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

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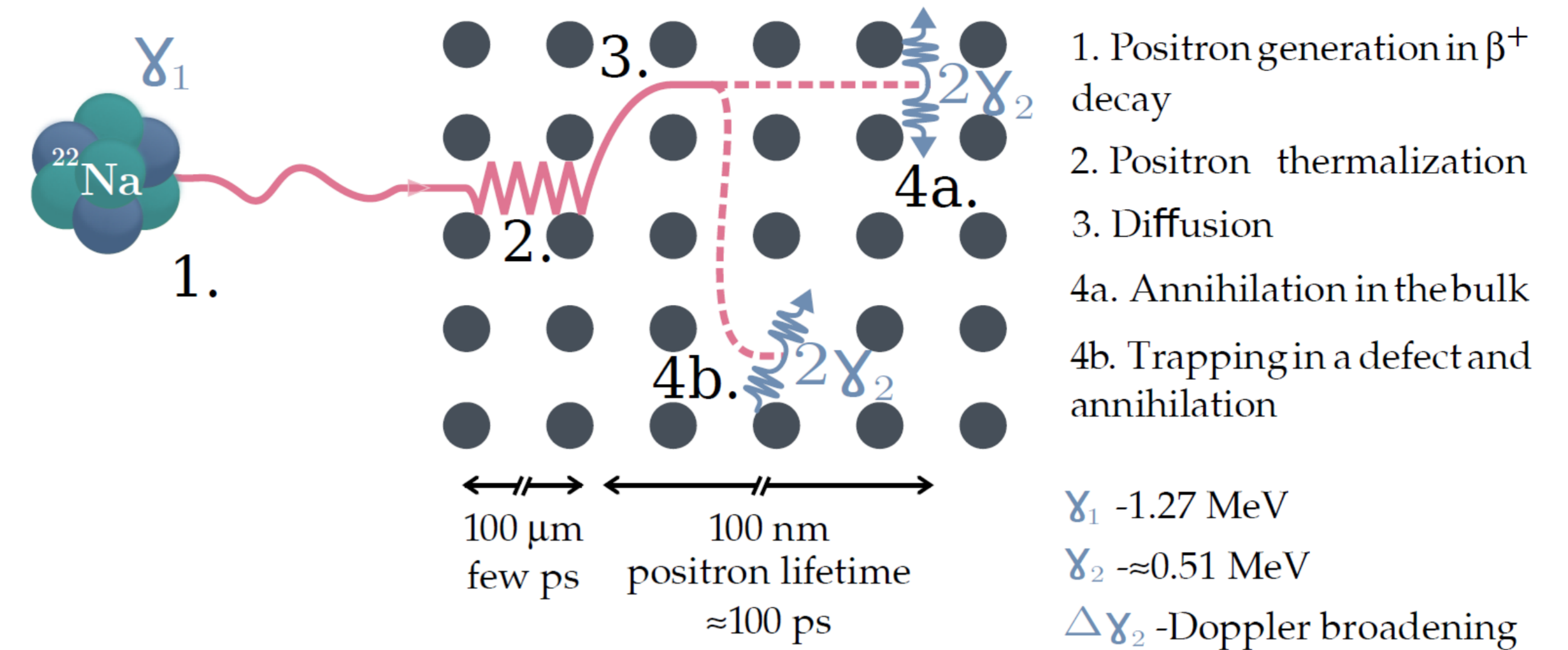
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# Coupled first-principles and experimental positron annihilation study of defects in actinide mixed oxides

## 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  $\text{UO}_2$  by coupling experiments and ab initio calculations [1]
- MOX fuels ( $\text{UPuO}_2$ ) will be the fuel of Gen IV reactors
- MOX fuels fabricated from spent fuels  $\rightarrow$  contains few % of minor actinide elements like Am
- Under irradiation, defects are created in the fuel and have strong impact on behaviour of fission products and evolution of microstructure

