Electron Emission Channeling for lattice location of radioactive isotopes in single crystals

Improvements from a Timepix 3 quad detector and new PyFDD data analysis software

E. David-Bosne [a], A. Costa [b], B. Bergman [a], J. Correia [c], M. da Silva [d], P. Burian [a, e], U. Wahl [c]

a) Institute of Experimental and Applied Physics, Czech Technical University in Prague, Czech Republic
b) KU Leuven, Quantum Solid-State Physics, Belgium
c) Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Portugal
d) CICECO, Institute of Materials, Universidade de Aveiro, Portugal
e) Faculty of Electrical Engineering, University of West Bohemia, Czech Republic

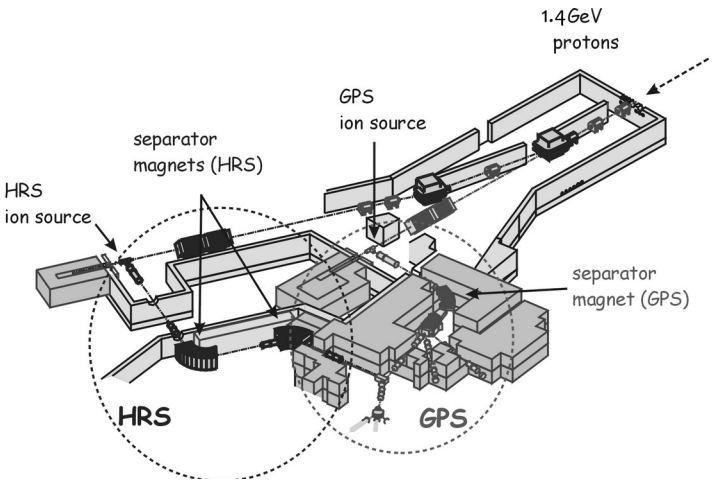
Emission channeling for lattice site location – Brief overview

- Emission Channeling (EC) is a nuclear technique for measuring the lattice site location of dopants in single crystals
- Understanding the lattice site occupied by dopants allows for better comprehension of the material's electrical, optical, and magnetic properties
- Annealing a sample at a specific temperature enables tuning of the dopant lattice site and by consequence manufacturing processes

Emission channeling for lattice site location – Brief overview

- Starts with: The implantation of radioactive isotopes in the crystal lattice
- Charged particle decay, usually beta or conversion electron, generates a source of channeling particles with origin in the lattice sites occupied by the impurity
- Usual energy range is, 3 keV to 10 MeV electrons
- A 2D channeling pattern is measured by a position sensitive detector in the vicinity of the crystal
- EC has parallels with Rutherford Backscattering Spectrometry with Channeling (RBS/C), but uses much lower dopant concentrations and is sensitive to atomic elements lighter than the host atoms

