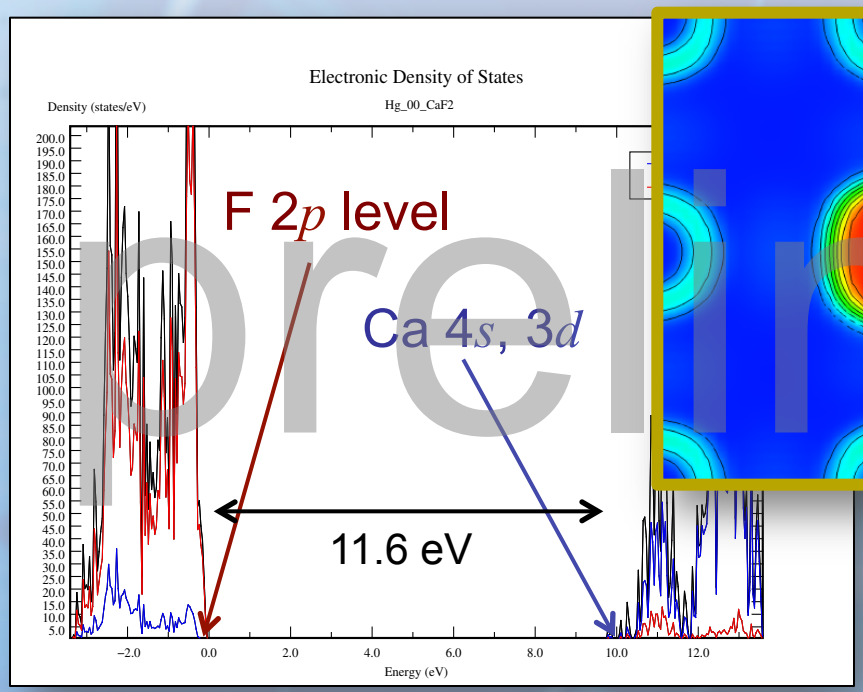


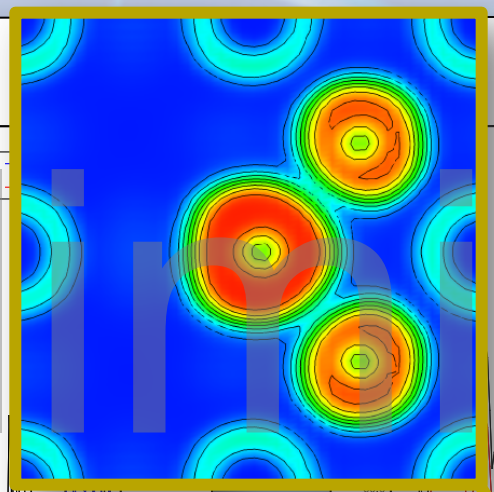
Th-doped CaF_2

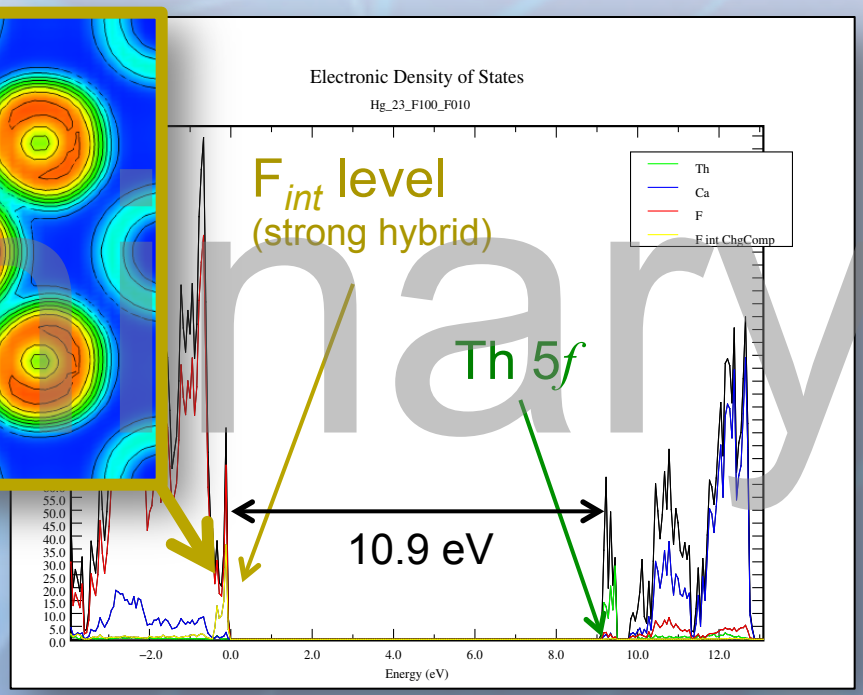
The Th:CaF_2 system
Step 2: electronic structure calculations