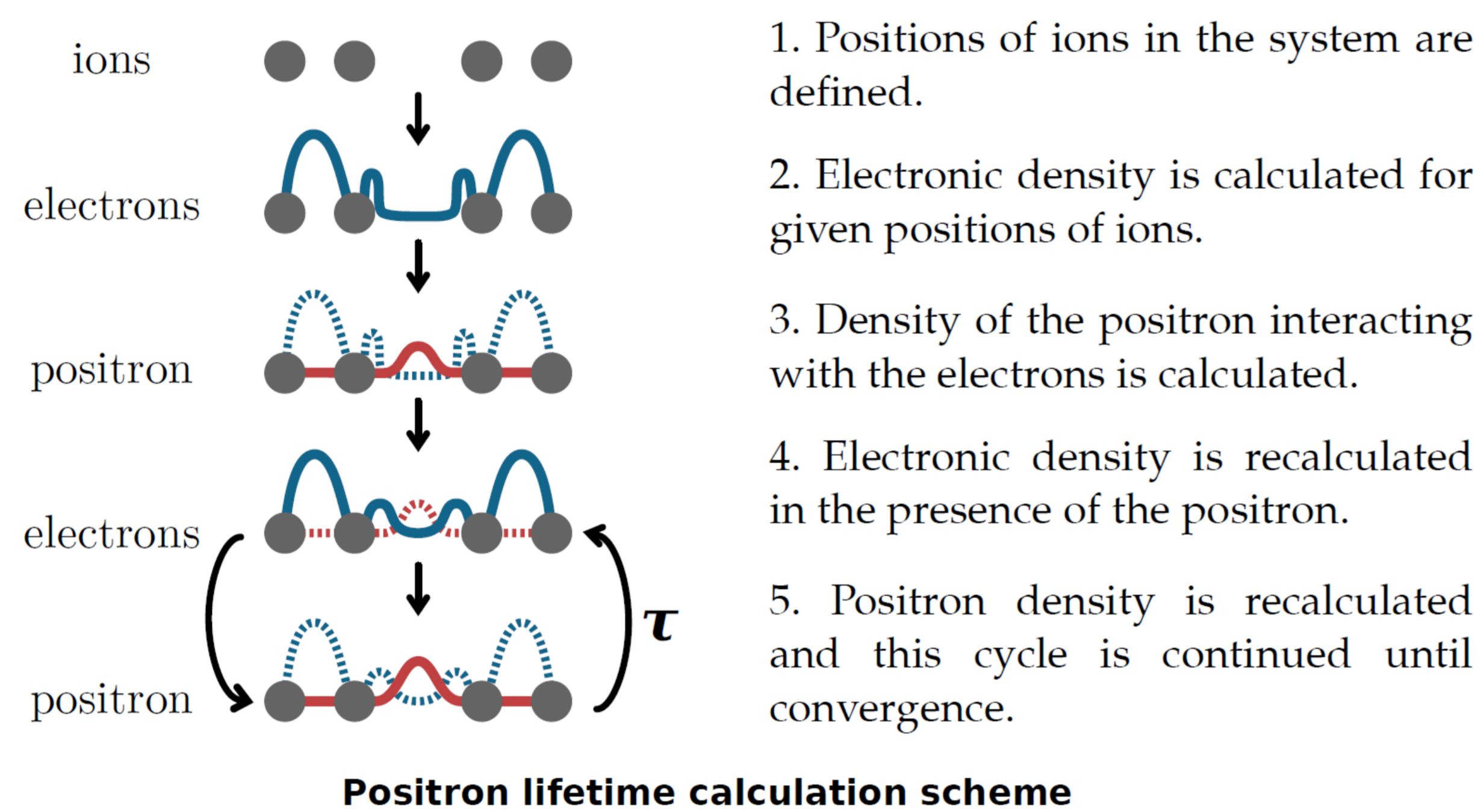


Scheme of the main positron-solid interactions

➔ Strong interest in characterizing vacancy defects created in actinides mixed oxides

