



The PowerPoint version including  
animations and videos can be found at:  
[https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329\\_atomic\\_collisions\\_in\\_bolometers.pptx?rlkey=0fdiy8tjiu2kg3aj7dw4wokwz&dl=0](https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329_atomic_collisions_in_bolometers.pptx?rlkey=0fdiy8tjiu2kg3aj7dw4wokwz&dl=0)

## Atomic collisions in cryogenic detectors

**G. Soum-Sidikov<sup>1</sup>, J.-P. Crocombette<sup>2</sup>, D. Lhuillier<sup>1</sup>**

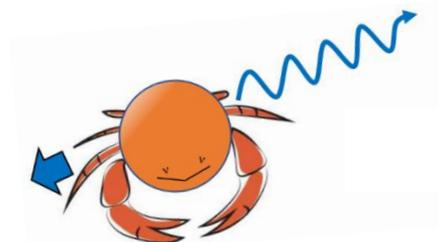
<sup>1</sup> CEA Saclay DRF/ IRFU/ Département de Physique Nucléaire

<sup>2</sup> CEA Saclay DES / ISAS/ Section de recherches de métallurgie physique

DRTBT 2024  
Aussois  
29 March 2024



irfu



# Outline

1/ The importance of atomic collisions

2/ Simulation of atomic collisions

3/ A detailed example: CRAB – accurate calibration of bolometers

Summary

# Energy loss of an atom in a crystal

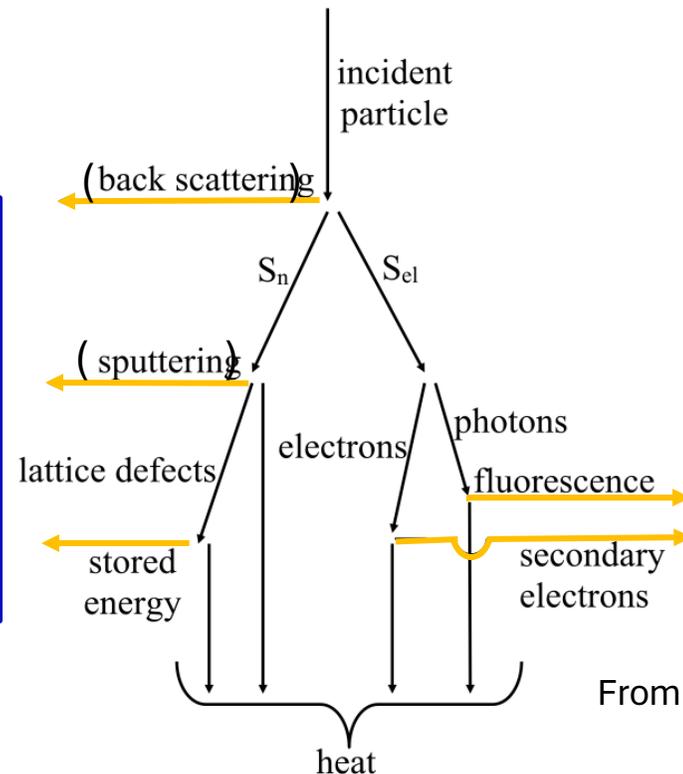
Complex process, depends on the atom energy, mass and on the target

Nuclear recoil in the target (f. g. neutron interaction), ion from radioactive source or ion beam

**Nuclear energy loss**

**Cascade of elastic collisions** with the atoms in the absorber of a cryogenic detector

Energy stored in crystal defects



**Electronic energy loss**

Dominant at high energy

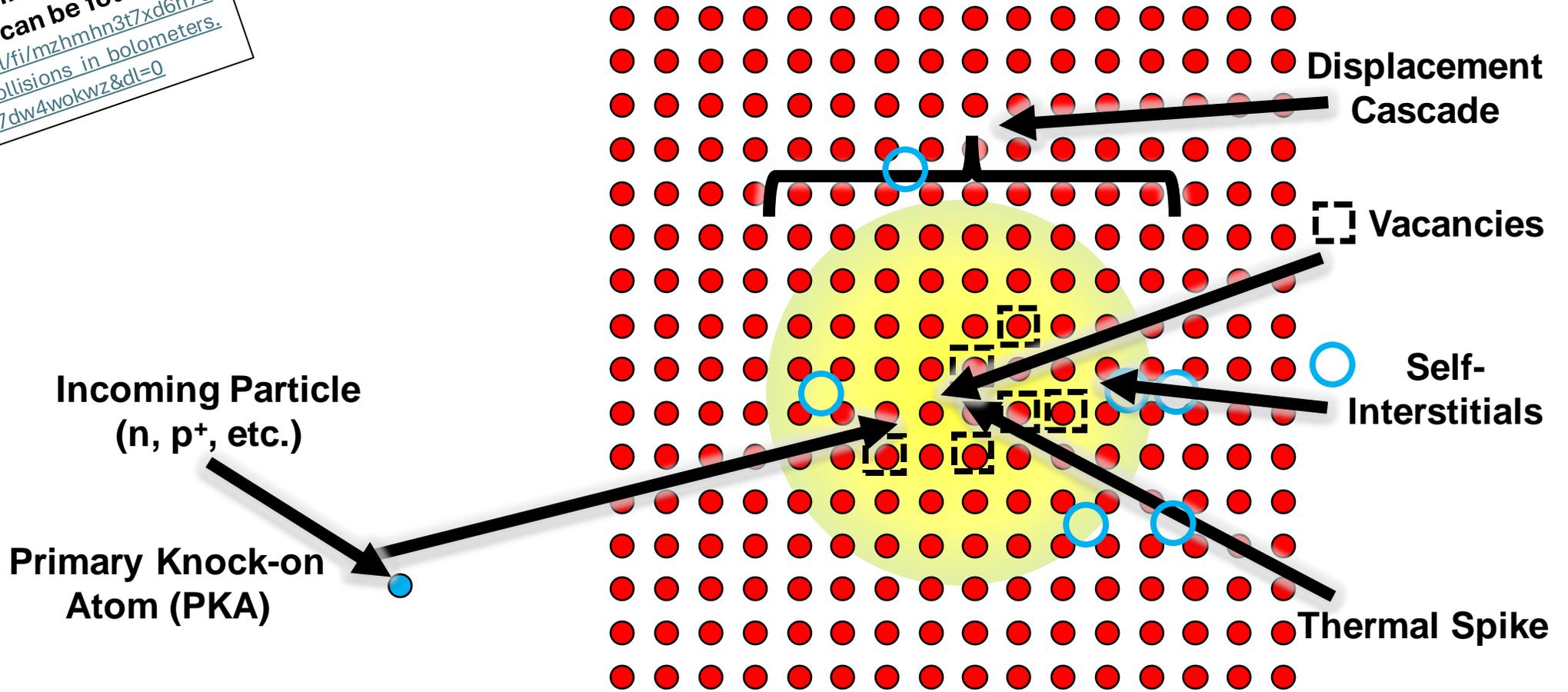
From P. Egelhof, S. Kraft-Bermuth, *Prog. Part. Nucl. Phys.* 132, 104031(2023)

**Simplified image of the energy flow in the absorber**

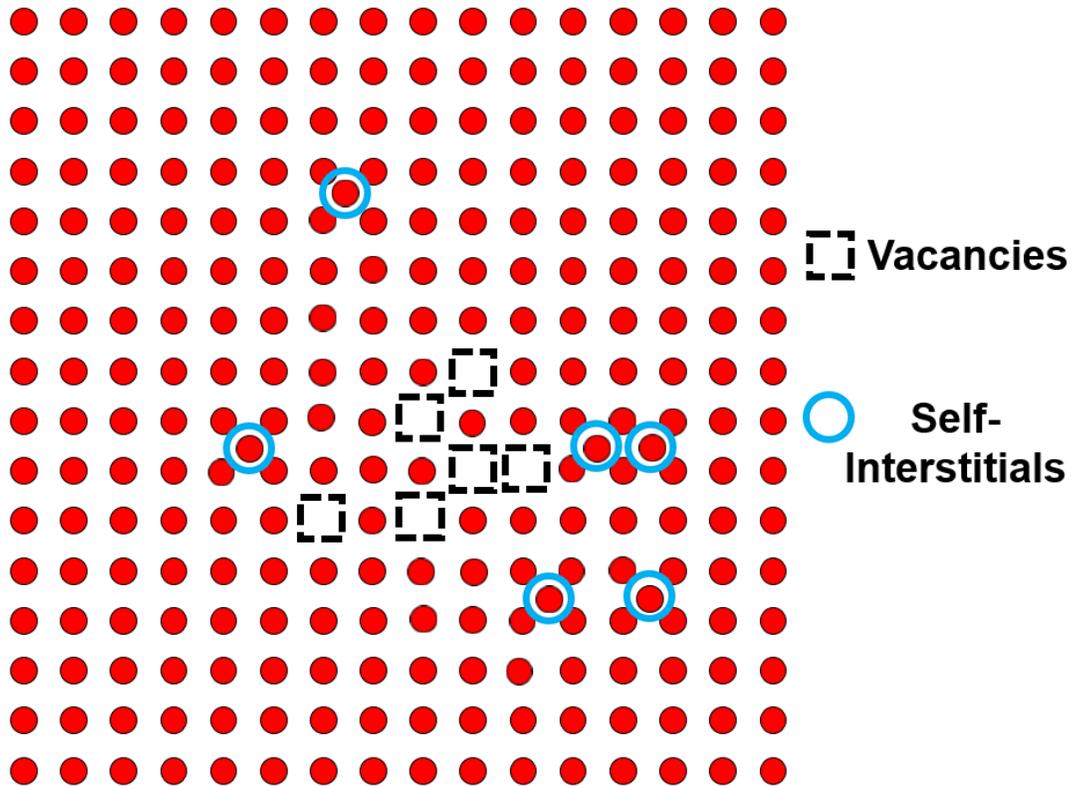
→ Energy not detected as heat

Duration: ~10ps

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[https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329\\_atomic\\_collisions\\_in\\_bolometers.pptx?rlkey=0fdiy8tjju2kg3aj7dw4wokwz&dl=0](https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329_atomic_collisions_in_bolometers.pptx?rlkey=0fdiy8tjju2kg3aj7dw4wokwz&dl=0)

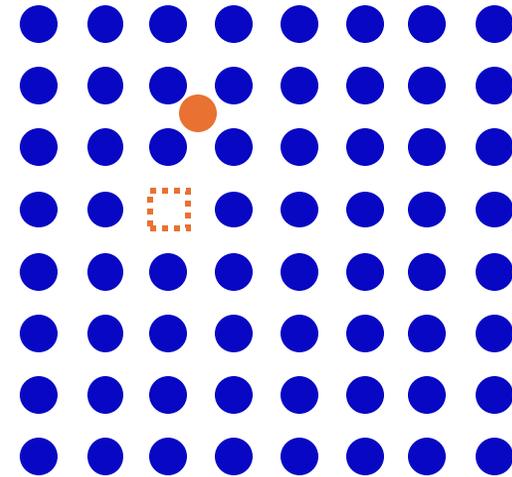


# Collision cascade and lattice defects



Duration: ~10ps

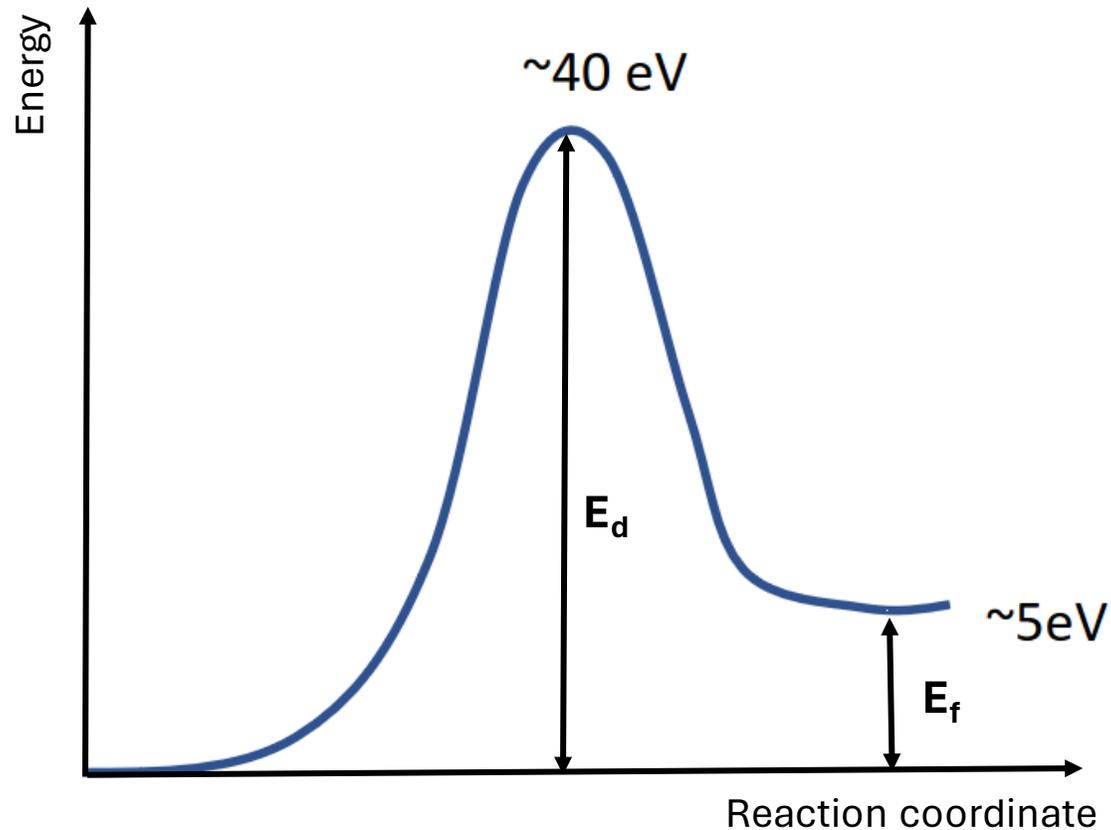
Frenkel pair: vacancy + interstitial



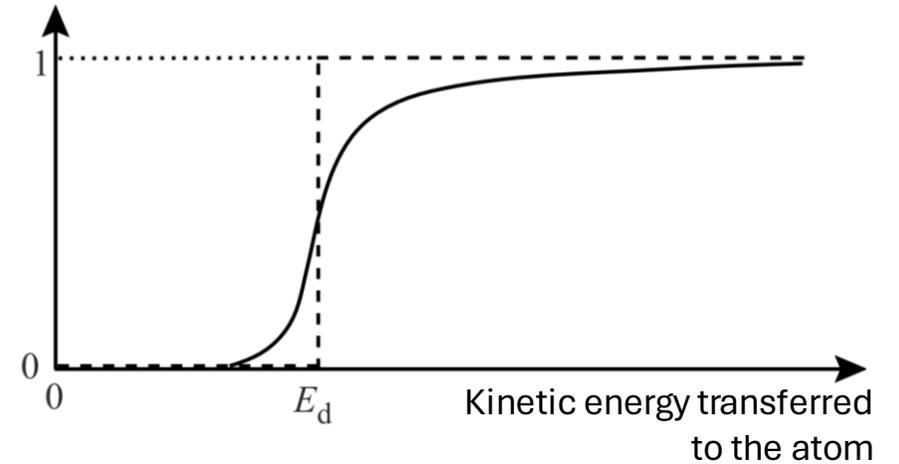
No recombination after the cascade  
at cryogenic temperatures

$$\text{Detected energy} = \text{kinetic energy of incident particle} - \text{energy stored in defects}$$

# Displacement of atoms



Probability of defect creation



From G. S. Was, [Springer \(2016\)](#)

## $E_d$ : threshold displacement energy

- Kinetic energy needed to create a defect
- Depends on the recoil direction
- Not a sharp threshold (atomic vibrations, impurities etc.)

## $E_f$ : formation energy

Energy of the defect

# Atomic collisions in bolometers

PHYSICAL REVIEW D **106**, 063012 (2022)

## Energy loss in low energy nuclear recoils in dark matter detector materials

Sebastian Sassi,<sup>\*</sup> Matti Heikinheimo,<sup>†</sup> Kimmo Tuominen,<sup>‡</sup> Antti Kuronen,<sup>§</sup>  
Jesper Byggmästar,<sup>||</sup> and Kai Nordlund<sup>¶</sup>

Department of Physics, University of Helsinki and Helsinki Institute of Physics,  
P.O. Box 64, FI-00014 Helsinki, Finland

Nader Mirabolfathi<sup>\*\*</sup>

Department of Physics and Astronomy and the Mitchell Institute for Fundamental Physics and Astronomy,  
Texas A&M University, College Station, Texas 77843, USA

(Received 20 June 2022; accepted 23 August 2022; published 15 September 2022)

Recent progress in phonon-mediated detectors with eV-scale nuclear recoil energy sensitivity requires an understanding of the effect of the crystalline defects on the energy spectrum expected from dark matter or

## Crystal Defects: A Portal To Dark Matter Detection

Fedja Kadribasic, Nader Mirabolfathi

Department of Physics and Astronomy, Texas A&M University, College Station, TX, USA

Kai Nordlund and Flyura Djurabekova

Helsinki Institute of Physics and Department of Physics, PB 43, University of Helsinki, Finland

(Dated: February 12, 2020)

We propose to use the defect creation energy loss in commonly used high energy physics solid state detectors as a tool to statistically identify dark matter signal from background. We simulate the energy loss in the process of defect creation using density functional theory and molecular dynamics methods and calculate the corresponding expected dark matter spectra. We show that in phonon-mediated solid state detectors, the energy loss due to defect creation convolved with the expected dark matter interaction signal results in a significant change in the expected spectra for common detector materials. With recent progress towards  $\sim 10$  eV threshold low-mass dark matter searches, this variation in expected dark matter spectrum can be used as a direct signature of dark matter interactions with atomic nuclei.

RESEARCH ARTICLE | AUGUST 27 2018

## Energy loss due to defect formation from $^{210}\text{Pb}$ recoils in SuperCDMS germanium detectors

R. Agnese, T. Aralis, T. Aramaki, I. Aronov, E. Aprile, W. Baker, S. Baker, D. Barker, D. A. Basler, T. Birler, M. A. Bowles, P. L. Brink, Y.-Y. Chang, J. Cooley, B. Cornell, E. Figueroa-Feliciano, C. W. Fisk, Z. Hong, E. W. Hoppe, L. Hsu, M. E. N. A. Kuzinsky, R. E. Lawrence, V. Mandic, N. Mast, E. H. Miller, N. S. M. Osier, W. A. Page, R. Partridge, A. Raju, T. Rein, T. Reynolds, A. A. Scarf, R. W. Schnee, S. Scott, H. A. Tanaka, D. Toback, R. Uleni, M. J. Wilson, J. Winchell, D. H. ...

Check for updates

Appl. Phys. Lett. **113**, 092101 (2018)  
<https://doi.org/10.1063/1.5041457>

CRUISE

## Impact of Crystal Lattice Defect Quenching on CEvNS at reactors

Excess Workshop 2022, Vienna

Thierry Lasserre & Chloé Goupy

CFA & TUM-SFB1258

## Lattice Damage in Superconducting Microcalorimeter Detectors

Robert D. Horansky, Katrina E. Koehler, Mark P. Croce, Gerd J. Kunde,  
Michael W. Rabin, Barry L. Zink, and Joel N. Ullom

There is currently significant interest in using superconducting detectors for measurement of ion kinetic energies. High resolution is possible with an order of magnitude over semiconductors. Superconducting detectors are free of the resolution limitations imposed by straggling caused by incoming ions. Here we will calculate resolution limits due to ion damage, as well as use atom simulation SRIM to compare results. Finally, on-going experiments will be made when possible.

Keywords—Lattice defects, microcalorimeter, Q spectroscopy, transition-edge sensor.

detectors. Then we will use the calculations of Andersen, Lindhard *et al.*, and Haines *et al.*, as well as simulations with SRIM to compare to expected results from our detectors [16]. Some comparisons to experimental results will also be discussed.

### II. CRYOGENIC MICROCALORIMETERS

Cryogenic calorimeters are made up of two basic elements: an absorber and a thermometer. In our group, we use super-

# The importance of atomic collisions

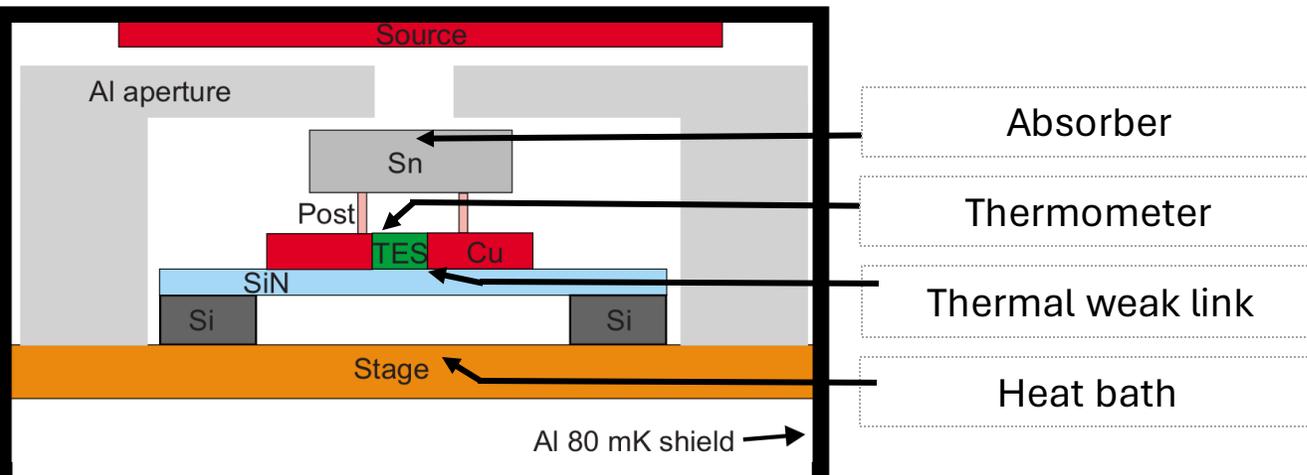
High energy cascades:  
 $\alpha$ -spectroscopy

# $\alpha$ -spectroscopy resolution

Identification of trace nuclear materials performed with  $\alpha$ -spectroscopy

Example: measurement of the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio indicate the intended purpose of a Pu sample

R. D. Horansky and *al.*, [Appl. Phys. Lett. 93, 123504 \(2008\)](#)

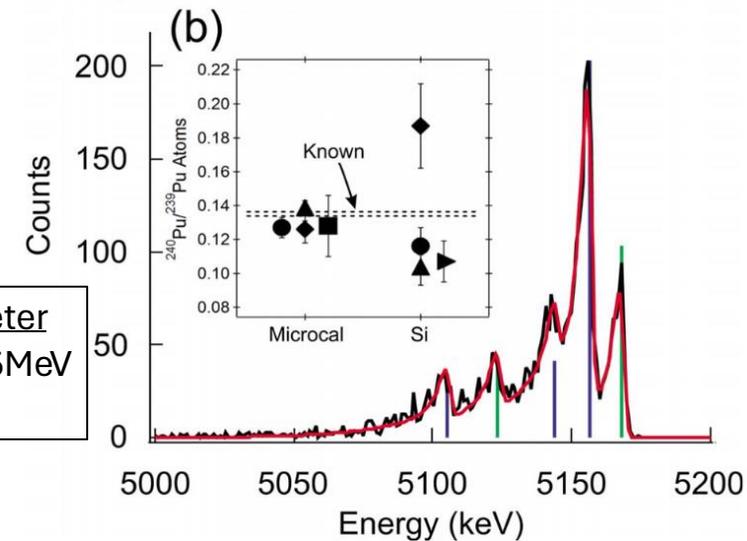
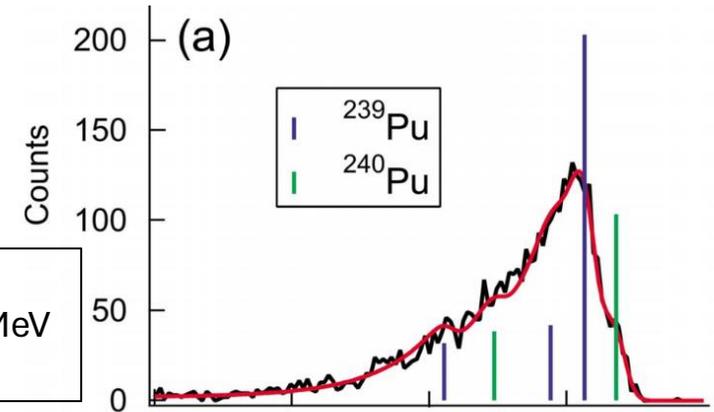


R. D. Horansky and *al.*, [J. Appl. Phys. 107, 044512 \(2010\)](#)

Mixed isotope Pu  $\alpha$  particle spectrum

With a Si diode  
Resolution  $\sim 8\text{keV}@5\text{MeV}$   
Straggling  $\sim 5\text{keV}$

With the microcalorimeter  
Resolution  $\sim 1.1\text{keV}@5\text{MeV}$   
Straggling  $\sim 1.7\text{keV}$



**Energy resolution is a key factor for nuclear materials identification with  $\alpha$ -spectroscopy**

# $\alpha$ -spectroscopy resolution limit

R. D. Horansky and *al.*, [J. Appl. Phys. 107, 044512 \(2010\)](#)

TABLE I. Resolution degradation mechanisms.