Results:

- Undoped CaF_2: large bandgap of 11.6 eV
- **Th-doped CaF_2: slightly reduced bandgap 10.9 eV** ☺
  - we also know the wave function at the fermi edge (field gradients)
  - input for calculations of the nuclear quantum levels…





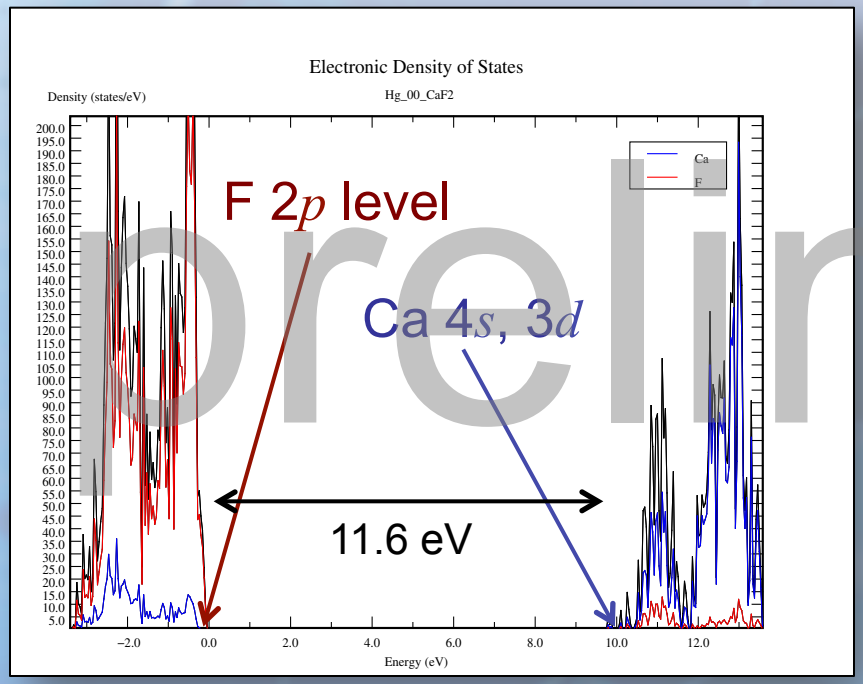


Undoped CaF_2 | Th-doped CaF_2

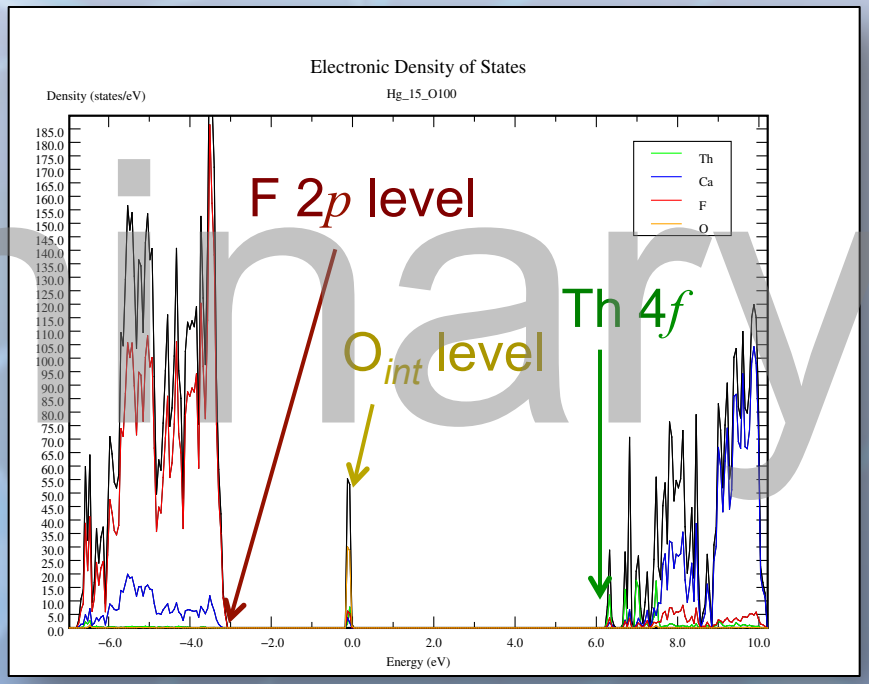
The Th:CaF_2 system
Step 2: electronic structure calculations

Results:

- Undoped CaF_2: large bandgap of 11.6 eV
- Th-doped CaF_2: slightly reduced bandgap 10.9 eV
- **O charge comp. in Th:CaF_2: kills transparency!** ☹
  (multi-step excitation via co-doping?)