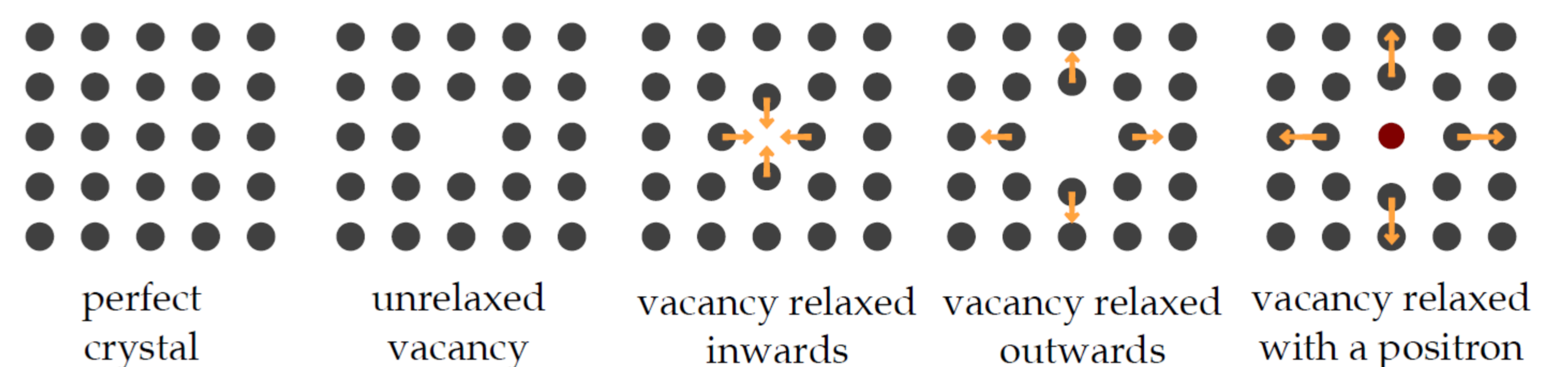
## Methods

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.

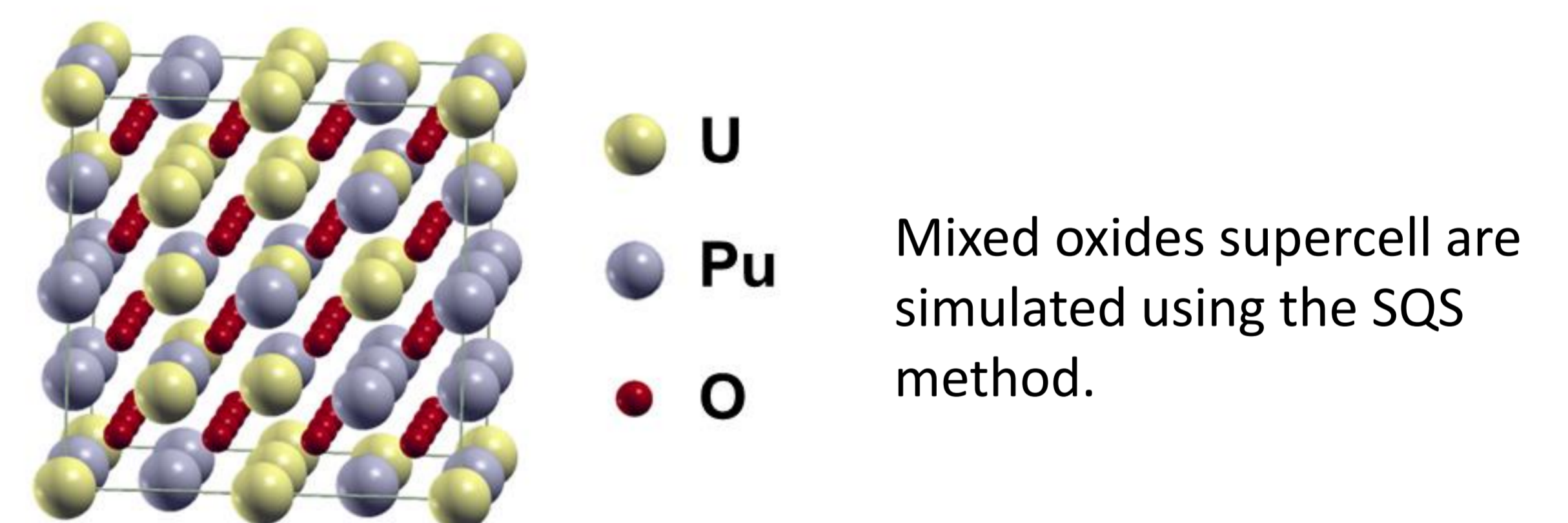


Positron lifetime calculation scheme

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



Calculations are performed with the ABINIT code [3] using 96 atom supercells. The DFT+U [4] method is used to take into account the strong correlations between the 5f electrons.