Mechanism	Energy fluctuation (eV)
Total measured resolution	1090
Thermodynamic	98
Room temperature amplification	365
Temperature fluctuations	134
Electron emission	80
Anomalous thermalization	320
Total degradation in quadrature	520
Unaccounted energy fluctuation	960

**Fluctuation of the energy stored in defects?**

1st estimation in H. H. Andersen, [NIM-B 15, 722 \(1986\)](#)

- fluctuation of the energy going to atomic movement  $E_n$
- fluctuation of the number of created defects for a same  $E_n$

**Limit of the resolution for 5MeV  $\alpha$ -particles: ~1keV**

Relies on the "NRT" formula for the number of created defects

$$n(E) = 0.42 \frac{E_n(E)}{E_{d,eff}}$$

M. Norgett, M.T. Robinson, I. Torrens, [Nucl. Eng. Design 33, 50 \(1975\)](#)

Cf. K. Nordlund, et al., [J. Nucl. Mat. 512, 450 \(2018\)](#) for a recent model

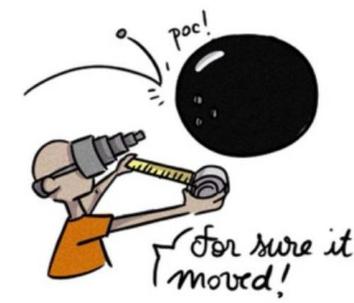
Comparison of calculations with SRIM simulations and available experimental results:

R. D. Horansky, et al., [IEEE Trans. Appl. Supercond. 23, 2101104 \(2013\)](#)

## The importance of atomic collisions

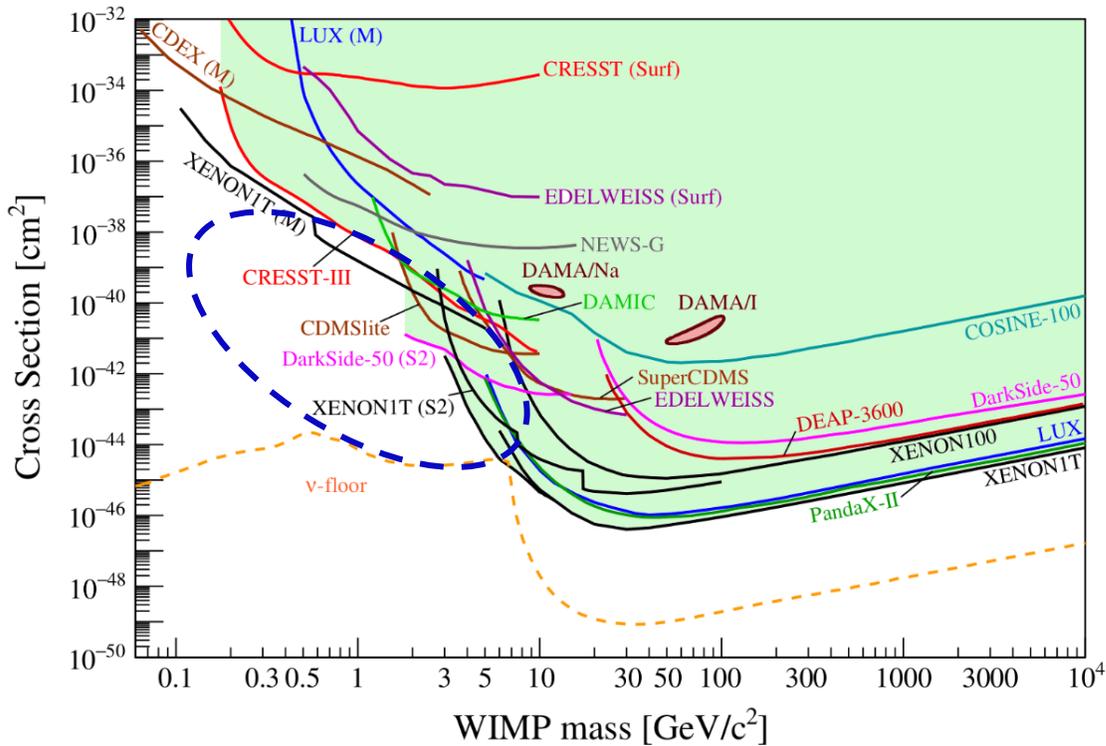
# Low energy nuclear recoils: DM and CEvNS

# Sub-keV nuclear recoils



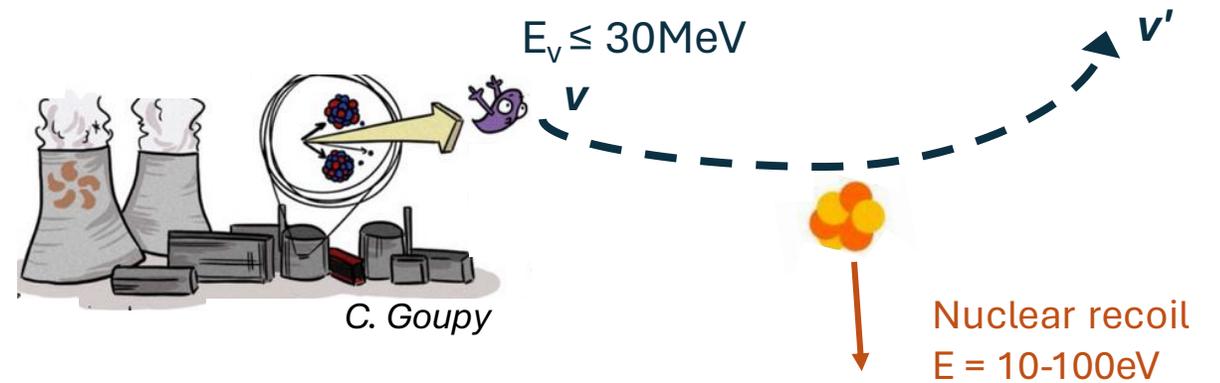
tiny nuclear recoils from [NUCLEUS comics](#)  
C. Goupy, CEA-IRFU

## Direct detection of light Dark Matter



J. Billard et al., [Rep. Prog. Phys. 85, 056201\(2022\)](#)

## Precision measurements with coherent neutrinos/nuclei scattering



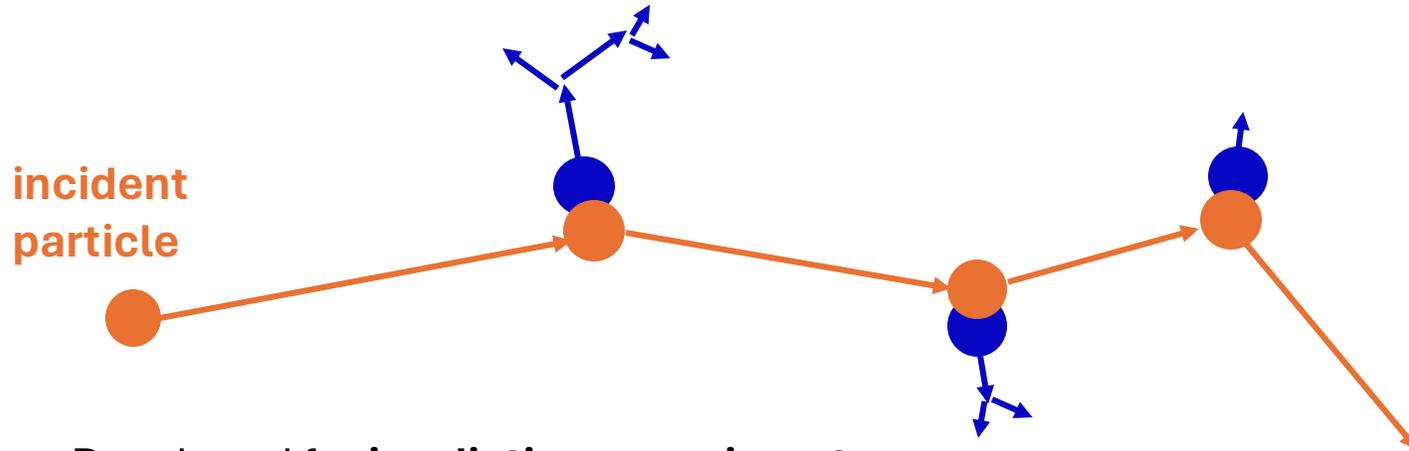
Complementary tests of Standard Model  
Reactor antineutrinos: few MeV

Nuclear recoils <1keV with low-threshold bolometers  
Sensitive to the displacement thresholds and energy stored in few created defects  
--> possible bias in energy reconstruction

## Simulation of atomic collisions

Binary Collision Approximation:  
SRIM etc.

# Binary Collision Approximation



- Developed for **irradiation experiments**
- Ion travel in matter approximated by
  - a sequence of **independent 2-body elastic collisions**
  - **straight path** between collisions (taking into account electronic stopping power)
  - Recursive tracking of secondary recoils
- Universal rough interatomic potential
- Most codes: random material (no crystal structure)

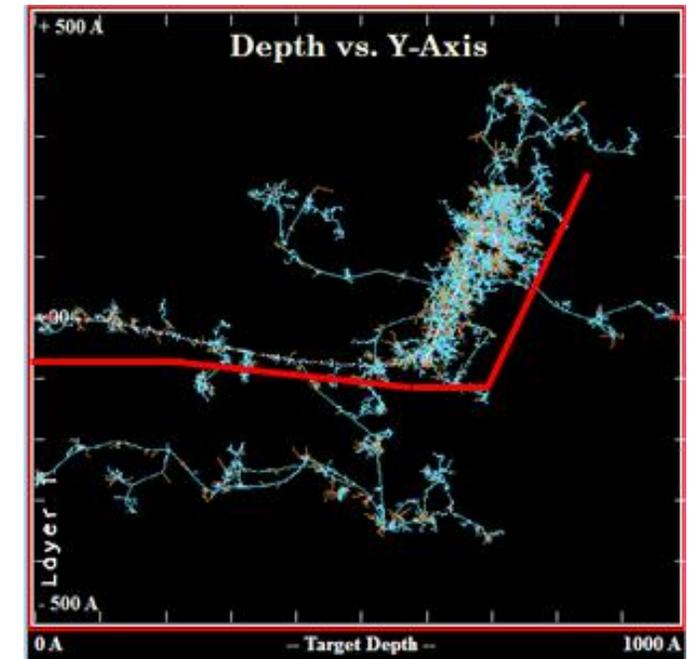


Image from a SRIM simulation

**SRIM:** vastly used, BCA benchmark but code is poorly documented

J.F. Ziegler, [www.srim.org](http://www.srim.org)

**IRADINA:** open source alternative, very fast

C. Borschel, C. Ronning, [Nucl. Instrum. Methods Phys. Res. Sect. B 269, 2133 \(2011\)](#)

J.-P. Crocombette and C. Van Wambeke, [EPJ Nuclear Sci. Technol. 5, 7 \(2019\)](#)

# Binary Collision Approximation

- Qualitative
- Very fast
- Modelling of high-energy collision cascades
- Commonly used to estimate implantation depth

- Qualitative
- Displacement threshold energy is an entry parameter
- Confusion between the number of displaced atoms & the number of defects
- No access to energy stored in defects

# α-spectroscopy resolution limit

R. D. Horansky and *al.*, [J. Appl. Phys. 107, 044512 \(2010\)](#)

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**Simulation of atomic collisions**

# Molecular Dynamics

# Goal: a movie of the cascade

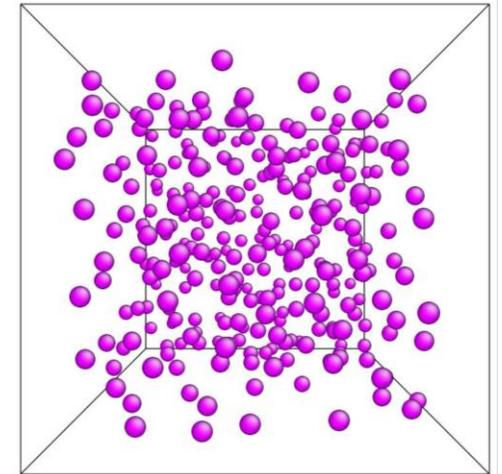
Simulate the cascade itself with the correct dynamics and energy: **Molecular Dynamics**

- step by step displacements with time discretization ( $\delta t \sim 10^{-15}\text{s}$ ):
- Newtonian Dynamics

$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{f}_i(t)$$

- Numerical method (Verlet integration algorithm)

$$\vec{r}_i(t + \delta t) = 2\vec{r}_i(t) - \vec{r}_i(t - \delta t) + \frac{(\delta t)^2}{m_i} \vec{f}_i(t) + O((\delta t)^4)$$

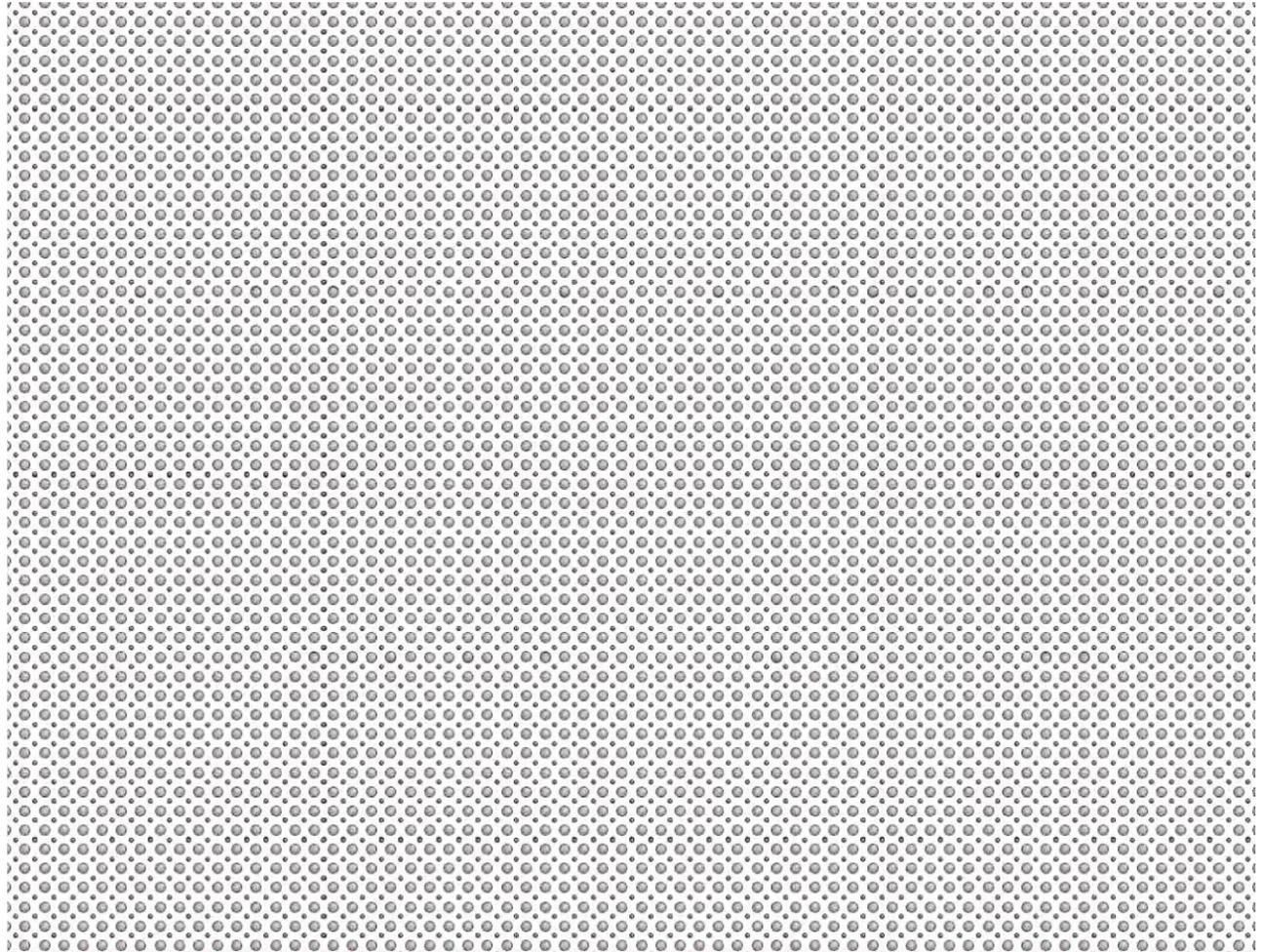


# Example: displacement cascade simulation

**Idea:** accelerate an atom in a big simulation box, follow the motion of all atoms, analyze final positions

80keV recoil in  $\text{UO}_2$   
Slice view

**The PowerPoint version including animations and videos can be found at:**  
[https://www.dropbox.com/scl/fi/mzhmhn3t7xd6n76ukve4x/20240329\\_atomic\\_collisions\\_in\\_bolometers.pptx?rlkey=0fdjy&tjiu2kg3aj7dw4wokvz&dl=0](https://www.dropbox.com/scl/fi/mzhmhn3t7xd6n76ukve4x/20240329_atomic_collisions_in_bolometers.pptx?rlkey=0fdjy&tjiu2kg3aj7dw4wokvz&dl=0)



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80keV recoil in  $\text{UO}_2$

global view

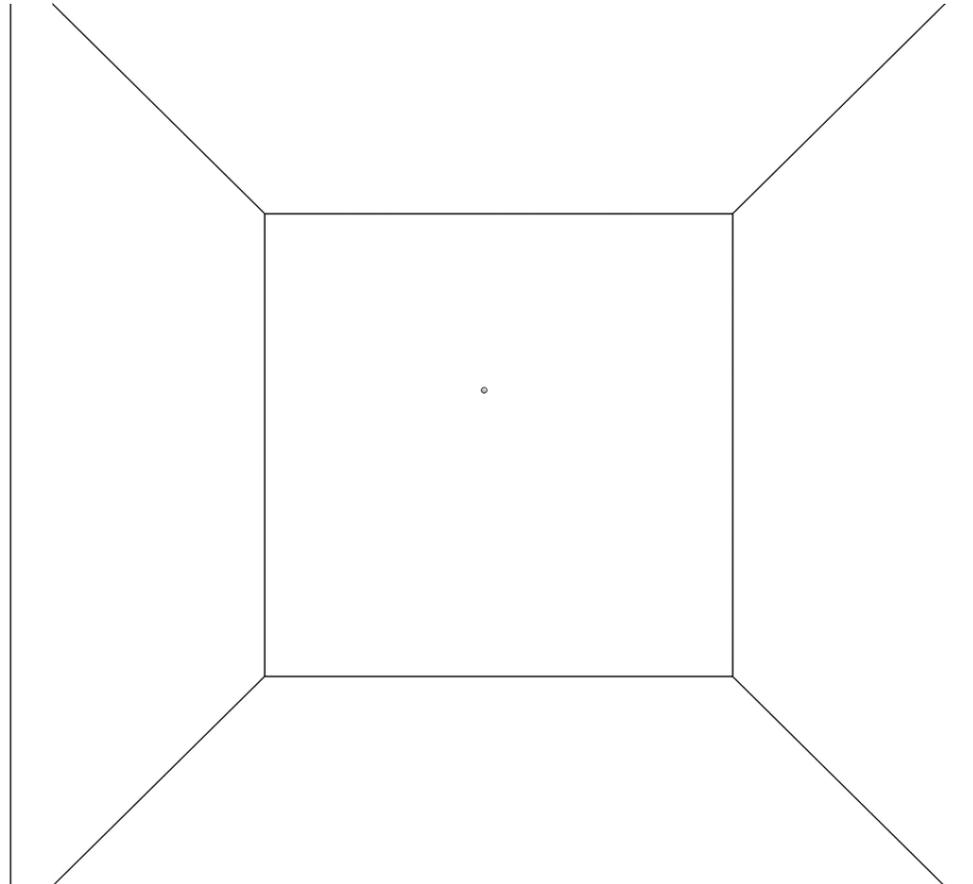
Only displaced atoms ( $>0.8 \text{ \AA}$ )

At the end:

26000 atoms displaced by more than  $2 \text{ \AA}$

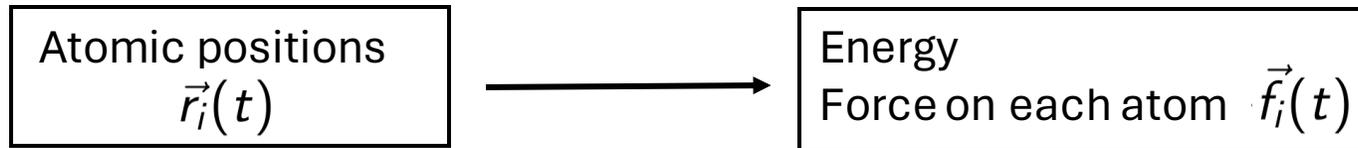
404 Frenkel pairs created

L. Van Brutzel, M. Rarivomanantsoa et D. Ghaleb,  
*J. Nucl. Mater.* 354, 28 (2006)



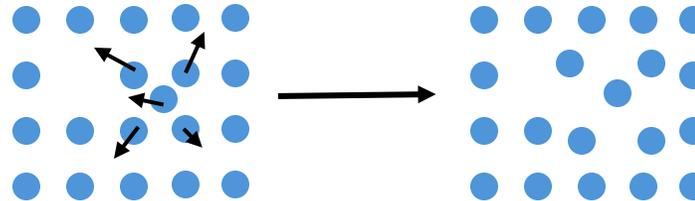
# Energy and force engines

- Molecular Dynamics simulations request an "**energy and force engine**":



- By product: **relaxation** of a structure to a local energy minimum

(e. g. minimization algorithm: conjugate gradient)



- **3 types of engines:**
  - "Ab initio": Density Functional Theory of electronic structure
  - Empirical interatomic potential
  - Machine Learning interatomic potential

## Simulation of atomic collisions

**Energy and force engine:**

"Ab initio": Density Functional Theory of electronic structure

# Electronic structure

Hartree units

$$\hbar = m_e = e = \frac{1}{4\pi\epsilon_0} = 1$$

Schrödinger equation:  $N$  nuclei and  $n$  electrons problem

$$\hat{H}\psi = E\psi$$

$$\hat{H} = \underbrace{-\frac{1}{2} \sum_{i=1}^n \nabla_i^2}_{\text{Kinetic energies}} - \underbrace{\frac{1}{2} \sum_{I=1}^N \frac{1}{M_I} \nabla_I^2}_{\text{Kinetic energies}} - \underbrace{\sum_{i,I} \frac{Z_I}{\|\vec{r}_i - \vec{R}_I\|}}_{\text{Nuclei - electrons attraction}} + \underbrace{\frac{1}{2} \sum_{i \neq j} \frac{1}{\|\vec{r}_i - \vec{r}_j\|}}_{\text{Electrons - electrons repulsion}} + \underbrace{\frac{Z_I Z_J}{2} \sum_{I \neq J} \frac{1}{\|\vec{R}_I - \vec{R}_J\|}}_{\text{Nuclei - nuclei repulsion}}$$

**First simplification: Born-Oppenheimer**  $m_i \ll M_A$

- Movement of electrons decoupled from movement of nuclei
- Electronic ground-state for each position of nuclei
- > nuclei are treated classically
- > positions of nuclei are parameters of the Hamiltonian (not variables)

# Density Functional Theory (DFT)

Ground-state energy of the system is a functional of the electronic density  $\rho(\vec{r})$  (Hohenberg-Kohn, 1964)

$$E[\rho(\vec{r})] = T[\rho(\vec{r})] - \sum_I \int \frac{Z_I \rho(\vec{r})}{\|\vec{r} - \vec{R}_I\|} d\vec{r} + \underbrace{\frac{1}{2} \int \frac{\rho(\vec{r}) \rho(\vec{r}')}{\|\vec{r} - \vec{r}'\|} d\vec{r} d\vec{r}'}_{\text{Classical part (Hartree)}} + \underbrace{E_{xc}}_{\substack{\text{Small quantum remainder} \\ \text{"exchange - correlation functional"}}} + \frac{1}{2} \sum_{I \neq J} \frac{Z_I Z_J}{\|\vec{R}_I - \vec{R}_J\|}$$

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???

$\rho(\vec{r})$

???

???

Small quantum remainder  
"exchange - correlation functional"

???

# Density Functional Theory (DFT)

## Simplifications:

- Monoelectronic wave functions  $\psi(\vec{r}_1, \dots, \vec{r}_n) = \prod_{i=1}^n \psi_i(\vec{r}_i) \longrightarrow \rho(\vec{r}) = \sum_{i=1}^n \|\psi_i(\vec{r})\|^2$
- Electronic Hamiltonian  $h(\vec{r})$  is a function of **electronic density**

Kohn-Sham  
equations

$$h(\vec{r})\psi_i(\vec{r}) = \varepsilon_i\psi_i(\vec{r}) \quad \text{with} \quad h(\vec{r}) = -\frac{1}{2}\nabla^2 - \sum_{I=1}^N \frac{Z_I}{\|\vec{r} - \vec{R}_I\|} + \int \frac{\rho(\vec{r}')}{\|\vec{r} - \vec{r}'\|} d^3r' + \boxed{V_{xc}[\rho](\vec{r})}$$

???

# Density Functional Theory (DFT)

## Simplifications:

- Monoelectronic wave functions  $\psi(\vec{r}_1, \dots, \vec{r}_n) = \prod_{i=1}^n \psi_i(\vec{r}_i) \longrightarrow \rho(\vec{r}) = \sum_{i=1}^n \|\psi_i(\vec{r})\|^2$
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???

**Exchange-correlation:** many many different available ways to approximate the functional

**LDA** (Local Density Approximation): Inhomogeneous electronic density modelled locally as an homogeneous electron gas

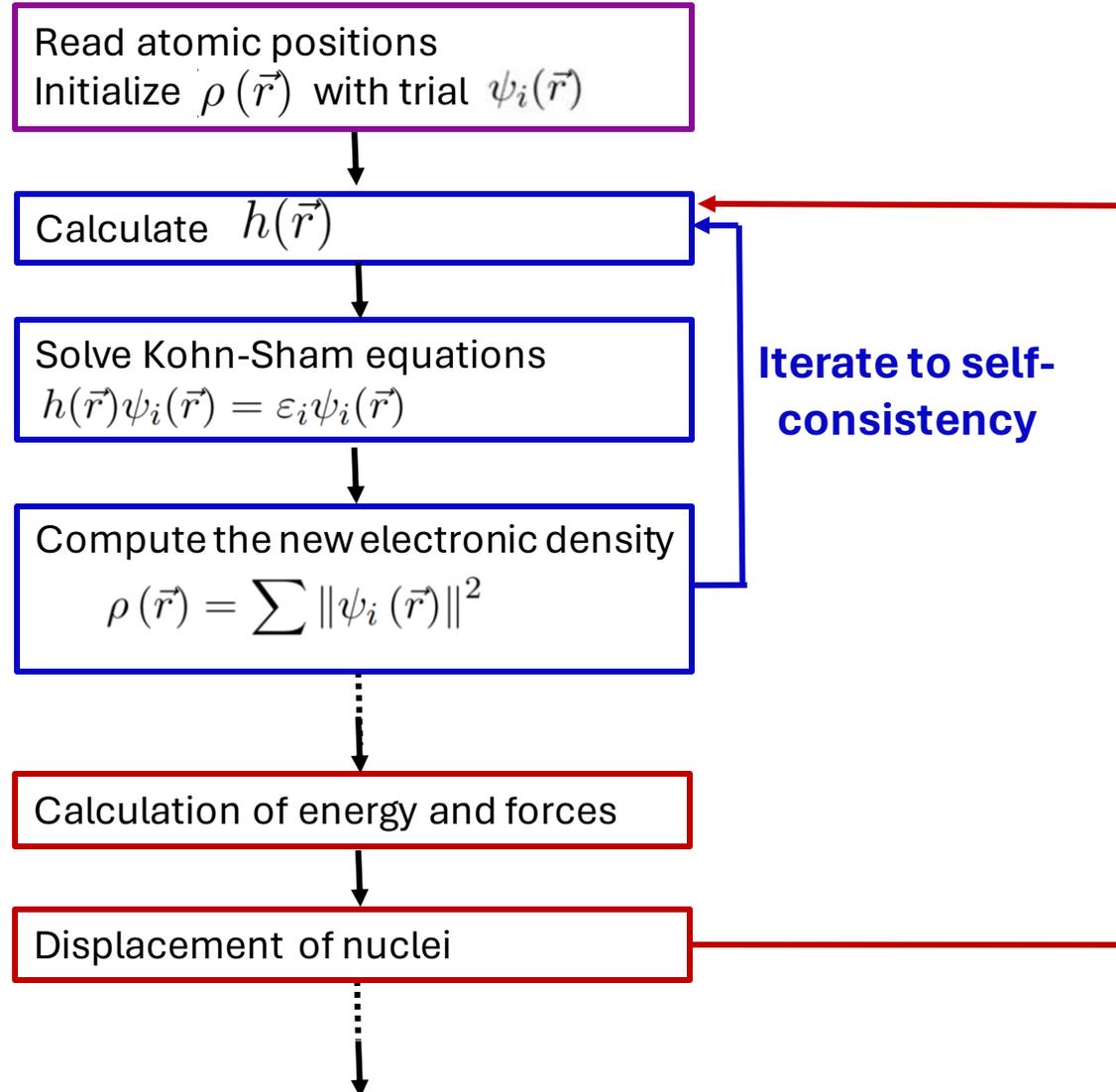
D. M. Ceperley and B. J. Alder, [Phys. Rev. Lett. 45, 566 \(1980\)](#)

**GGA** (Generalized Gradient Approximation): Group of methods based on the local electronic density + the gradient of the electronic density

Ex: J. P. Perdew, K. Burke, and M. Ernzerhof, [Phys. Rev. Lett. 77, 3865 \(1996\)](#)

# DFT in practice

## Simplified algorithm:



# DFT in practice

Many possible choices for *basis sets, functionals, diagonalization schemes etc.*

+ many different **codes!**



For most solid-state DFT codes:

- Periodic systems with sampling of the reciprocal space
- Valence electrons + ions
- Minimum radius around the atoms where the description starts to fail --> atoms too close do not interact correctly
- **$O(n^3)$  scaling**

- Best description available
- Can compute energy stored in defects through structure relaxation
- Can be used to build a database for ML potentials

- $O(n^3)$  scaling: limited to ~1000 atoms and 1000 steps
- Not suitable to simulate cascades...

