Threshold Displacement Energies of Oxygen in $YBa_2Cu_3O_7$: A Multi-Physics Analysis

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Background

The radiation environment of a nuclear fusion reactor is hostile...

- Many future reactors aim to use High Temperature Superconducting (HTS) magnets (e.g. $YBa_2Cu_3O_7$, YBCO).
- High neutron fluence impinging on magnets.
- Simulations reveal significant damage on relatively short time-scales...

Key points:

- These estimates are based on analytical damage models, which take Threshold Displacement Energy (TDE) as an essential input.
- **Goal**: Obtain TDE for oxygen atoms in YBCO using an existing empirical potential[1], then validate with *Born-Oppenheimer* Molecular Dynamics simulations.
- TDE then used to determine, required shielding and material lifetime.

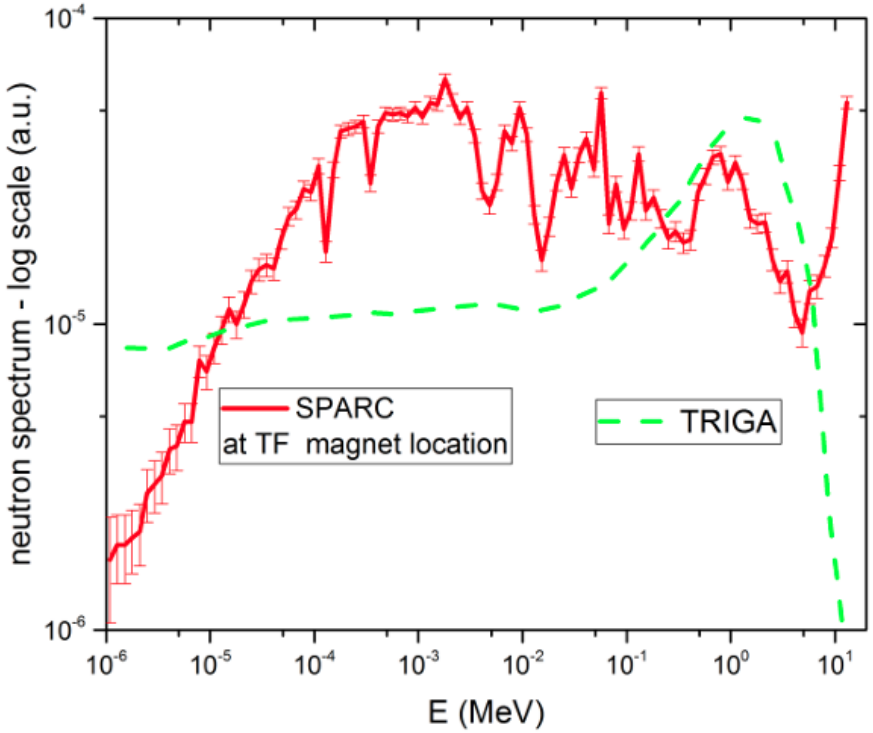


Figure 1: Neutron spectrum impinging on the YBCO TF magnet (solid red line) compared to the TRIGA spectrum (green dashed line). T., Daniele, et al. Supe. Sci. Tech. 36.1 (2022): 014003.

[1] *R L Gray et al 2022 Supercond. Sci. Technol. 35 035010*

REBCO

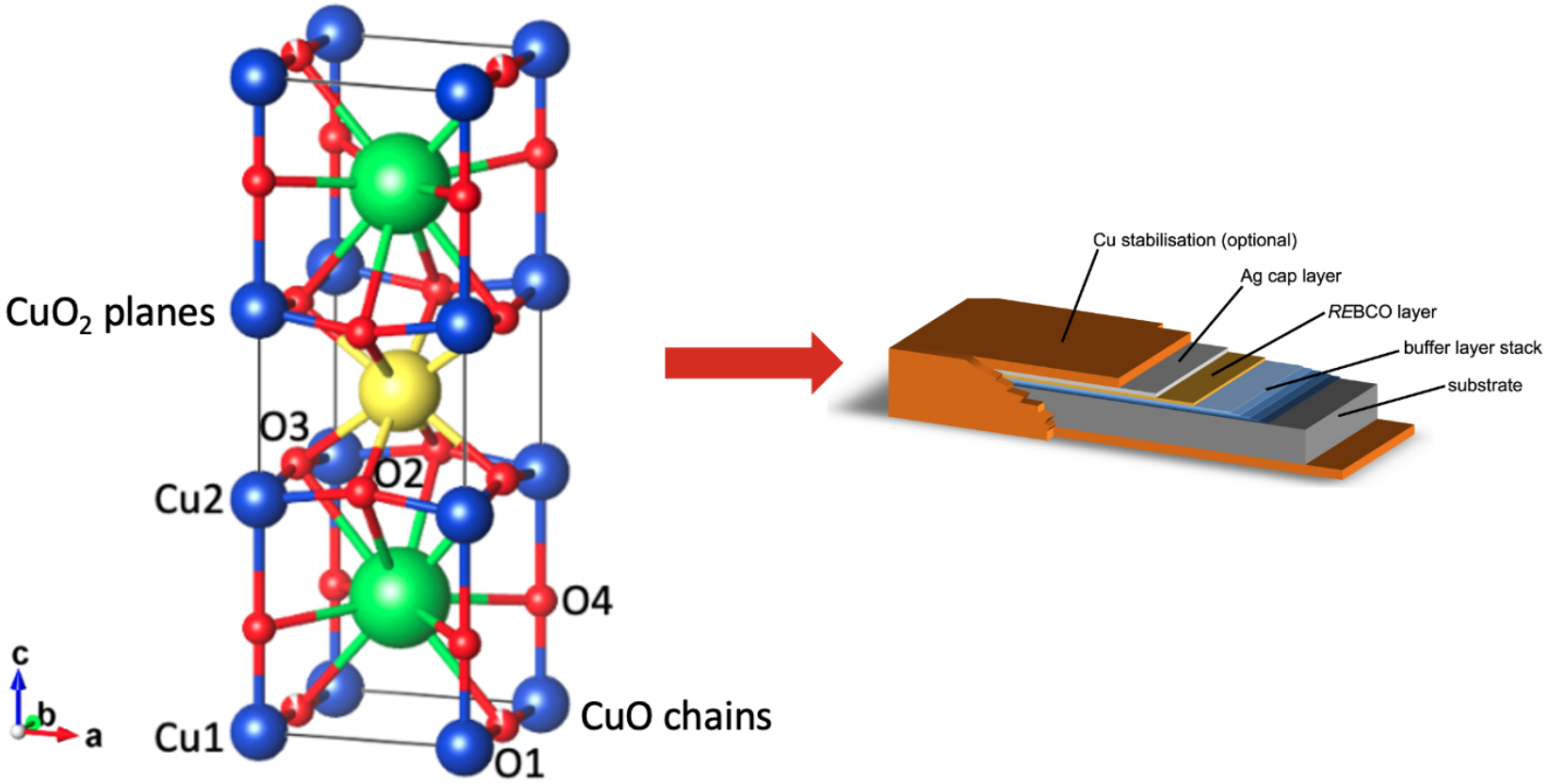


Figure 2: Left) REBCO crystal structure. Red O, blue Cu, green Ba, yellow Rare-Earth element (e.g. Y/ Gd). Right) HTS REBCO tape structure. C Barth et al. 2015 Supercond. Sci. and Tech., 28 045011, 2015.