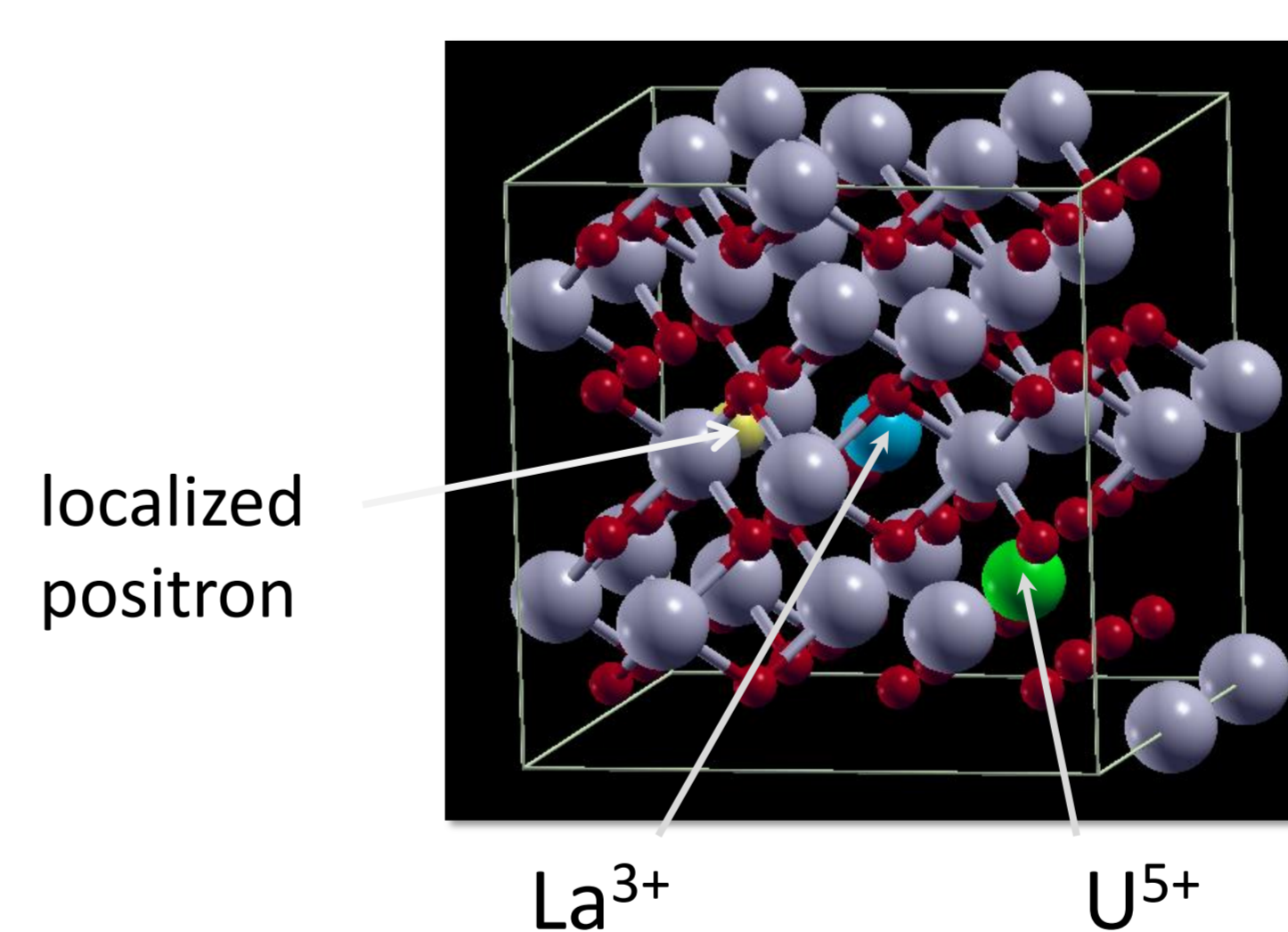


## Preliminary results : unexpected localization of positron in bulk actinide oxides

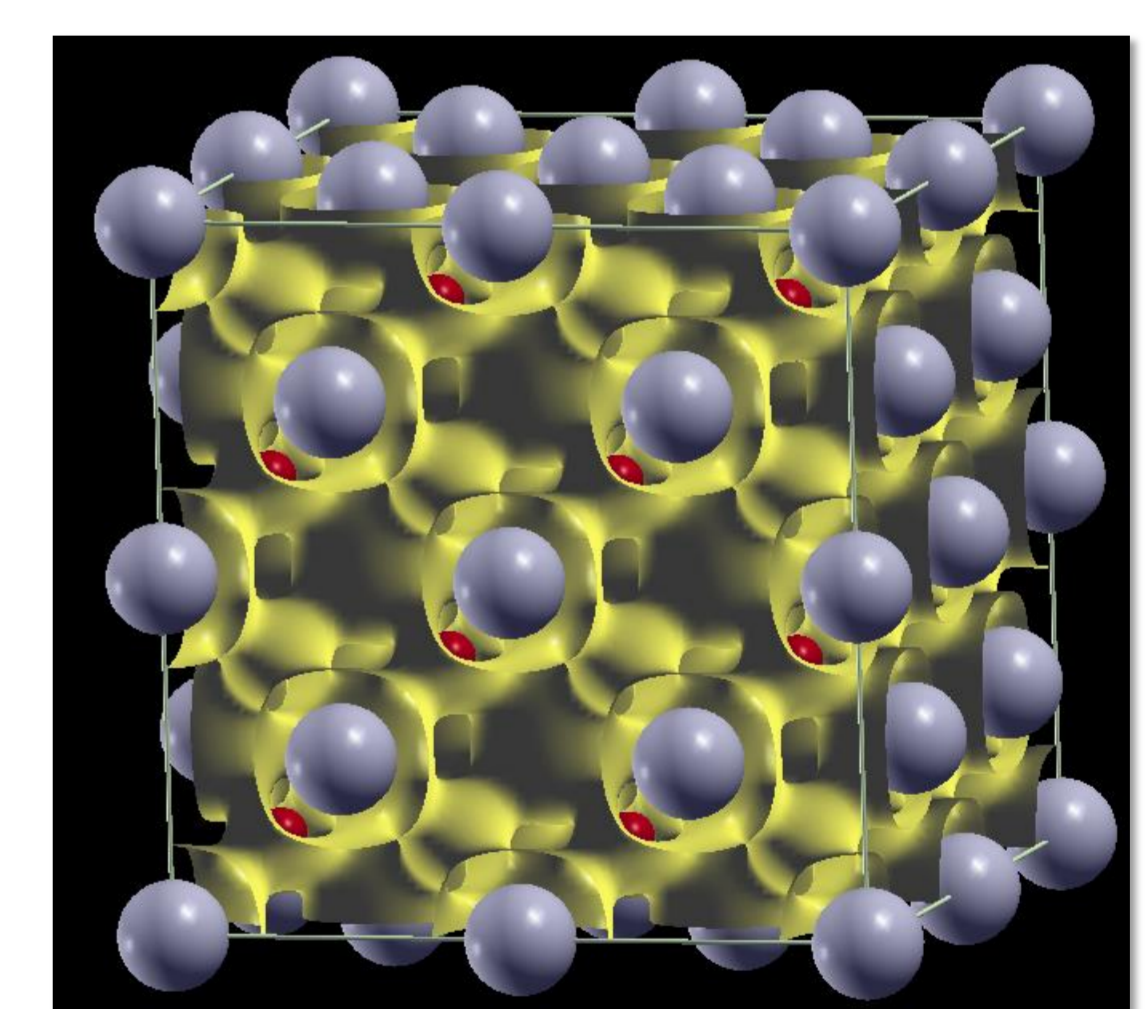
4 compounds investigated :  $\text{UCeO}_2$ ,  $\text{UPuO}_2$ ,  $\text{UAmO}_2$  and  $\text{ULaO}_2$

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

compounds	$\tau_{\text{GGGC-GGA}}$ (ps)	$\tau_{\text{CONV-GGA}}$ (ps)
$\text{ULaO}_2$ 3,125 at%	199	168
$\text{ULaO}_2$ 6,25 at%	201	168
$\text{ULaO}_2$ 12,5 at%	202	168
$\text{ULaO}_2$ 25 at%	206	168
$\text{ULaO}_2$ 50 at%	211	168
$\text{UAmO}_2$ 6,25 at%	200	168
$\text{UCeO}_2$ 25 at%	168	168
$\text{UPuO}_2$ 6,25 at%	168	168
$\text{UO}_2$ $\tau_{\text{exp}}=169\pm 1\text{ps}$	168	168



In  $\text{ULaO}_2$  and  $\text{UAmO}_2$ , positron localized in the vicinity of  $\text{La}^{3+}$  and  $\text{Am}^{3+}$  respectively.

