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Gérald Jomard, I. Cheik Njifon, Ms. Talla-Noutack, M. Freyss, M. Bertolus, et al.. Coupled first-principles and experimental positron annihilation study of defects in actinide mixed oxides. AC-TINIDES 2017, Jul 2017, Sendai, Japan. ACTINIDES 2017, 2017. hal-02417746

#### HAL Id: hal-02417746 https://hal.science/hal-02417746

Submitted on 18 Dec 2019

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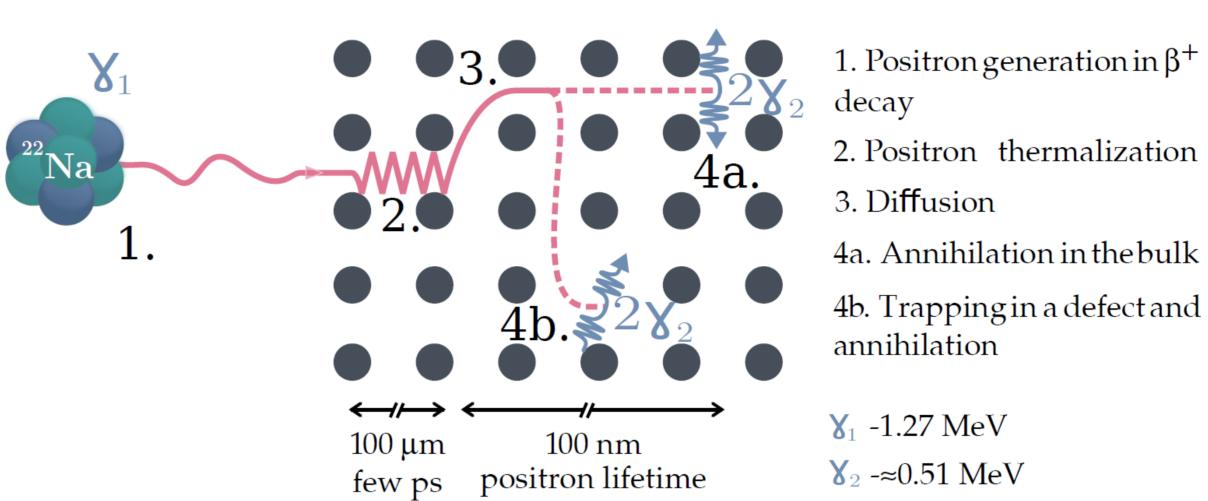
# Coupled first-principles and experimental positron annihilation study of defects in actinide mixed oxides

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## Context

- > Positron Annihilation Spectroscopy (PAS) is a powerful technique to detect vacancy-type defects in materials
- We demonstrated its capability to identify irradiation-induced defects in UO<sub>2</sub> by coupling experiments and ab initio calculations [1]
- MOX fuels (UPuO<sub>2</sub>) will be the fuel of Gen IV reactors  $\triangleright$
- MOX fuels fabricated from spent fuels  $\rightarrow$  contains few % of minor actinide  $\succ$ elements like Am
- Under irradiation, defects are created in the fuel and have strong impact  $\triangleright$





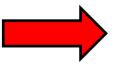


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## Scheme of the main positron-solid interactions

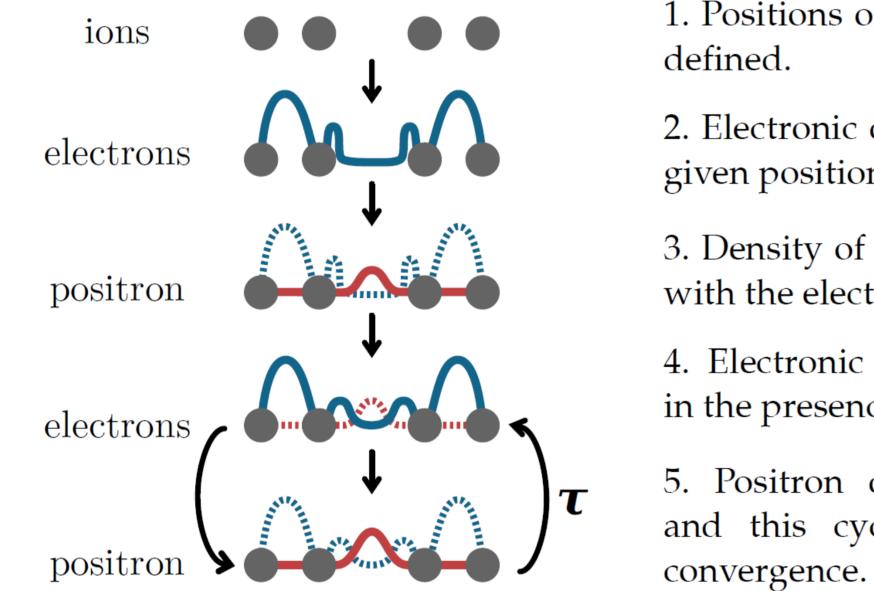


Strong interest in characterizing vacancy defects created in actinides mixed oxides

# **Methods**

Positron lifetimes are computed using electronic structure calculations in the two component density functional theory (TCDFT) [2].

