Investigation of the lattice location of chromium in uranium dioxide single crystals with using PIXE/C analysis

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OUTLINE

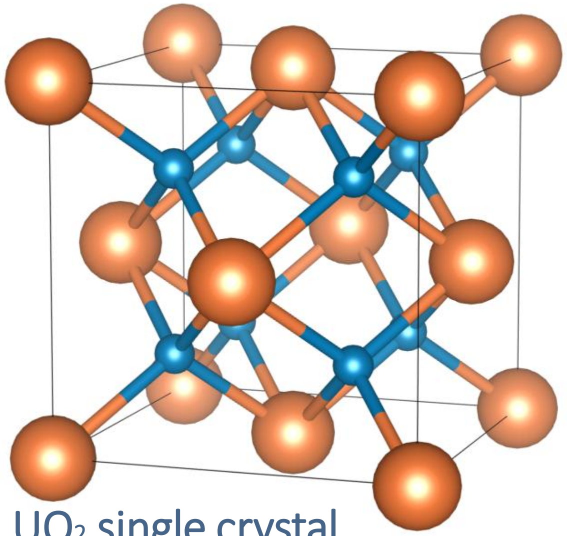
1. Introduction
   - Cr-doped UO2 as an leading Accident Tolerant Fuel (ATF)
2. Experimental and simulation methods
   - Experimental condition
   - Particle-induced X-ray emission in channeling mode (PIXE/C)
   - Wavelet Transform for background removal
   - Peak analysis
   - The PIXEK code
3. Results
   - Illustration of background-removal processing for a Cr-doped UO_2 spectrum
   - Peak analysis of the processed spectrum
   - Angular scan measurements
4. Conclusions

Cr-doped UO_2 as an leading Accident Tolerant Fuel (ATF)

Key Advantages of Cr-doped UO_2:

- **Enhanced Grain Growth** – larger grains improve fission gas retention.
- **Reduced Internal Pressure** – lower risk of cladding failure during accidents (e.g., LOCA).
- **Superior Oxidation Resistance** – better performance in high-temperature steam environments.
- **Increasing safety margins and efficiency in nuclear reactors.**

Cr lattice location in Cr-dopped UO_2 is required to understand the structural evolution of irradiated nuclear fuel in atomic scale. The role played by Cr and its location in the UO_2 fluorite-type structure remain yet open questions. Ion channeling methods coupled to IBA techniques are invaluable tools for lattice site determination.



UO_2 single crystal

Delafoy C. et al., *'Benefits of Framatome's E-ATF evolutionary solution: Cr-coated cladding with Cr_2O_3-doped fuel'*, TopFuel2018,
Milena-Pérez A. et al., *'Oxidation resistance of high-burnup Cr-doped UO2 accident tolerant fuel and comparison with irradiated UO_2'*, Journal of Nuclear Materials, 2025,
Massih, Ali & Jernkvist, Lars. (2021). *Effects of additives on UO2 fuel behavior: expanded edition*.

Characterisation of UO_2 and Cr-doped UO_2 single crystals

- The experiment was performed with using UO_2 and Cr-doped UO_2 single crystals.
- Cr-doping was performed by ion implantation:

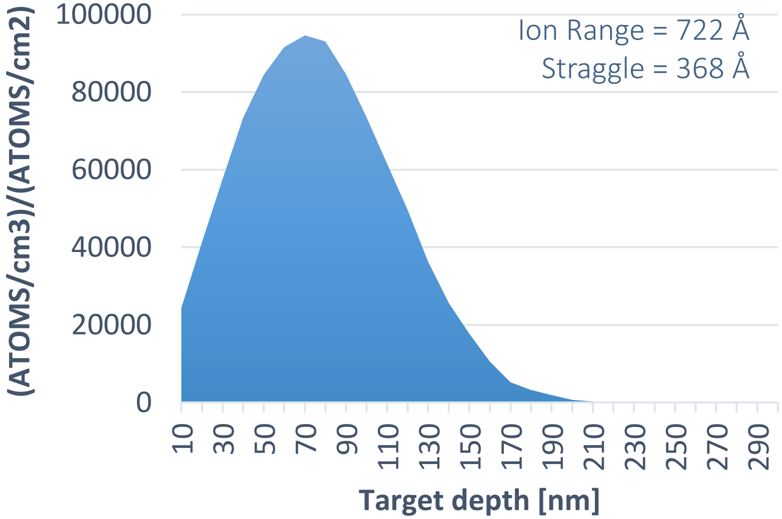
$170\ \mathrm{keV}\ \mathrm{Cr}^{+}$ ions with fluence $1 \cdot 10^{16}\ \mathrm{cm}^{-2}$ (detection test),

$1.5 \cdot 10^{15}\ \mathrm{cm}^{-2}$ (corresponding to 0.16 wt% Cr at max)