Damage in REBCO

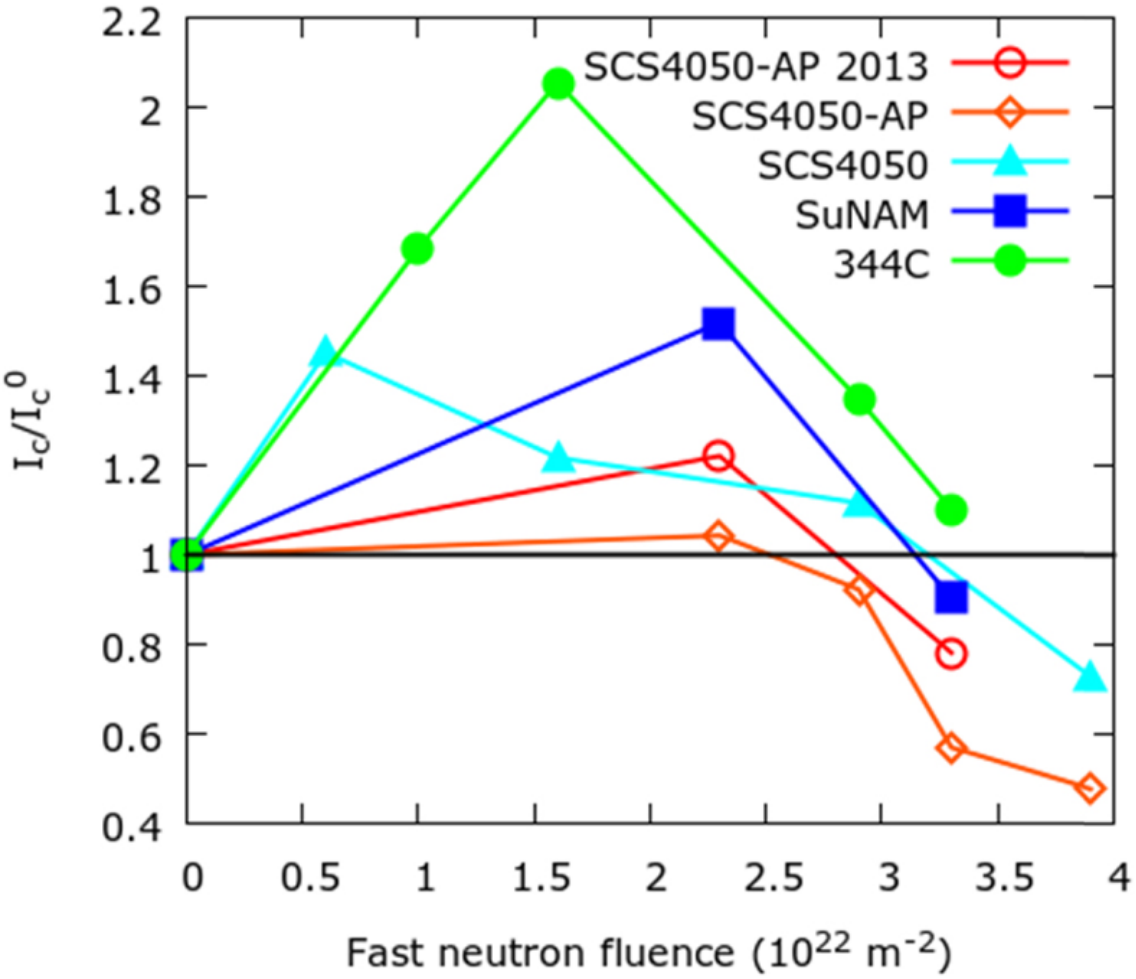
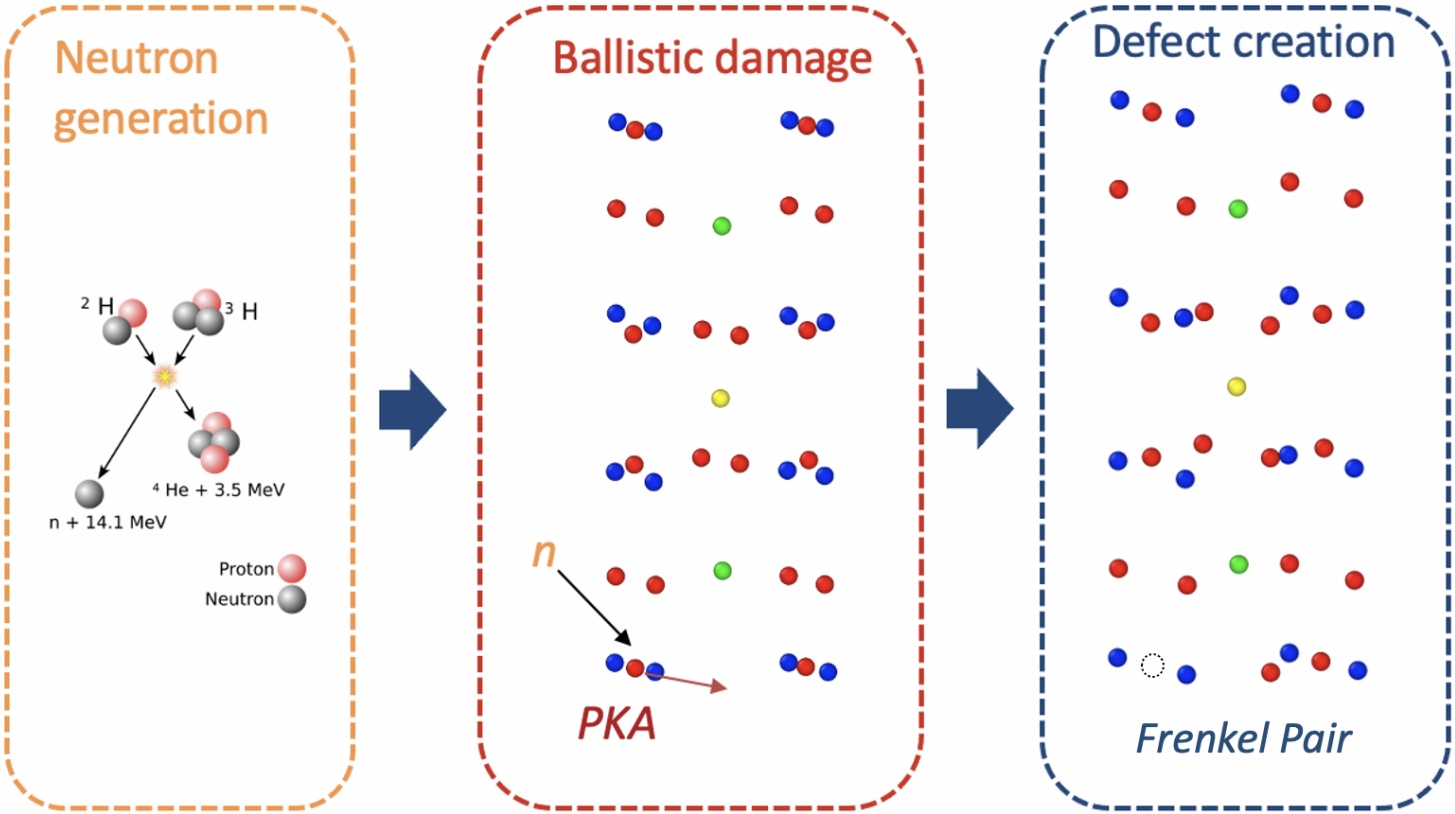


