Lattice Location of Th in CaF_2 Using Channeling techniques: Towards a Nuclear Clock

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Why Th-doped CaF_2?

^{229,m}Th

The extremely low-energy ^{229}Th isomer state (8.28 ± 0.17 eV) [1] can potentially be used to construct a nuclear clock [2]. Such a nuclear clock is expected to outperform current electronic-shell-based atomic clocks. The isomeric state has two possible decay channels towards the ground state: radiative decay and internal conversion (IC), the latter being dominant. Therefore, blocking the IC decay channel is critical for future high-precision measurements of the transition energy.

CaF_2

A possibility to block the IC decay channel is to have Th dopants in CaF_2 crystals. These crystals are transparent in the VUV regime (direct band gap of 11,6 eV – 12,1 eV) and can be grown to high quality. Density functional theory (DFT) calculations [3] predict that the band gap remains larger than the isomer energy, thus not allowing IC decay, if Th^{4+} dopants occupy Ca^{2+} substitutional lattice positions (see figure to the right). Therefore, these crystals can be used for VUV spectroscopy of the thorium isomer and fluorescence spectroscopy could potentially be used to construct a CaF_2:Th solid-state nuclear clock [4].

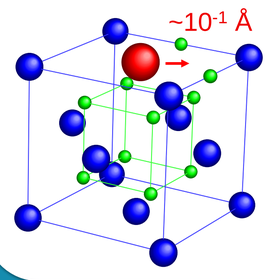
Doping approaches

- Doping **during crystal growth** up to 1% Th at TU Wien: equilibrium process. Expecting ground state configuration.
- Doping by **ion implantation** (of Th or parent isotope) at ISOLDE, CERN: out-of-equilibrium process. Superior control over dopant concentration and depth profile, but possible non-substitutional or no charge compensated configurations that allow IC.

Goal: determine Th dopant configurations in CaF_2

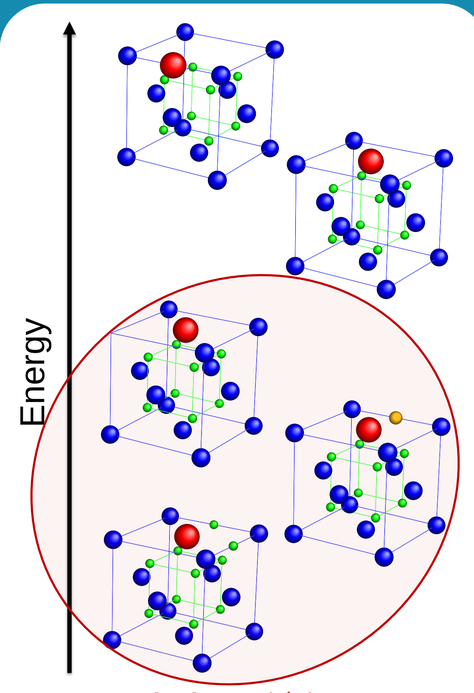
Th dopant configurations in CaF_2

The lowest-energy dopant configuration (figure below) is Th^{4+} in a Ca^{2+} substitutional lattice position accompanied by two interstitial F^- as charge compensation mechanism. Higher energy configurations (figure to the right) can have a different charge compensation mechanism (O^{2-} interstitial Ca^{2+} vacancy, …) and still preserve the band gap. However, no charge compensation, a different Th charge state or an interstitial Th configurations are not capable of bocking the IC decay channel. The goal is to determine which configurations are present in Th doped CaF_2 crystals.



Each charge compensation mechanism has a specific displacement of the Th dopant, that together with the lattice position, can be determined by channeling techniques. Therefore, providing information on the full dopant configuration.