Experimental Setup – ISOLDE

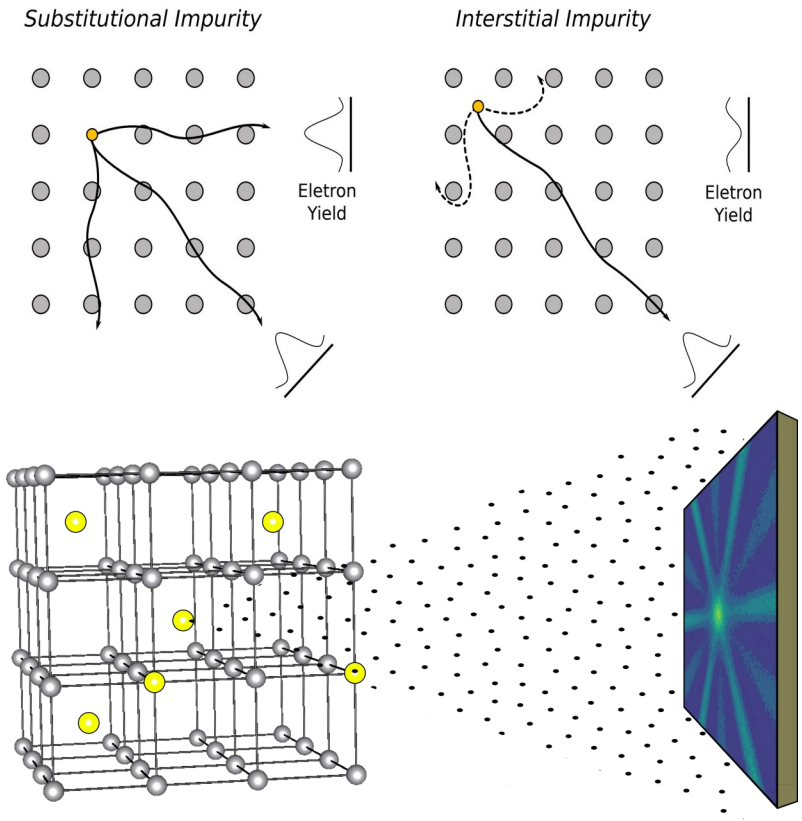


- Implantation of radioactive probes at ISOLDE (CERN):
  - Radioactive isotopes are produced by 1.4 GeV protons hitting a target with heavy elements.
  - Isotopes are ionized and accelerated. Then selected by separator magnet.
  - Isotopes travel in an experimental beam-line into an implantation chamber

Experimental Setup – Implantation

- Isotopes implanted into single crystal decay with the emission of a charged particle (beta, alpha, CE)
- Emitted particles will channel or block depending on their relative position on the lattice and traveling direction
- The channeling effect causes an anisotropic emission of charged particles around crystallographic axes, indicative of the lattice location
- 2-dimensional (2D) position sensitive detectors are used to measure the angular emission yield (channeling pattern)
- Quantitative analysis of patterns require comparison with simulations

Examples of beta channeling



Pattern analysis – Simulations

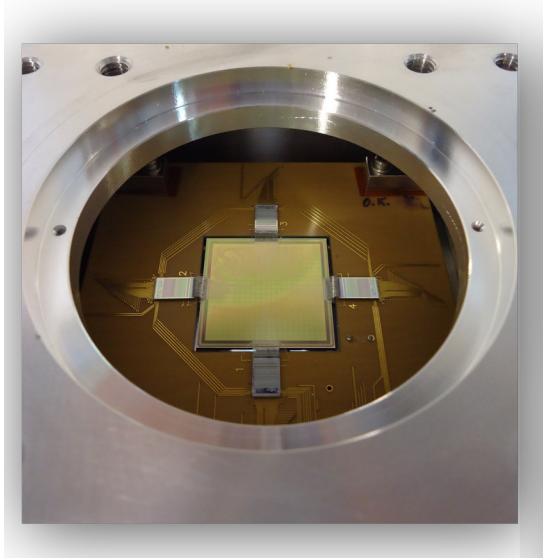
- 2D channeling patterns are simulated prior to the data analysis
- Simulations are done using the many beam approach
- For each point in the simulated pattern a new simulation is done considering:
  - The full emission spectrum of the isotope
  - The expected depth profile from the implantation
  - De-channeling effects
  - The RMS vibration of the dopant in the lattice
- This process is repeat for all major lattice position and relevant displacements
- Very computationally intensive

Detector and Data analysis

Experimental Setup – Detectors

- Characteristics:
  - Si sensor - 500 µm thick
  - 22 x 22 pixels of 1.3 mm
  - 3500 hits per second (low pile-up)
  - Energy sensitive
  - Low position resolution
  - Aging and no longer produced