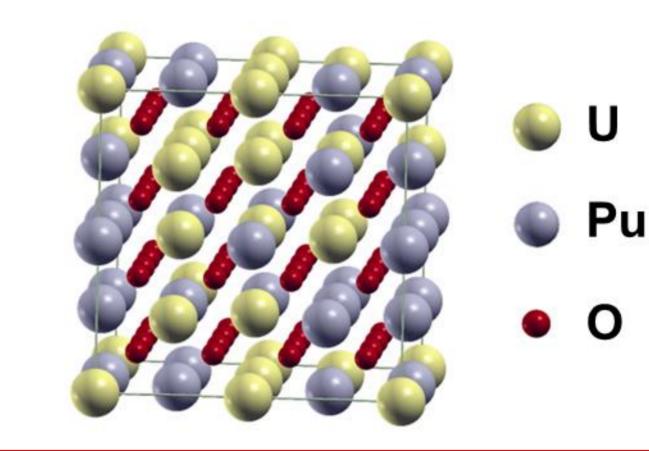
Self-consistent calculations are performed to obtain the electron and positron densities.



- 1. Positions of ions in the system are
- 2. Electronic density is calculated for given positions of ions.
- 3. Density of the positron interacting with the electrons is calculated.
- 4. Electronic density is recalculated in the presence of the positron.
- 5. Positron density is recalculated and this cycle is continued until

In our study the relaxation effect due to the vacancy creation and the positron localized in the defect is taken into account.

perfect crystal	unrelaxed vacancy	5	cy relaxed vacancy relaxed wards with a positron	
Calculations are performed with the ABINIT code [3] using 96 atom supercells. The DFT+ <i>U</i> [4] method is used to take into account the strong correlations between the 5 <i>f</i> electrons.				



Mixed oxides supercell are

**Positron lifetime calculation scheme** 

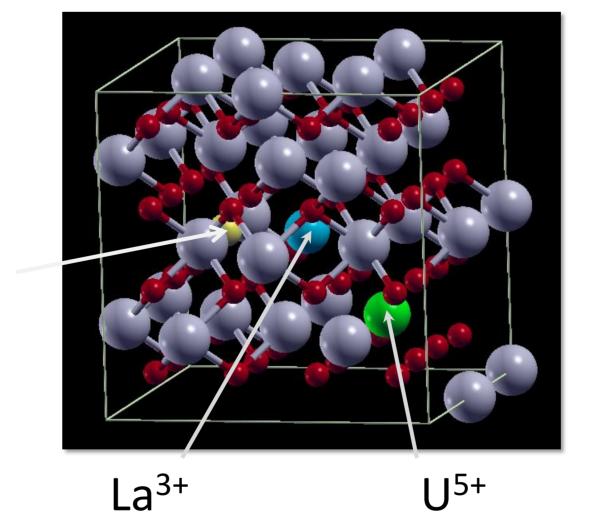
simulated using the SQS method.

**Preliminary results : unexpected localization of positron in bulk actinide oxides** 4 compounds investigated : UCeO<sub>2</sub>, UPuO<sub>2</sub>, UAmO<sub>2</sub> and ULaO<sub>2</sub>

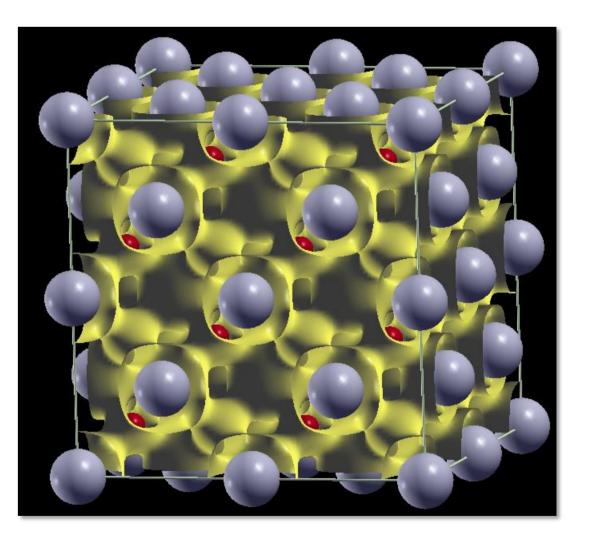
For pure tri-valent elements, bulk lifetime within the GGGC-GGA scheme is much higher than expected.