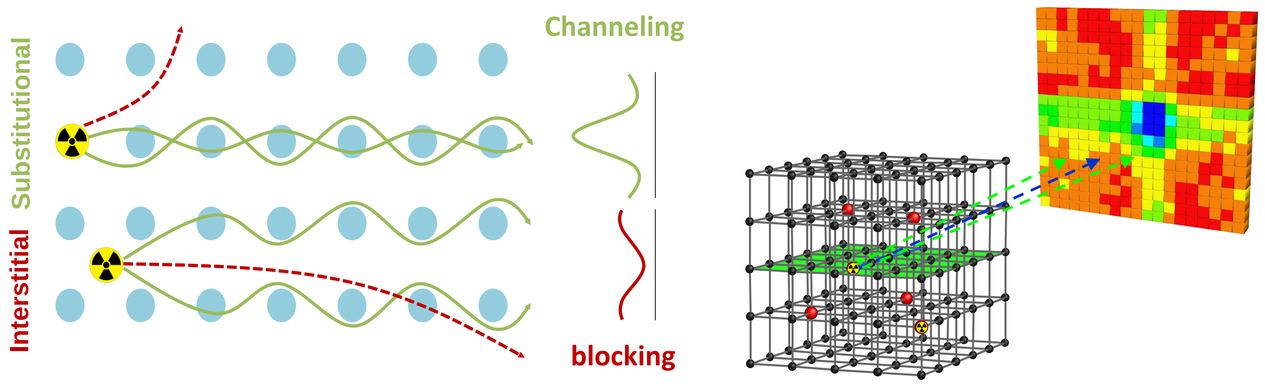
Determine lattice location: electron and ion channeling techniques



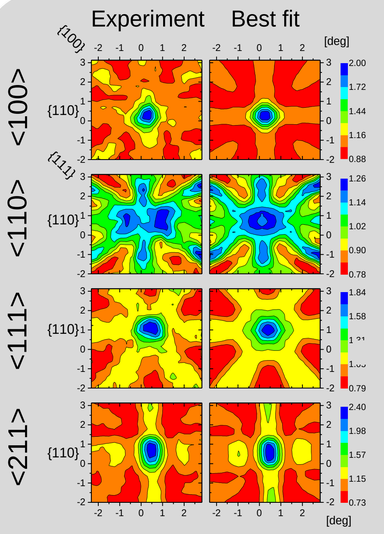
Block IC: Th^{4+} in Ca substitutional position

Channeling Techniques

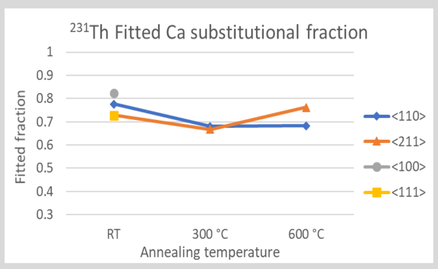
Electron emission channeling

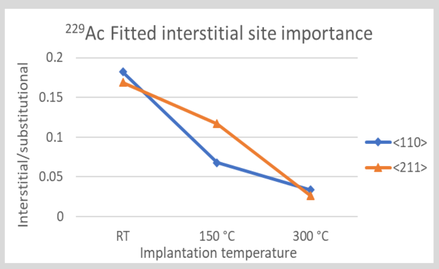


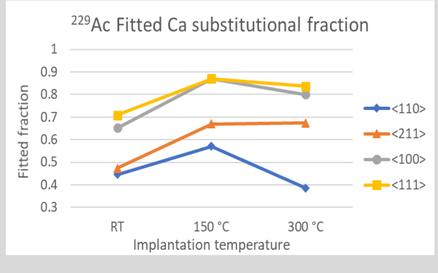
Radioactive ^{229}Ac and ^{231}Ac are mass separated and implanted into a CaF_2 single crystal at the ISOLDE facility at CERN. The emitted β^- particles upon decay follow anisotropic paths and are measured along 4 different crystal directions using a position-sensitive detector: ^{229}Ac ($T_{1/2}$ = 63 m) at the on-line EC-SLI setup [5] and the longer lived ^{231}Th ($T_{1/2}$ = 25.5 h) at the off-line setup [6]. 2D electron emission patterns are simulated for various high-symmetry lattice sites [7]. Linear combinations of these simulated patterns are fitted [8] to measured patterns, allowing to identify the occupied lattice sites and their corresponding fractions.



Experimental β^- emission channeling patterns from ^{231}Th in CaF_2 following room temperature implantation and their corresponding best single site fits.