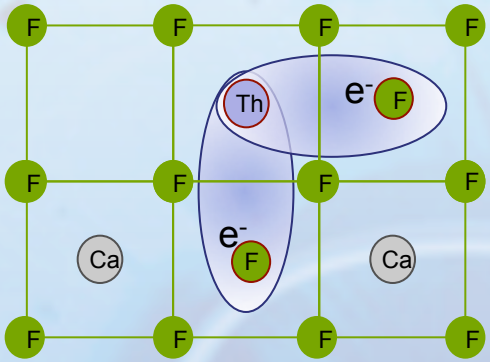


Undoped CaF_2



Th-doped CaF_2 with O charge compensation

The Th:CaF_2 system
Step 3: Quantum states of the nucleus



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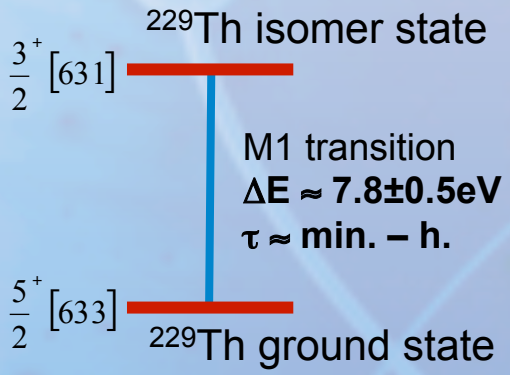


W.G. Rellergert, et al.,Phys. Rev. Lett. 104, 200802 (2010)
Kazakov et al, New. J. Phys. 14, 083019 (2012)

Nuclear-lattice interaction (via electronic states)

$$H = \cancel{H_{FS}} + H_{HFS}$$

no unpaired electrons
➔ No fine interaction

$$H_{HFS} = H_{E0} + H_{M1} + H_{E2} + \ldots$$

multipole expansion of the hyperfine interaction

H_{E0}: electric monopole shift (DC field at nucleus pos.)
- order GHz (exact calculation under way)
- identical for all Th ions up to temperature (**≈ 10 kHz/K**)

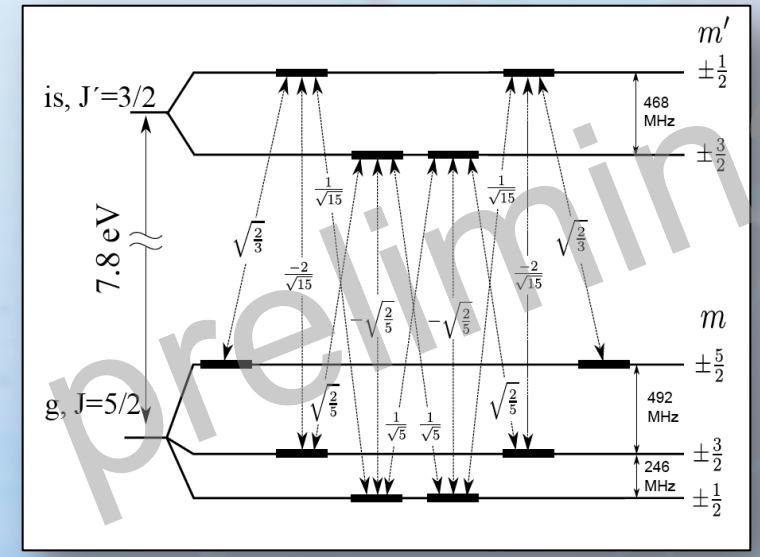
H_{E2}: electric quadrupole shift (gradient at nucleus pos.)
- 1.52×10^{18} V/cm^2 for calculated electronic structure
- gives splittings on order **100 MHz**
- inhom. broadening **≈ 100 Hz**

H_{M1}: magnetic dipole interaction
- randomly oriented neighboring nuclei (F-ions, 2.63 μ_N)
- inhom. broadening **≈ 1 - 2 kHz** (dominant broadening)

+ 2nd order Doppler effect
- global shift: 10 Hz/K (**700 Hz** at liquid Nitrogen)
- broadening: 7 Hz/K (**500 Hz** at liquid Nitrogen)