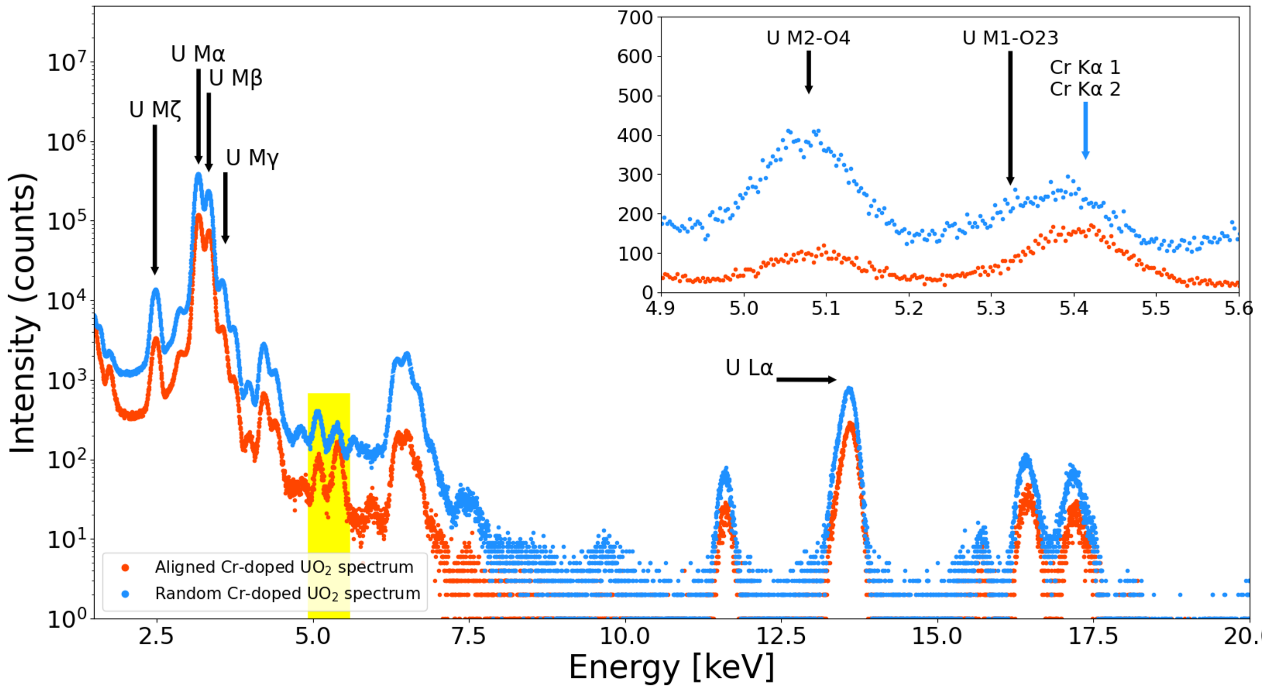
- RBS/C and PIXE/C:

$3.07\ \mathrm{MeV}\ \mathrm{He}^{2+}$ ions — to sense both U and O sublattices through $^{16}O(^{4}He,\ ^{4}He)^{16}O$ elastic nuclear reaction

ION RANGES calculated by SRIM



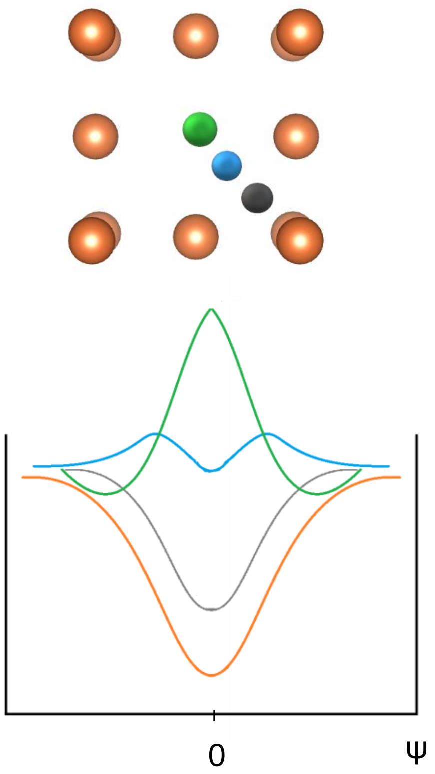
Particle-induced X-ray emission in channeling mode (PIXE/C)



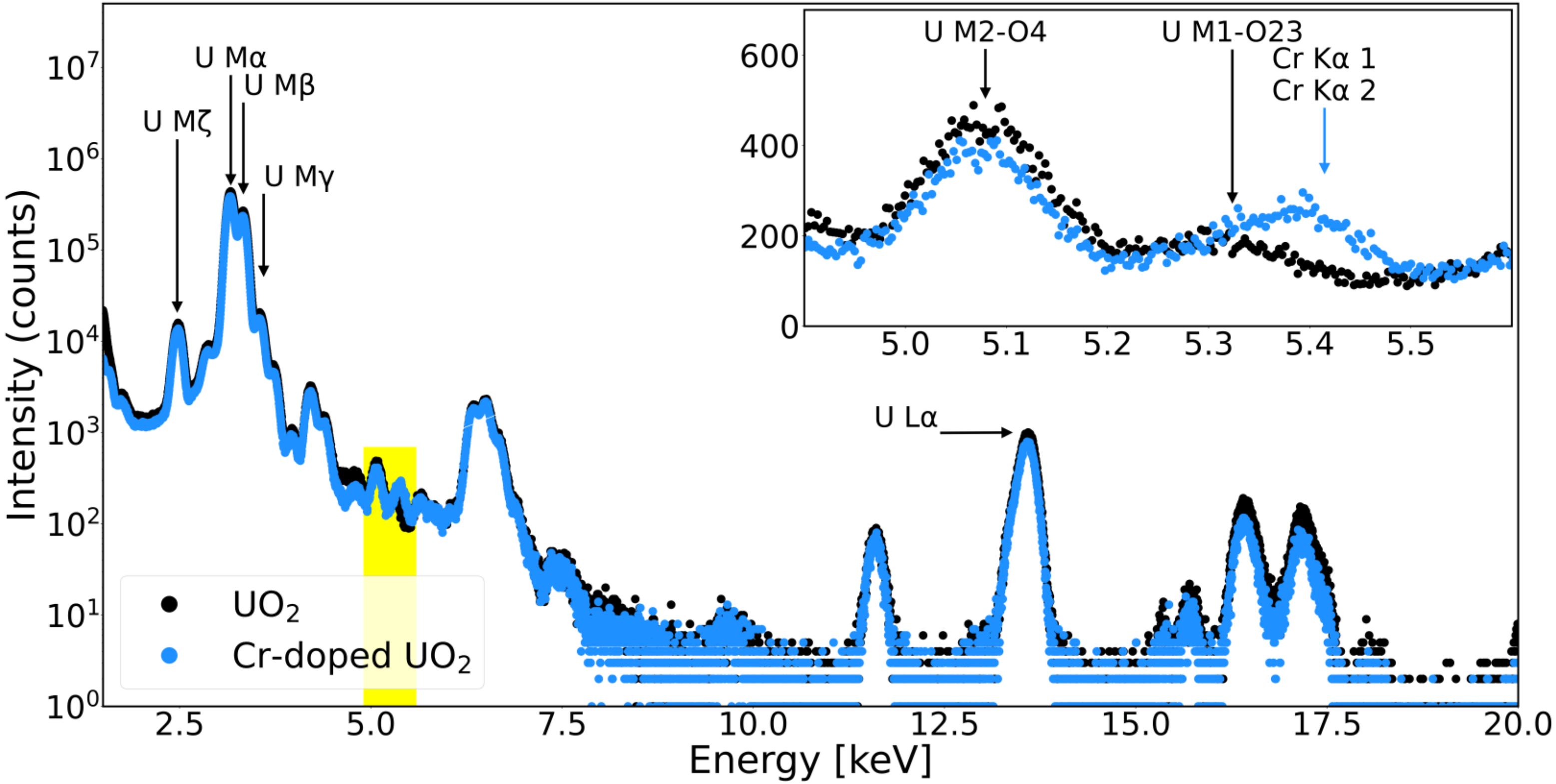
PIXE spectra recorded on Cr-doped UO_2 single crystals using 3.075 MeV He as probing ions. Spectra were collected for the [110]-oriented sample aligned along the [011] crystallographic direction (orange dots) and in random orientation (blue dots). Highest intensity peaks are labeled: U Mζ, Mα, U Mβ, U Mγ and U Lα. The region where U M2-O4, U M1-O23 and Cr Kα peaks are located is marked with a yellow rectangle and a zoom is shown in the inset located at the top right of the plot. Note that the main plot is presented in logarithmic scale, while the inset is in linear scale.