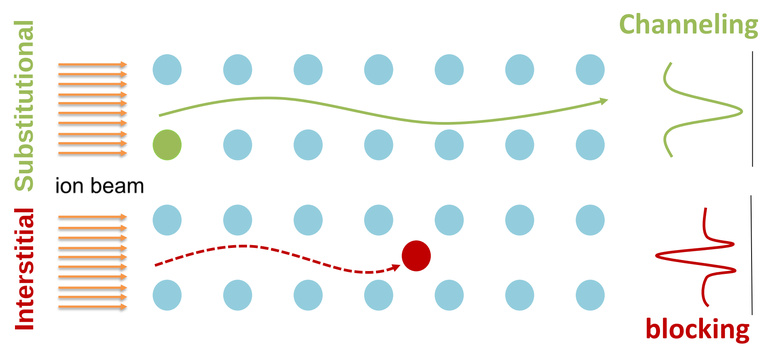




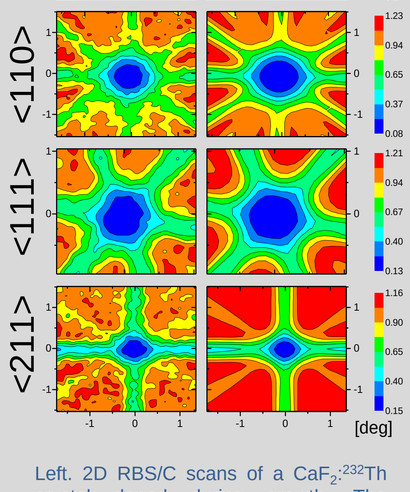


- ^{229}Ac and ^{231}Th primarily occupy Ca substitutional positions.
- ^{231}Th fractions will be corrected upward for backscattered electrons.
- Significant interstitial fraction, only distinguishable along <110> and <211> crystal directions, for ^{229}Ac parent nucleus.

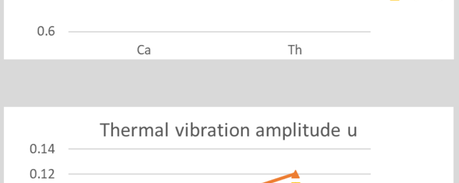
Rutherford backscattering and channeling spectrometry (RBS/C)

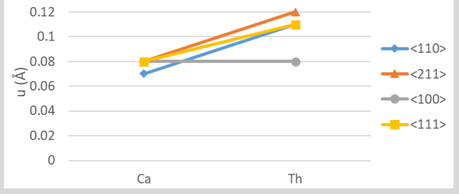


A collimated He^+ ion beam (~ MeV) from a linear accelerator impinges on the CaF_2 crystal and interacts with the periodic Coulomb potential of the lattice. Along the trajectory in the crystal, He^+ ions scatter from Ca, F and Th atoms (with a kinetic energy characteristic to Ca, F or Th) and backscattered ions are measured using a point detector. 2D patterns are measured by rotating the sample with respect to the incident beam. Like the analysis of emission channeling, 2D patterns are simulated for various high symmetry lattice sites [9] and used for fitting, thereby identifying occupied lattice sites and corresponding fractions. A major advantage is that the lattice position of Th dopants can be directly compared to the lattice position of the host Ca atoms.



Left. 2D RBS/C scans of a CaF_2:^{232}Th crystal doped during growth. The number of counts per pixel is the sum over the channels corresponding to the Th signal from the top 400 nm of the sample. Right. Corresponding best single site fits.





- ^{232}Th primarily occupies Ca substitutional positions.
- Compared to Ca host atoms: $\frac{Fr_{Th}}{Fr_{Ca}} = \mathbf{1,00 \pm 0,07}$. Th occupies host lattice position.
- Slight increase in fitted thermal vibration amplitude for Th dopants. There is Additional disorder, potentially a charge compensation mechanism.

Conclusions & Outlook

- CaF_2 crystals doped during growth: Ca substitutional Th with small additional displacement, in agreement with the ground state configuration.
- Ion implanted CaF_2 crystals: Primarily Ca substitutional Th. However, significant interstitial fraction for Ac parent nucleus.
- Compare fitted displacements to DFT calculated displacements to narrow down the number of possible charge compensation mechanisms.
- Electron paramagnetic measurements are being performed to determine the Th dopant charge state.
- Study the Th dopants' chemical environment via Extended X-ray Absorption Fine Structure (EXAFS) spectroscopy and X-ray absorption near edge structure (XANES).

References

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Acknowledgments

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