Figure 3: Fluence dependent critical current enhancement factor of different REBCO coated conductors at 30 K, 15 T. Initial rise in critical current before sharp loss. D Fischer et al. Superconductor Science and Technology, 31(4):044006, 2018.

Threshold Displacement Energy



Threshold Displacement energy (TDE) is minimum energy at which a defect is formed.

-Angularly anisotropic
- Temperature dependent
- Material / atom specific

Molecular Dynamics

Molecular Dynamics (MD) represents a tool that is used to simulate dynamics of systems in sub-picosecond time-scales.

$$M_n \nabla^2 \mathbf{R}_n = -\nabla_n V_{classical}(\mathbf{R})$$

This can be integrated over time to propagate nuclear positions with respect to the classical interaction potential $V_{classical}$.

Similarly, we can consider the interaction potential quantum mechanically. Electrons are represented as an electron density (DFT). By extension, the wavefunction Ψ is defined by this density. The nuclear positions are clamped (BO approximation):

$$M_I \nabla^2 \mathbf{R}_I(t) = -\nabla_I \langle \Psi_0 | H_{BO} | \Psi_0 \rangle$$

Evaluate instantaneous Ψ for $t \rightarrow$ use solution to propagate nuclear positions by classical mechanics.

What Does a Displacement Event Look Like?

Lancaster University

Figure 4: DFT Displacement cascade of O1 PKA with an energy of 10 eV at 25 K. O is red, Ba is green, Y is yellow, and Cu is blue.

Analytical Damage Models and why DFT?

- **Why obtain the TDE?**
  - TDE is used to estimate material damage by use of analytical damage models (e.g. NRT model).
  - How much shielding is required for the magnets? What is their expected lifetime under radiation fluence?

$$N_d(T_d) = \left\{ \begin{array}{ll} 0, & T_d < E_d \\ 1, & E_d < T_d < \frac{2E_d}{0.8} \\ \frac{0.8T_d}{2E_d}, & \frac{2E_d}{0.8} < T_d < \infty \end{array} \right\}$$

- **Why DFT?**
  - DFT can describe charge transfer, and more explicitly considers the physics of the system.
  - We use DFT to validate an empirical potential. TDEs in other materials show drastic differences (in some cases) between DFT and interatomic potentials...