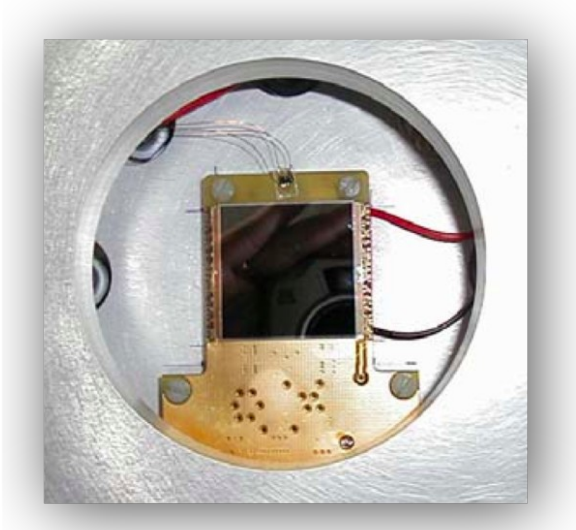
Si PAD detector 22x22



PAD detector
22x22 = 484 pads
1.3 x 1.3 mm²
< 5 kHz

Timepix Quad trial

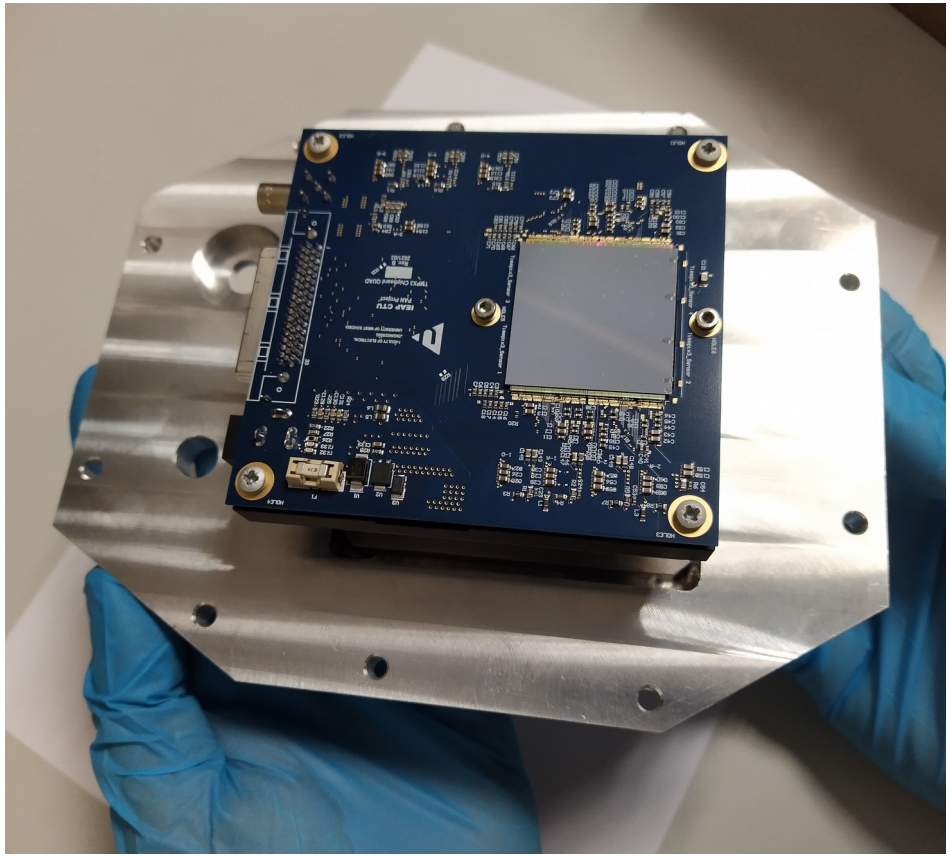
- Characteristics
  - Si sensor - 300 µm thick
  - 512 x 512 pixels of 55 µm
  - 4000 hits per second (low pile-up)
  - Frame based readout
  - ~50% dead time for 0.1s frames
- High improvement in position resolution
- Slow readout impeded full adoption for EC



Timepix-quad detector

Timepix3 quad upgrade

- Provided by IEAP in Prague to the Emission Channeling team at ISOLDE
- 512 by 512 pixels of 55um
- Fast readout up to 16M pixel hits/s
- Active development of software and readout