Triangulation method

shape of angular scan recorded on impurity according to its lattice location



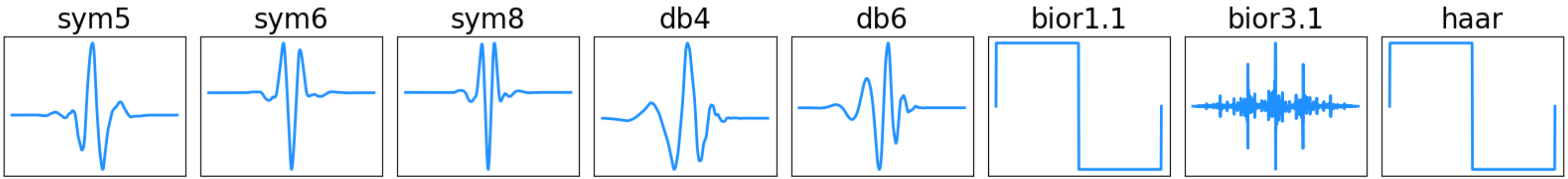
Particle-induced X-ray emission in channeling mode (PIXE/C)



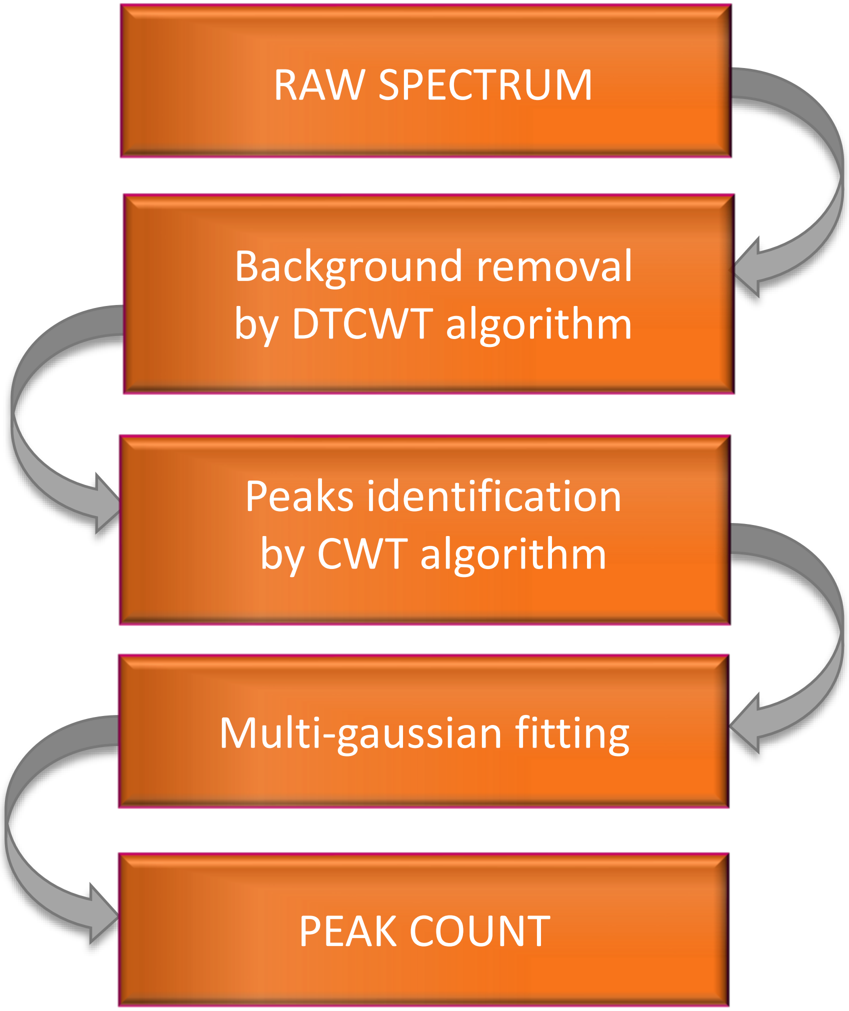
PIXE spectra recorded on UO2 (black dots) and Cr-doped UO2 (blue dots) single crystals using 3.075 MeV He as probing ions. Spectra are collected in random direction. Highest intensity peaks are labeled: U Mζ, Mα, U Mβ, U Mγ and U Lα. The region where U M2-O4, U M1-O23 and Cr Kα peaks are located is marked with a yellow rectangle and a zoom is shown in the inset located at the top right of the plot. Note that the main plot is presented in logarithmic scale, while the inset is in linear scale.