DFT vs Empirical Potential

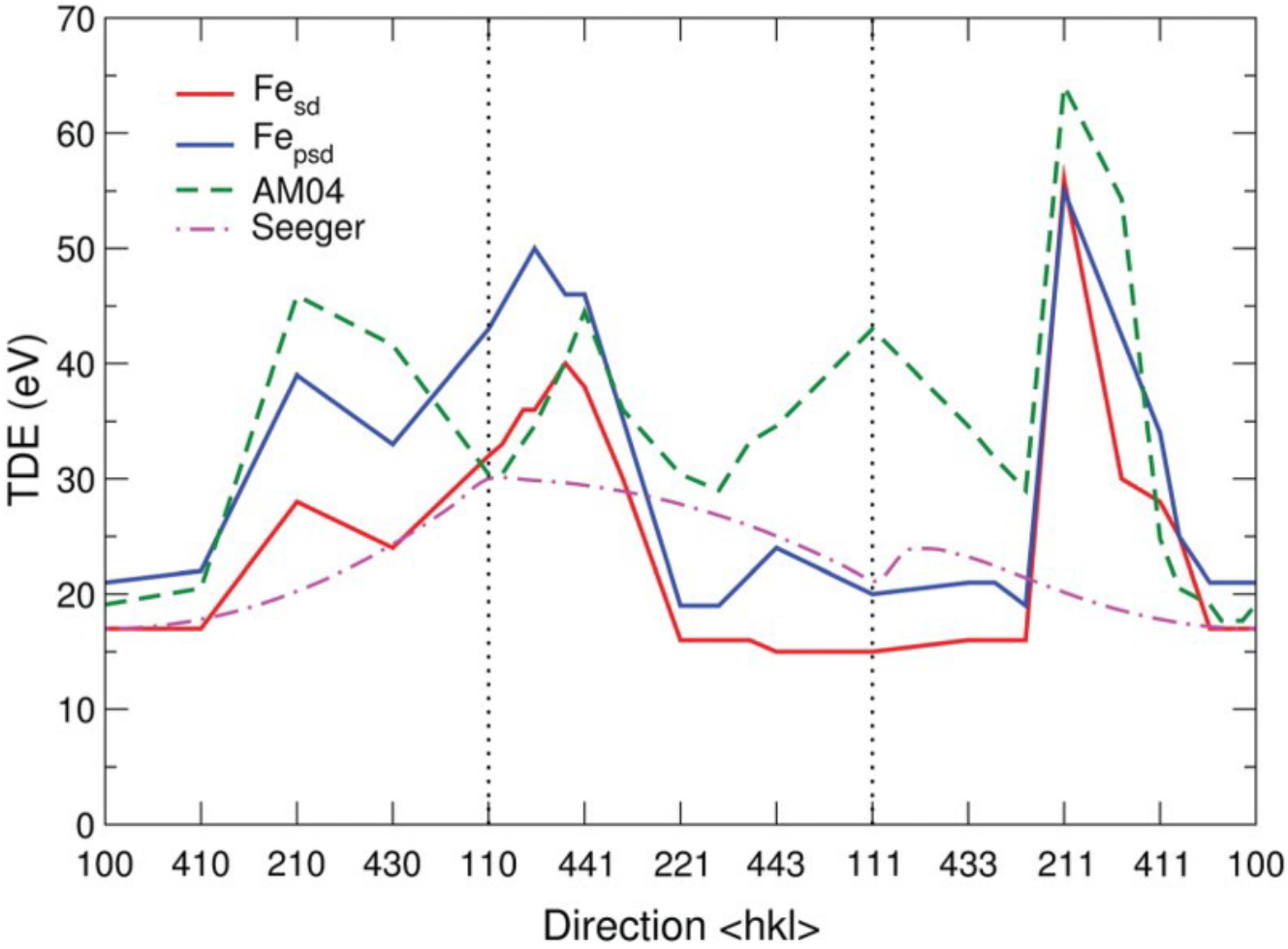
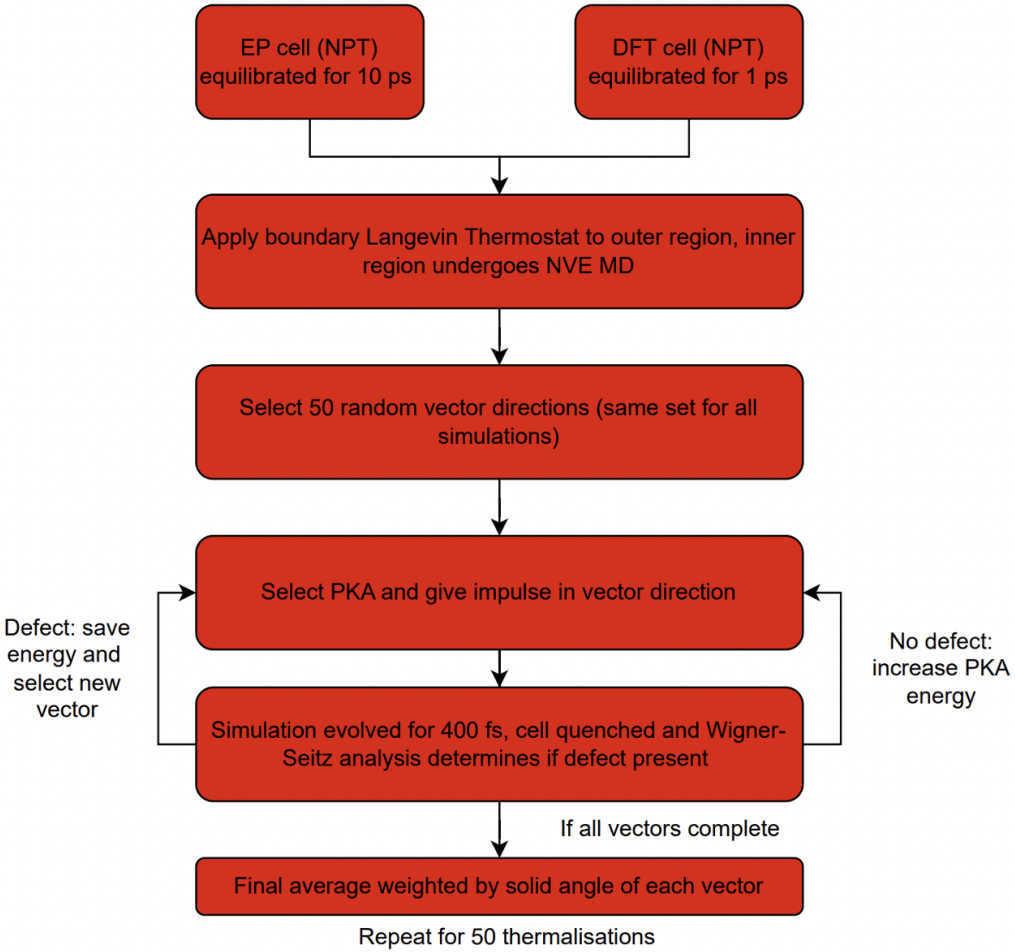
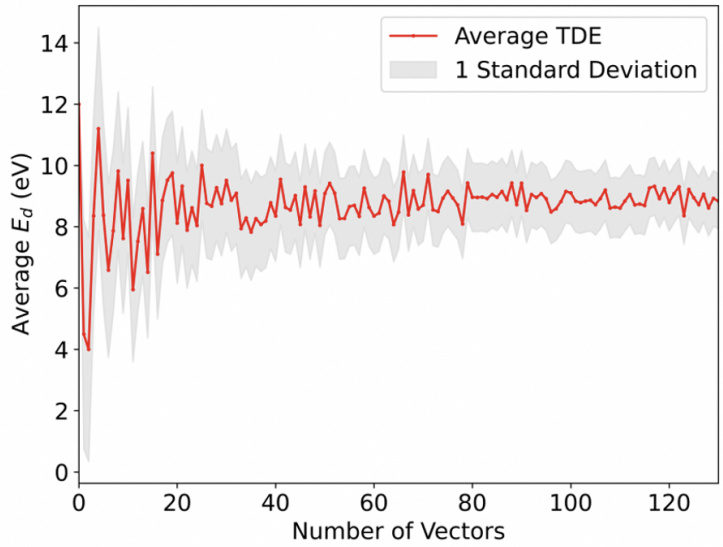


Figure 5: Comparison of direction specific TDEs in Fe. Two different DFT pseudopotentials are used (red and blue). AM04 is an EAM type potential, and the Seeger data is a fitted curve to experimental TDEs. P. Olsson et al.(2016) Mat. Res. Let., 4(4), 219–225.

Methodology





$$E_{d,ave}^{av} = \frac{\int_0^{2\pi}\int_0^{\pi} E_d^l(\theta,\phi)\sin\theta d\theta d\phi}{\int_0^{2\pi}\int_0^{\pi}\sin\theta d\theta d\phi}$$