The Th:CaF_2 system
Step 3: Quantum states of the nucleus

Quantum states of the nuclear transition inside the CaF_2 crystal



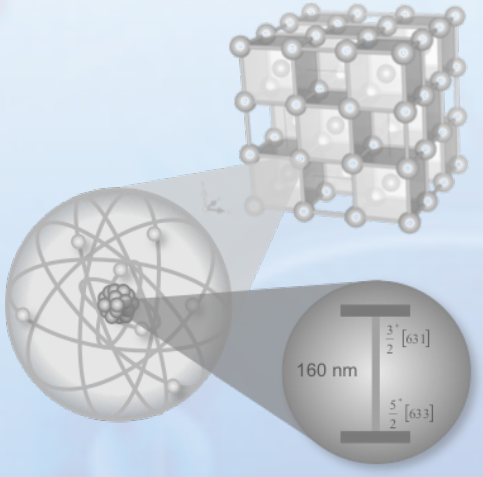
- **Global shift** due to electric monopole interaction (≈1 GHz, calculation under way)
- **Hyperfine splitting** due to interaction with electron orbital of the charge compensation
- **Broadening + dephasing** (≈ 1.5 kHz) due to magnetic interaction to nearby nuclei

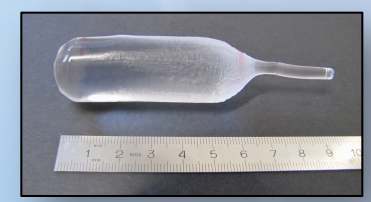
➔ **expected ensemble linewidth: 1.5 kHz**

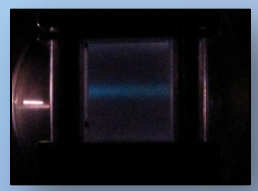
Consequences:

- Direct **nuclear spectroscopy seems feasable** in the solid-state approach (bandgap only slightly reduced by doping)
- Solid-state environment **reduces Q-factor to 1 x 10^{13}**
- Rapid dephasing (order ms) makes **„coherent“ interrogation schemes impossible!**

Towards a “Solid-state optical nuclear clock” Outline







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- fluorescence interrogation scheme
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Poster Kazakov

Part 3 - Experimental efforts

- ^{232}Th-doped CaF_2 crystals:
  - bulk characterization
  - transmission + luminescence

Part 4 - Conclusions and outlook

- Towards homegrown crystals
- Towards a UV frequency comb

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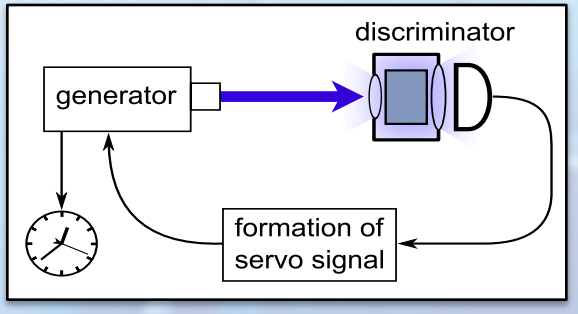
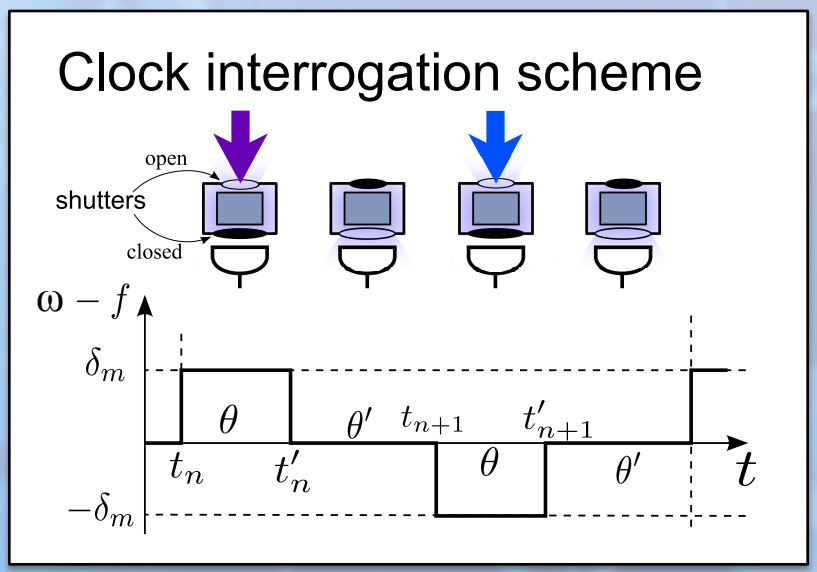
The Th:CaF_2 system
How to build a clock from it?