Wavelet Transform (WT):

- A mathematical method used to analyze signals at **different scales (frequencies) and positions (time).**
- It decomposes a signal into **small wave-like functions called wavelets**.
- Similar to the Fourier Transform, which decomposes a signal into sinusoids of different frequencies.
- Wavelet transform provides **both time and frequency localization**.
- Used to detect **local features**, analyze **temporal changes**, and **remove background and noise**.
- Examples of wavelets: **Symlets** (sym5, sym6, sym8), **Daubechies** (db4, db6), **Biorthogonal** (bio1.1, bior3.1), and **Haar**.



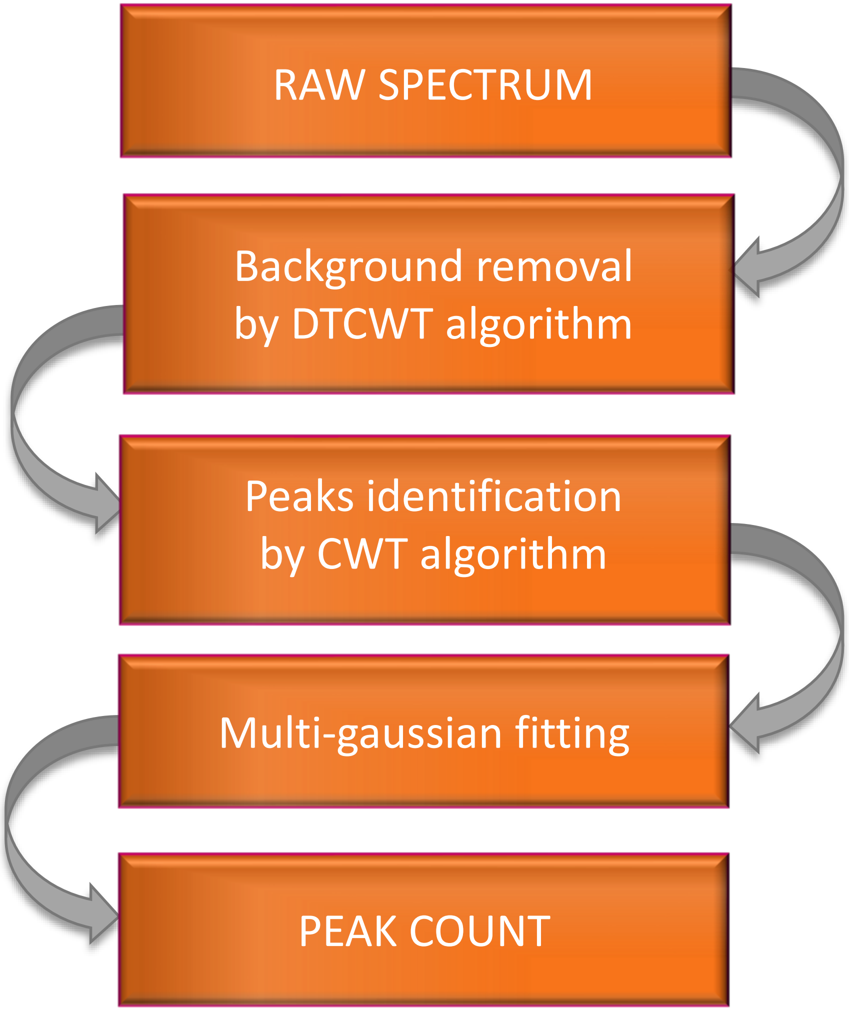
Wavelet Transform for background removal & peak analysis



Background removal by Dual-Tree Complex Wavelet Transform (DTCWT) algorithm:

- DTCWT is an advanced extension of the **Discrete Wavelet Transform**.
- It uses **two parallel wavelet filter banks (dual trees)** to generate **complex-valued coefficients (real and imaginary parts)**.
- The **number of decomposition levels (DL)** controls the scale separation and the ability to capture background features without distorting local peaks.
- The transform **divides a signal into frequency bands**, with low levels capturing high-frequency details, high levels capturing low-frequency background components, and the approximation providing a smoothed, noise-free baseline.
- DTCWT is used to reconstruct the **low-frequency component of the signal**, which represents the **background** and is then subtracted from the original signal.