## Simulation of atomic collisions

**Energy and force engine:**  
Empirical interatomic potentials

# Empirical potentials

- Quantum mechanical / electronic effects are **hidden** in an empirical potential of **interatomic** interactions
- Attractive part (model the bond) + repulsive part at short distance
- **Assumed shape** based on the **type of chemical bonding**
- Depend on atomic positions + a **few parameters** (~2-100)
- **Fitted on physical properties:** crystallographic structure, elastic constants, formation energy of defects etc.

• Based on the "physics of the material"

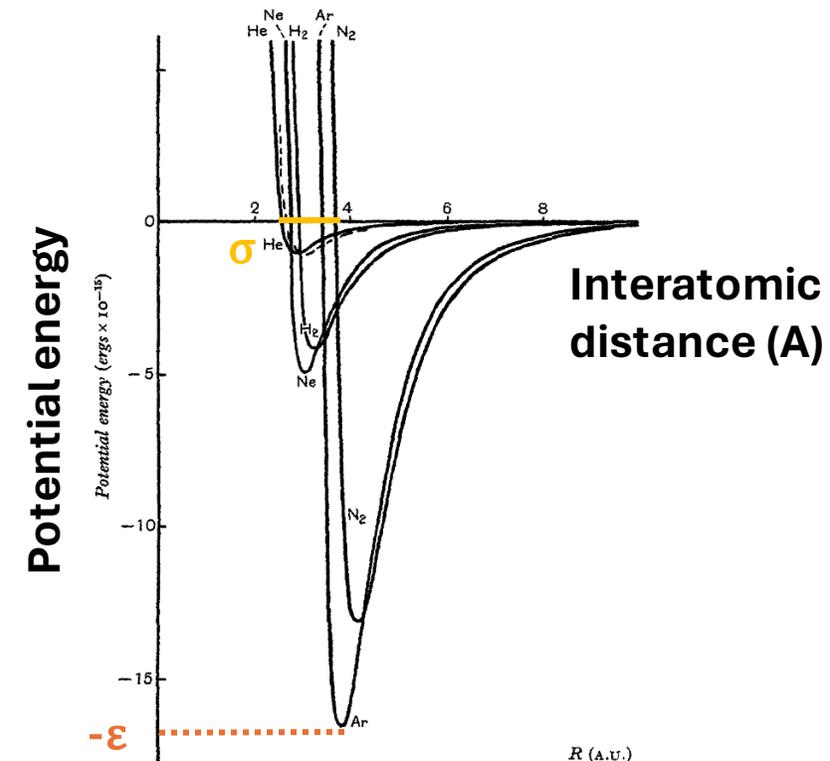
• Very fast: adapted for MD simulations of cascades

• Qualitatively accurate at best

• Lack of transferability

**Lennard-Jones potential** for van der Waals interactions in noble gas crystals  
(pair potential: depends only on the interatomic distance)

$$E_{LJ}(r) = 4\epsilon \left( \left[ \frac{\sigma}{r} \right]^{12} - \left[ \frac{\sigma}{r} \right]^6 \right)$$



The potential energy of pairs of inert gas atoms as a function of their distance apart in Å.

J. E. Lennard-Jones, *Proc. Phys. Soc.* **43**, 461(1931)

# MD simulations with empirical potentials

S. Sassi, et al., *Phys. Rev. D* 106, 063012 (2022)

Box of 4096 atoms @40mK

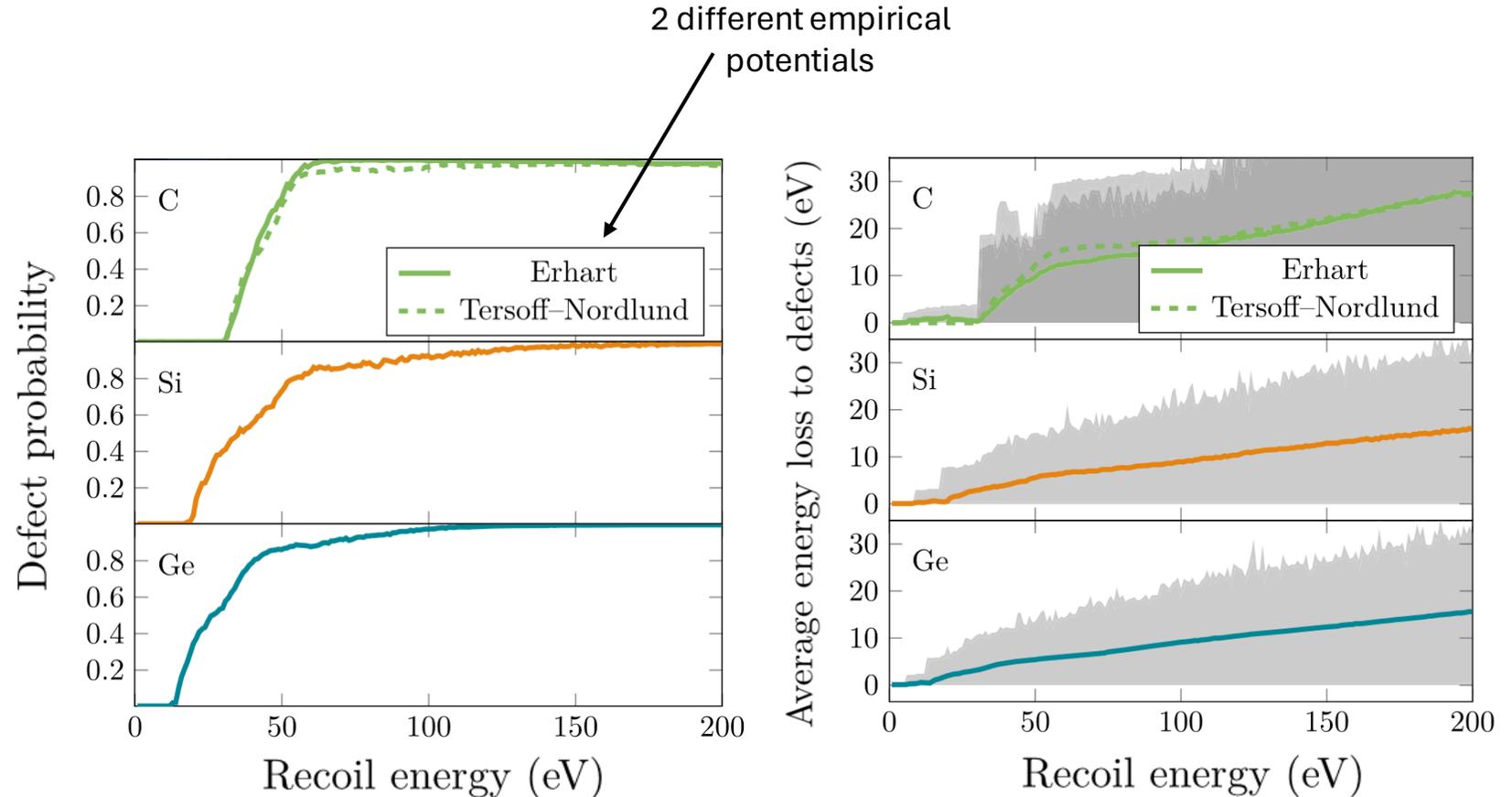
## Recoils:

- Atom in the central unit cell
- Random direction
- Energy 1-200eV (step: 2eV)

Border region with temperature control @40mK

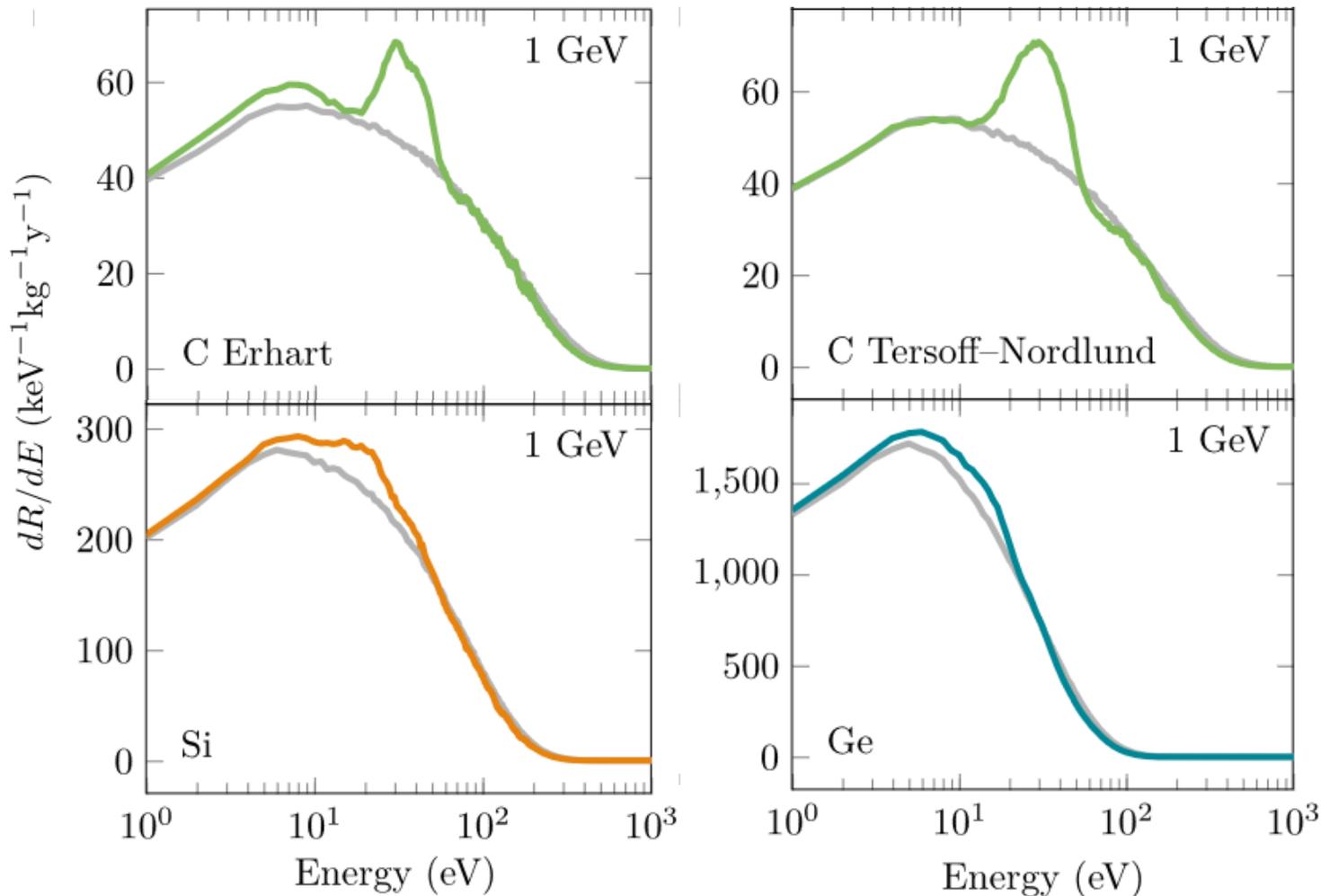
Simulation for 20ps

>1000 directions



# Impact on DM energy spectra

S. Sassi, et al., *Phys. Rev. D* 106, 063012 (2022)



For a **1 GeV** DM particle, assuming a spin-independent interaction

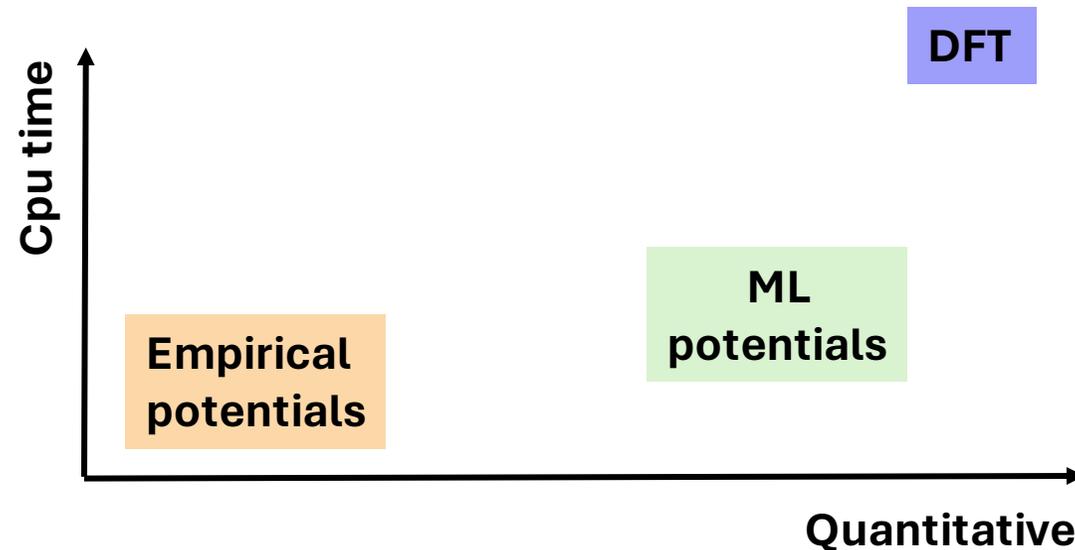
In **gray**, rate **without** energy stored in defect  
 In **color**, rate **with** energy stored in defect

**Distortion of the spectrum shape** at low energy

- In C (diamond detector)  
 Sharp displacement energy threshold --> peak
- For other materials:  
 Smooth distortion, degenerescence with research of Beyond Standard Model effects in CEvNS spectra

# Simulation of atomic collisions

## Energy and force engine: Machine Learning interatomic potentials

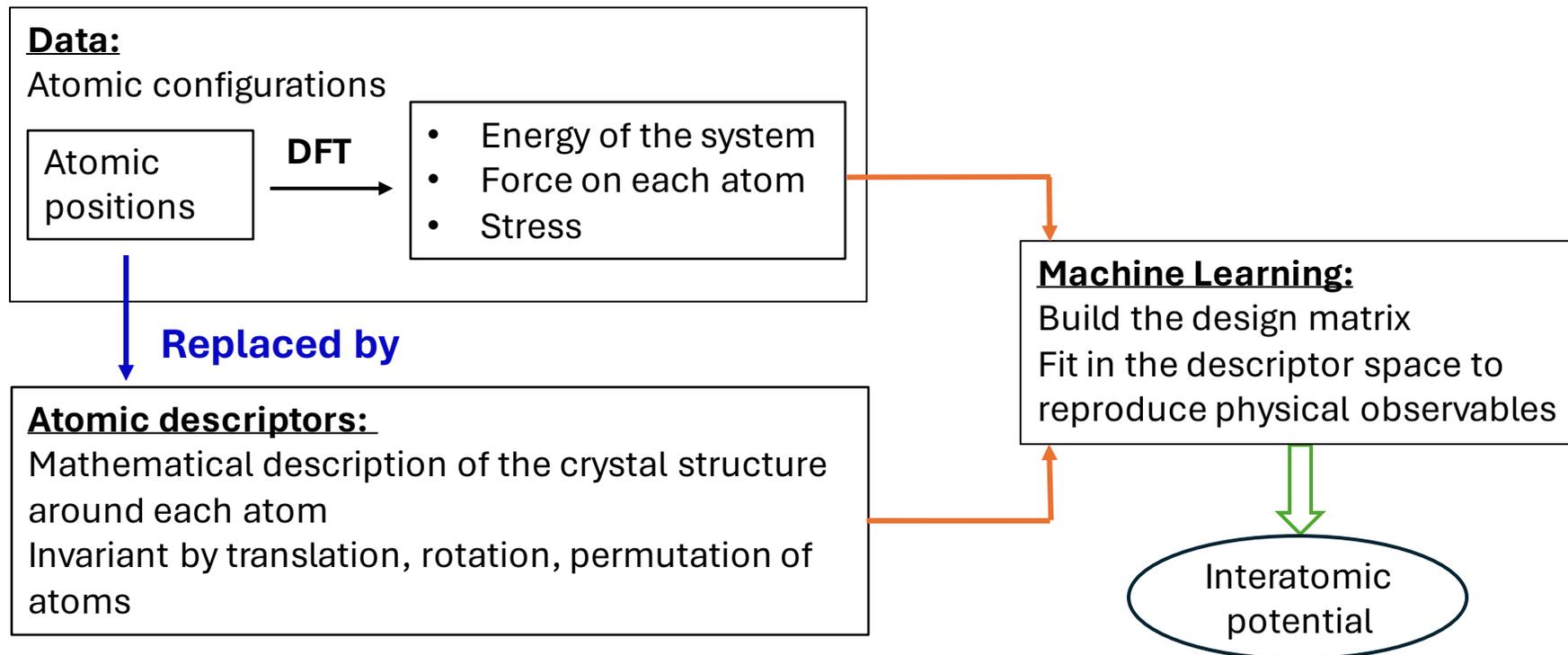


# Machine Learning Potentials

Gap between fast+less accurate empirical potentials and  $O(n^3)$  more accurate DFT methods

Idea: use database of atomic positions and forces (calculated with DFT) to design/fit a ML potential

Performance and accuracy determined by 3 equally important components



# ML Potentials: database

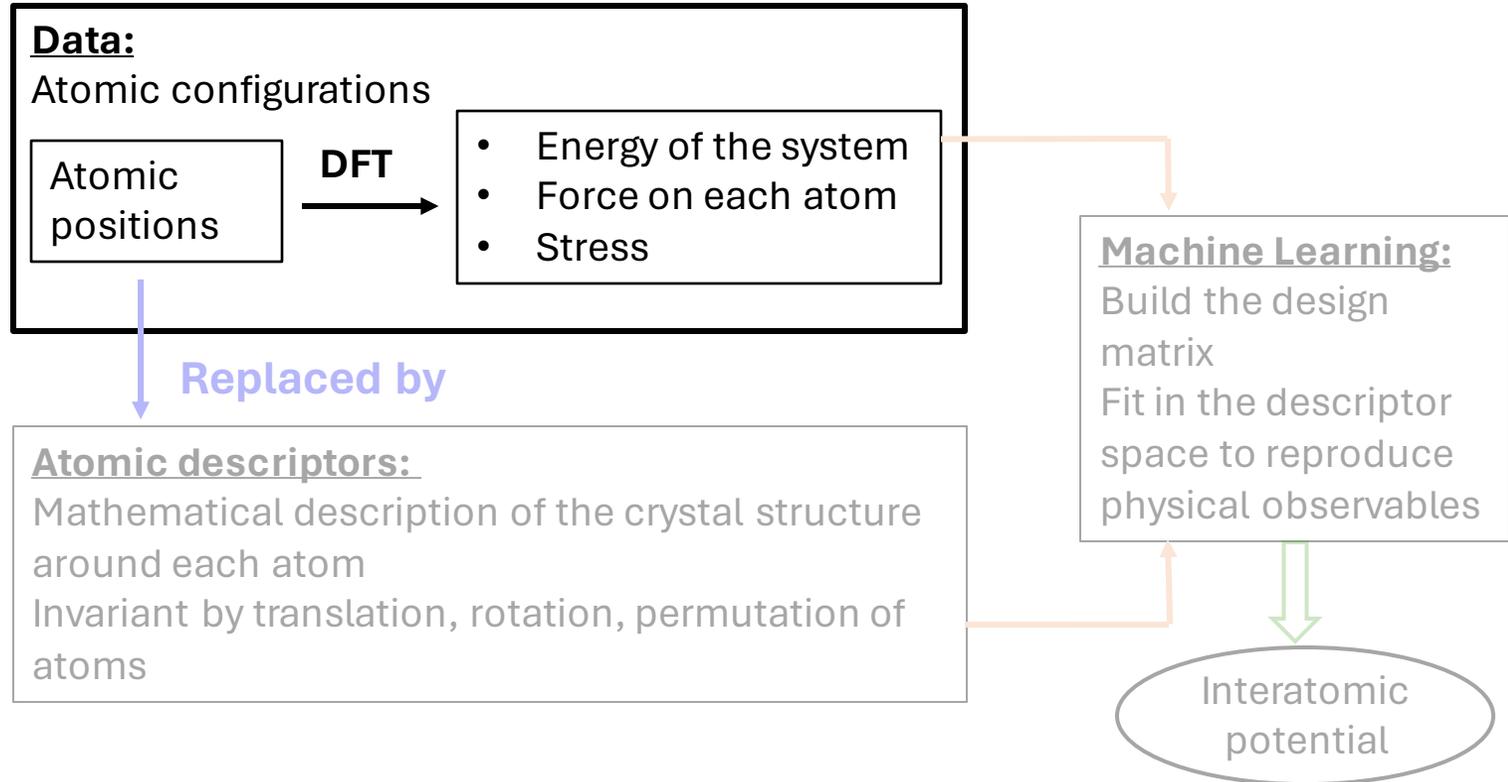
Content should be **adapted to the intended use of the potential**

*"art and science"* (Bartók)

Examples:

- Database with only diatomic molecules  
---> potential will depend only on the distance
- For **cascades**, database needs to include configurations with **very close atoms**

Lists of cartesian coordinates can look very different but represent very similar structures (e. g. through a symmetry operation)

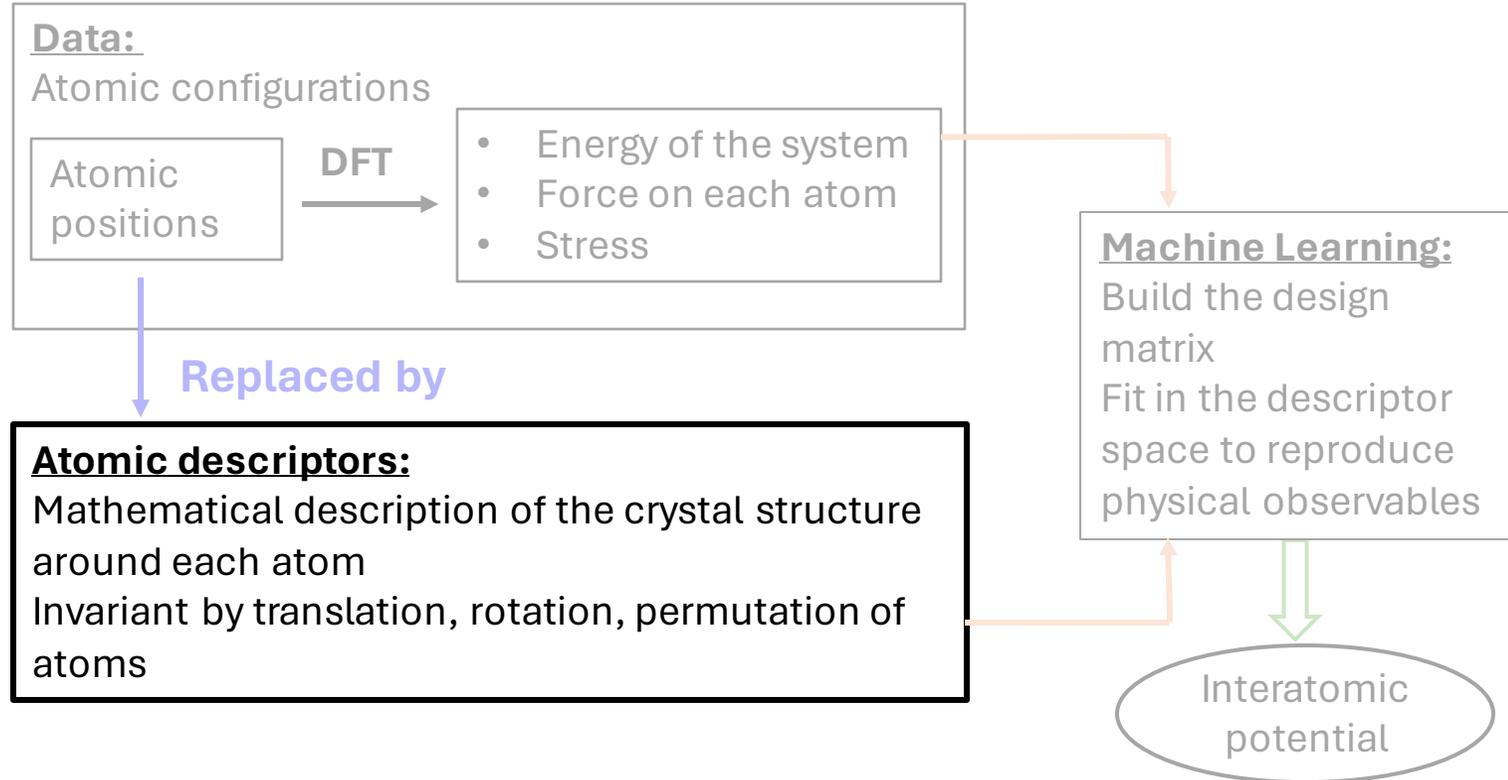


# ML Potentials: atomic descriptors

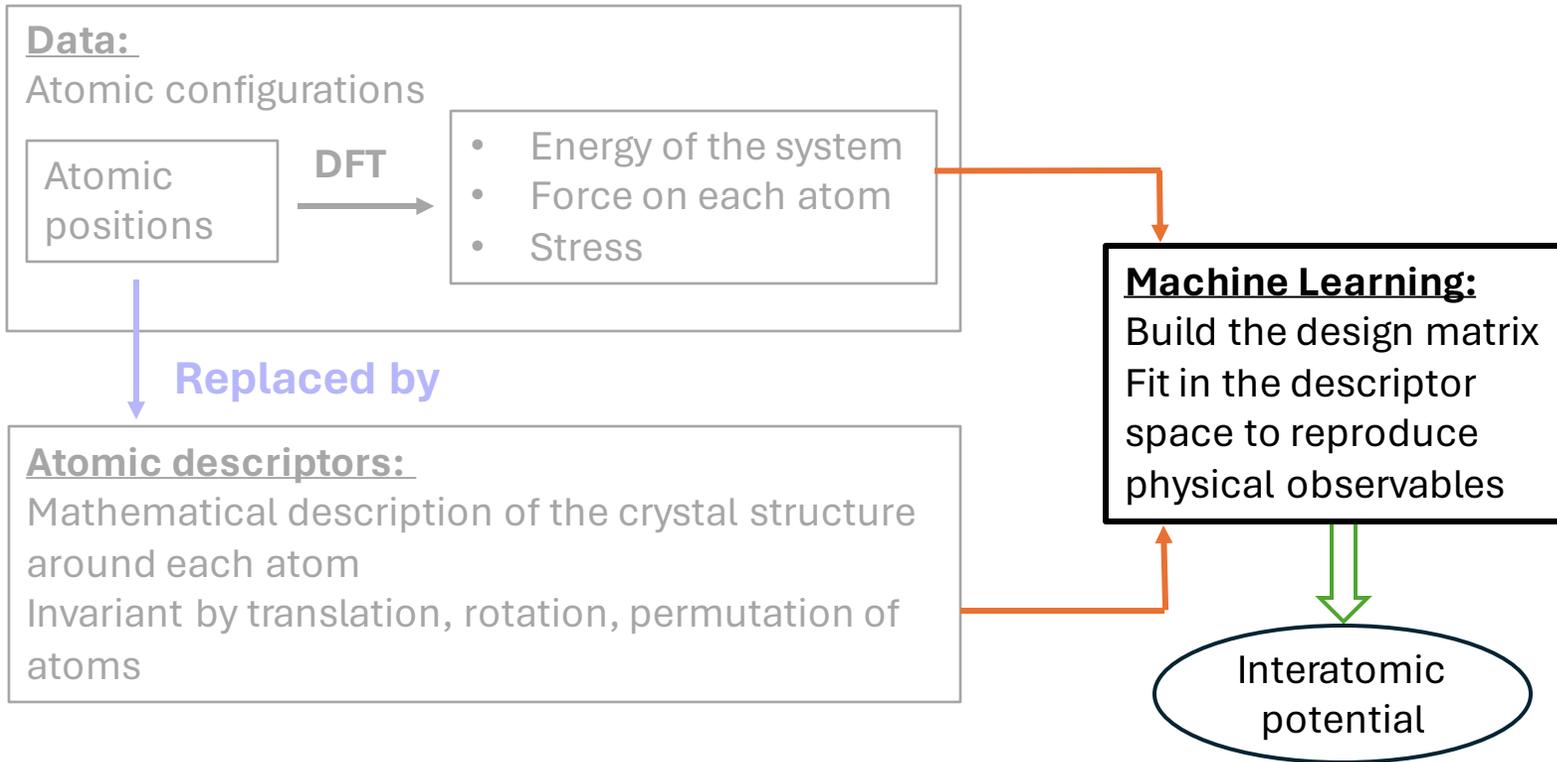
Describes each local atomic environment in each configuration