Empirical Potential TDE Results

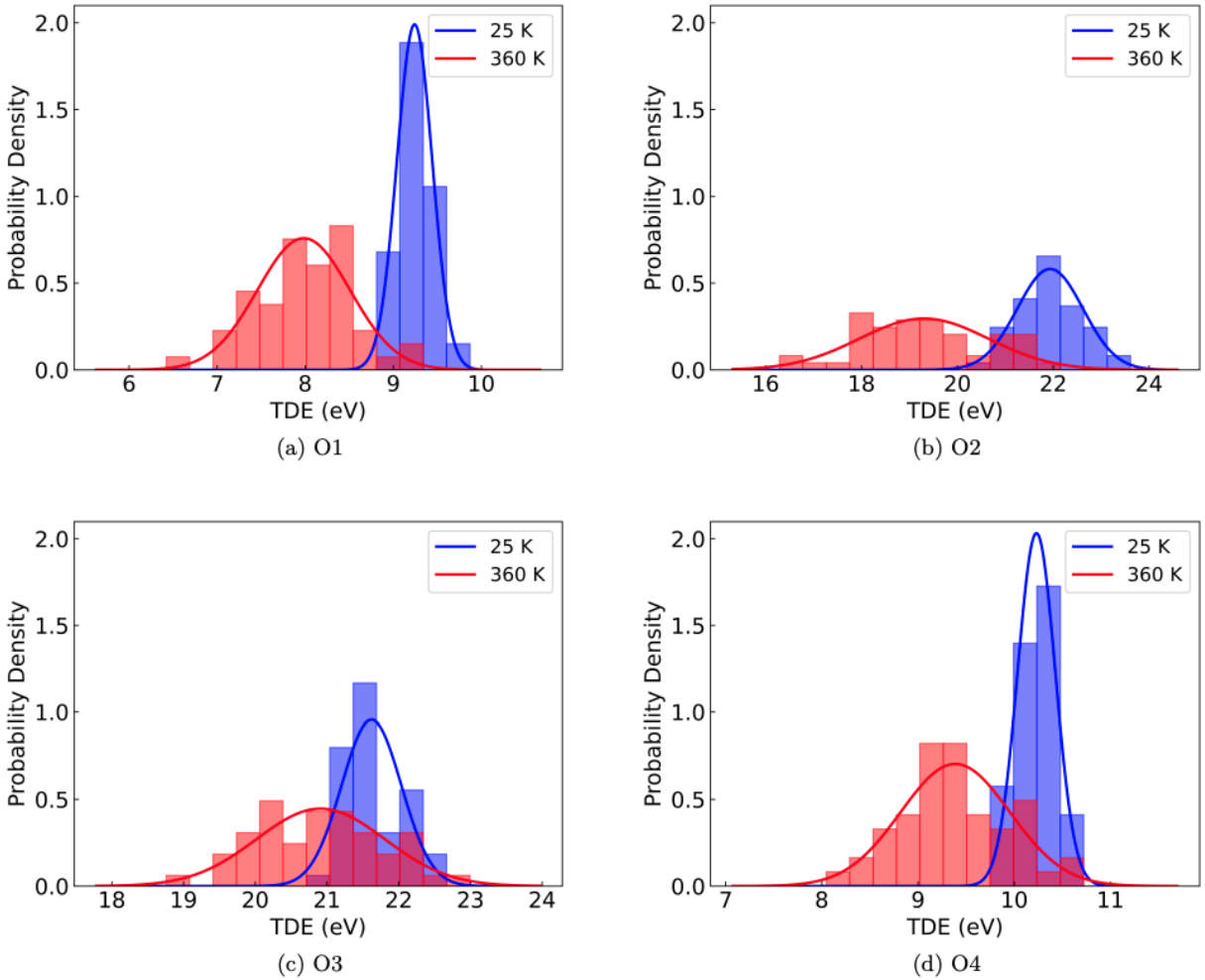


Figure 6: Distributions of TDE ($E^{av}_{d,ave}$) for EP simulations at 50 different thermalisations. The histogram of values is overlaid with a fitted Gaussian distribution. Red distribution corresponds to simulations at 360 K, and blue corresponds to 25 K.

Empirical Potential TDE Results (II)

- Results support the popular presumption that oxygen atoms in the chain region are easier to displace than those in the planes.
- In all cases, the average TDE is reduced for higher temperatures, presumably due to the increased lattice spacing.
- Observation of a decrease in TDE with increasing temperature is consistent with other work that considers the lower bound of the TDE.
- Temperature and PKA energy both increase the breadth of the TDE distribution.

Table 1: Mean (μ) and variance (σ) of $E_{d,ave}^{av}$ for different oxygen PKAs at 360 and 25 K (using EP).

	25 K		360 K	
PKA	μ (eV)	σ (eV)	μ (eV)	σ (eV)
O1	9.24	0.04	7.99	0.28
O2	21.94	0.47	19.30	1.82
O3	21.62	0.17	20.91	0.80
O4	10.23	0.04	9.39	0.33

Empirical Potential Defects

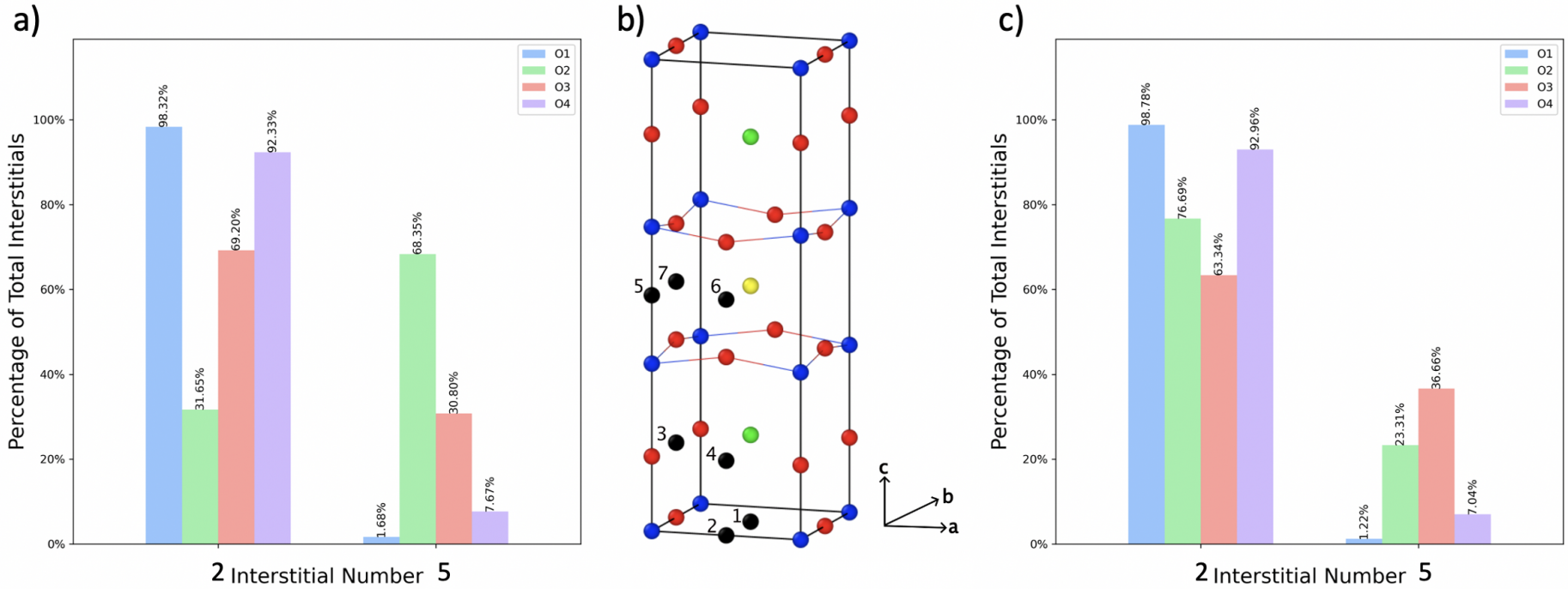


Figure 7: Percentage of different interstitial types observed for a given oxygen PKA at 25 K (a) and 360 K (c). Any interstitials with total occupancy below 1.5 % are not included. b) Interstitial positions in YBCO.

