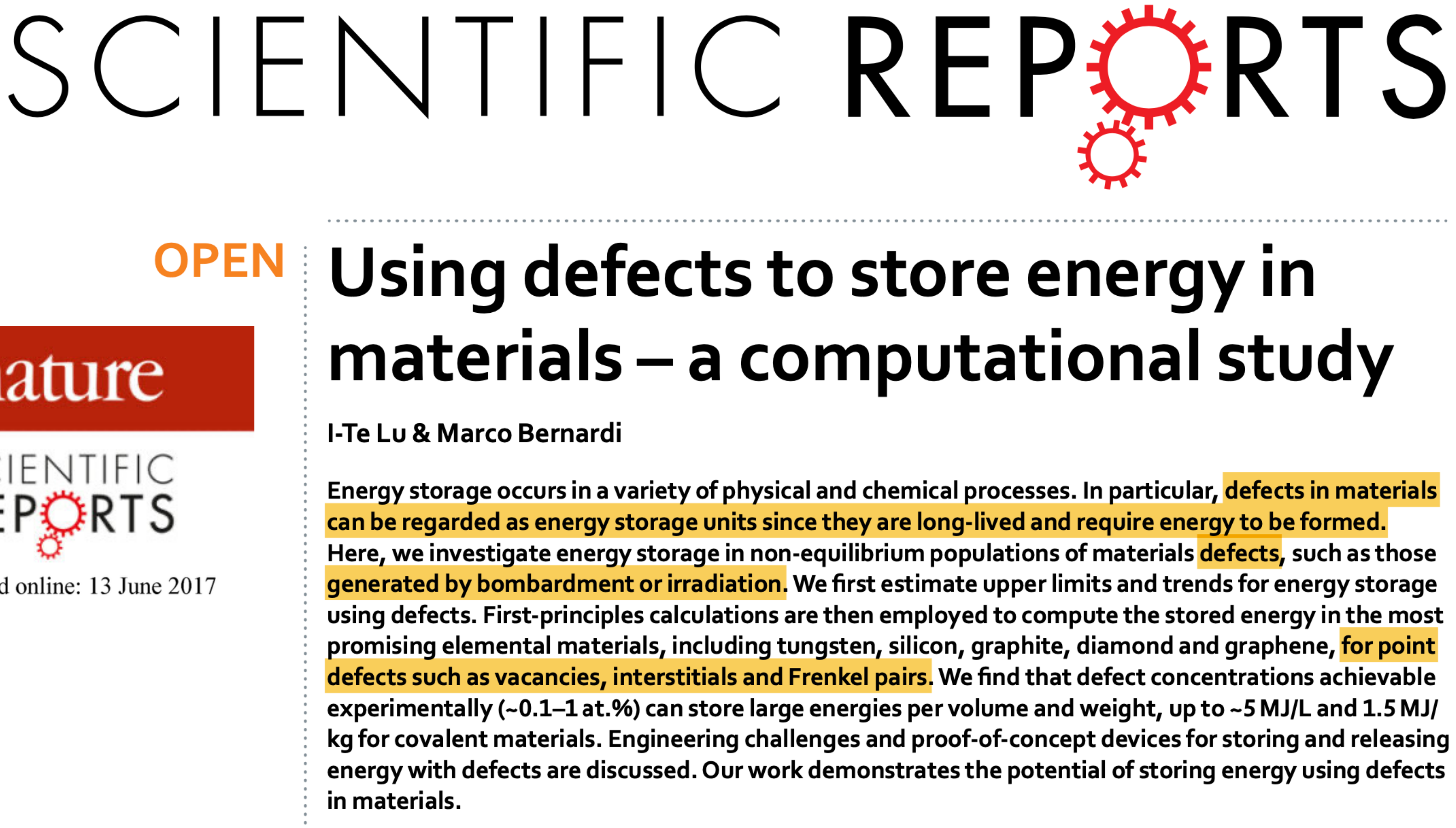
Impact of Crystal Lattice Defect Quenching on CEνNS at reactors

Excess Workshop 2022, Vienna

Thierry Lasserre & Chloé Goupy

CEA & TUM-SFB1258



SCIENTIFIC REPORTS

OPEN

Using defects to store energy in materials – a computational study

I-Te Lu & Marco Bernardi

Energy storage occurs in a variety of physical and chemical processes. In particular, defects in materials can be regarded as energy storage units since they are long-lived and require energy to be formed. Here, we investigate energy storage in non-equilibrium populations of materials defects, such as those generated by bombardment or irradiation. We first estimate upper limits and trends for energy storage using defects. First-principles calculations are then employed to compute the stored energy in the most promising elemental materials, including tungsten, silicon, graphite, diamond and graphene, for point defects such as vacancies, interstitials and Frenkel pairs. We find that defect concentrations achievable experimentally (~0.1–1 at.%) can store large energies per volume and weight, up to ~5 MJ/L and 1.5 MJ/kg for covalent materials. Engineering challenges and proof-of-concept devices for storing and releasing energy with defects are discussed. Our work demonstrates the potential of storing energy using defects in materials.