Many different possible descriptors:

- Distances/angles between atoms
- Spectral analysis of neighbor density
- Etc.
- Hybridization possible



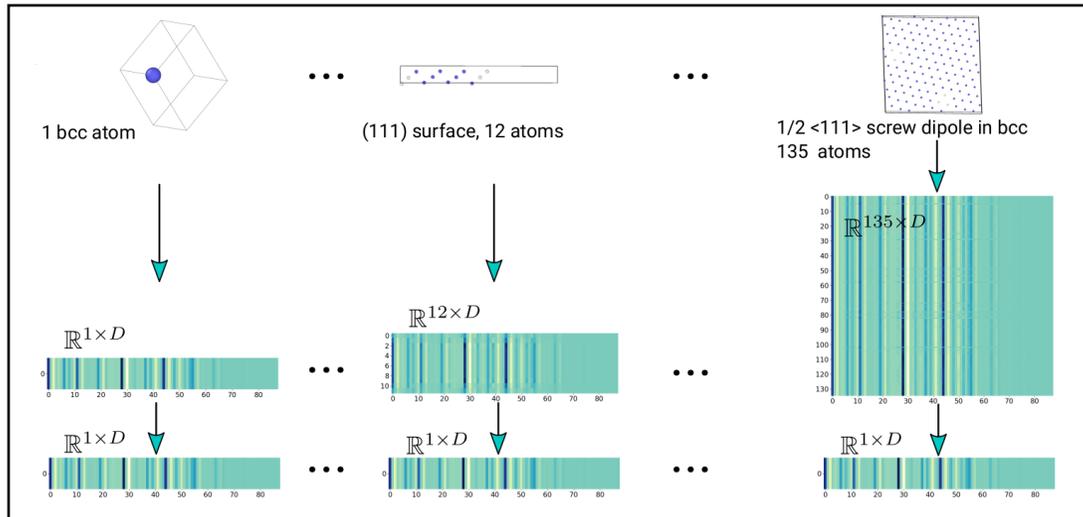
# ML Potentials: ML magics



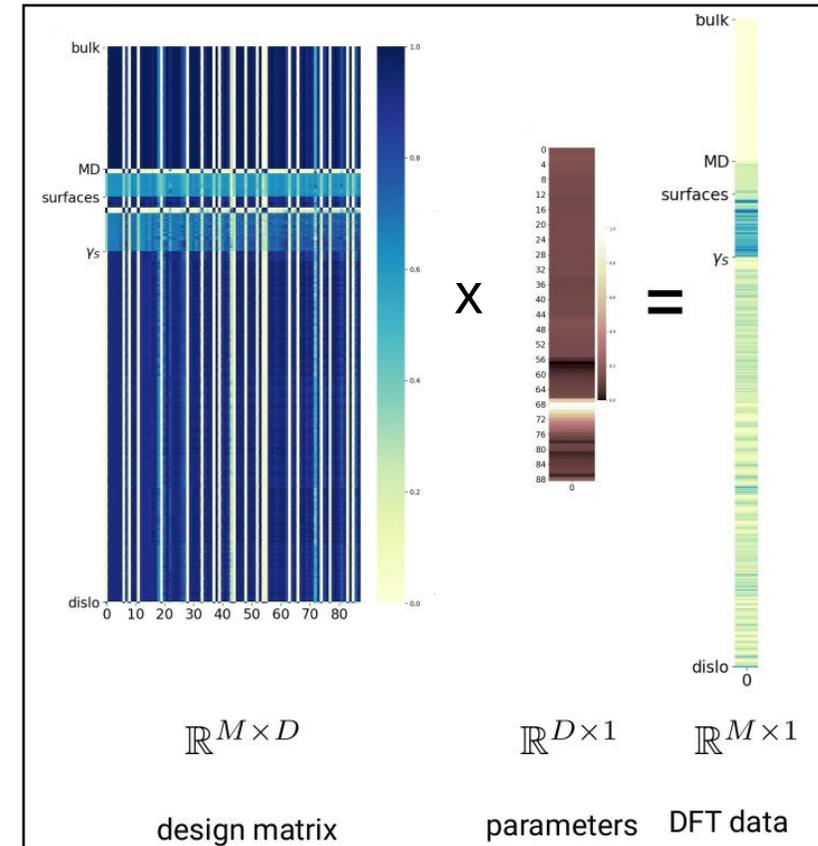
- Non-linear methods: "Kernel" methods, Neural networks
- Linear Machine Learning potentials:
  - > simpler, quicker
  - > training/test error higher
  - > good trade-off between accuracy and computational efficiency
  - > good choice for MD simulations

# Linear machine learning (LML) potential

## Summation of local atomic descriptors



## Linear regression in the descriptor space

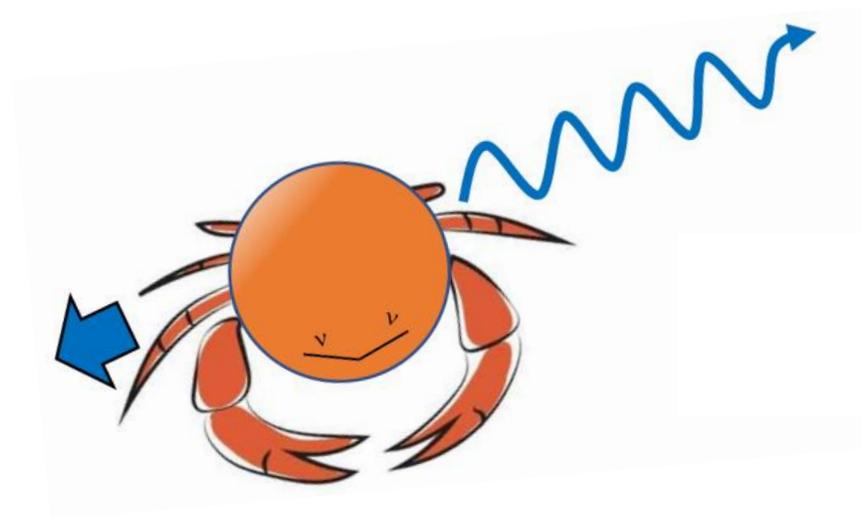


- Much faster than DFT
- Allow MD simulations
- Quantitative

- Slower than empirical potentials
- Lack of transferability
- Tricky

From M. C. Marinica, A. Goryaeva, et al.,  
CEA internal document, 2021

**A detailed example: CRAB**

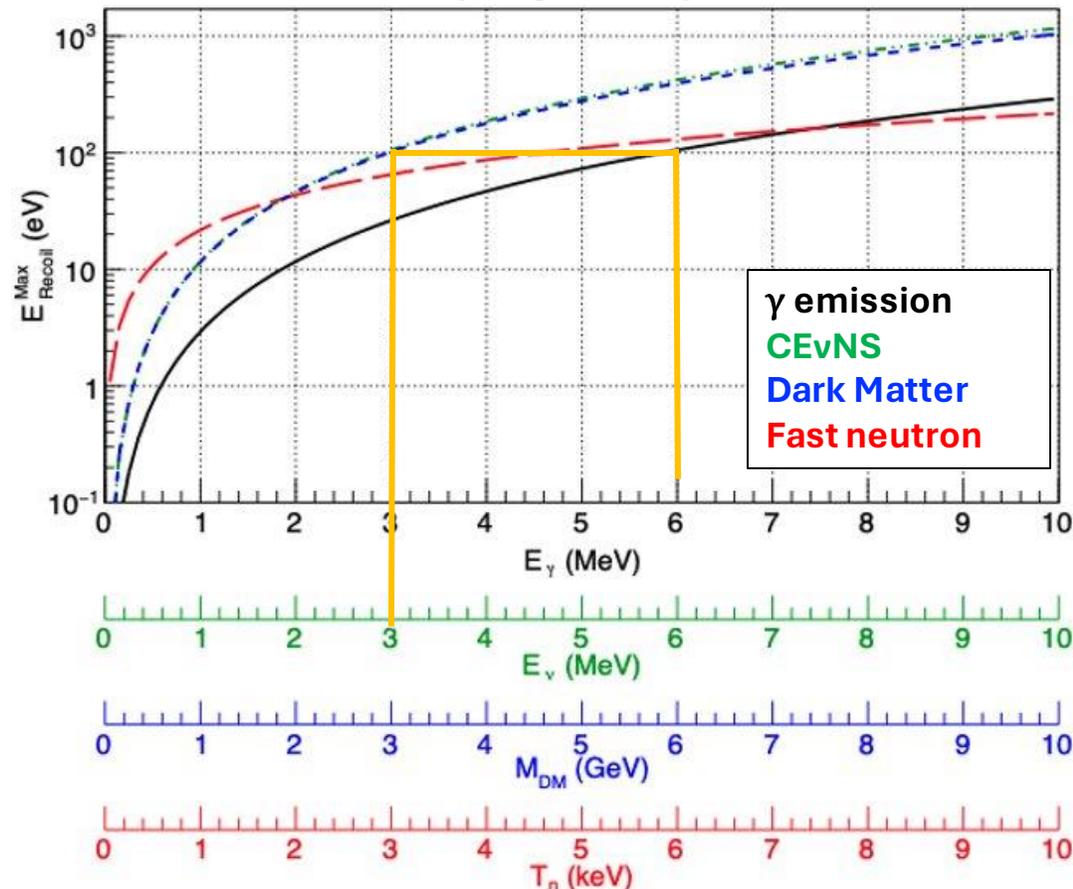


# The CRAB method

**Calibrated Recoils for Accurate Bolometry**

# 100eV nuclear recoils

Maximal recoil energies for various processes  
(target = W)



Sub-keV nuclear recoils are the expected signal for:

- **CEvNS with MeV neutrinos**
- **Direct detection of low-mass O(GeV) DM**



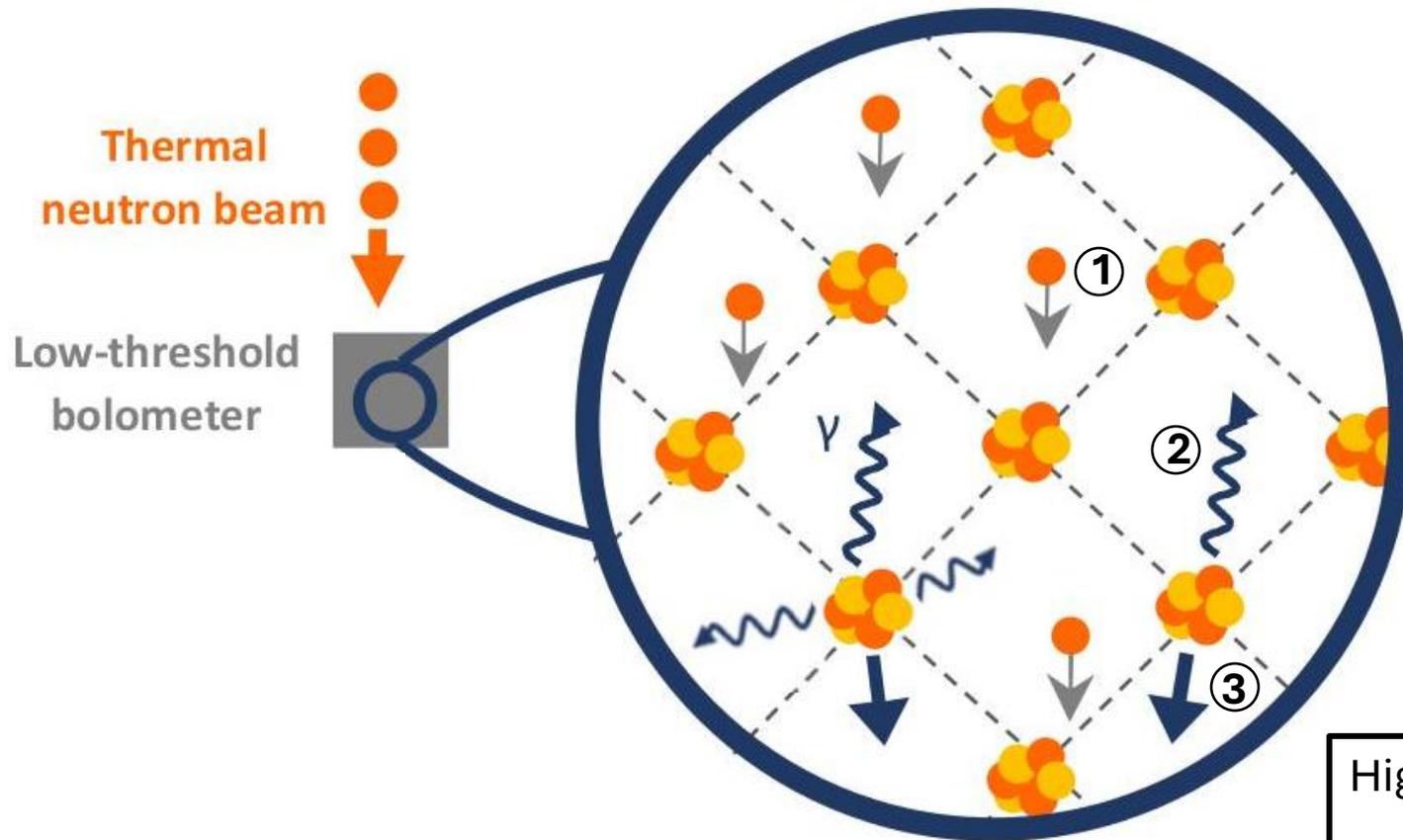
Understanding the sub-keV nuclear recoil signal is crucial for upcoming experiments searching for new physics

Calibrated sub-keV nuclear recoils from:

- **Elastic scattering of keV neutrons**
- **Nuclear recoils following MeV  $\gamma$  emission**



# CRAB calibration method



① **Thermal (25 meV) neutron capture**

② **Emission of a single- $\gamma$**  with energy  $S_n \sim 5-8\text{MeV}$   
Leaves the cm-size detector without energy deposition

③ **Well-defined recoil energy**  
(two-body kinematics)

$$\frac{E_{\gamma}^2}{2Mc^2} \sim 100\text{eV}-1\text{keV}$$

High-precision & all specifications of physics:

- Pure nuclear recoils
- In the bulk of the detector
- In the sub-keV region

# CRAB targets

Cryo-det crystal	Target Isotope	Nuclear Recoil (eV)	F.O.M. (Ab*σ*I*N)
Ge	<sup>70</sup> Ge	416.2	538
	<sup>74</sup> Ge	303.2	238
Si	<sup>28</sup> Si	1330	180
	<sup>28</sup> Si	989.9	589
Al <sub>2</sub> O <sub>3</sub>	<sup>27</sup> Al	1145	2871
CaWO <sub>4</sub>	<sup>182</sup> W	112.5	9608
	<sup>183</sup> W	160.3	1053
	<sup>186</sup> W	85.8	360

Suitable candidates have

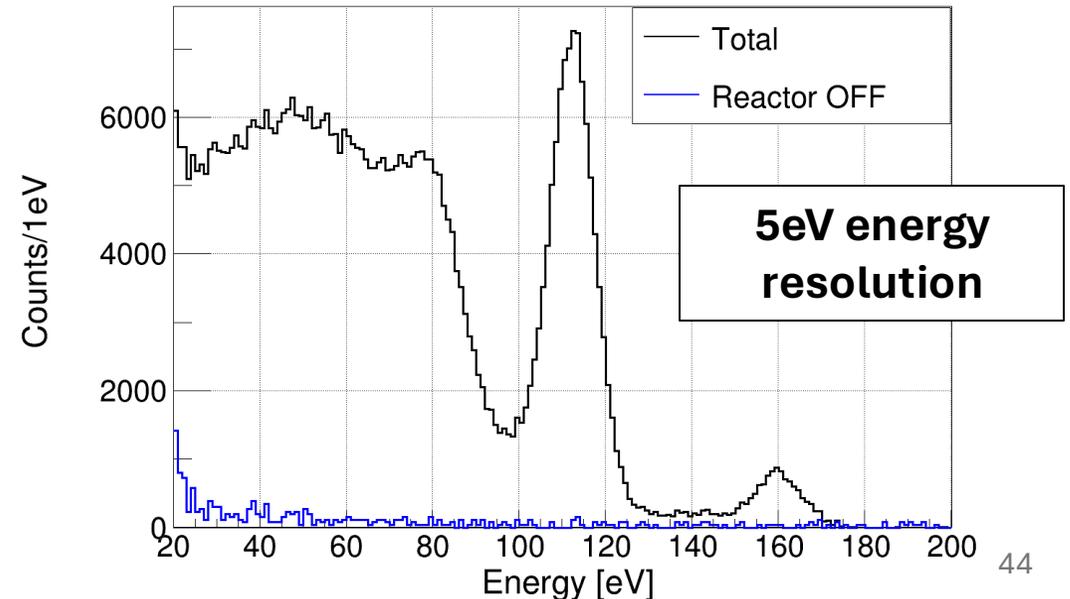
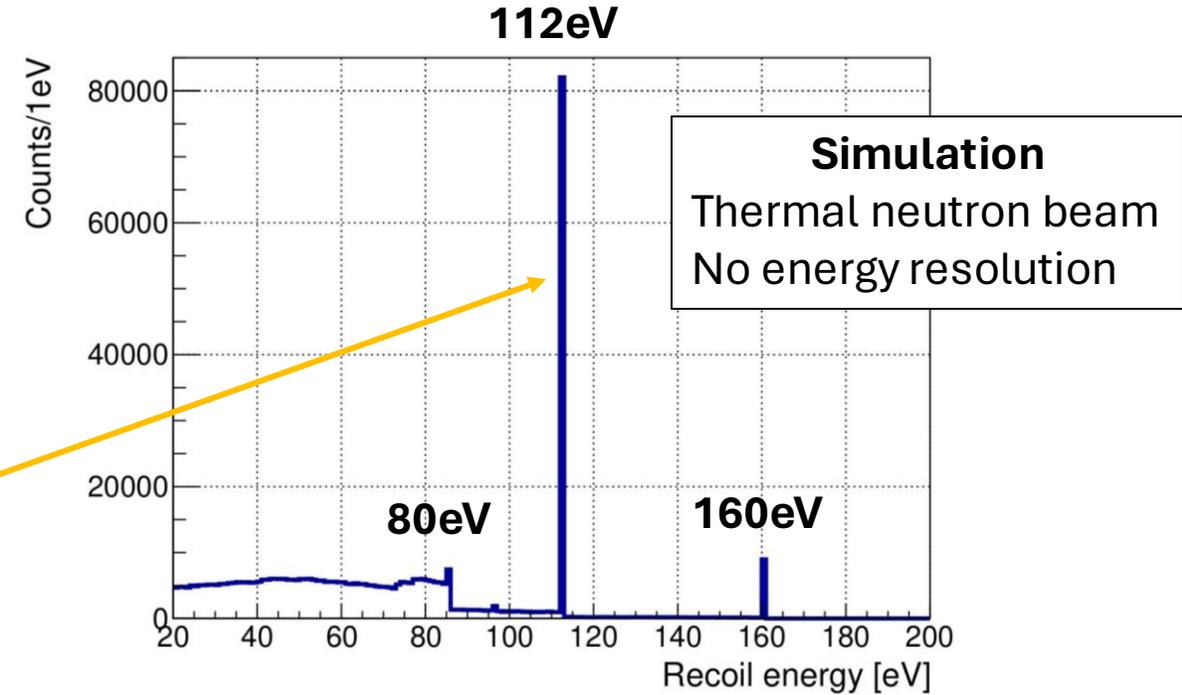
- High natural abundance
- Large neutron capture cross-section
- High branching ratio for single-γ transition
- Volumic number of target atoms in the detector

**The four main cryodetectors used in the DM/CevNS communities could be calibrated with CRAB**

# CRAB in CaWO<sub>4</sub>

Cryo-det crystal	Target Isotope	Nuclear Recoil (eV)	F.O.M. (Ab*σ*I)
CaWO <sub>4</sub>	<sup>182</sup> W	112.5	9608
	<sup>183</sup> W	160.3	1053
	<sup>186</sup> W	85.8	360

**Calibration peak @112eV prominent enough for a proof of concept measurement with defavorable background conditions**



# CRAB in CaWO<sub>4</sub>

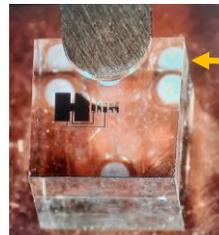


## Experimental validation

Significant fast neutron background  
 Agreement with simulation at  $6\sigma$   
 **$3\sigma$  significance**

0.75g **NUCLEUS**  
 CaWO<sub>4</sub> crystal

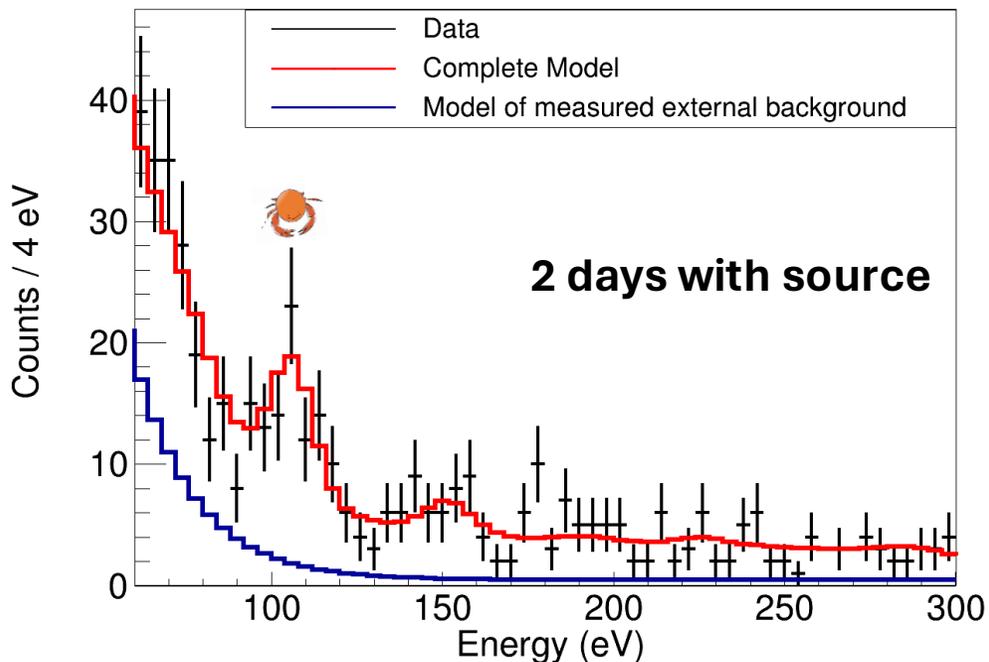
TES



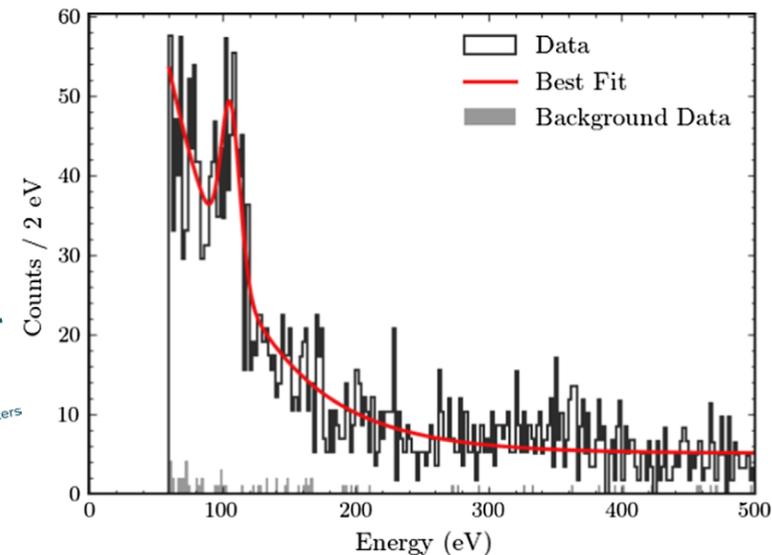
Baseline resolution:  $\sim 6.5\text{eV}$