DFT TDE

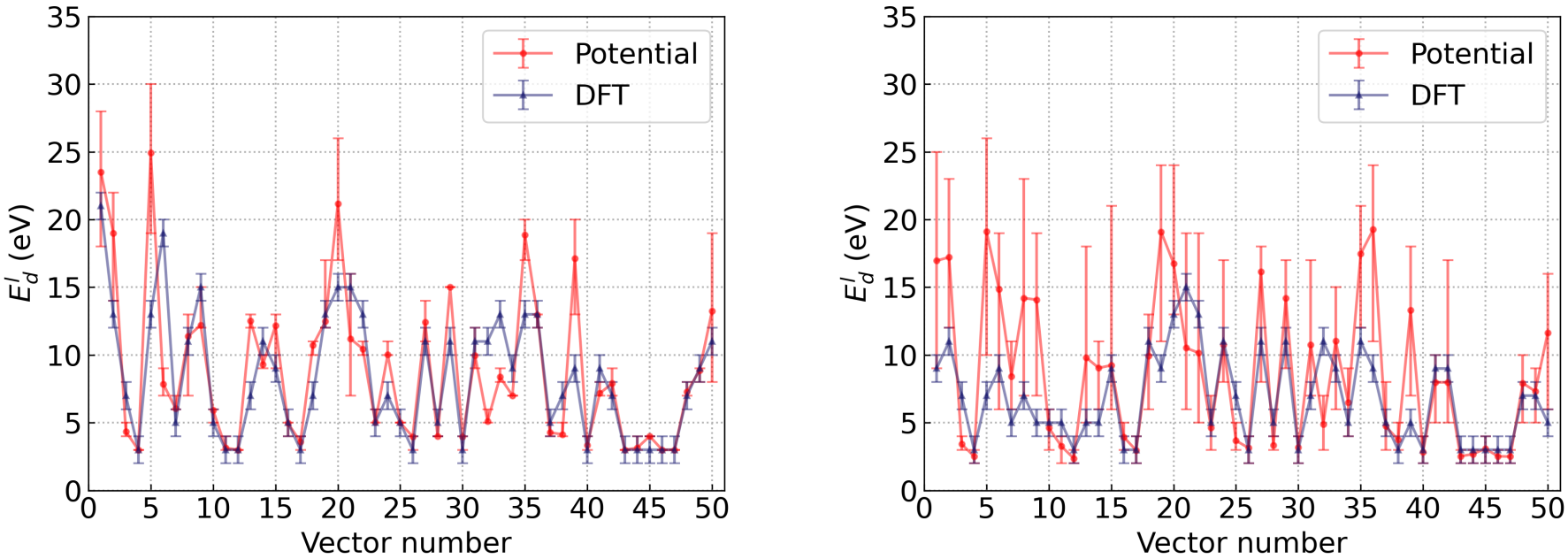


Figure 8: Comparison of TDE ($E_d^l(\phi, \theta)$) values from EP and DFT simulations at 25 K (left) and 360 K (right). Vector numbers correspond to one vector in a list of 50 random vectors. The error bars for the DFT results represent the PKA discretisation error, and the error bars for the potential represent the range of values obtained due to thermalisation.

- Directionally averaged TDE for the potential is 9.24 eV and 7.98 eV for 25 K and 360 K, respectively.
- Directionally averaged TDE for our DFT study is 7.88 eV and 7.25 eV for 25 K and 360 K, respectively.

DFT Defects

- The defect chemistry observed from our DFT simulations is very similar to that from the EP. The only difference is a small number of more complex split interstitials.
- These peroxide-like defects have not yet (to our knowledge) been reported.
- DFT has a tendency to over-stabilise peroxides.

Table 2: Proportion of different defect types from DFT TDE simulations.

Interstitial type	Occurrence (%) 25 K	Occurrence(%) 360 K
2	82	74
5	0	6
6	2	0
Other	16	20

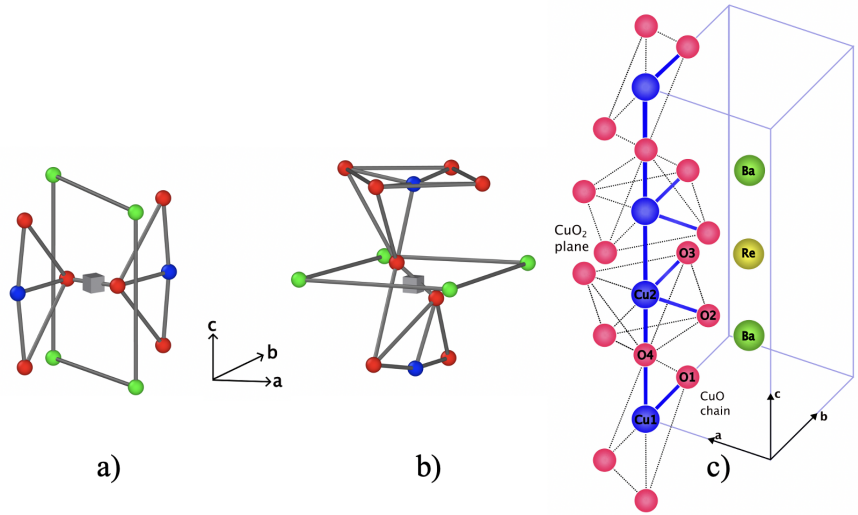


Figure 9: Morphology of different defect structures observed from DFT TDE simulations in YBCO. Cu atoms are blue, Y is yellow, Ba is green and O is red. The panels correspond to the following defects: a) O_2 peroxide dumbbell, sat in ab plane next to O1 vacancy in CuO chain b) O_2 peroxide dumbbell next to O4 vacancy. c) Representative structure of REBCO.

Conclusions and Further Work

- We recommend averaged oxygen TDE values of 16.8 and 15.3 eV, for temperatures of 25 and 360 K, respectively.
- DFT can be used to determine a statistically significant TDE.
- TDE of O PKA is lower than previous estimates.

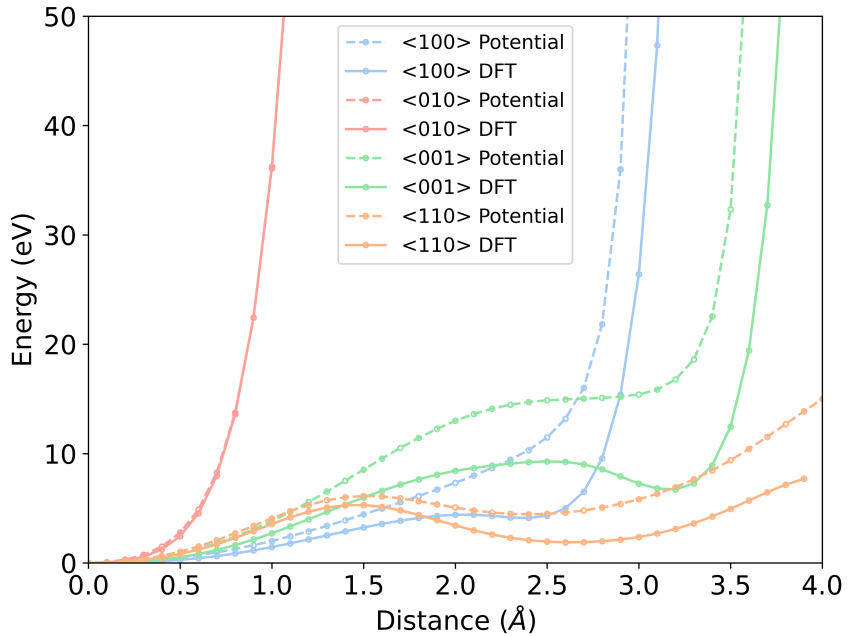
Acknowledgements

- UKAEA for funding and guidance.
- ARCHER2 UK National Supercomputing Service.
- Lancaster University's High-End Computing facility.
- MARCONI supercomputing service

Thank you for your attention.
Any questions?