„Nuclear fluorescence spectroscopy“



Problem: Coherence time ≈ 1 ms
➔ We have to rely on spontaneouse decay
Next problem: lifetime of nuclear state: $1/\gamma \approx 1000$ s



- detune red by δ from nominal frequency
- illuminate for time θ with a pumping rate R
- count photons for time θ'
- repeat for blue detuning
- correct frequency offset f
- start again (**continuous operation!**)

cycle determined by **4 parameters δ, θ, θ', R**

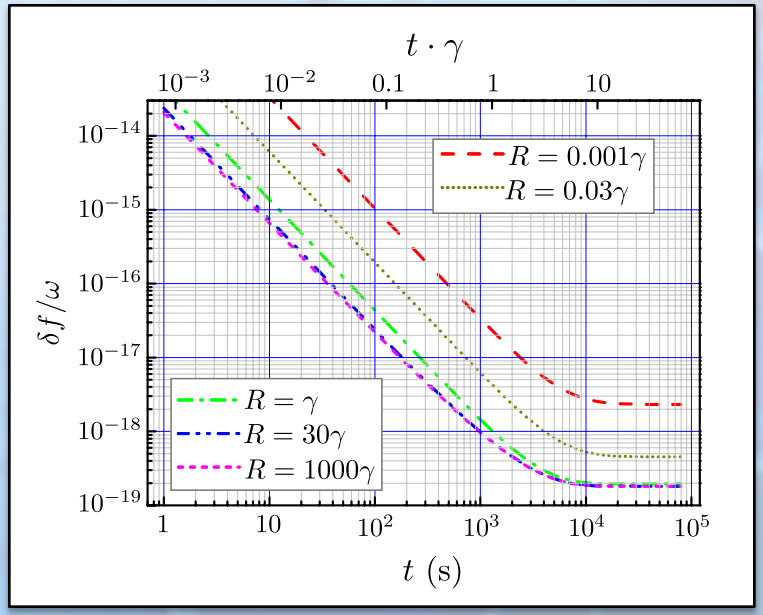
- can be optimized to minimize the error δf on f (numerical optimization, photon shot noise limit)
- optimal parameters maintain a constant number of excited nuclei

Kazakov et al, New. J. Phys.14, 083019 (2012)

The Th:CaF_2 system
Projected clock performance

Fractional frequency error per interrogation cycle

Kazakov et al, New. J. Phys.14, 083019 (2012)



fractional clock instability
for a series of N measurements:

$$\sigma_{opt}(N) = \frac{\delta f}{\omega\sqrt{N}} = \frac{1.6\times10^{-17}\, s^{-1/2}}{\sqrt{\tau}}$$

(for $R \approx 1/\gamma$)

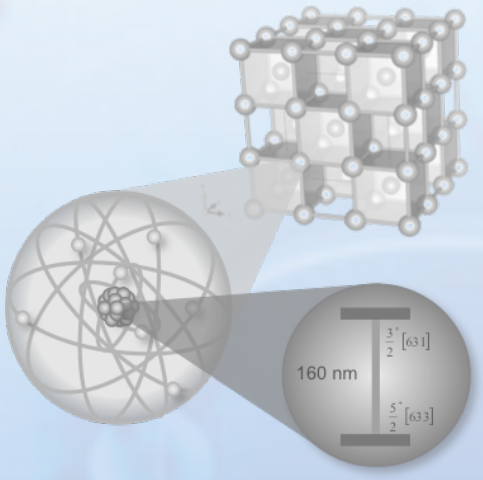
should reach **10^{-19} fractional instability**
within a few hours of averaging
(similar to trapped ion approach?)

NOT included here (...the real world):

- temperature stability of the sample (2nd order Doppler@10 Hz/K)
- properties of the laser (linewidths, frequency/amplitude stability)
- external fields
- non-nuclear luminescence effects (background)

+ this is **never an accurate clock!** ➔ secondary standard?

Towards a “Solid-state optical nuclear clock” Outline



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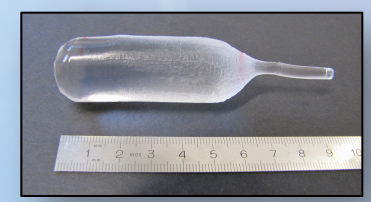

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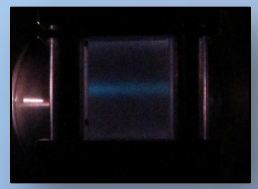
  - transmission + luminescence