Portable "thermal" neutron source:  
 3MBq <sup>252</sup>Cf fission source in moderators



**Independent confirmation**  
 On 3 CaWO<sub>4</sub> detectors  
 Significance  $>5\sigma$

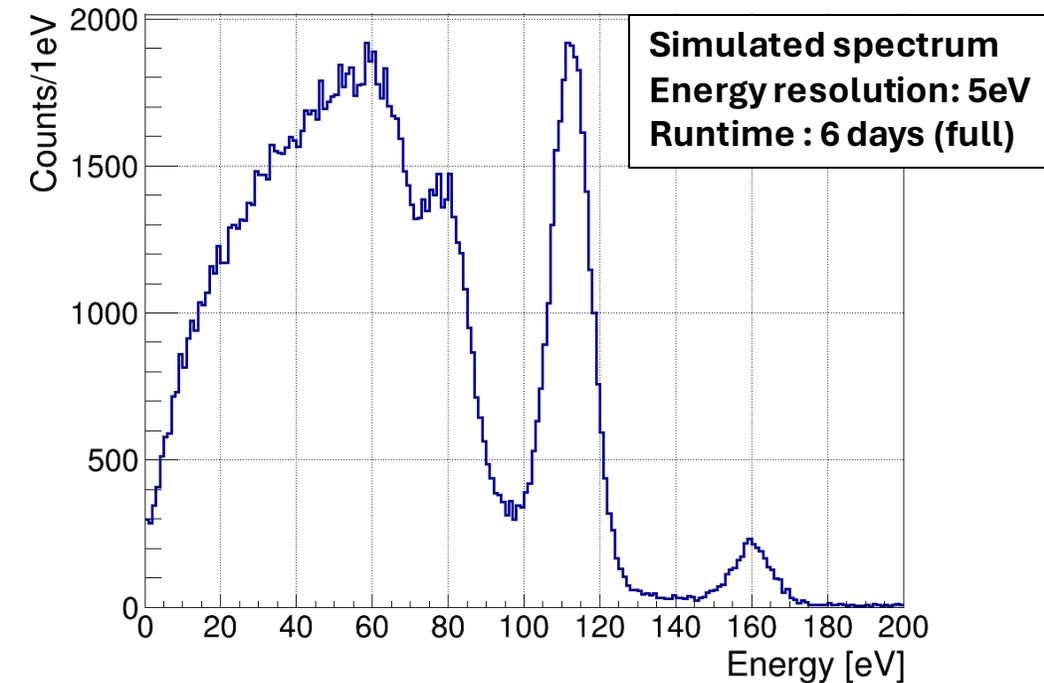
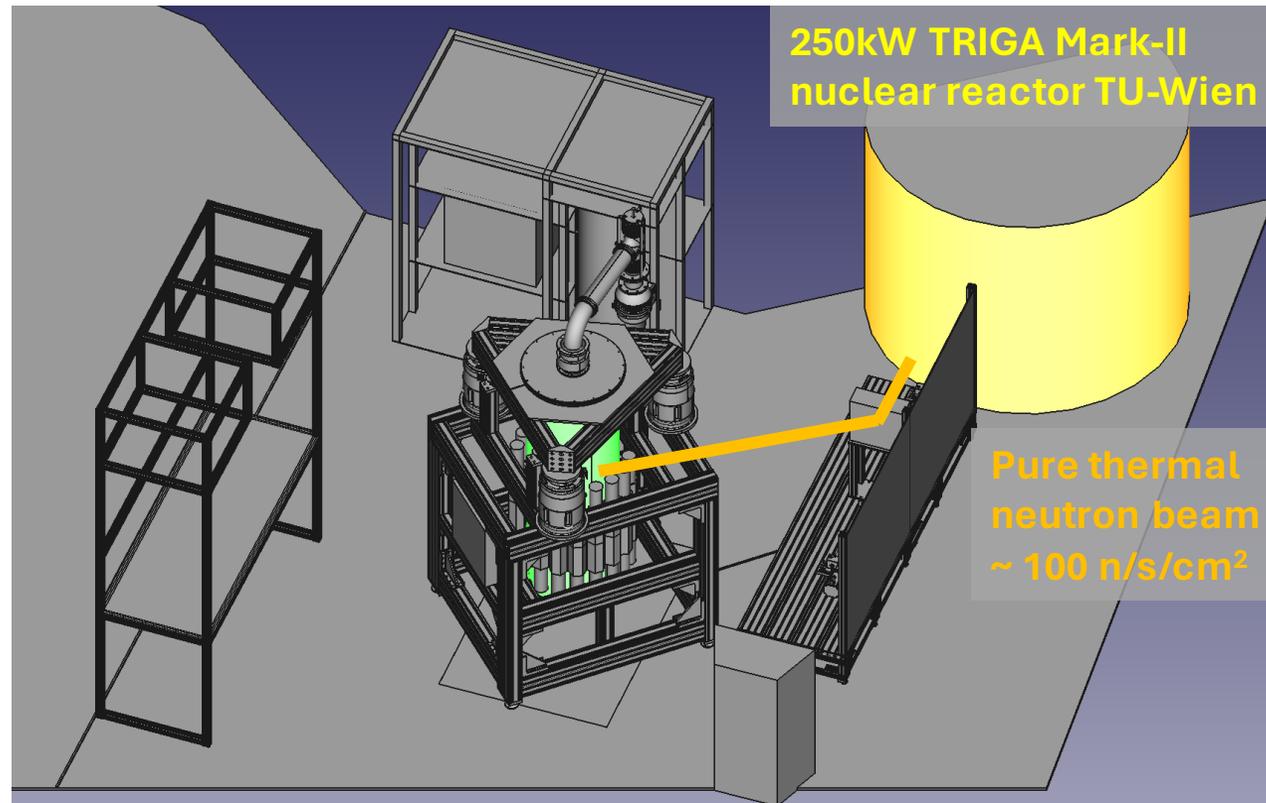


G. Angloher *et al.* (CRESST),  
*Phys. Rev. D* 108, 022005 (2023)

H. Abele *et al.* (CRAB&NUCLEUS), *Phys. Rev. Lett.* 130, 211802 (2023)

# High-precision CRAB on $\text{CaWO}_4$

*Later this year in Vienna*



- No fast neutrons background
  - Counting rate dominated by the CRAB process
- > high-precision and full use of the method potential

## A detailed example: CRAB

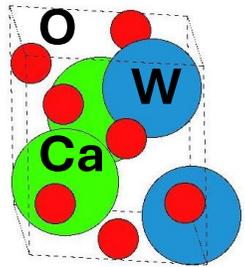
Towards MD simulations of 100eV nuclear recoils in  $\text{CaWO}_4$

# Calculate the energy stored in defects

For CRAB  $\sim 100$  eV recoils, we expect the created defects to be **close Frenkel pairs**

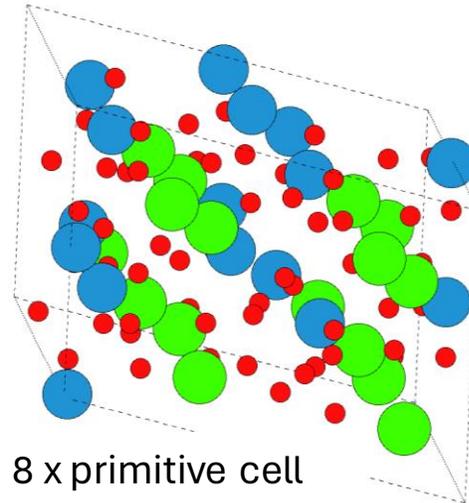
Idea:

- Build close Frenkel pairs in  $\text{CaWO}_4$  simulation "boxes"



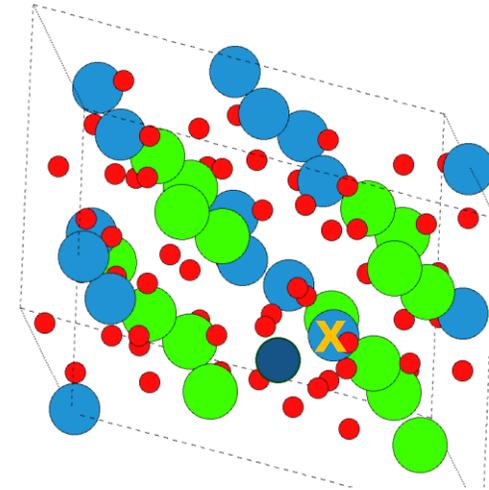
$\text{CaWO}_4$  primitive cell:  
2 **W**, 2 **Ca**, 8 **O**

Repeat in space  
→



8 x primitive cell

Place interstitial  
and vacancy  
→



**W interstitial** and neighbor **X** to remove to form a close Frenkel pair (distance: 2.9 Å)

- Calculate the energy of the box using DFT structure relaxation

Energy stored  
in defect

=

Energy of relaxed  
box with defect

-

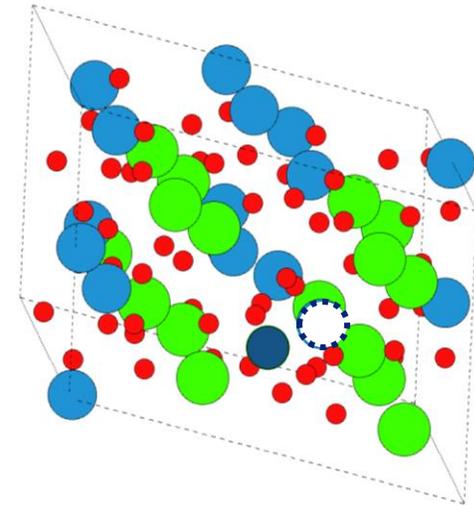
Energy of relaxed  
box without defect

# DFT formation energies

G. Kresse and J. Furthmüller, *Phys. Rev. B* 54, 11169 (1996)  
<https://www.vasp.at/>

Energy stored in the defect calculated "ab initio": DFT with VASP

Close Frenkel pairs	Ca	W	O
Ef (eV)	5.1	5.6 - 8.6	4.7 - 5.2

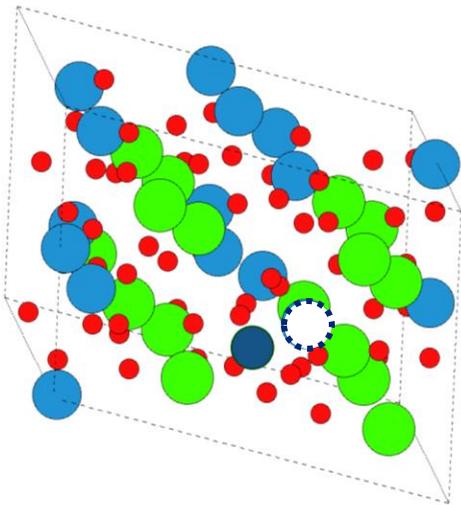


Not sufficient to derive the impact on the CRAB spectrum...  
 How many defects are created? FP?  
 --> MD simulations of CRAB recoils

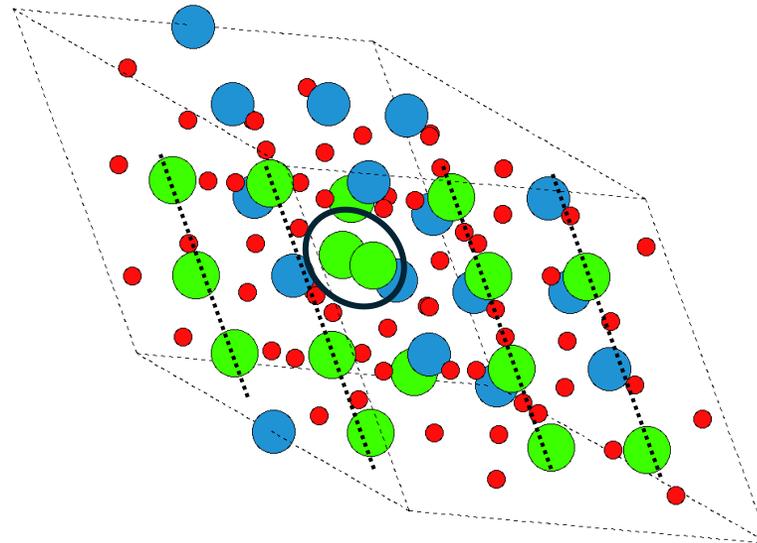
# Build a database to train a LML potential

**Database** from DFT calculations: 348 configurations

Configurations with minimized forces (structure relaxation)  
+ configurations before the relaxation (non-zero forces)

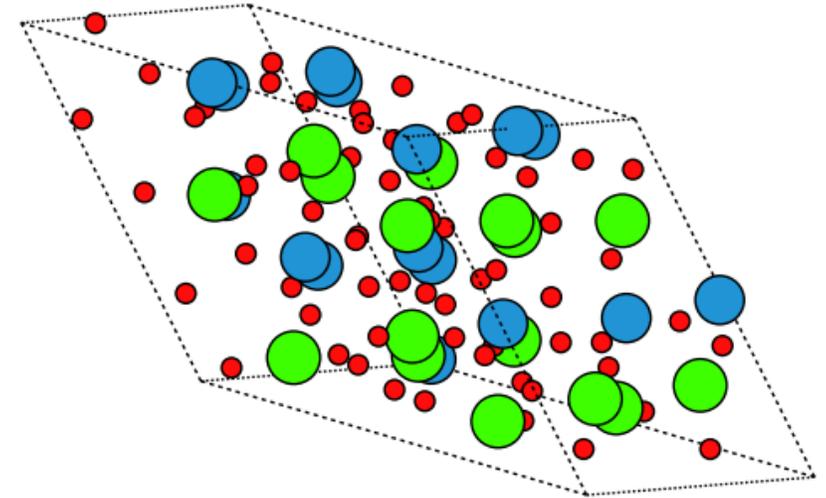


Close Frenkel pairs in a 96-atom box/ in a 324-atom box



Pairs of close atoms in a 96-atom box

Molecular Dynamics @2000K  
for the 96-atom box



# Short-range interactions

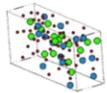
- VASP description fails at very short distances (around 1-1.5 Å)  
--> database does not describe short-range interactions
- For cascades, the LML potential is coupled at very short distances with the "ZBL" potential (2-body)

$$E_{ZBL}(r_{ij}) = \frac{Z_i Z_j}{r_{ij}} \sum_{k=1}^4 c_k \times \exp\left(-\frac{b_k r_{ij}}{a}\right)$$

# First tests of the potential in MD

## Simulation

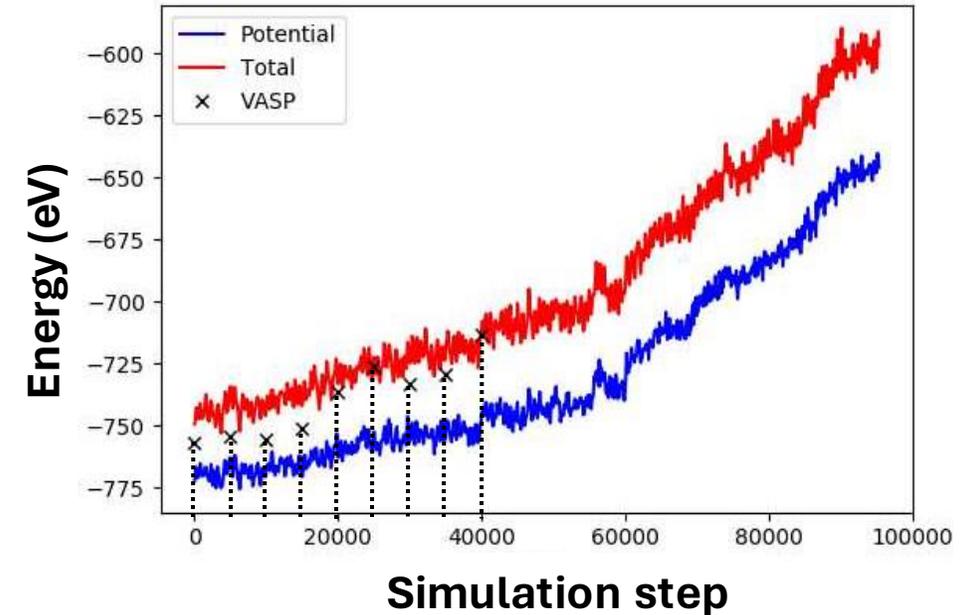
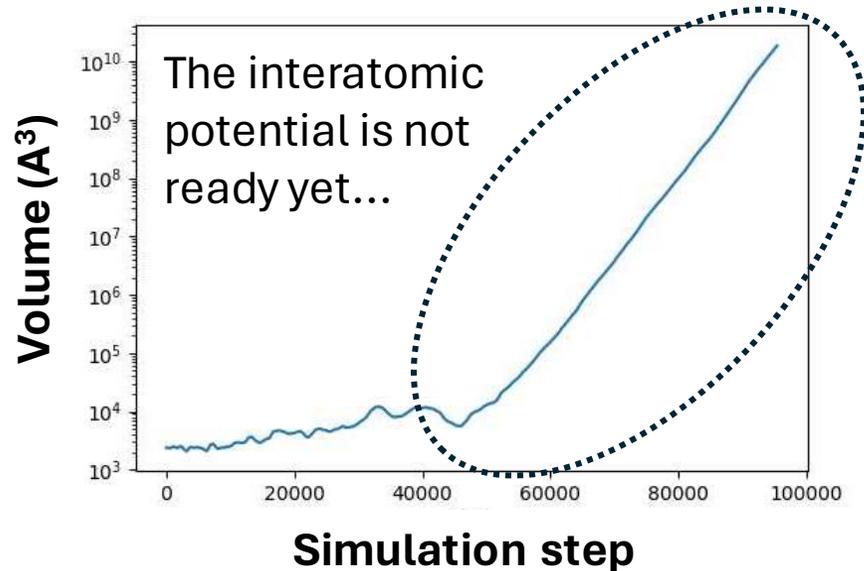
- Box with 96 atoms
- Equilibrium for 100ps @2000K (*not shown*)
- **Temperature increased from 2000K to 5000K** in 300ps



# First tests of the potential in MD

## Simulation

- Box with 96 atoms
- Equilibrium for 100ps @2000K (*not shown*)
- **Temperature increased from 2000K to 5000K in 300ps**



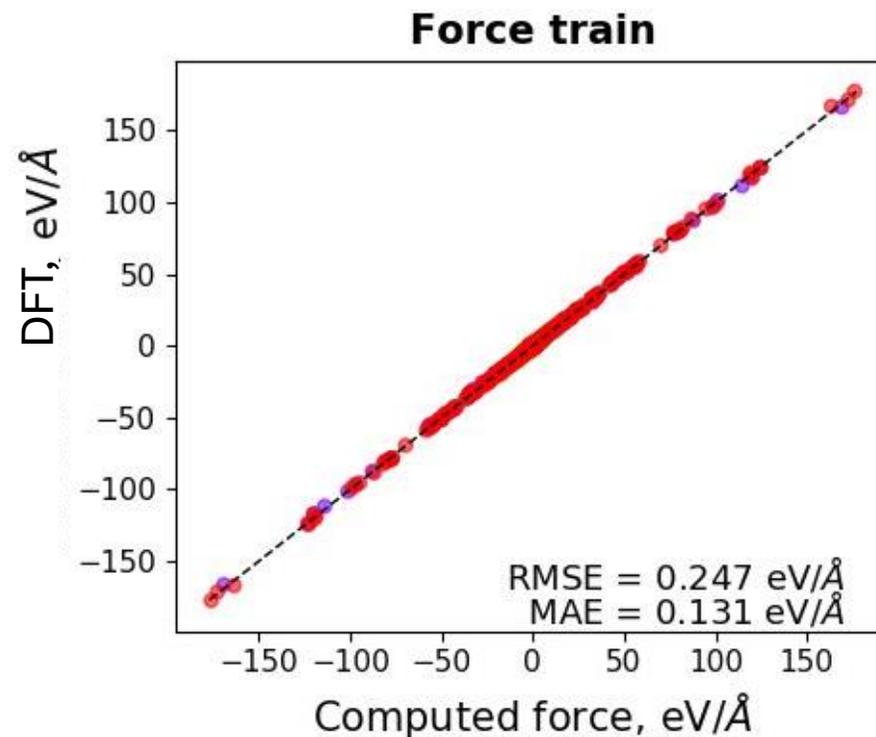
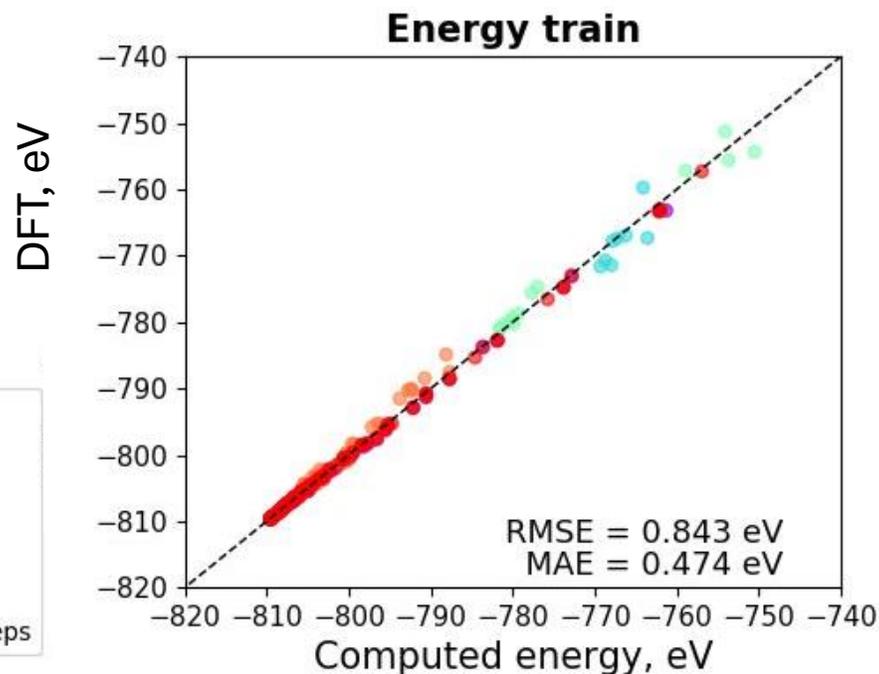
9 configurations extracted from the simulation  
Energy/forces recalculated in DFT

--> added to the training database  
--> test of the new potential

# Quality of the final potential

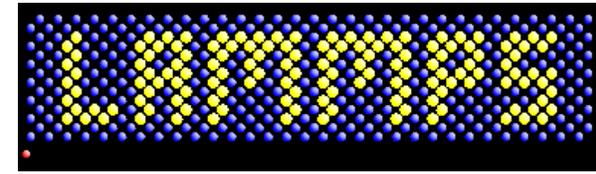
Good fit of the database

Mean errors of the order of magnitude expected for a LML potential

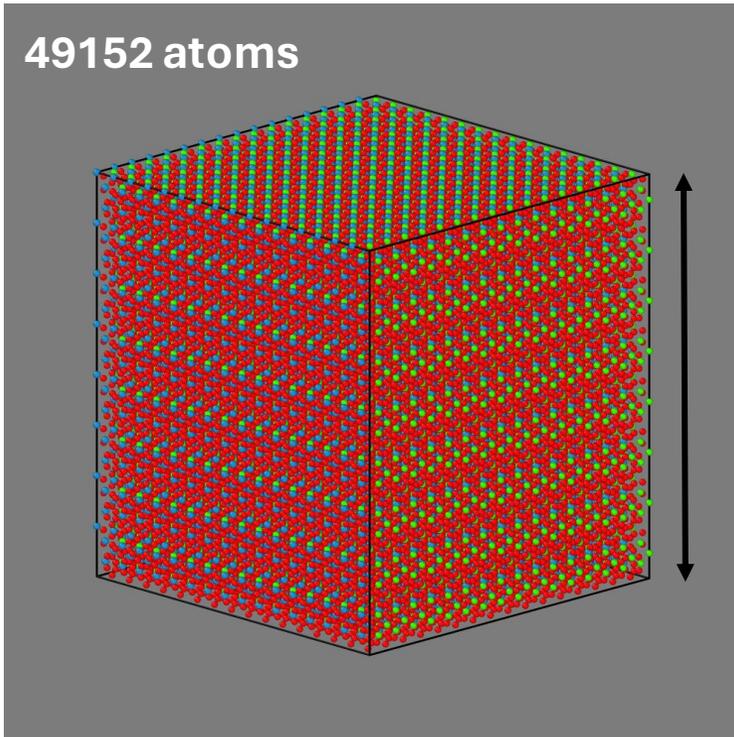


Mean test errors are similar

# Cascades in molecular dynamics



A. Thompson, et al., *Comput. Phys. Commun.*, 271:108171 (2022)



~ 80Å

1. Give an initial velocity to the central W atom corresponding to
  - **81eV**
  - **112eV**
  - **160eV**(main CRAB peaks)

2. Follow the movement of all atoms for ~8ps  
Final thermal heating: ~8K (for 112eV)

3. Relaxation of the structure

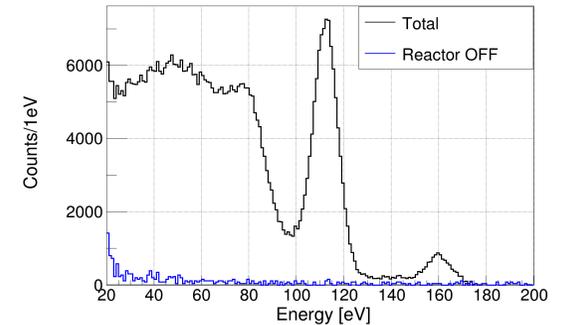
Stored energy  
due to cascade

=

Energy of relaxed  
box

-

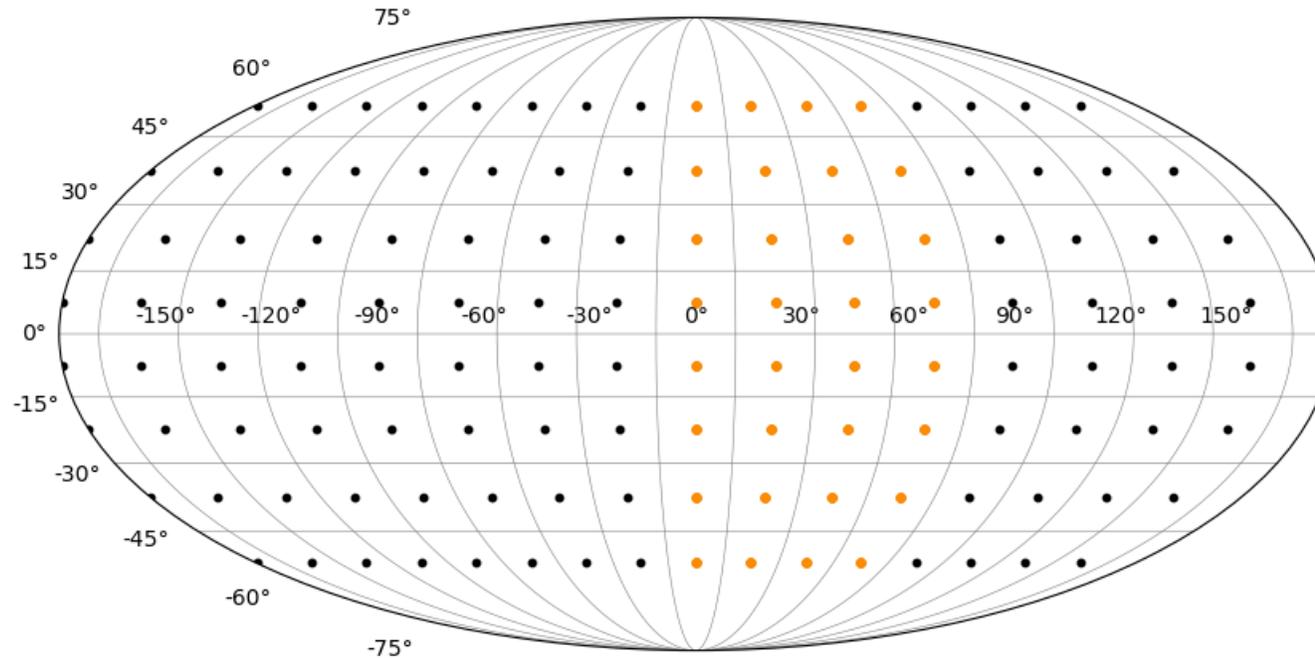
Energy of box before  
cascade



**Zero-point energy** is taken into account :

- through a classical equivalent : non-zero temperature J. Li, [PhD thesis, MIT, 2000](#)  
 $3/8 \times T_D = 133K$  for  $CaWO_4$
- simulation @ 133K and save 10 intermediate configurations  
= 10 sets of initial positions for each cascade