Quasi-Static Drag

- Quasi-static drag simulations allow one to probe the energetic response of different models to local compression.
- Results indicate that the DFT pseudopotential sufficiently describes close atomic approaches in our case, as PKA energies do not exceed 30 eV.
- The DFT is (in general) softer than the EP.



QSD calculations for displacement of an O1 atom along $< 100 >$, $< 010 >$, $< 001 >$, *and* $< 110 >$ *directions. Performed for both the DFT and EP models.*

DFT Lattice Parameters

Lattice Parameter	DFT (Å)	Experimental (Å)
a	3.869	3.827
b	3.958	3.893
c	11.854	11.699

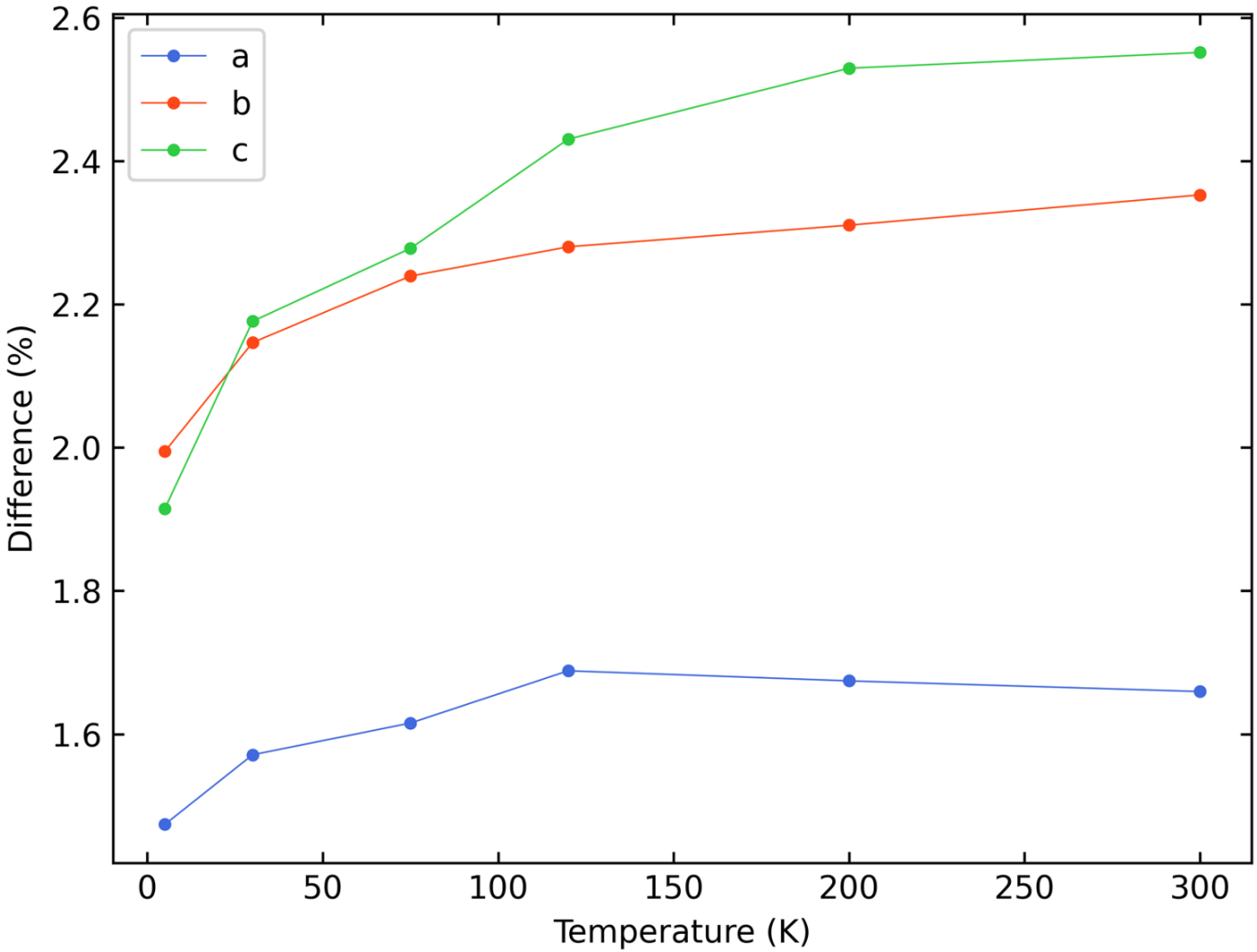
YBCO lattice parameters after relaxation with our DFT model. Compared with experimental crystallographic data from ''G Calestani and C Rizzoli. Nature, 328(6131):606–607, 1987."

DFT Vacancy Formation Energies

Defect formation energy (eV)		
Vacancy type	DFT	Literature DFT
V_{O1}	0.94	0.83
V_{O2}	1.61	1.75
V_{O3}	1.57	1.74
V_{O4}	1.27	1.36

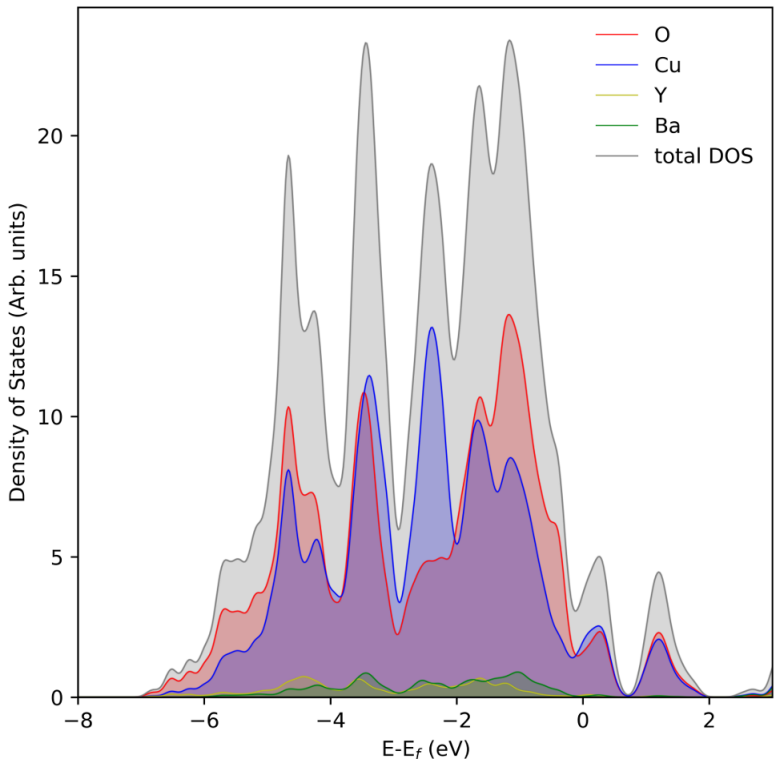
YBCO defect formation energies for our DFT model, compared with values from '' G Liu et al. RSC advances, 13(27):18927–18933, 2023."