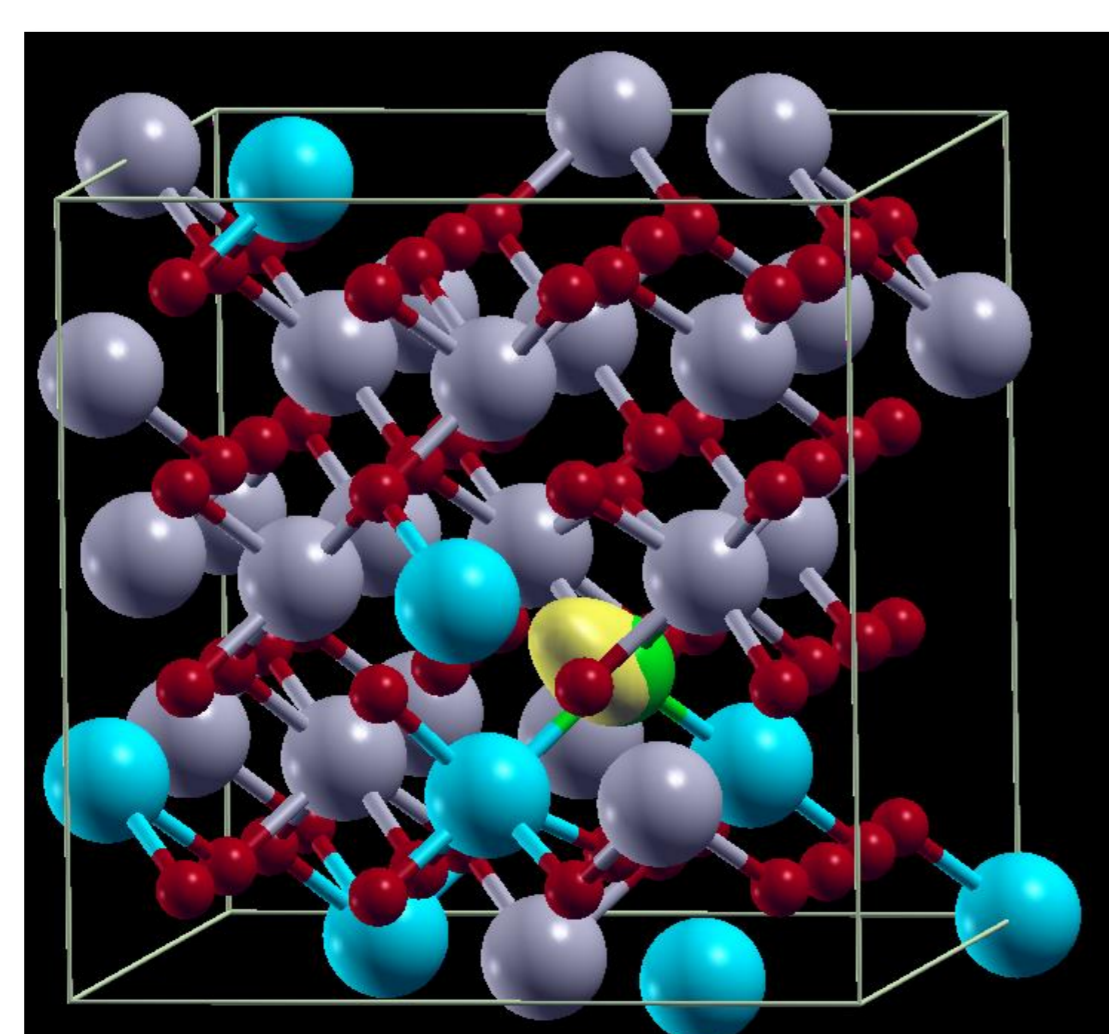


Delocalized positron in bulk  $\text{UO}_2$ ,  $\text{UCeO}_2$  and  $\text{UPuO}_2$ .