Timepix 3 clusters

- Particles deposit energy in several pixels of the Timepix due to their small size
- Connected pixels need to be clustered into single hit imprints
- The Cluster shape signature allows for some particle type classification

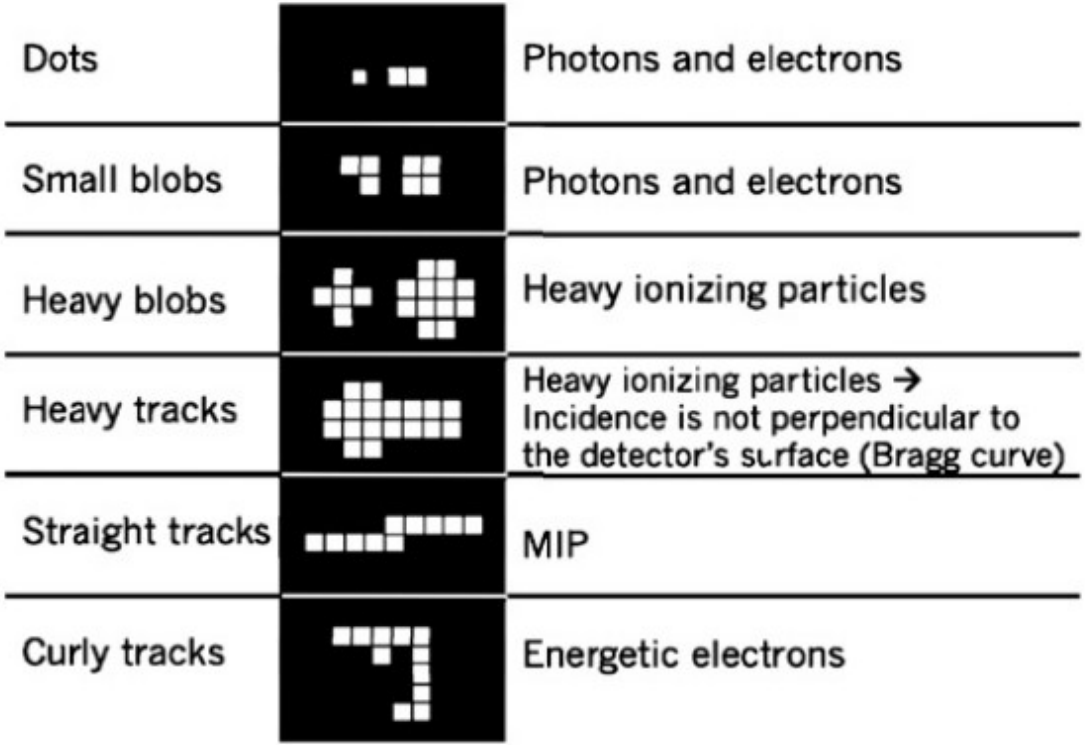


Image from: Bouchami J., et al.
Nucl. Instrum. Methods Phys. Res. A, 633 (2011), pp. S187-S189

IEAP readout and software

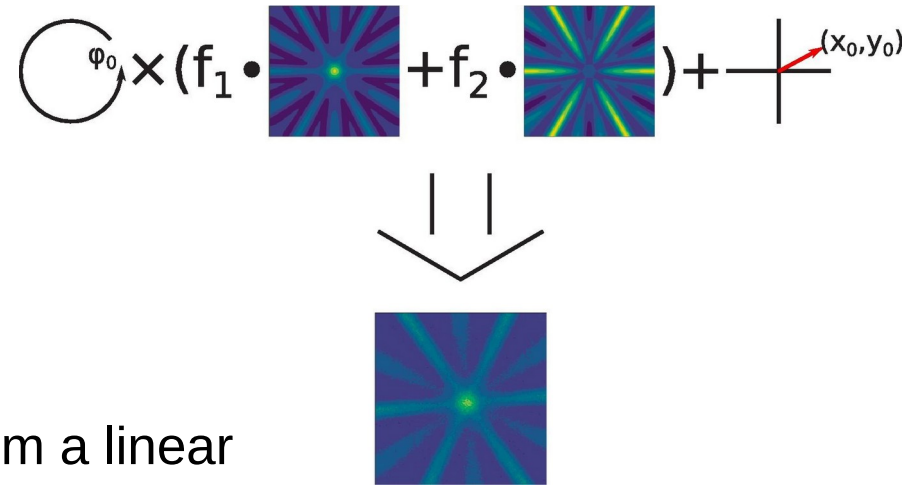
- Support from the Institute of Experimental and Applied Physics in Prague, and the West Bohemian University in Pilsen
  - Readout hardware: Katherine Gen2
    - Timepix3 Quad
    - 16MHit/s (ethernet), 40MHit/s (usb3)
    - Bias range - +1kV