Wavelet Transform for background removal & peak analysis

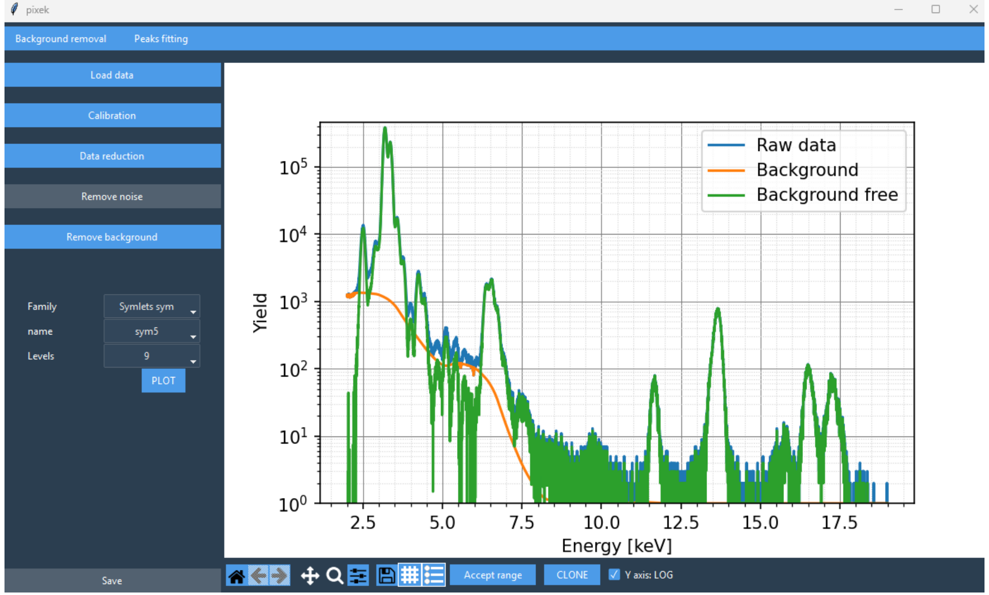


Peak identification by Continuous Wavelet Transform (CWT) algorithm:

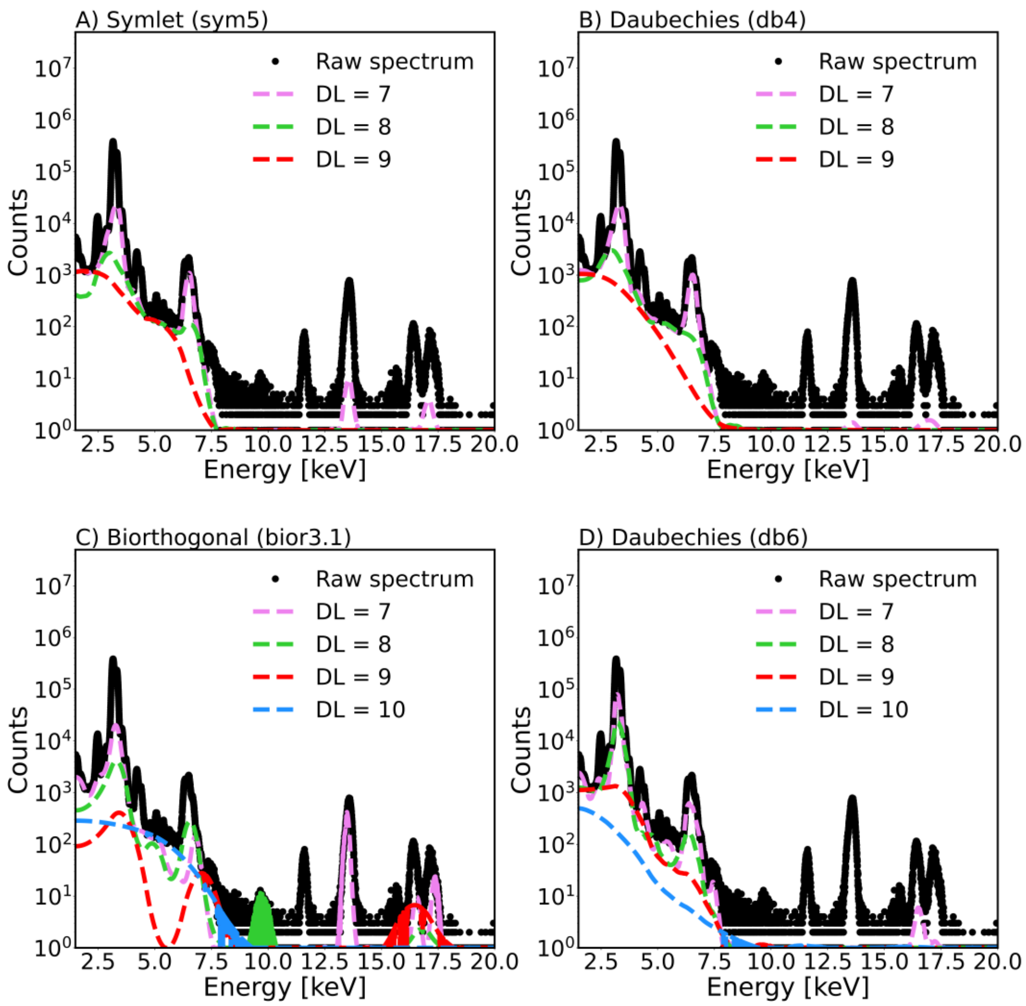
- CWT analyzes the spectrum at **multiple scales**.
- It uses **continuous scaling and shifting of a mother wavelet** to examine the signal’s time–frequency content.
- The transform **enhances local maxima** corresponding to spectral peaks.
- Peaks are identified by detecting **significant wavelet coefficients** in the wavelet transform.
- The **method improves accuracy and reliability of peak detection** in the spectrum.

The **PIXEK code** developed for PIXE/C analysis

- **Python-based code**
- **DTCWT – Dual-Tree Complex Wavelet Transform** was implemented for background removal, while **CWT - Continuous wavelet transform** was used for peaks identifitation.
- For each spectrum the code provides the intensity of the relevant signal peaks.
- Multiple spectra can be analyzed in the same time.
- The final result is angular scan for the selected energy spectrum.
- Important feature ability to operate efficiently with minimum user intervention.