compounds	τ <sub>GGGC-GGA</sub> (ps)	τ <sub>conv-gga</sub> (ps)
ULaO <sub>2</sub> 3,125 at%	199	168
ULaO <sub>2</sub> 6,25 at%	201	168
ULaO <sub>2</sub> 12,5 at%	202	168
ULaO <sub>2</sub> 25 at%	206	168
ULaO <sub>2</sub> 50 at%	211	168
U <b>Am</b> O <sub>2</sub> 6,25 at%	200	168
UCeO <sub>2</sub> 25 at%	168	168
UPuO <sub>2</sub> 6,25 at%	168	168
$UO_2 \tau_{exp} = 169 \pm 1ps$	168	168

localized positron



In ULaO<sub>2</sub> and UAmO<sub>2</sub>, positron localized in the vicinity of La<sup>3+</sup> and Am<sup>3+</sup> respectively.



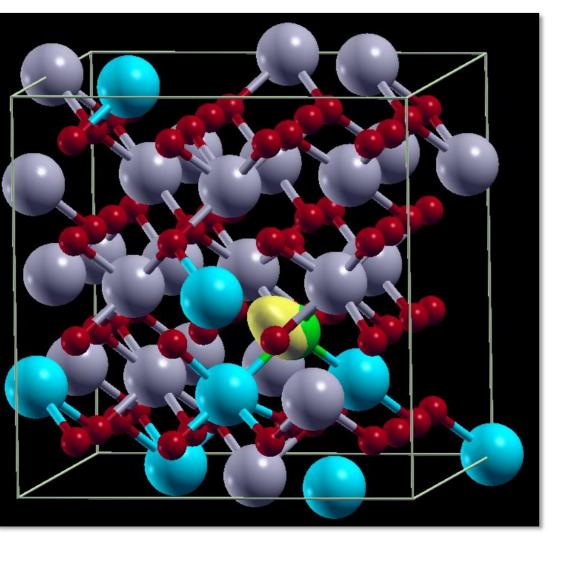
Delocalized positron in bulk  $UO_2$ , UCeO<sub>2</sub> and UPuO<sub>2</sub>.

## $UO_2 \tau_{exp} = 169 \pm 1ps$

## For oxygen mono-vacancy :

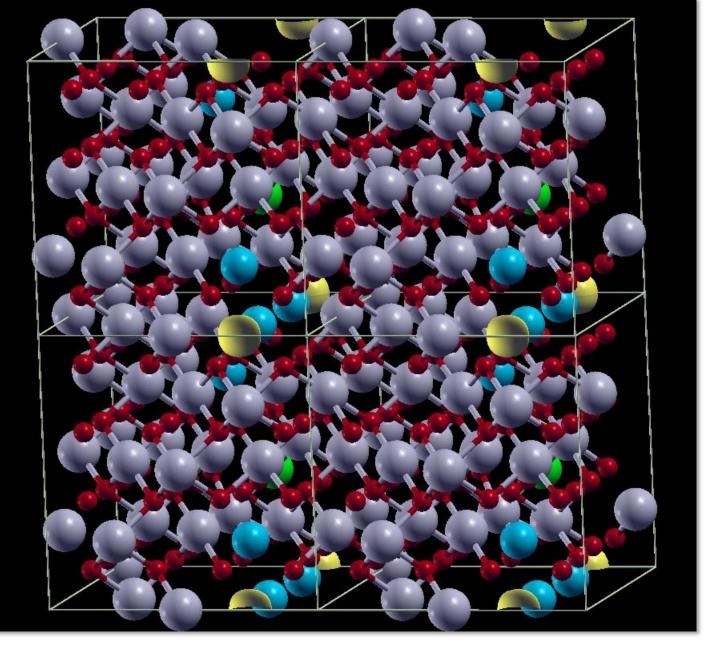
Positron localization in the vacancy for UCeO<sub>2</sub> and UPuO<sub>2</sub>

 $\tau_{V_O^{-UCeO_2}} = 227 ps$ 



Same positron localization as in the bulk for ULaO<sub>2</sub> and UAmO<sub>2</sub>

 $\tau_{V_O^{-ULaO_2}} = 202 ps$ 



Experimental work in progress on UCeO<sub>2</sub> and ULaO<sub>2</sub> at CNRS/CEMHTI : DFT predictions soon confirmed?

<u>**References:**</u> [1] J. Wiktor, G. Jomard, M. Torrent et al., Phys. Rev. B 90, 184101 (2014) [3] X. Gonze et al., CPC 205 (2016) 106

[2] R. M. Nieminen, E. Boronski and L. J. Lantto, Phys. Rev. B 32, 1377 (1985) [4] B. Dorado, B. Amadon, M. Freyss and M. Bertolus, Phys. Rev. B 79, 235125 (2009)