Part 4 - Conclusions and outlook

- Towards homegrown crystals
- Towards a UV frequency comb

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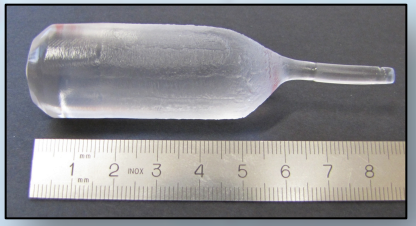
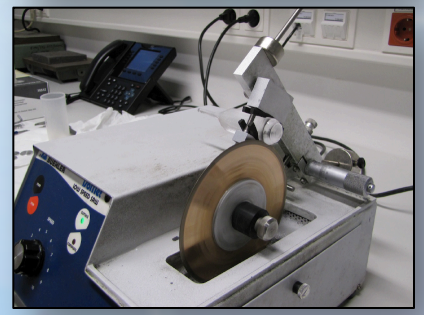
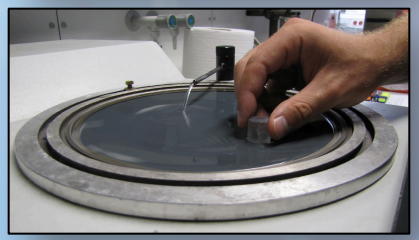
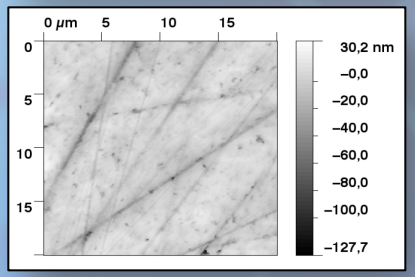
First ^{232}Th:CaF_2 crystal …doping really works!









AFM after polishing

Bulk material properties

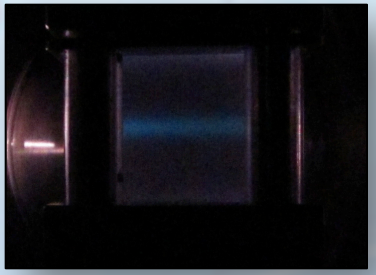
- Grown at IKZ Berlin using Czochralski method (2 weeks total)
- Measured Thorium concentration:
- 200-300 ppm (2 x 10^{18} /cm^3), homogeneous over the crystal, crystal-melt distribution 1:3 (30% efficiency)
- ICP mass spectrometry, neutron activation analysis, gamma spectroscopy (identical for us;-)

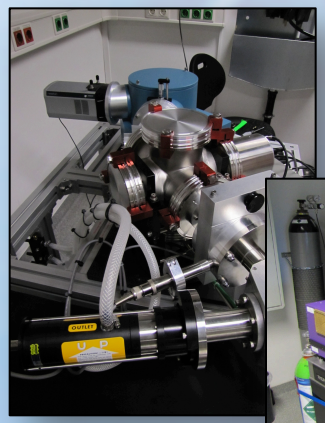
➔ **CaF chemically accepts the Thorium, doping concentration under control**

Surface properties

- Cut and polished at TU Wien (radiation safety area)
- Corrugation after polishing: 30 nm RMS, 300 nm P-P
- Parallelism an issue

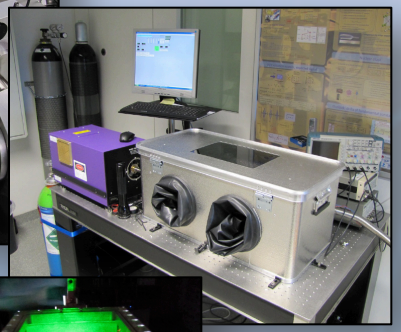
First ^{232}Th:CaF_2 crystal optical properties: transmission

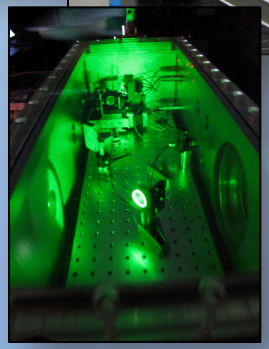




broadband UV spectrometer

157 nm Excimer Laser





160 nm fs pulse laser @MBI Berlin

Transmission measurements:

In-house UV spectrometer (deuterium lamp)

- smooth transmission down to 150 nm
- no additional absorption bands compared to CaF_2

In-house F_2-Excimer laser

- 157 nm fixed, narrow band (?)
- 8 ns pulse duration, 200 Hz reprate
- peak photon flux: 4 x 10^{21} $s^{-1}mm^{-2}$ (7 x 10^{14} $s^{-1}mm^{-2}$ CV eq.)
- **> 80% transmission** (9% surface loss)
- weak 2-photon processes, no visible damage