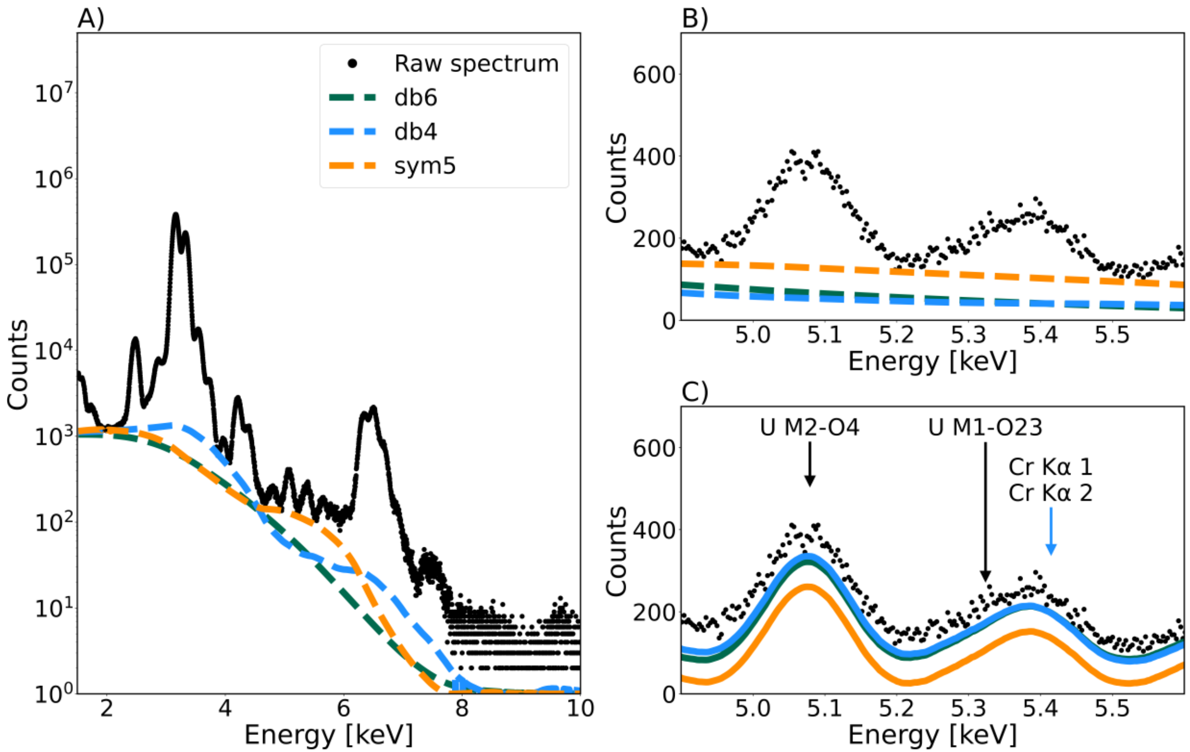
Wavelet Transform for background removal



Background removal on 3.075 MeV He PIXE spectra recorded on a Cr-doped UO2 crystal analyzed with different wavelet families: Biorthogonale (bior3.1), Symlet (sym5) and Daubechies (db4, db6) and decomposition levels (from 7 to 9 or 10). Maximum level that can be used for each wavelet depends on the signal length and wavelet filter length, for db4 and sym5 maximum level is 9, while for bior3.1 and db6 it is possible to use level 10.

E. Kucal, F. Garrido, K. Skrobas, C. Mieszczyński, C. Bachelet, J. Bourçois, F. Pallier, *Wavelet-based analysis for PIXE method—Study of Cr-doped UO2 spectrum,* Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2026, doi:10.1016/j.nimb.2026.166087

Wavelet Transform for background removal



A) Application of DTCWT algorithms to the background removal on PIXE spectrum recorded on Cr-doped UO2 using 3.075 MeV He ions. The background has been approximated with Daubechies ’db4’, ’db6’ wavelets (green and blue dashed lines, respectively) and Symlet ’sym5’ wavelet (orange dashed line). B)Zoom of the plot A limited to energies ranging between 4.9 keV to 5.6 keV and plots of various background estimation. C) Spectra after background subtraction using ’db4’, ’db6’ and ’sym5’ wavelets (green, blue and orange lines, respectively) with labels U M2-O4, U M1-O23 and Cr Kα of PIXE peaks.

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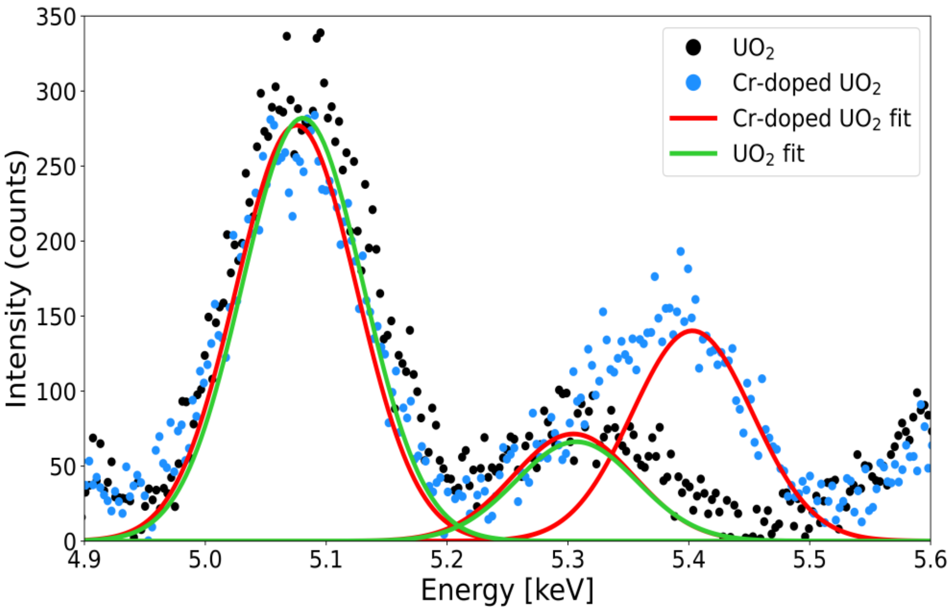
Peak analysis