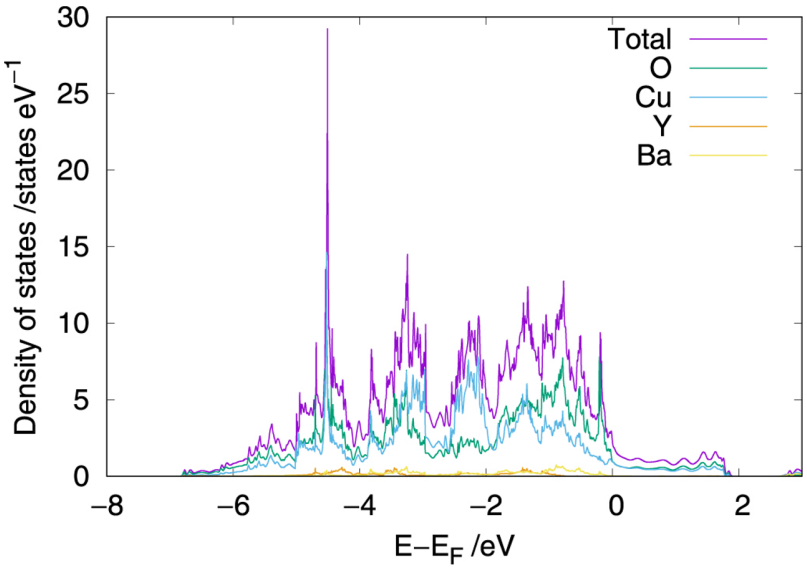
DFT Thermal Expansion



Percent difference between experimental and computed lattice parameters at different temperatures.

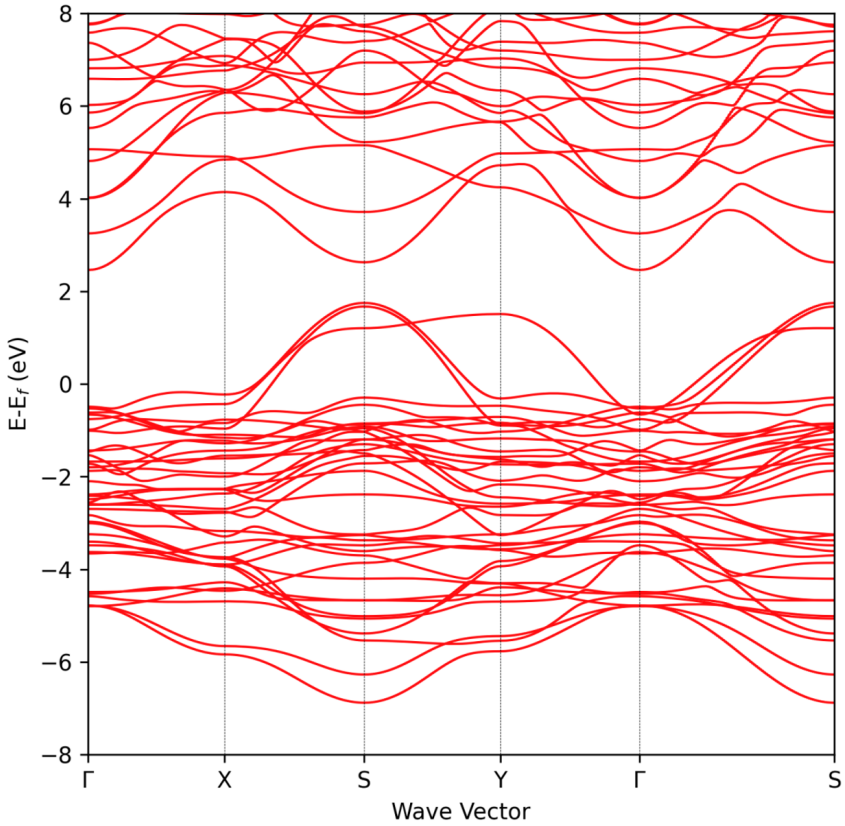
DFT Density of States

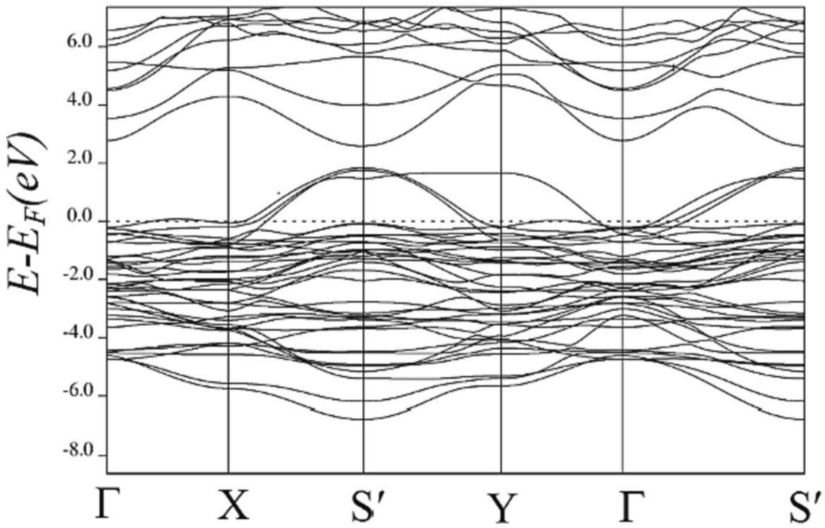




Total and partial DOS for YBCO. Normalised so the Fermi level sits at 0. (left) Our work, (right) S. Murphy. Journal of Physics Communications, 4(11):115003, 2020.

DFT Band Structure





Band structure of YBCO. Fermi level at zero. (left) Our work, (right) C. Cheong and S. Chen. Materials Today: Proceedings, 96:94–99, 2024.

Methodology

- 936 atom supercell of YBCO (6x6x2) simulated in CP2K.
- Gaussian-plane wave method employed, as provided in the Quickstep module within CP2K.
- DZVP basis used with PBE functional.
- Plane-wave cutoff set to 650 Ry.
- Native Goedecker-Teter-Hutter (GTH) Pseudopotentials.
- Simulations performed at the Γ point.
- CP2K was used due to its quasi-linear scaling with system size.
- All other simulations use LAMMPS (same cell size).

1. Calculate average TDE of all O atoms over 50 directions with EP at 25 K and 360 K, at 50 different thermalisations for each.
2. Calculate average TDE of O1 atom over 50 directions for 1 thermalisation with DFT.
3. Does DFT agree well?