      https://doi.org/10.1088/1748-0221/12/11/C11001
  - Readout control and data analysis: Track Lab
    - Acquisition and Control: pixel detectors, photomultipliers, digitizers etc.
    - High-performance analysis and live visualization. Customizable!

      https://software.utef.cvut.cz/tracklab/

Pattern analysis – Fitting

- Fitting aims at
  - Identify occupied lattice sites
  - Compute site occupation fractions
  - Estimate confidence intervals
- Fitting works by
  - Constructing theoretical patterns from a linear combination of simulated yields
  - Adjusting these patterns for rotation and translation
  - Finding the combination of sites that best represents the measurement

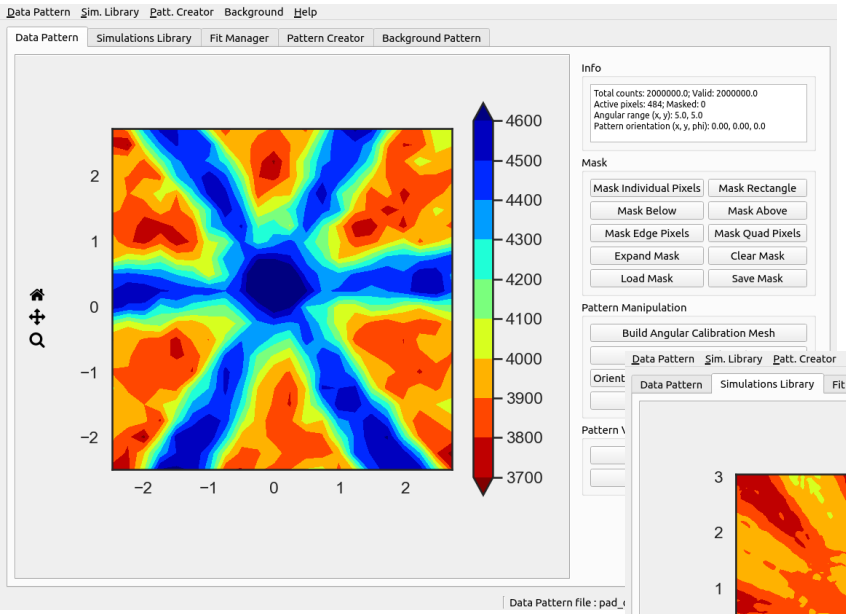


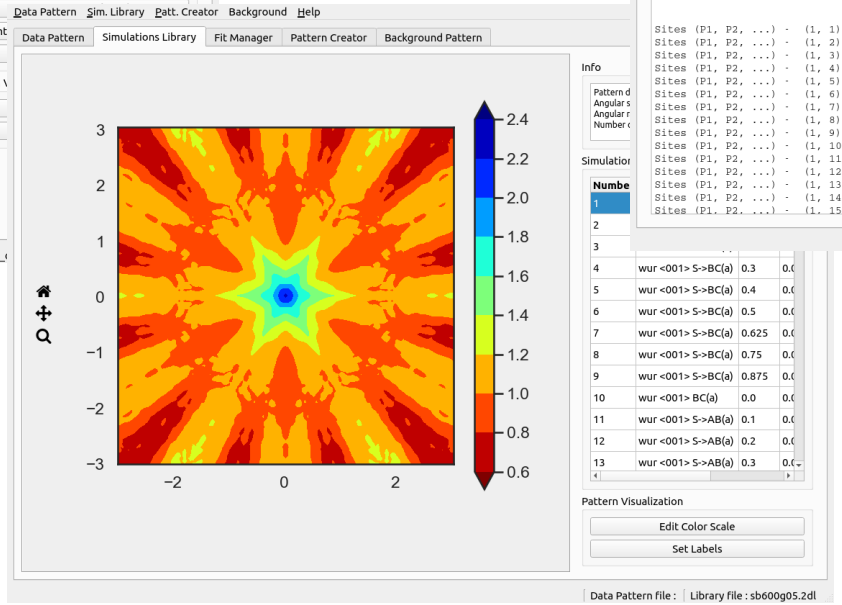
Pattern analysis – PyFDD software

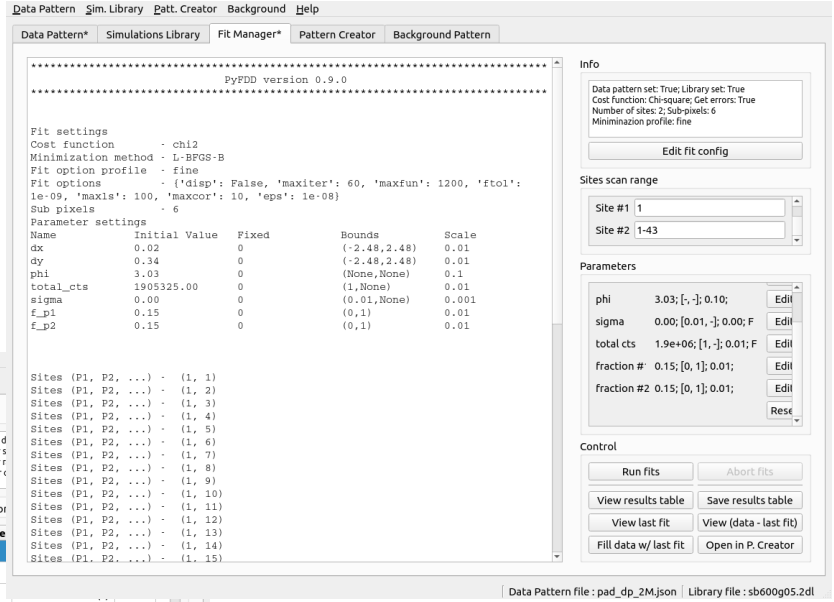
- PyFDD – Python software for Fitting 2D channeling patterns
- Started during my PhD and is still being improved
- Main features:
  - Can visualize and prepare measurements for fitting
  - Visualize previously simulated yield patterns
  - Fit patterns and provide statistical errors
  - Generate expected patterns from simulations
  - Include gamma background measurements in the fits