Peak areas recorded on U M2-O4, U M1-O23 and Cr Kα signals with the calculated limits of quantification LOQ for Cr Kα

0	Peak	Raw	'db6'	'db4'	'sym5'
Cr-doped UO_2	U M2-O4	16677	13845	14438	11362
UO_2	U M2-O4	17385	13522	13910	11456
Cr-doped UO_2	U M1-O23	11021	5916	6240	2928
UO_2	U M1-O23	7231	4653	4383	2690
Cr-doped UO_2	**Cr Kα**	8774	9157	9089	5746
Cr Kα - LOQ			1179	1190	1259

	Raw	'db6'	'db4'	'sym5'
R_{UO_2}	0.416	0.344	0.315	0.235
+/-	0.005	0.013	0.013	0.016
$R_{Cr\text{-}UO_2}$	0.526	0.427	0.432	0.258
+/-	0.008	0.013	0.013	0.016
δ [%]	26	24	37	10

Calculated ratios of U M1-O23 to U M2-O4 peaks area for UO2 (RUO2) and Cr-doped UO2 (RCr−UO2) and percentage difference δ of this two ratios.



Decomposition of PIXE signals recorded on UO2 and Cr-doped UO2 crystals after background removal using WF: 'sym5'. The components peaks of the UO2 spectrum are plotted by green lines, while the peaks that were parts of fitted Cr-doped UO2 spectrum are red lines.

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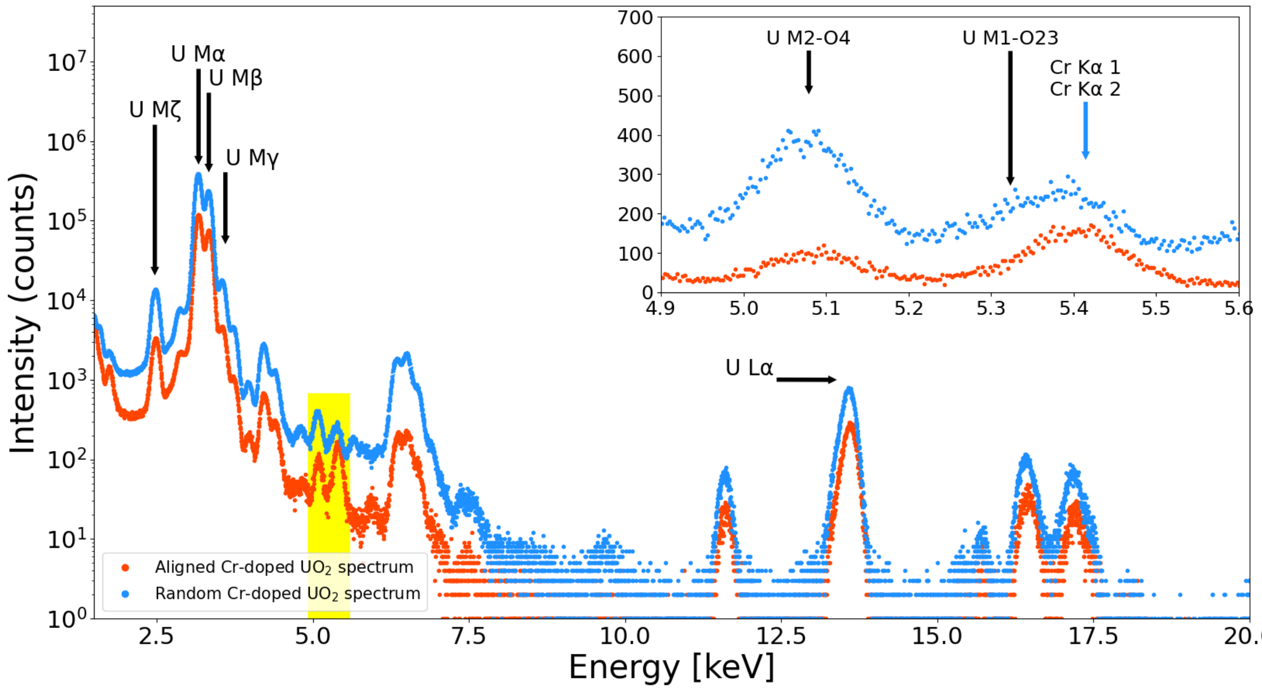
Ion channeling methods for lattice site determination

Particle-induced X-ray emission in channeling mode (PIXE/C)