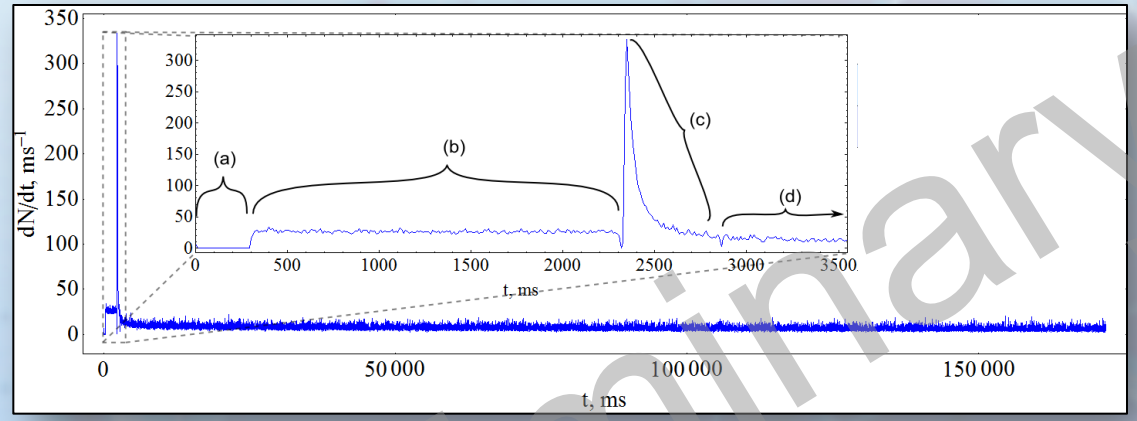
MBI Berlin femtosecond system

- 160 nm tuneable (?), 1 nm linewidth
- 50 fs pulse duration, 1 kHz reprate
- peak photon flux: 4 x 10^{24} $s^{-1}mm^{-2}$ (2 x 10^{14} $s^{-1}mm^{-2}$ CV eq.)
- **> 65% transmission, clear 2-photon processes**
- slight reversible damage for focused beam

We suspect: laser-induced damage for short pulses

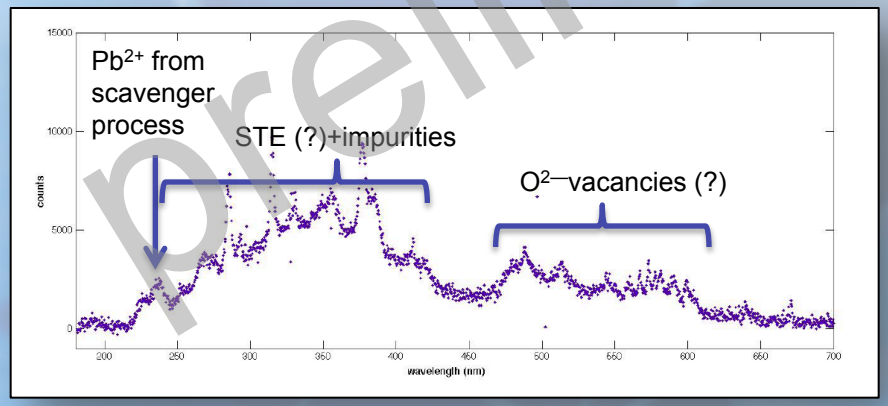
First ^{232}Th:CaF_2 crystal optical properties: luminescence

luminescence AFTER exposure by femtosecond pulse:



- very long „afterglow“ (up to secondes!) after illumination with laser (only femtolaser so far)
- luminescence shows (at least) three exponential decay timescales (different processes?)
- build-up of luminescence over several illuminations

luminescence spectrum after nanosecond pulse:

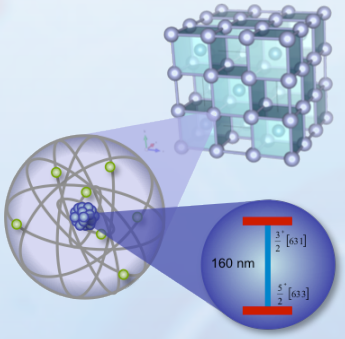


Luminescence timescale collides with expected lifetime of nuclear state!

We need to:

- clearly **identify impurities** (e.g. Pb^{2+}, Yt^{3+}, Eu^{2+}, Eu^{3+})
- measure timescales of the respective processes
- filter by time, frequency, spatial direction, ...
- compare other host crystals

Towards a “Solid-state optical nuclear clock”
Summary



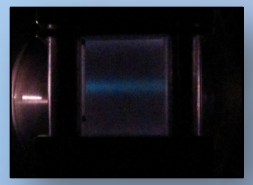
Part 1 – Solid-state theory

- Identified “microscopics” of Th:CaF_2 doping complex
  (charge compensation, geometry)
- Th-doped CaF_2 maintains large band gap (impurities problematic)
- main source of de-coherence (≈ 1ms): neighboring F-nuclei