https://doi.org/10.1016/j.nimb.2019.10.029

PyFDD Graphical interface

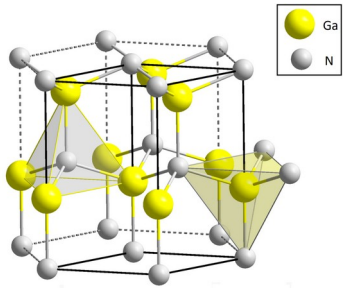




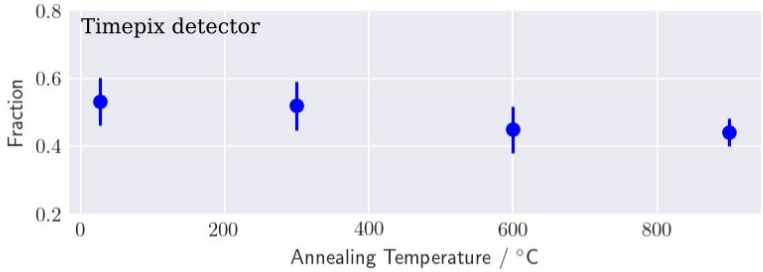


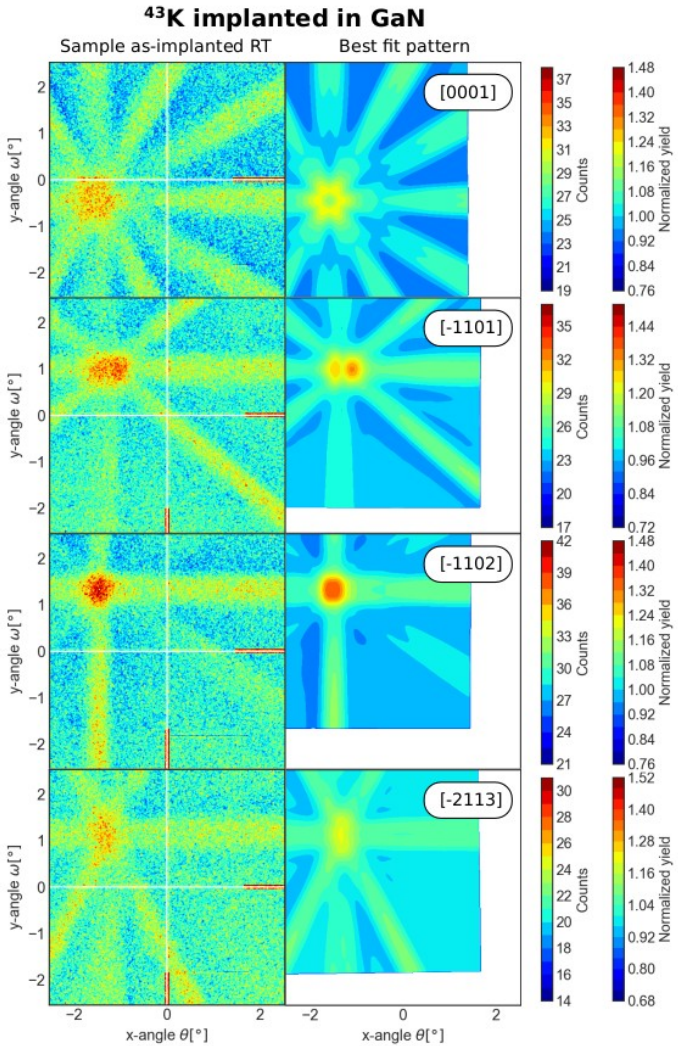
Measurement examples

Potassium in GaN



- Demonstrative measurement of four crystal directions with a Timepix Quad
- 43K, 304 keV mean energy, $t_{1/2}$ = 22.3 h
- Fits give best results for the Ga substitutional site
- Lower than 100% total fraction could be from, damage, unresolved sites





Studies of Quantum Color centers

- Sn and Mg split vacancy complexes in diamond raise interest as quantum color centers (Dopant located in the bond centered site (BC), between two C vacancies)
- ^{121}Sn, 400 keV mean energy, $t_{1/2}$ = 27.06 h
  - After implantation at RT: Sn shows occupancy distributed between the Substitutional and BC sites
  - After annealing at 920°C: Sn shows clear occupancy of both the substitutional,79±6%, and BC site 32±5 %