For oxygen mono-vacancy :

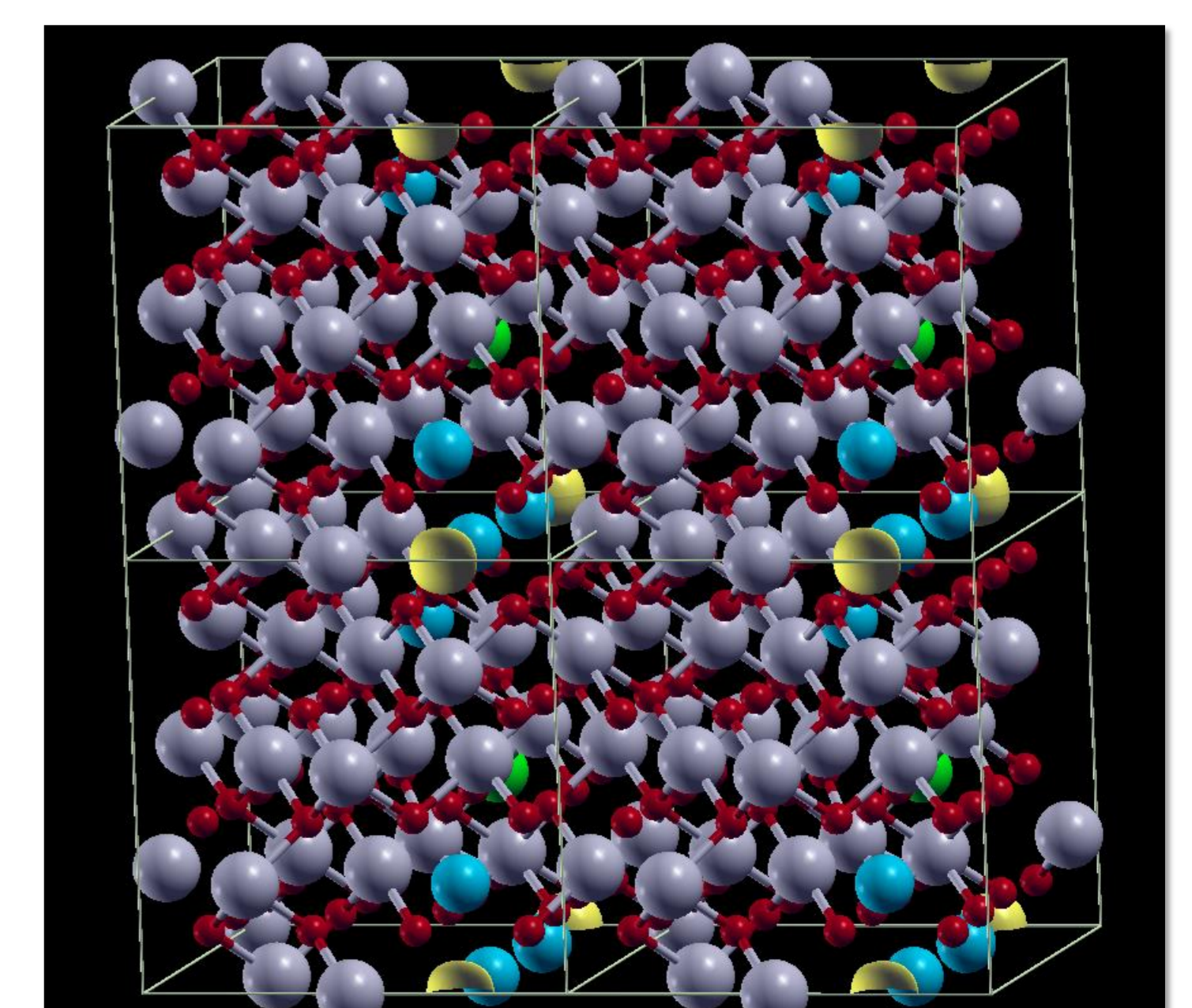
Positron localization in the vacancy for  $\text{UCeO}_2$  and  $\text{UPuO}_2$

$$\tau_{\text{V-O-UCeO}_2} = 227\text{ps}$$



Same positron localization as in the bulk for  $\text{ULaO}_2$  and  $\text{UAmO}_2$

$$\tau_{\text{V-O-ULaO}_2} = 202\text{ps}$$



Experimental work in progress on  $\text{UCeO}_2$  and  $\text{ULaO}_2$  at CNRS/CEMHTI : DFT predictions soon confirmed?