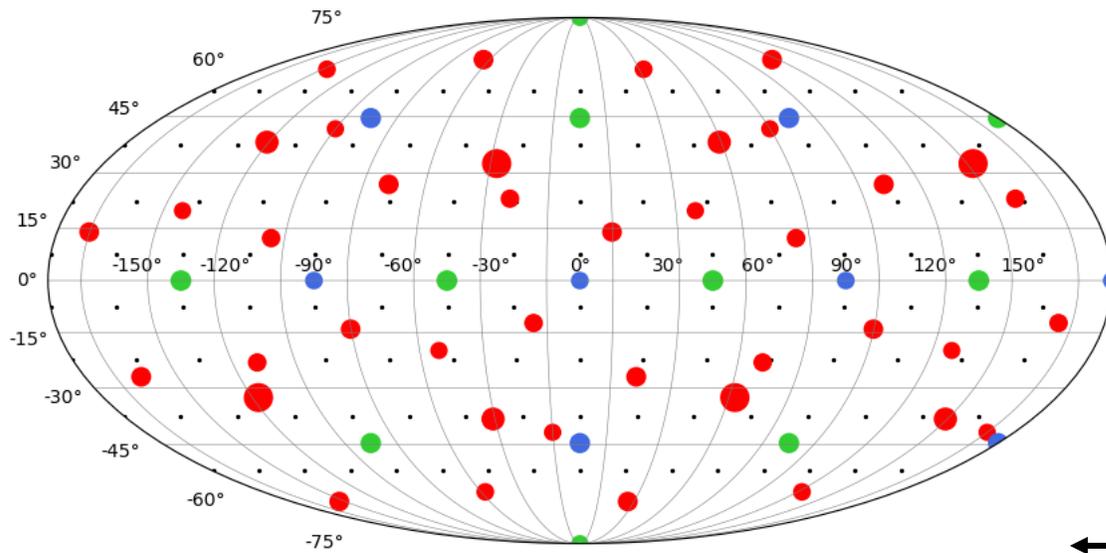
# Cartography of the energy stored in defects



- W on a position with point symmetry  $\bar{4}$  --> reduces the study to  $\frac{1}{4}$  of the sphere
- Solid angle (almost) regularly sampled with **32 recoil directions**

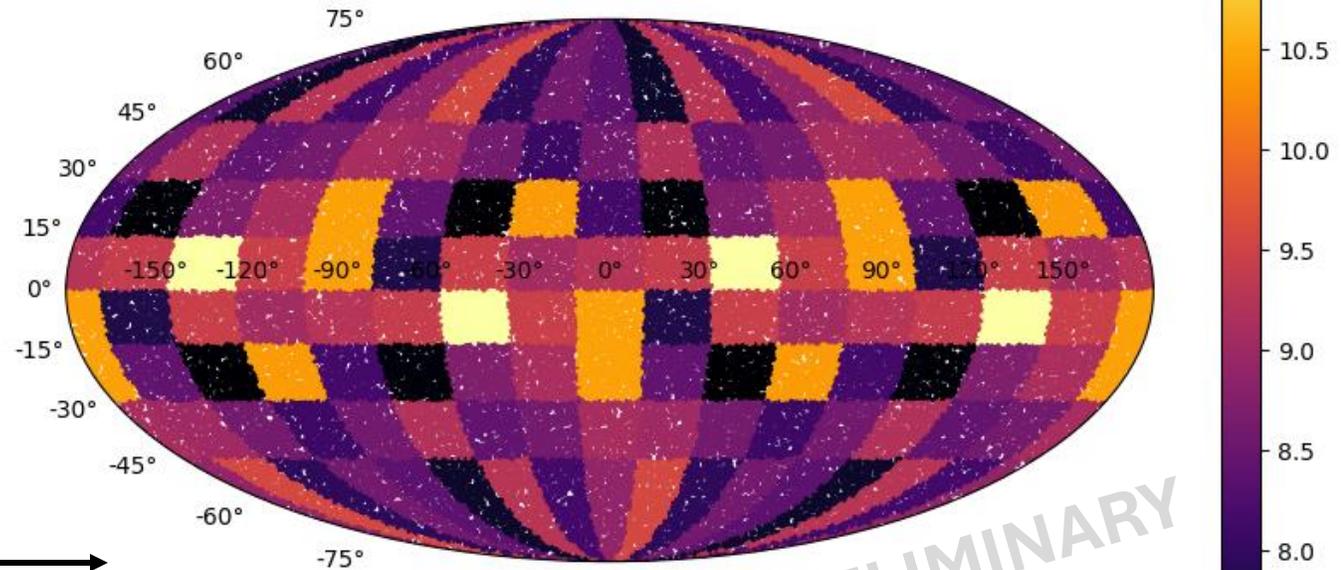
# Cartography of the energy stored in defects

Mollweide projection of the **crystal structure** of the ball centered at the projectile (W) and with radius 6 Å



Colors **Ca** **W** **O**  
Size  $\propto 1/d$

Average (10 initializations) energy stored in defects for each initial direction  
**112eV** cascade

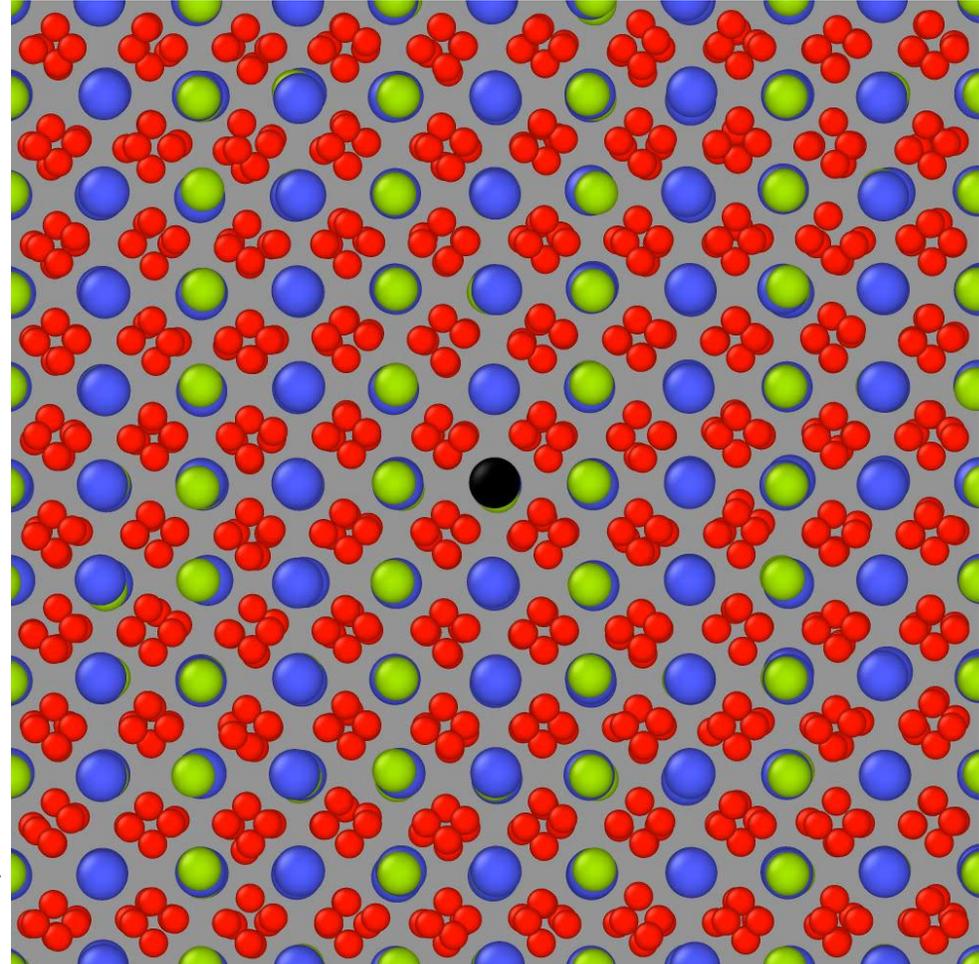
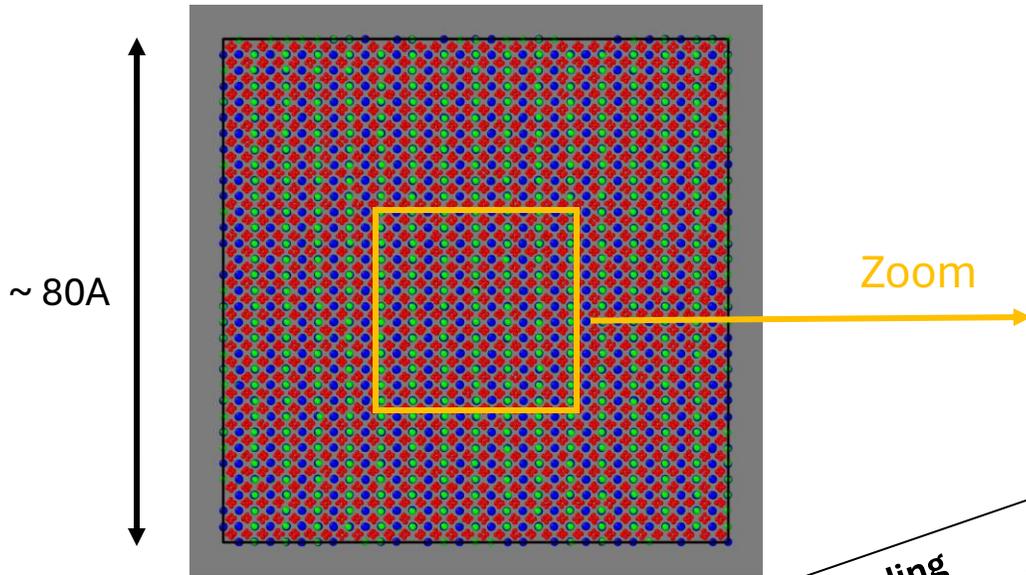


↔  
No clear  
relationship

PRELIMINARY

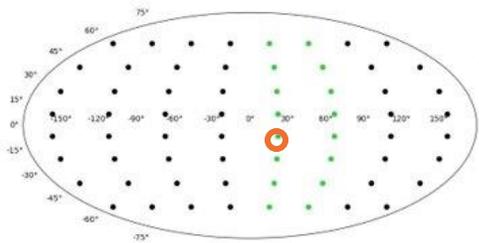
# 8.5ps movies: A 112eV cascade in CaWO<sub>4</sub>

Sectional view around the  
PKA location (depth: 20 Å)



With the PKA in black

Ca W O

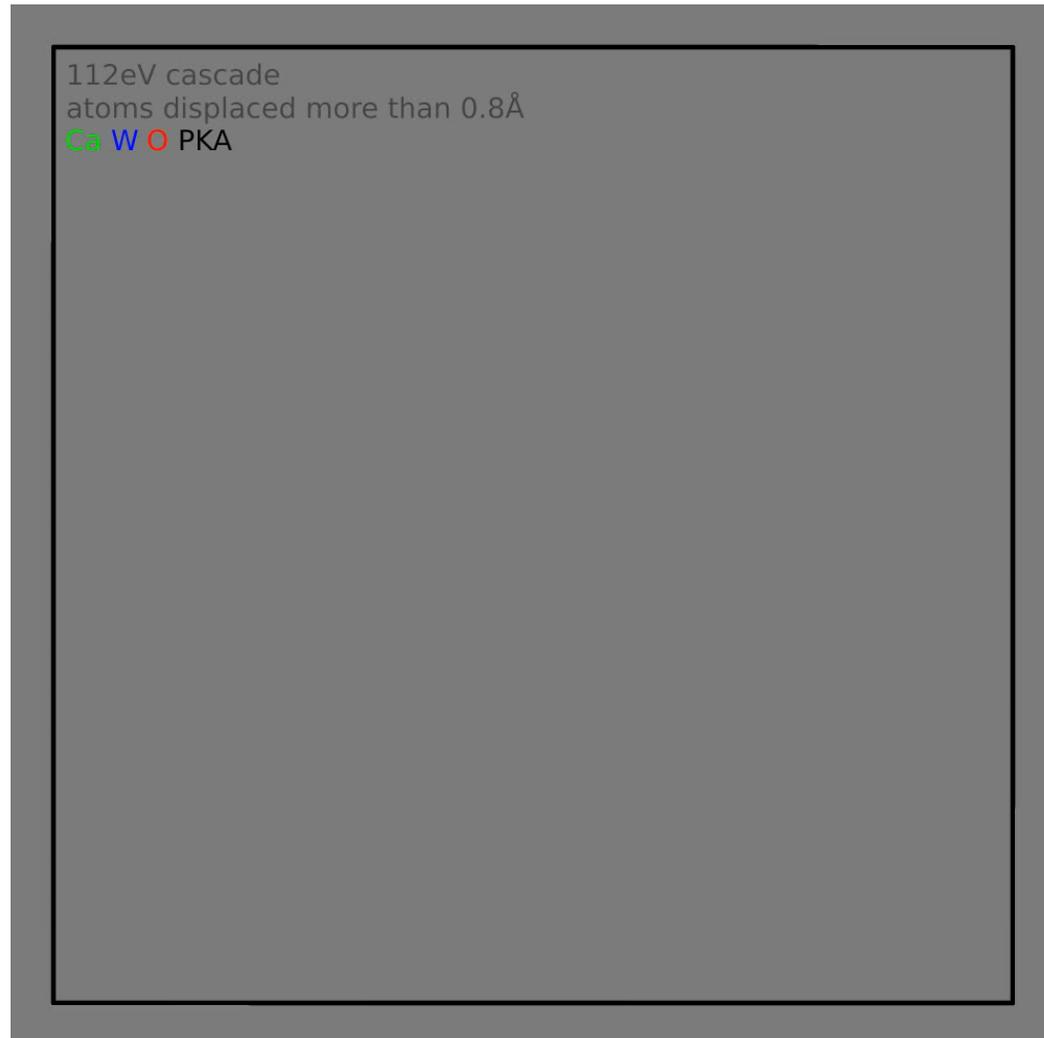


The PowerPoint version including  
animations and videos can be found at:  
[https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329\\_atomic\\_collisions\\_in\\_bolometers.pptx?rlkey=0fdiy&tjiu2kg3aj7dw4wokvz&dl=0](https://www.dropbox.com/sc/fi/mzhmhn3t7xd6n76ukve4x/20240329_atomic_collisions_in_bolometers.pptx?rlkey=0fdiy&tjiu2kg3aj7dw4wokvz&dl=0)

# 8.5ps movies: A 112eV cascade in $\text{CaWO}_4$

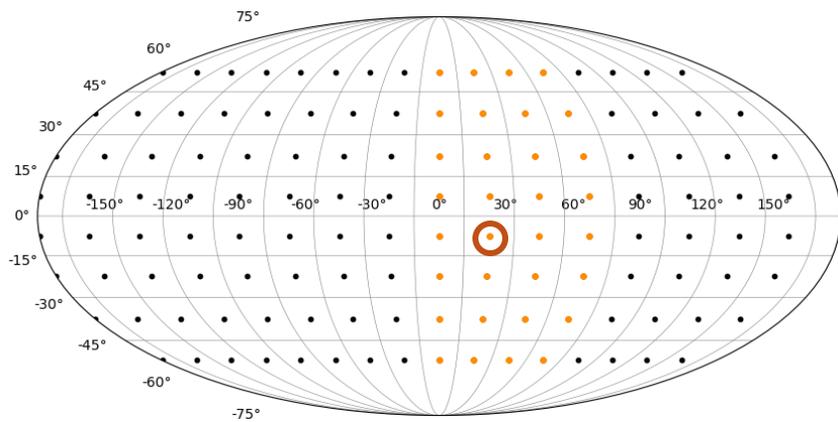
Same cascade  
Showing only the atoms displaced  
by more than 0.8Å

**The PowerPoint version including  
animations and videos can be found at:**  
[https://www.dropbox.com/scl/fi/mzhmhn3t7xd6n76ukve4x/20240329\\_atomic\\_collisions\\_in\\_bolometers.pptx?rlkey=0fdjy8tjiu2kg3aj7dw4wokwz&dl=0](https://www.dropbox.com/scl/fi/mzhmhn3t7xd6n76ukve4x/20240329_atomic_collisions_in_bolometers.pptx?rlkey=0fdjy8tjiu2kg3aj7dw4wokwz&dl=0)



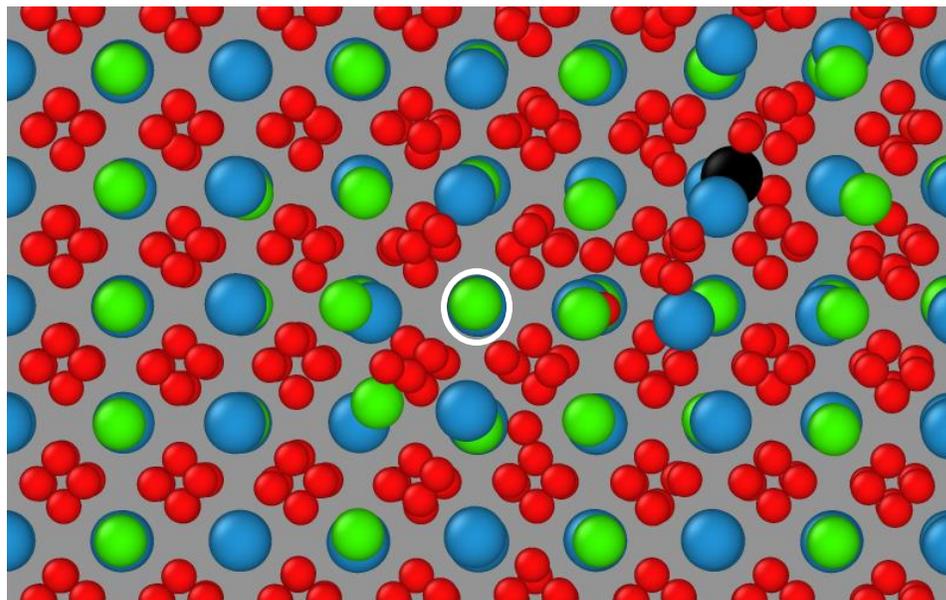
# Final positions : an example

112eV cascade → 7.1eV stored

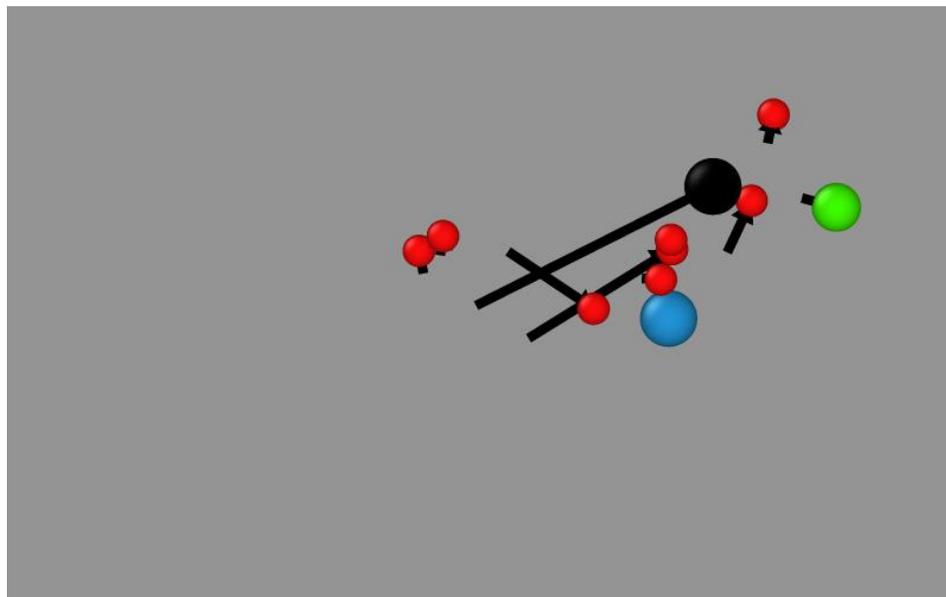


- Frenkel pair W
- But atoms are also quite disturbed around

Ca W O

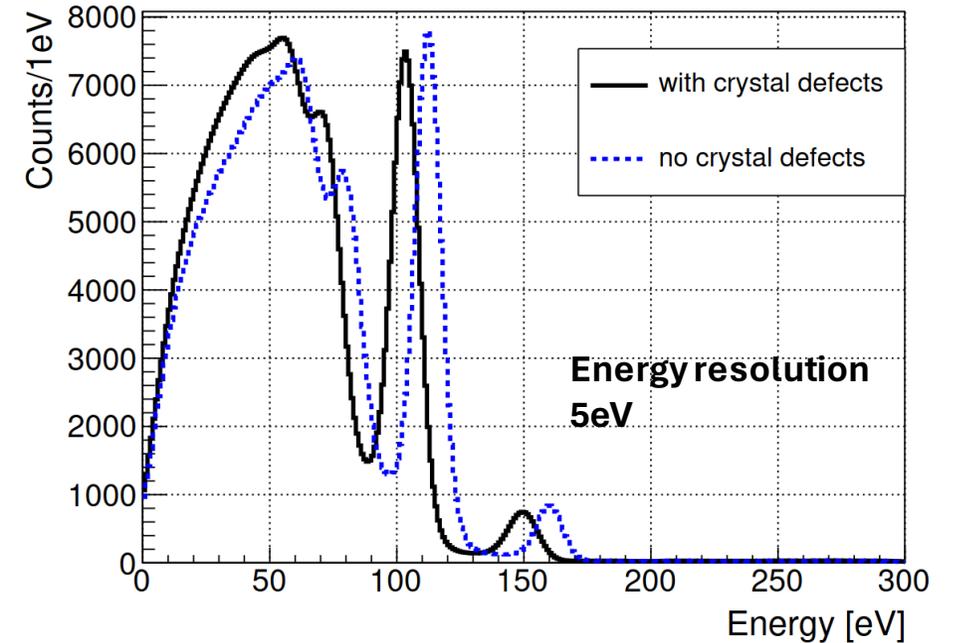
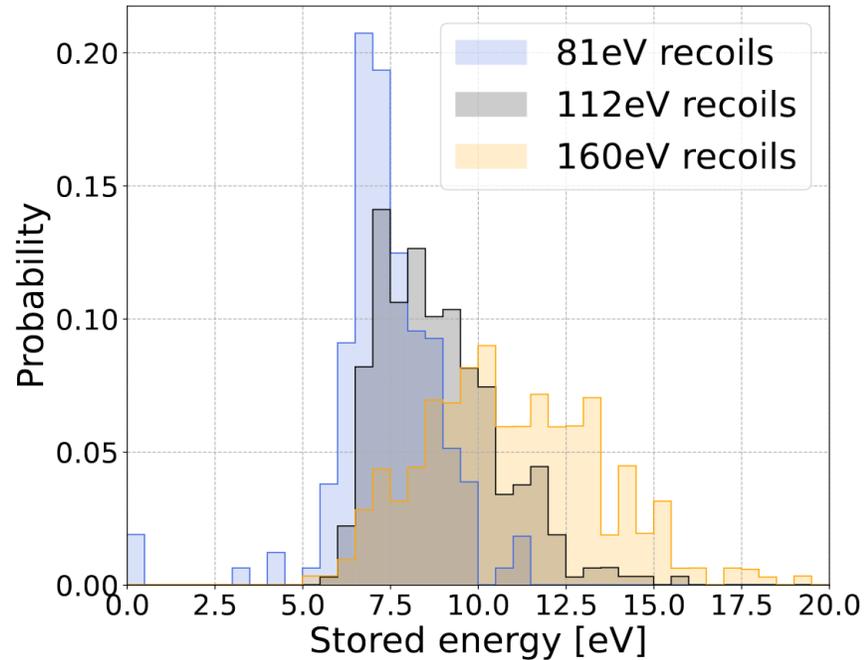


Sectional view  
around the **PKA**  
location (depth: 20 Å)



Atoms displaced by  
more than 0.8Å  
(with displacement vectors)

# Effect on the CRAB spectrum



<b>Nuclear recoil energy</b>	<b>81 eV</b>	<b>112 eV</b>	<b>160 eV</b>
<b>Average energy stored in defects</b>	<b>7.4 eV</b>	<b>8.9 eV</b>	<b>11.0 eV</b>

Non-linearity  
(defect creation thresholds)

Creation of crystal defects --> non-linearity of the energy scale between the three peaks