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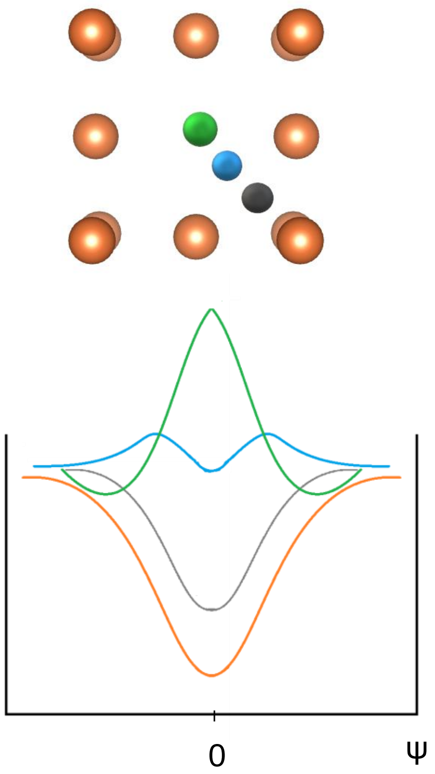
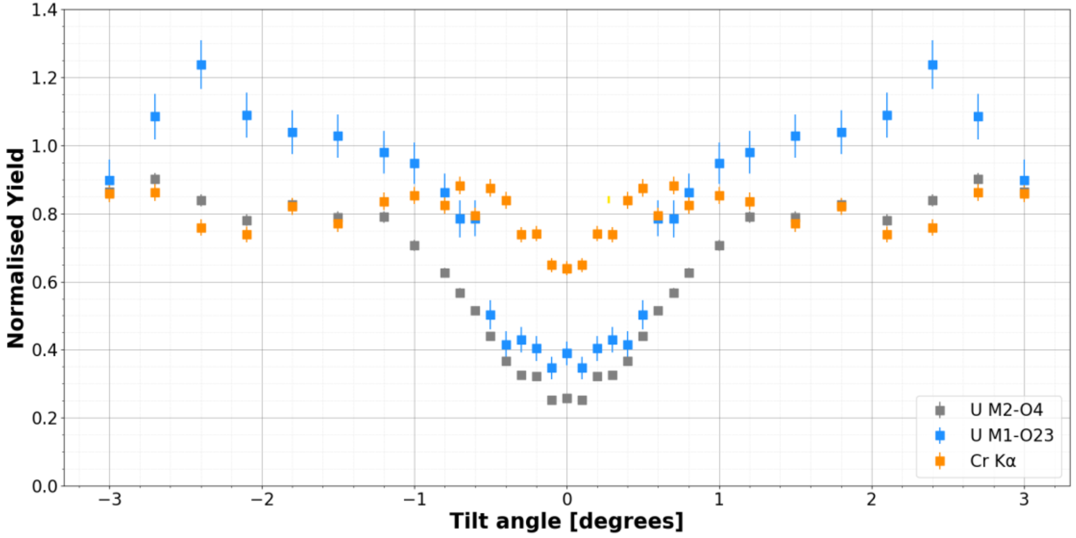
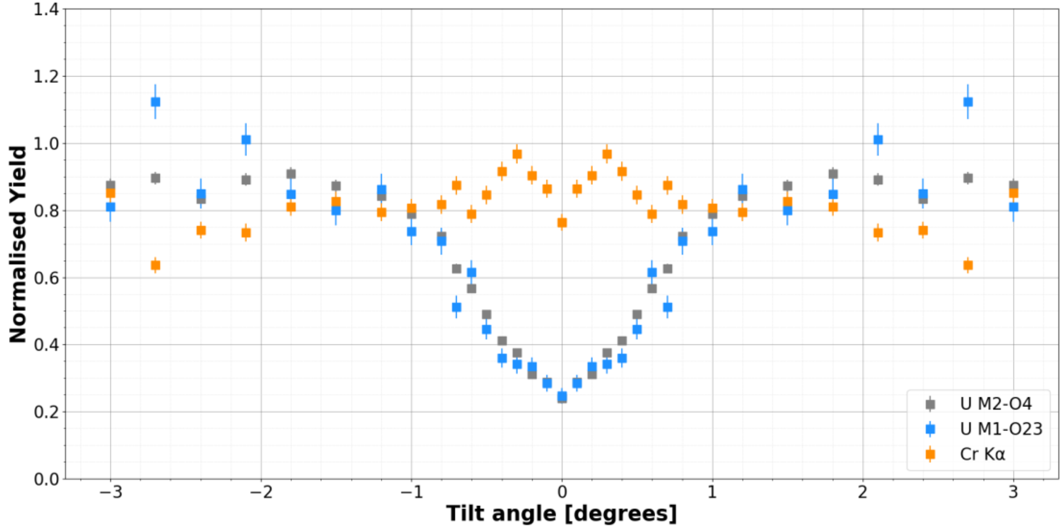


Angular scans – Cr- doped UO_2

Scan across [110] along (011)



Scan across [110] along (001)



Calculated ratios of U M1-O23 to U M2-O4

		0°	0.1°	0.2°	0.3°	0.4°	0.5°	0.6°	0.7°	0.8°	1.0°	1.2°	1.5°	1.8°	2.1°	2.4°	2.7°	3.0°
110-011	**R**	0.417	0.380	0.346	0.366	0.315	0.316	0.422	0.383	0.381	0.371	0.344	0.361	0.348	0.386	0.408	0.333	0.288
	+/-	8.1 %	8.7 %	8.0 %	7.6 %	7.9 %	7.0 %	5.2 %	5.2 %	4.8 %	4.6 %	4.5 %	4.3 %	4.4 %	4.2 %	3.9 %	4.2 %	4.8 %
110-001	**R**	0.377	0.363	0.395	0.334	0.321	0.335	0.398	0.301	0.360	0.343	0.377	0.337	0.343	0.417	0.375	0.461	0.341
	+/-	9.6 %	8.8 %	7.9 %	7.9 %	7.5 %	6.4 %	5.1 %	5.9 %	4.7 %	4.6 %	4.2 %	4.4 %	4.2 %	3.7 %	4.2 %	3.5 %	4.4 %

SUMMARY

- PIXE/C techniques were used to investigate the lattice location of Cr in UO_2.
- For the PIXE/C analysis, special attention was given to background reduction.
- A dedicated code (the PIXEK code) was developed based on wavelet analysis.
- The angular scans were recorded on the various main crystallographic directions, along major planes.
- The precise lattice location of Cr is currently being investigated by performing MC simulations by assuming various possible locations of Cr in the UO_2 cell and comparing the expected Cr signal with the experimental one.

Merci pour votre attention!

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