Part 2 - Consequences for a clock

- fluorescence interrogation scheme required
- still $<10^{-17}$ performance can be achieved
  (@ liquid N temp., neglecting ALL technical constrains)

Part 3 - Experimental efforts



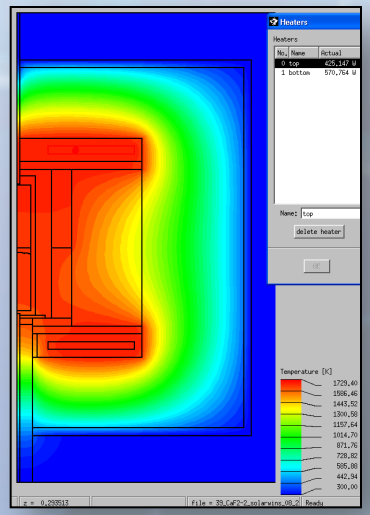
- ^{232}Th doping performed successful
- transmission indicates good transparency (avoid high pulse intensity)
- luminescence shows long-term component (more studies needed)

…next steps…
do the same with the “real” ^{229}Th isotope

Produce ^{229}Th:CaF_2 crystals...

will have to be done in-house...

assembled vessel



temperature simulations

Collaboration with Fraunhofer Institute IISB Erlangen

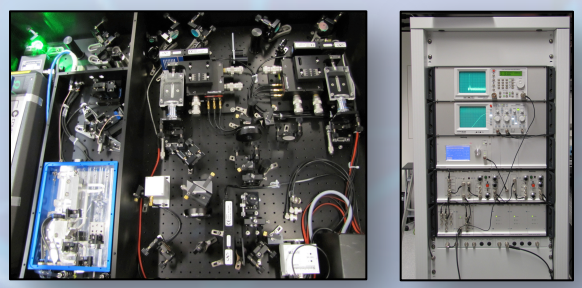
See posters of:

Matthias Schreitl

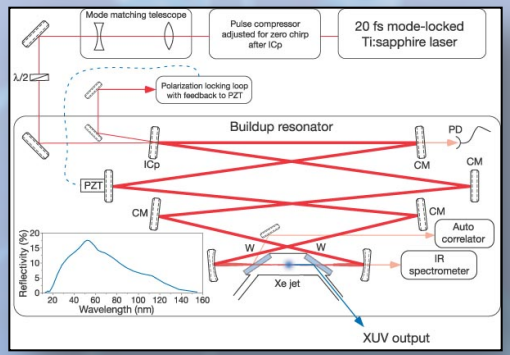
Georg Winkler

Perform VUV spectroscopy

in-house VUV frequency comb



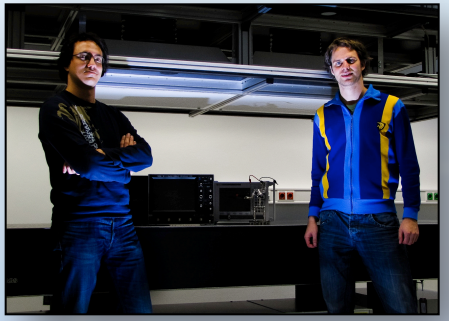
help by MENLO SYSTEMS & MPQ



High-harmonic generation in a passive build-up cavity
(following Hänsch/Gohle, Ye...)

Towards a "Solid-state nuclear clock"
The Thorium **team + collaborators**

The Thorium team:

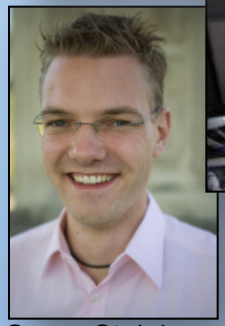


Georg Winkler Matthias Schreitl

Wolfgang Schlichtner

Bianka Ullmann

Georgy Kazakov



Georg Steinhauser

Thorsten Schumm

Collaborators:

- Crystal growing
  - IKZ Berlin
  - IISB Erlangen
  - University of Pisa
- Crystal characterization
  - Inorganic Chemistry University of Siegen
  - Kristallographie Universität Bremen
- Crystal simulations
  - Keele University
  - CMS Vienna (P. Mohn, P. Dessovic)
- Excitation sources
  - Max Born Institute Berlin
  - MPQ Munich
- Thorium extraction/handling
  - ATI radio chemistry group

More info: www.thorium.at

Towards a "Solid-state nuclear clock"
The Thorium **team + collaborators**

The Thorium team:

Georg Winkler

Matthias Schreitl

Wolfgang Schlichtner

Benjamin Ullmann

Georgy Kazakov

Georg Steinhauser

Thorsten Schumm

PostDoc Wanted!

Thorium Wanted!

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