**Observation at  $>3\sigma$  for CRAB Phase II?**

--> detector resolution is key

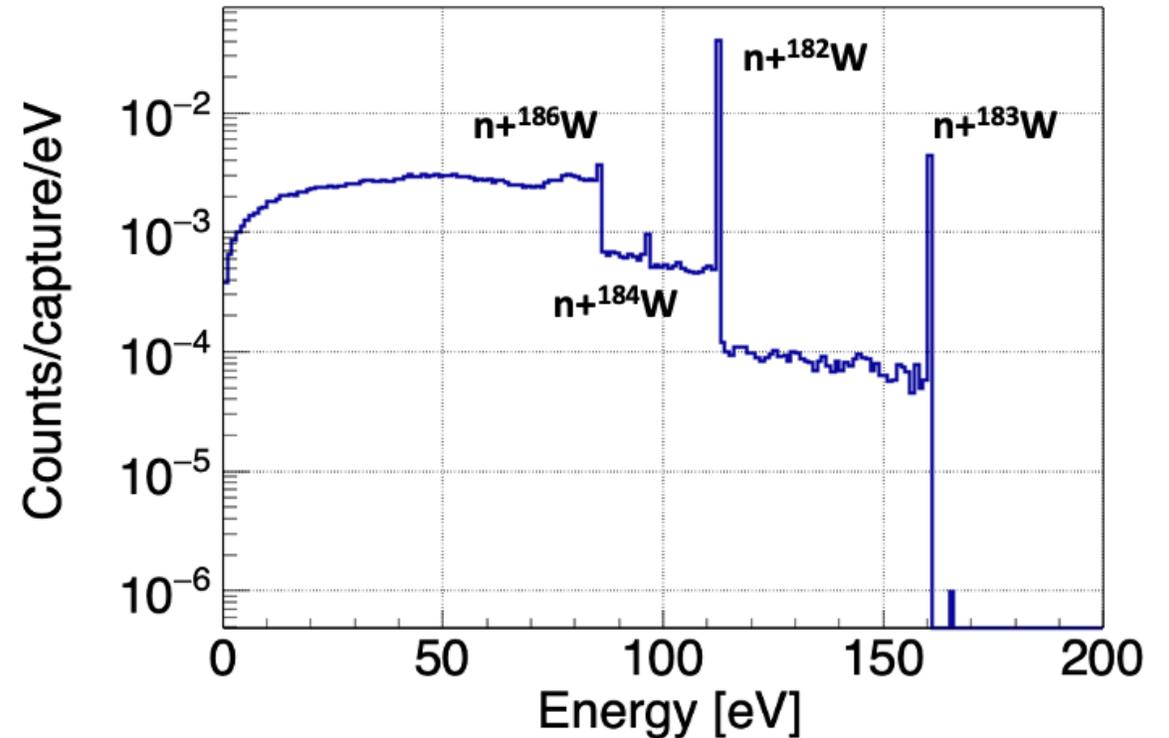
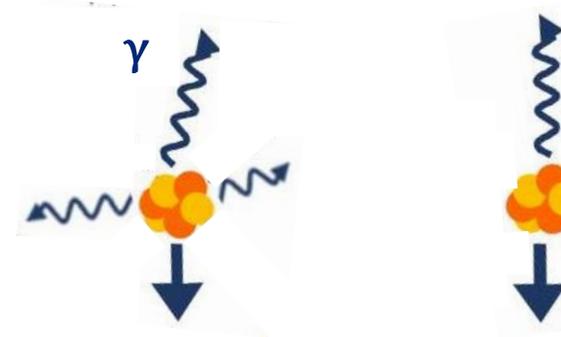
--> control of the experimental non-linearities is crucial

**A detailed example: CRAB**

# Timing of atomic collisions

# Back to the CRAB method

- $1\gamma$  de-excitations : calibration **peak**
- Multi  $\gamma$  de-excitations : recoil energy depends on
  - $\gamma$  energies and relative directions
  - **timing** of the  $\gamma$ -cascade vs timing of the recoil in the crystal



Recoil energy spectrum for  $\text{CaWO}_4$

# Study of collisions

## Prompt hypothesis

$$\tau_\gamma \ll \tau_{\text{recoil}}$$



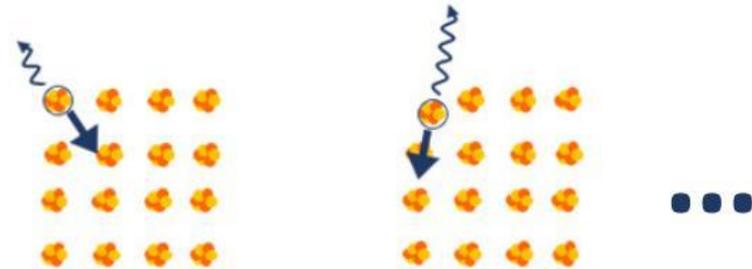
All  $\gamma$  are emitted  
The nucleus recoils

Continuum of  
possible recoil  
energies

$$E_{\text{recoil}} = \left( \sum_{\gamma} \vec{p}_{\gamma} \right)^2 / 2M_{\text{nucleus}}$$

## Slow hypothesis

$$\tau_\gamma \gg \tau_{\text{recoil}}$$



First  $\gamma$  is emitted  
The nucleus recoils  
and stops

Second  $\gamma$  is emitted  
The nucleus recoils and  
stops

$$E_{\text{recoil}} = \sum_{\gamma} p_{\gamma}^2 / 2M_{\text{nucleus}}$$

**A single** recoil energy  
per  $\gamma$ -cascade

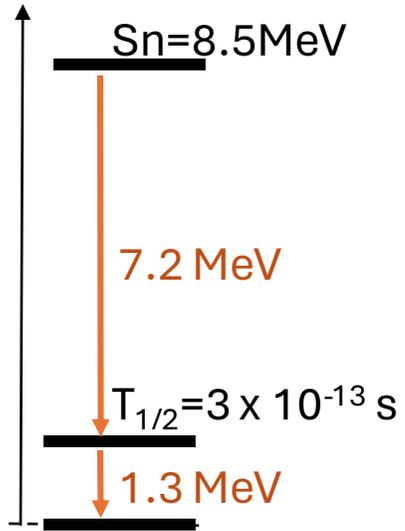
**IRADINA** (Binary Collision Approximation code) coupled to  
**FIFRELIN** (fission modelling) to simulate in-flight  $\gamma$  emission

- Timing changes the energy deposited in the bolometer
- Single- $\gamma$  calibration peaks are not affected

G. Soum-Sidikov et al. (CRAB  
colloration), [Phys. Rev. D 108,  
072009 \(2023\)](#)

# Timing effects in Silicon

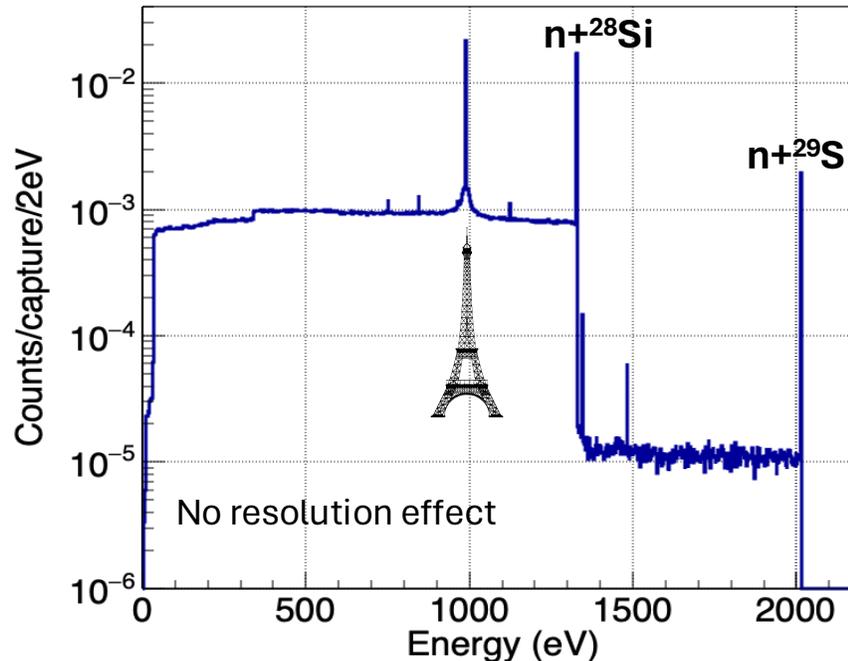
Probable 2 $\gamma$ -cascade involving a metastable nuclear level



Typical recoil durations:  
 $10^{-14}$ - $10^{-12}$ s

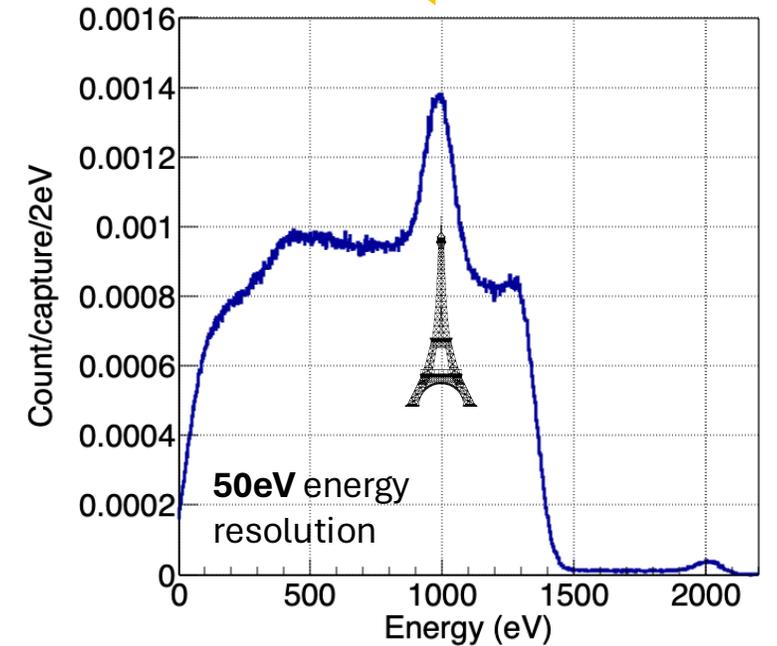
## Special feature

Present only when in-flight  $\gamma$ -emission is considered



## Calibration feature

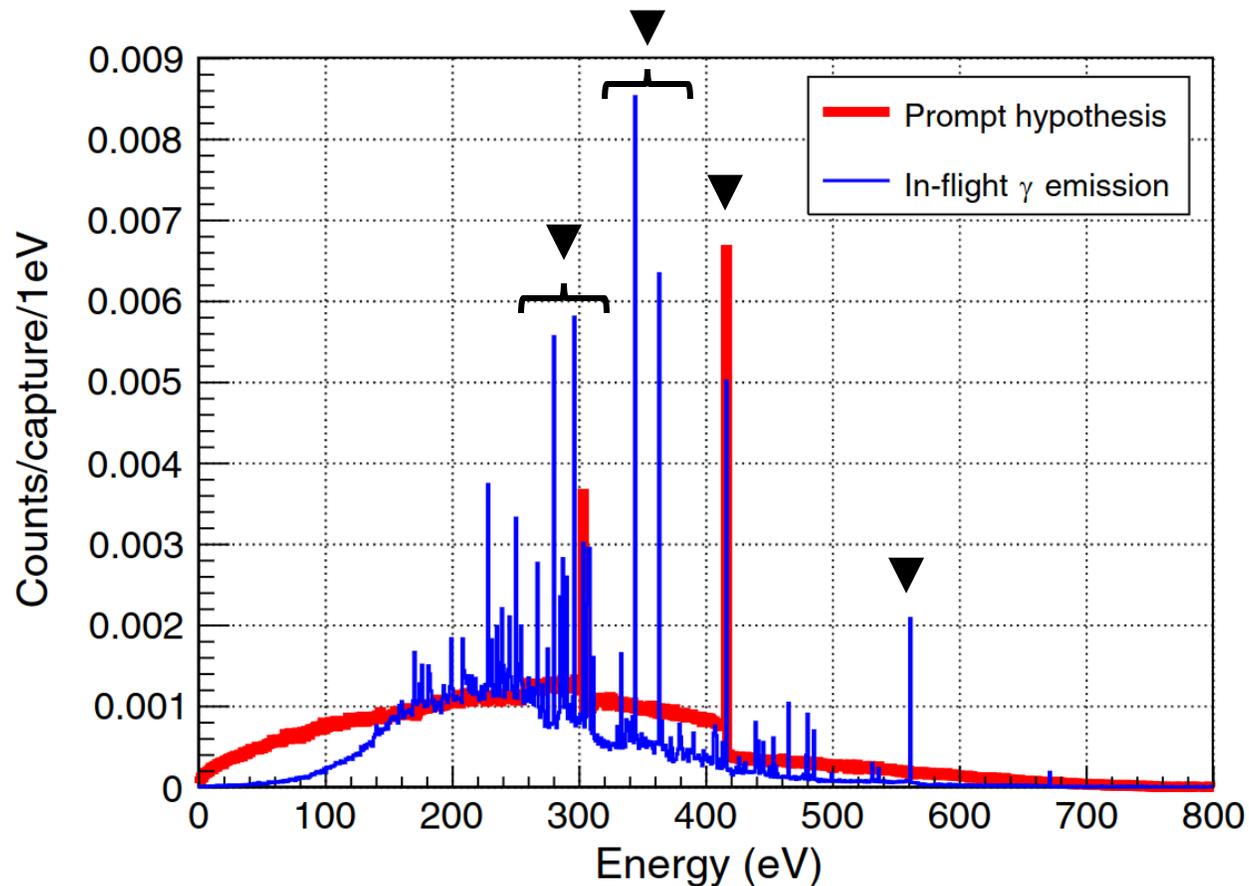
Sensitive to the interplay between the  $\gamma$ -cascade timing and the recoil timing



Recoil energy spectrum in a Si cryodetector

1st experimental evidence: A.N. Villano et al., [Phys.Rev.D 105,083014 \(2022\)](https://doi.org/10.1103/PhysRevD.105.083014)

# Timing effects in Germanium



Several calibration features would be available on Ge  
--> very interesting physics case

## Perspectives:

- development of a ML interatomic potential for Ge
  - MD simulations of cascades in Ge including emission of  $\gamma$  in flight
- > better estimation of timing effects

# Conclusion

- **Experimental progress** in cryogenic calorimeters makes them **sensitive** to effects of **atomic cascades**
- Various **methods** from solid-state physics can simulate these effects. Be aware of their potential and their limits
  - **BCA:** fast, qualitative, no access to energy stored in defects after a cascade
  - **Molecular Dynamics:** requires a good energy/force engine, allows quantitative predictions, possible calculation of energy stored in defects after a cascade
- **Impact** of the energy stored in defects **on the shape of expected spectra for DM, CEvNS** (low energy nuclear recoils)
- **Calibration** with nuclear recoils similar to the expected signal to provide experimental reference points: **CRAB method**
  - Suitable for  $\text{CaWO}_4$ , Ge, Si,  $\text{Al}_2\text{O}_3$  detectors
  - High-precision campaign on  $\text{CaWO}_4$  at the end of the year: preparation for a **calibration facility**

# Additional references

- G. S. Was, *Fundamentals of Radiation Materials Science: Metals and Alloys*, Springer (2016)

## **DFT:**

- R. M. Martin, *Electronic structure: basic theory and practical methods*, Cambridge University Press (2004)
- VASP workshop lectures: [https://www.vasp.at/wiki/index.php/VASP\\_workshop](https://www.vasp.at/wiki/index.php/VASP_workshop)

## **ML potentials:**

- A. Bartók-Partay, *The Gaussian Approximation Potential: an interatomic potential derived from first principles quantum mechanics*, Springer Theses (2010)
- A. P. Bartók, R. Kondor, and G. Csányi, *On representing chemical environments*, [Physical Review B, 87\(18\), 184115](#) (2013)
- A. M. Goryaeva, J. B. Maillet, and M. C. Marinica, *Towards better efficiency of interatomic linear machine learning potentials*, [Computational Materials Science, 166, 200-209](#), (2019)

# Back-up slides

# ML Potentials: ML magics

Key concept: energy per atom

- Not in the database
- Total energy = sum of energies per atom
- Forces = sums of derivatives of energies per atom

Link between energies per atom and atomic descriptors, fit of the database?

• Non-linear methods:

- "Kernel" methods: numerical cost scales with the number of local atomic environments in the training database
- Neural networks: limited application for MD simulations

• Linear Machine Learning potentials:

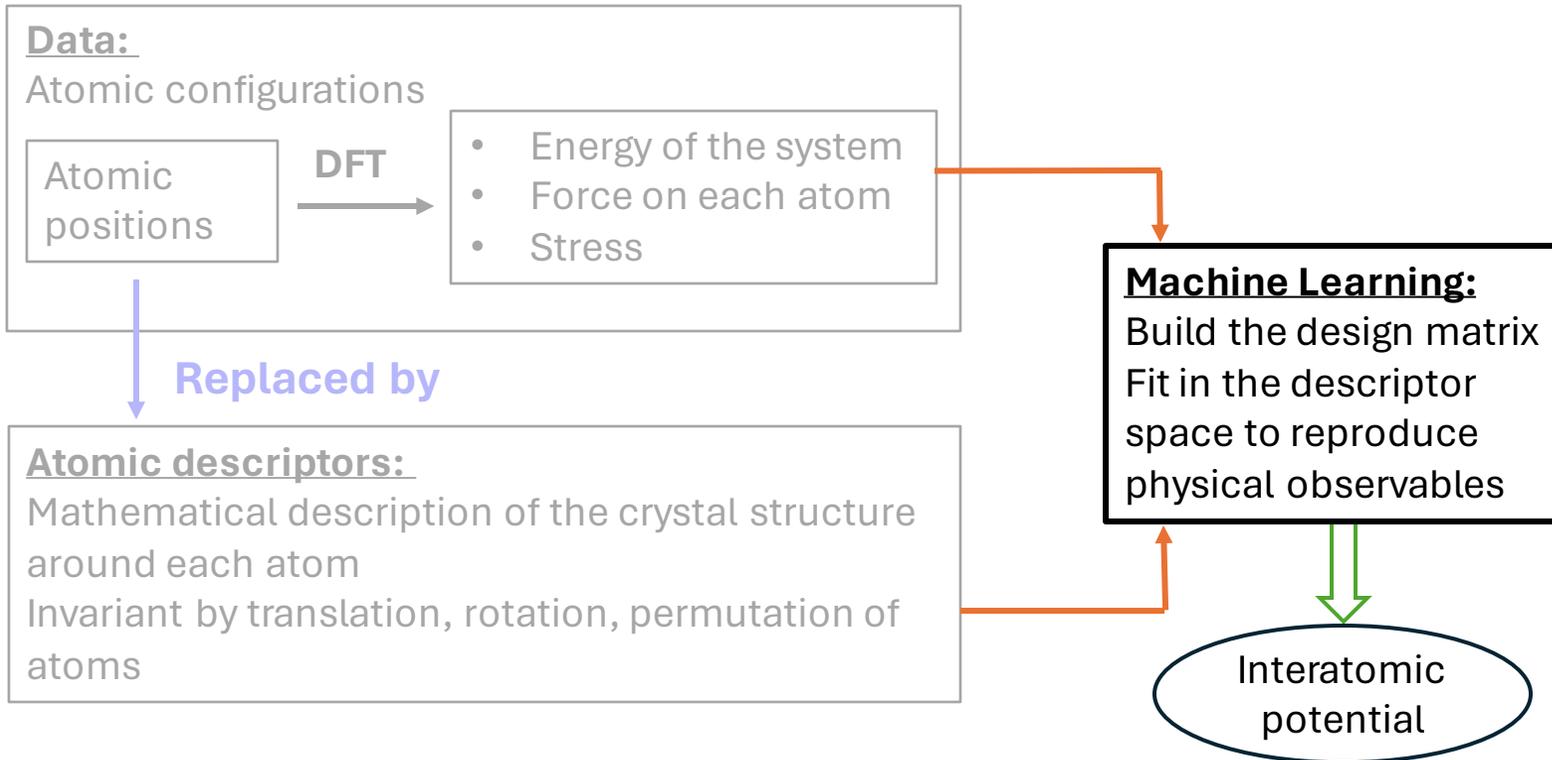
linear relationship between local energies and atomic descriptors, weighted linear regression to fit database

--> simpler, quicker

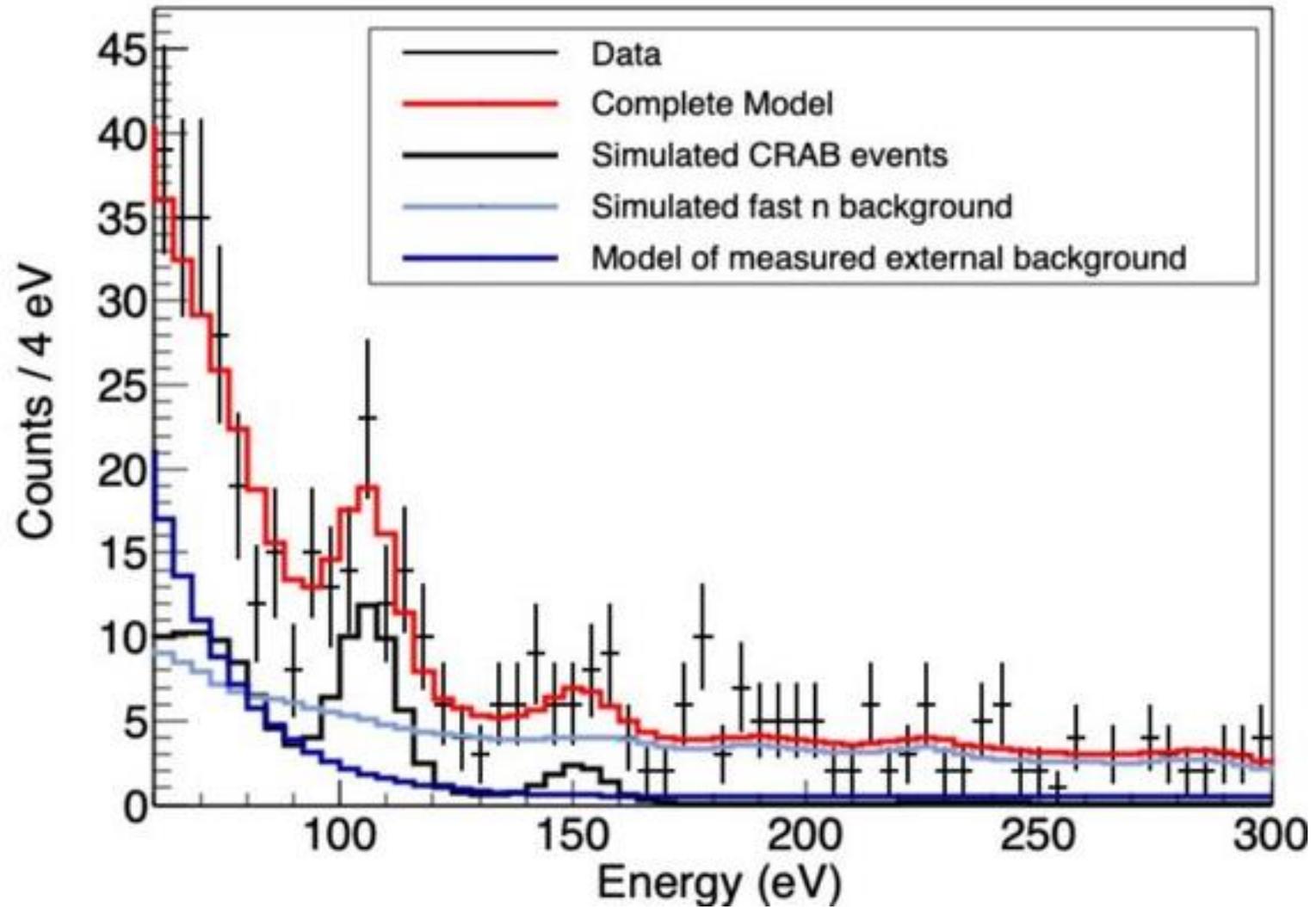
--> training/test error higher

--> good trade-off between accuracy and computational efficiency

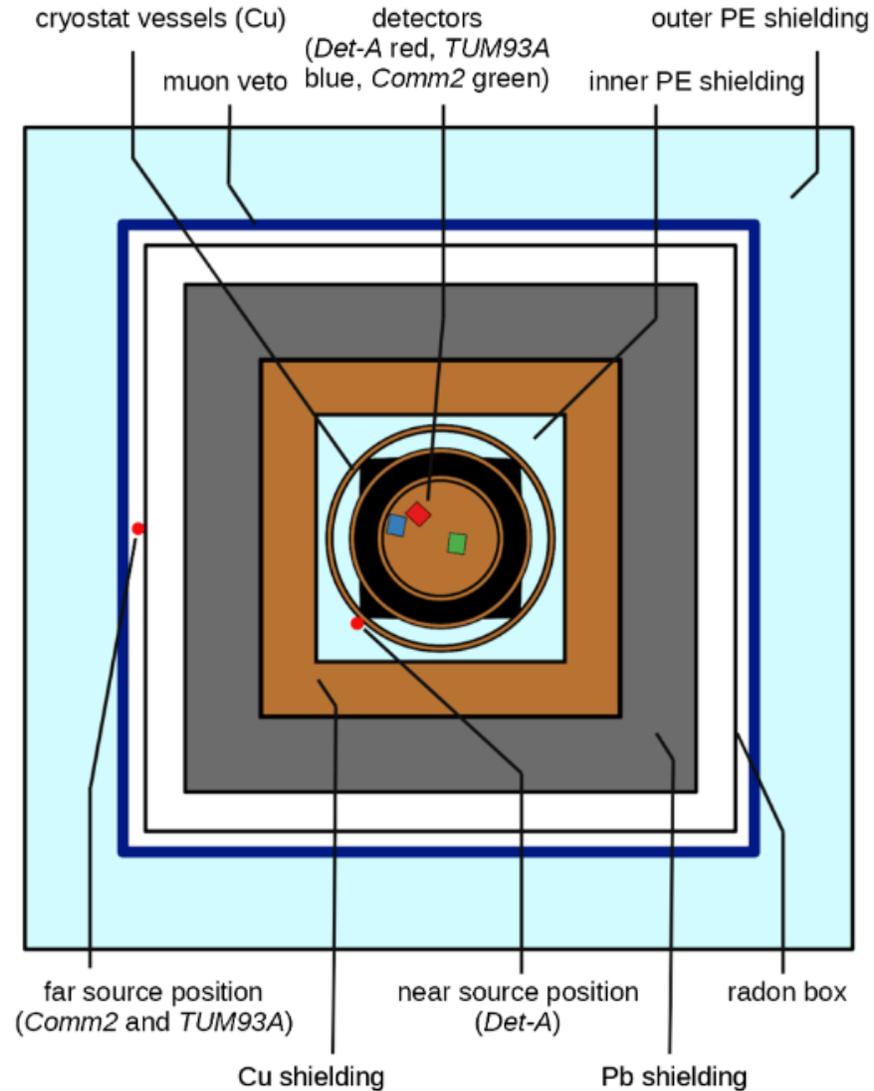
--> good choice for MD simulations



# CRAB Phase 1



# CRESST CRAB-like measurement



G. Angloher *et al.* (CRESST),  
[Phys. Rev. D 108, 022005 \(2023\)](#)

# CRESST CRAB-like measurement

TABLE II. Energy threshold  $E_{\text{thr}}$ , baseline energy resolution  $\sigma_{\text{BL}}$ , and raw measurement time  $T$  for the three datasets.