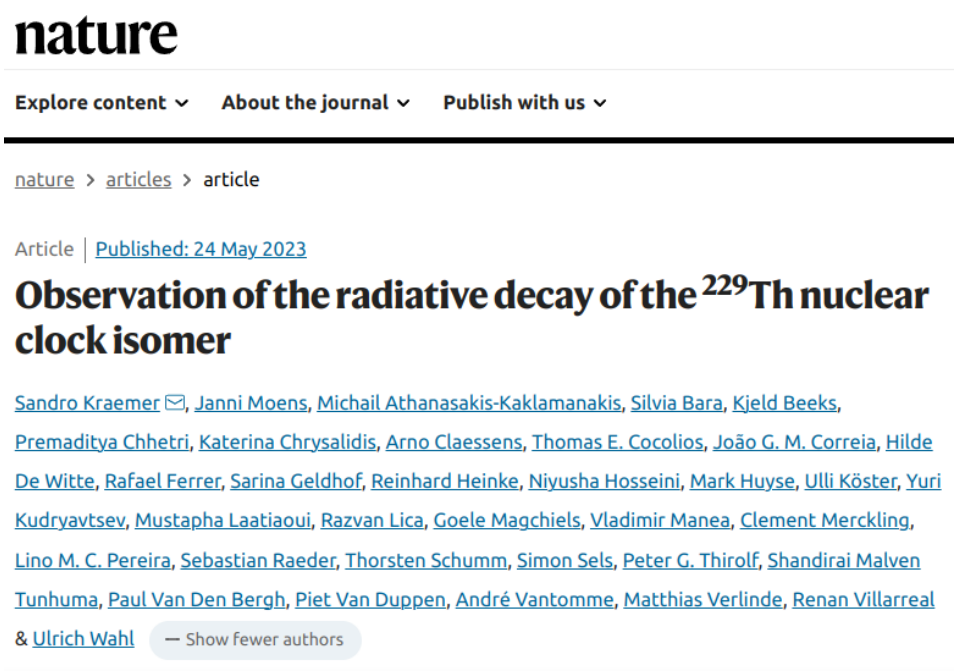
    https://doi.org/10.1103/PhysRevLett.125.045301
- ^{27}Mg, 700 keV mean energy, $t_{1/2}$ = 9.45 m
  - Implanted at RT: 42% BC
  - Implanted at 800°C: 30% BC
  - BC fractions much higher than expected from Photo Luminescence suggests that Mg centers are not optically active

    https://doi.org/10.1021/acsphotonics.2c01130

Observation of the radiative decay of the ^{229}Th nuclear clock isomer

- There is indication that Th lattice site and local defects can suppress the conversion-electron decay and thus allow observing the radiative decay of the ^{229m}Th isomer
- Measurement of the lattice site location of ^{231}Th in CaF_2, as proxy
- Best fit for substitutional calcium
- Substitutional occupancy between 77(4)% and 100%
- Lower than 100% fraction, indicates the existence of local defects

https://doi.org/10.1038/s41586-023-05894-z


nature

Explore content | About the journal | Publish with us

nature > articles > article

Article | Published: 24 May 2023

Observation of the radiative decay of the ^{229}Th nuclear clock isomer

Sandro Kraemer, Janni Moens, Michail Athanasakis-Kaklamanakis, Silvia Bara, Kjeld Beeks, Premaditya Chhetri, Katerina Chrysalidis, Arno Claessens, Thomas E. Cocolios, João G. M. Correia, Hilde De Witte, Rafael Ferrer, Sarina Geldhof, Reinhard Heinke, Niyusha Hosseini, Mark Huyse, Ulli Köster, Yuri Kudryavtsev, Mustapha Laatiaoui, Razvan Lica, Goele Magchiels, Vladimir Manea, Clement Merckling, Lino M. C. Pereira, Sebastian Raeder, Thorsten Schumm, Simon Sels, Peter G. Thirolf, Shandirai Malven Tunhuma, Paul Van Den Bergh, Piet Van Duppen, André Vantomme, Matthias Verlinde, Renan Villarreal & Ulrich Wahl — Show fewer authors

Conclusions

- Emission Channeling is a unique technique for lattice location
- IEAP developed hardware and software tools are of great value for the use of Timepix3 detectors
- PyFDD is a new tool for fitting 2D channeling patterns. Not only for Emission Channeling but also Rutherford Backscattering Spectrometry with channeling
- Tools used in the broader channeling community could help move the technique forward

Thank you!