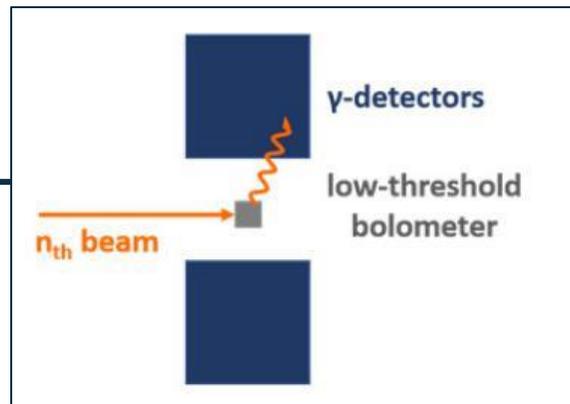
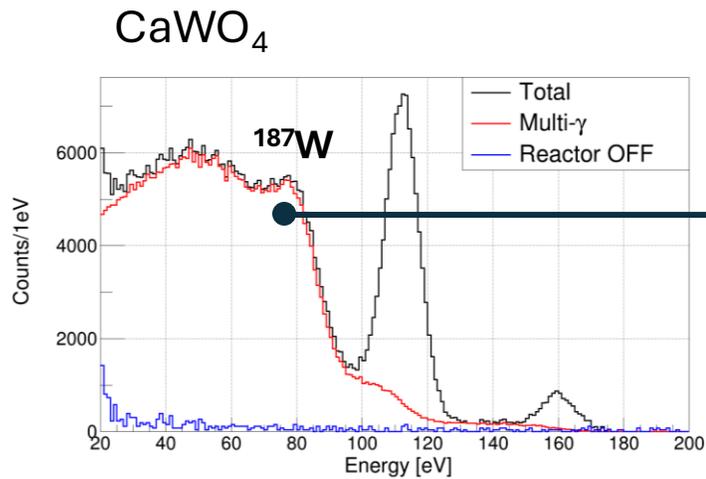
Module	$E_{\text{thr}}$ (eV)	$\sigma_{\text{BL}}$ (eV)	$T$ (h)
<i>Det-A</i>	30.6	$4.83 \pm 0.02$	671
<i>Comm2</i>	29.8	$4.95 \pm 0.02$	862
<i>TUM93A</i>	54.0	$7.92 \pm 0.03$	862

**Standard CRESST-III detectors:  
20 × 20 × 10 mm<sup>3</sup> CaWO<sub>4</sub> (~24 g)**

TABLE III. Best fit parameters for the three datasets together with the statistical significance for the peak.

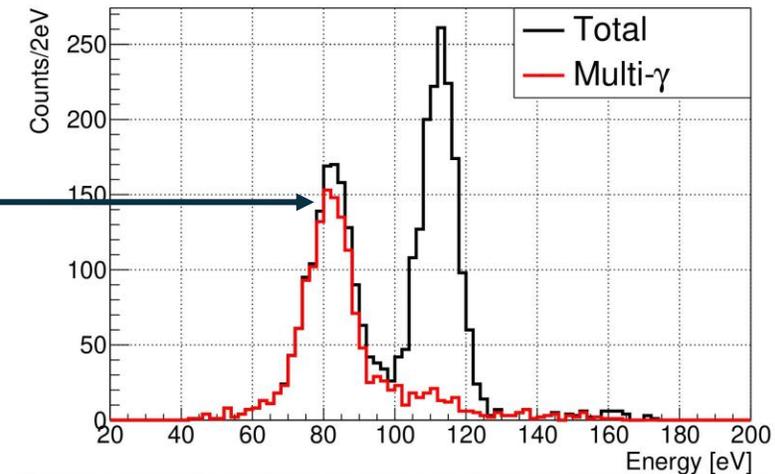
Detector	$R_{\text{sig}}$ (1/d)	$\mu$ (eV)	$\sigma$ (eV)	$R_{\text{exp}}$ (1/d)	$E_0$ (eV)	$R_{\text{flat}}$ (1/d)	Significance
<i>Det-A</i>	$1.6 \pm 0.6$	$113.3 \pm 1.4$	$3.5 \pm 0.8$	$25.1 \pm 2.6$	$52.0 \pm 9.3$	$36.8 \pm 2.4$	$3.6\sigma$
<i>Comm2</i>	$6.7 \pm 1.5$	$107.8 \pm 1.3$	$5.5 \pm 1.1$	$58.7 \pm 3.6$	$58.4 \pm 5.4$	$48.0 \pm 3.1$	$5.8\sigma$
<i>TUM93A</i>	$5.5 \pm 1.2$	$106.2 \pm 1.8$	$7.0 \pm 1.4$	$38.0 \pm 2.4$	$60.4 \pm 5.3$	$34.6 \pm 2.0$	$6.6\sigma$

# $\gamma$ -cryodetector coincidence



With coincidence

$$E_\gamma = 5.47 \pm 0.20 \text{ MeV}$$



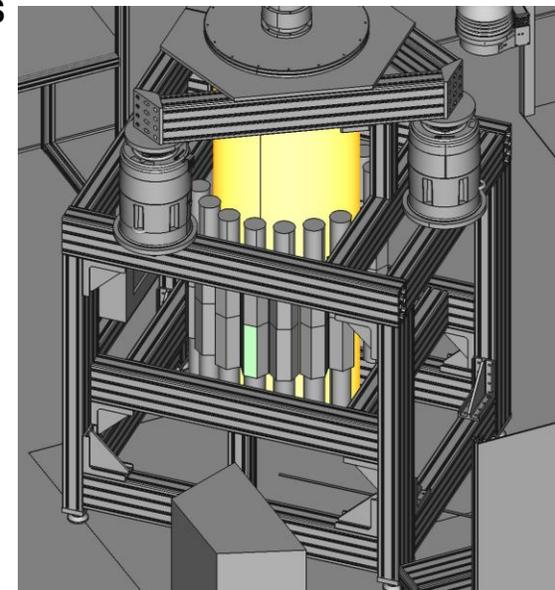
Third calibration peak @80eV

Detailed description of  $\gamma$  cascades using FIFRELIN

O. Litaize et al., Eur. Phys. J. A (2015) 51: 177

## Rejection of multi- $\gamma$ continuum

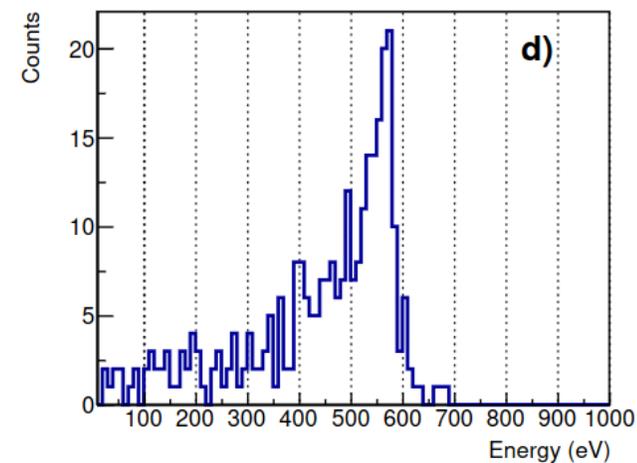
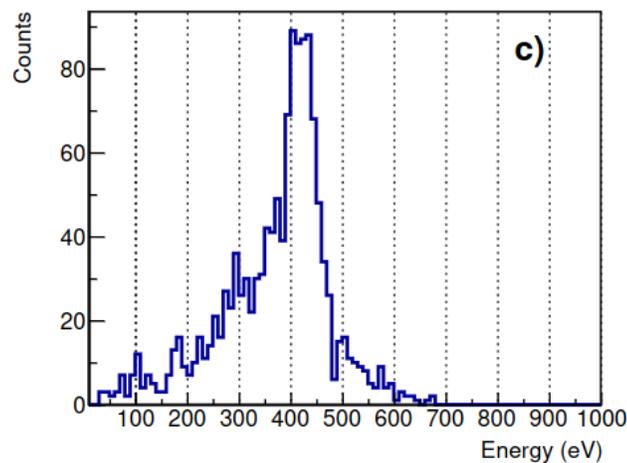
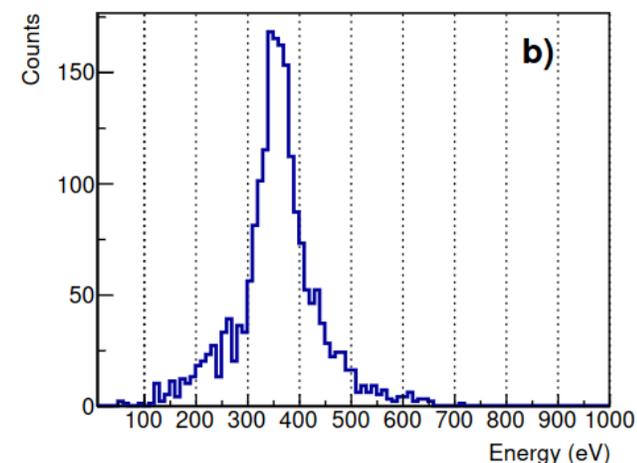
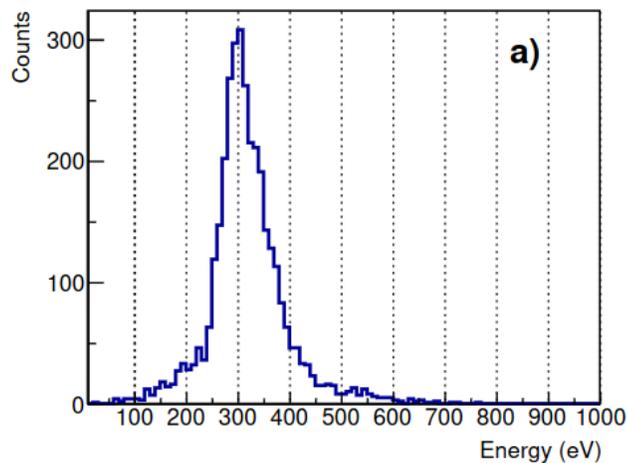
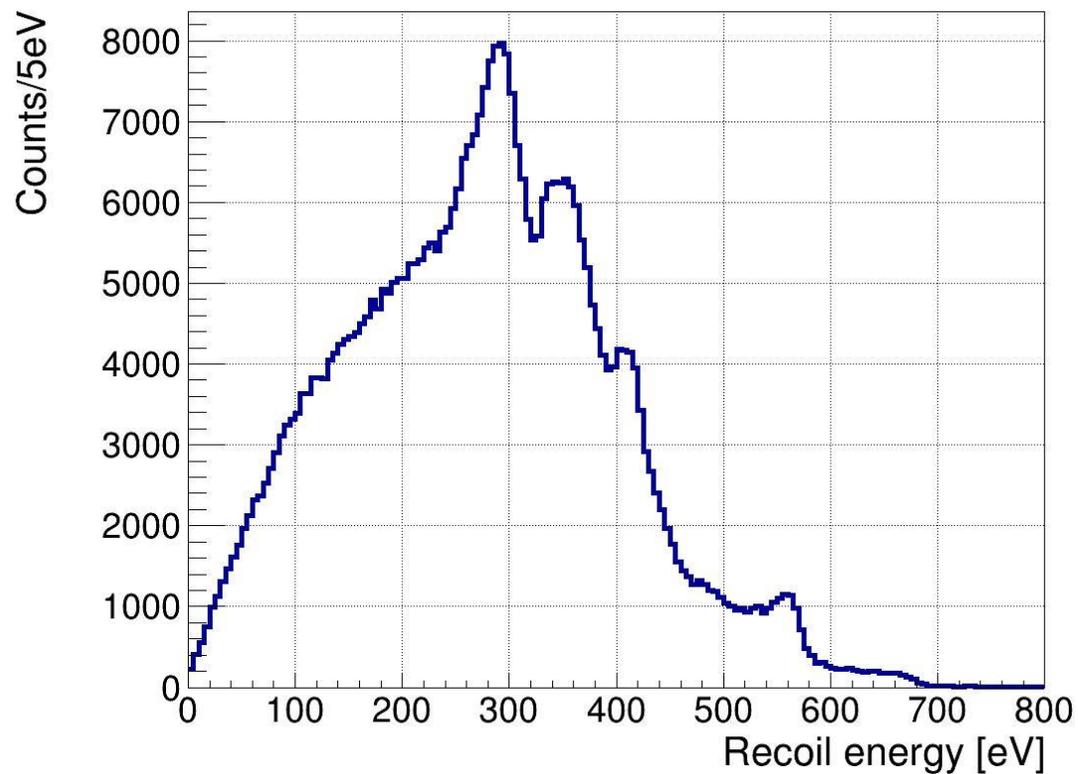
Large segmented  $\gamma$ -detectors (BaF<sub>2</sub>) outside the cryostat



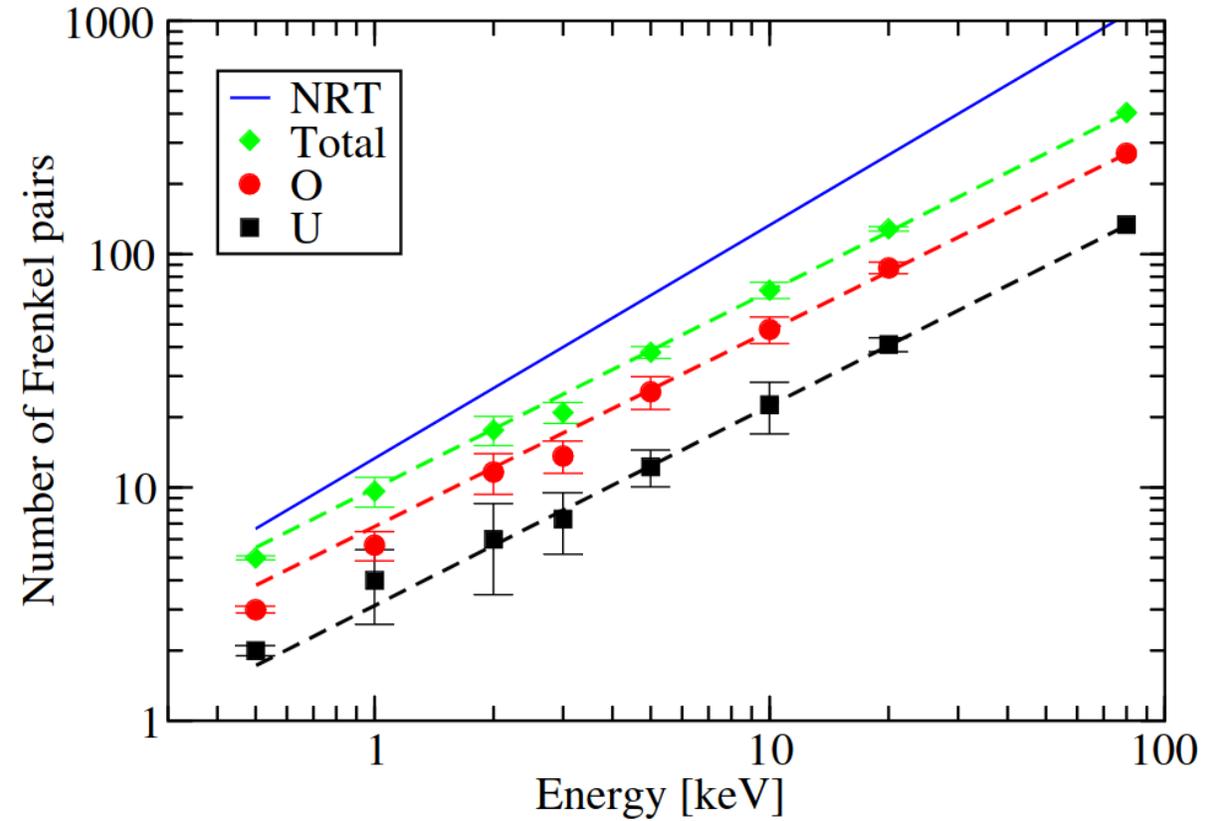
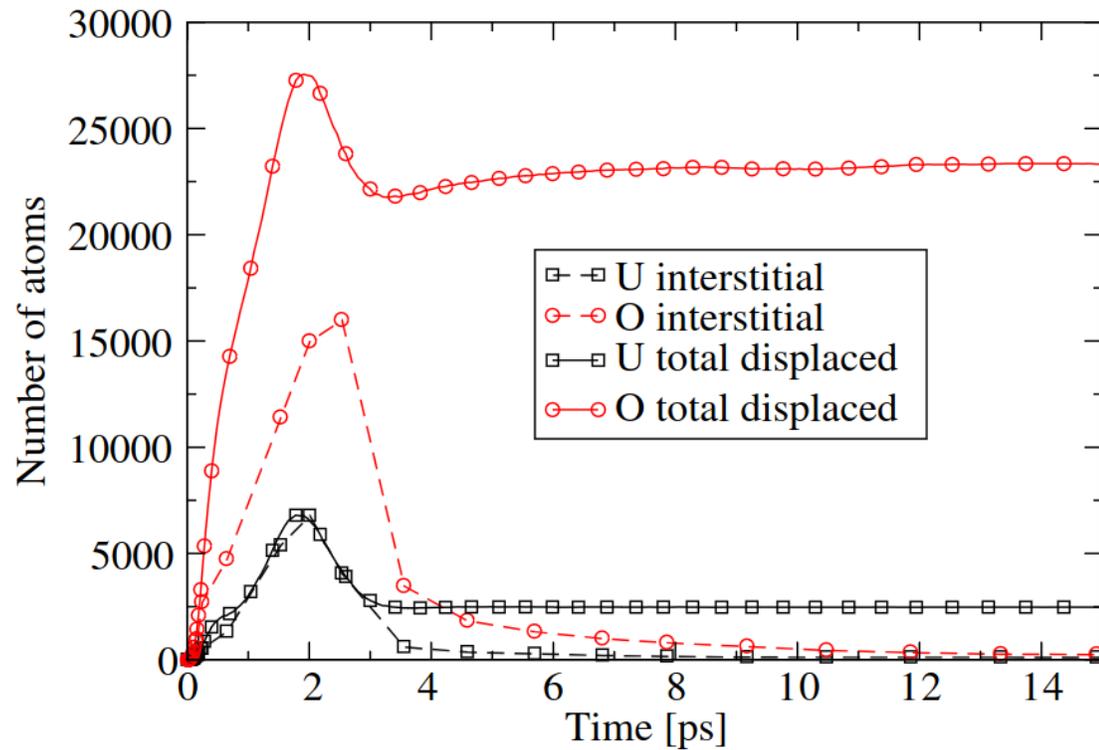
# $\gamma$ -tagging on Germanium

Res 20eV

Res 10eV

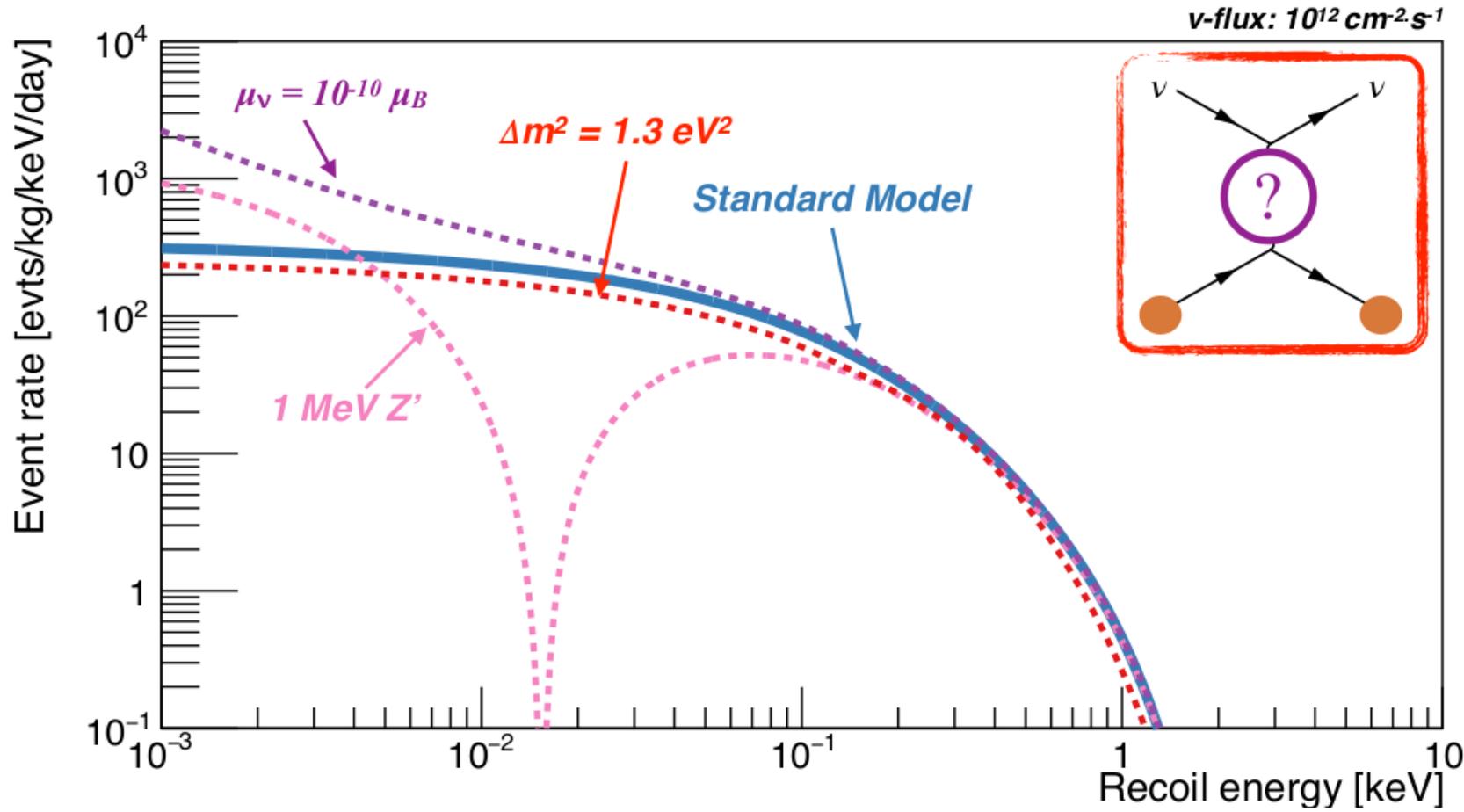


# MD simulation in UO<sub>2</sub>



# CEvNS: test for new physics

from A. Juillard's lecture



J. Billard, J. Johnston and B. Kavanagh, JCAP (2018)

New physics signatures will arise at the lowest energies :

→ **Calls for very low-energy thresholds:  $O(10) \text{ eV}$**

# Low Energy Excess (example of CRESST)

- ▶ **Low Energy Excess also seen by ALL cryogenic detectors !!**
  - Radiogenic bkg more expected to be flat and at the 1-100 dru level :
  - **LEE orders of magnitude higher !**

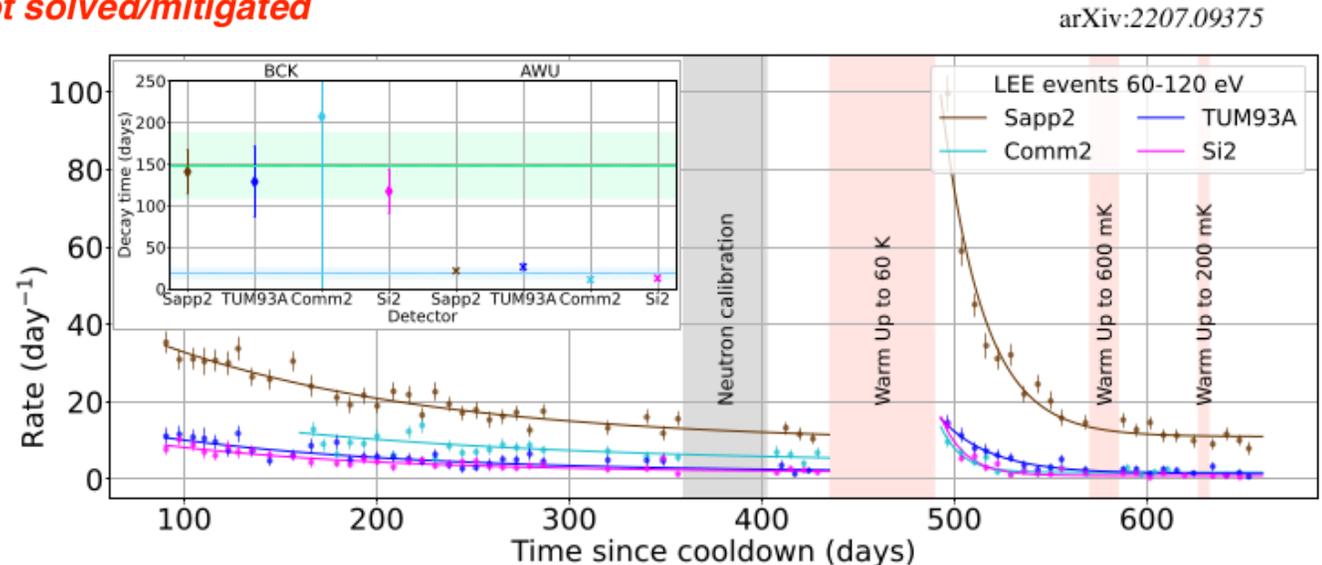
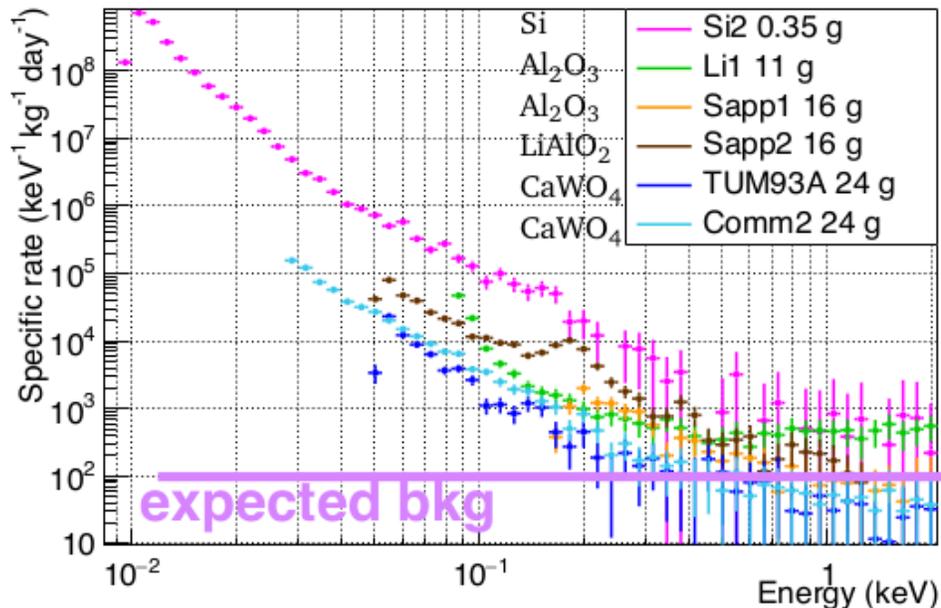
## Origins under investigation:

- Sensor related events
- Relaxation of holding-induced stress
- Intrinsic crystal effects

**Major issue today.**

**Will limit most of the science cases if not solved/mitigated**

from A. Juillard's lecture



arXiv:2207.09375