nature
SCIENTIFIC REPORTS

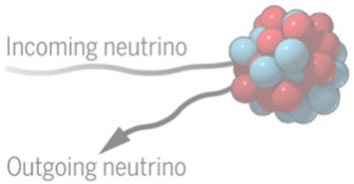
Published online: 13 June 2017

Crystal Lattice Defect Quenching

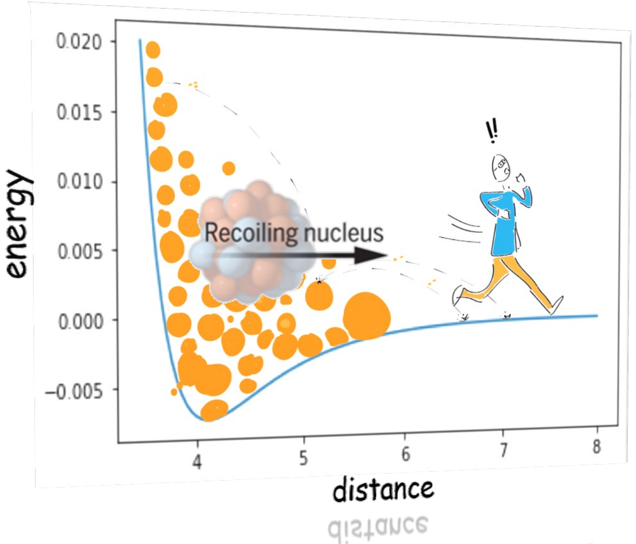
(CLDQ)

CEVNS interaction in a crystal lattice

- **Step 1) the neutrino scatters off the nucleus, transferring a defined momentum q**
  - Timescale << picosecond – spatial range: point-like



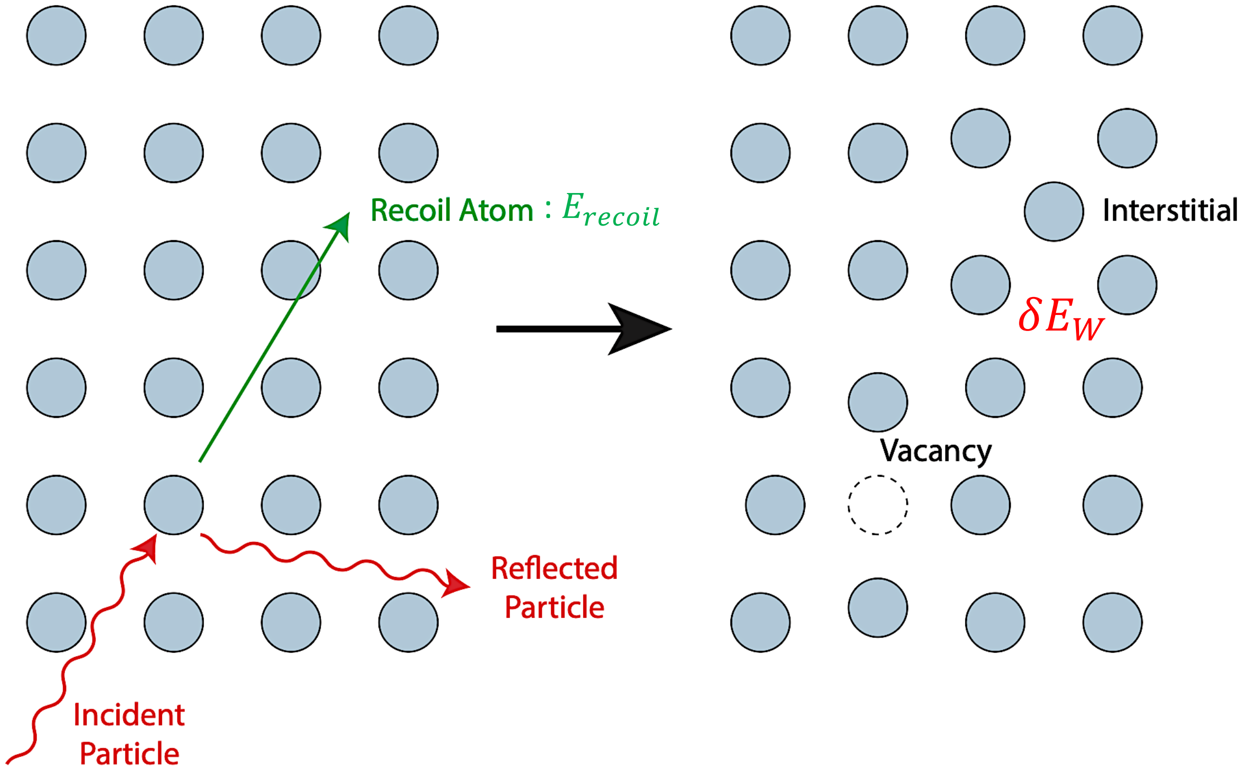
- **Step 2) the nucleus oscillates in the lattice's potential well.**
  - Case a) The nucleus scans it potential and dissipate energy but relaxes back to its initial state
    - Dissipation of phonons
      - Dependence on the material and momentum q
      - Mild dependence on direction of the signal upon incoming direction
  - **Case b)** The nucleus escape from its potential and create a vacancy
    - Creation of a lattice vacancy (defect)
      - Some energy may be used (quenched) for this process
      - Direction dependent (in the crystal frame)
      - Depends on the material and momentum $q>q_{min}$
    - Subsequent dissipation of phonons



- **Step 3) the primary recoil atom (PKA) may start moving other atoms of the lattice**
  - Timescale : 100 ps – range : few nm

Crystal defect induced energy loss

- Crystal defects can be formed whenever incident radiation strikes an atom, transferring sufficient energy to remove the atom from its lattice site, resulting in a vacancy



Incident energy

$$E_{recoil} = E_{phonon} + \delta E_W$$

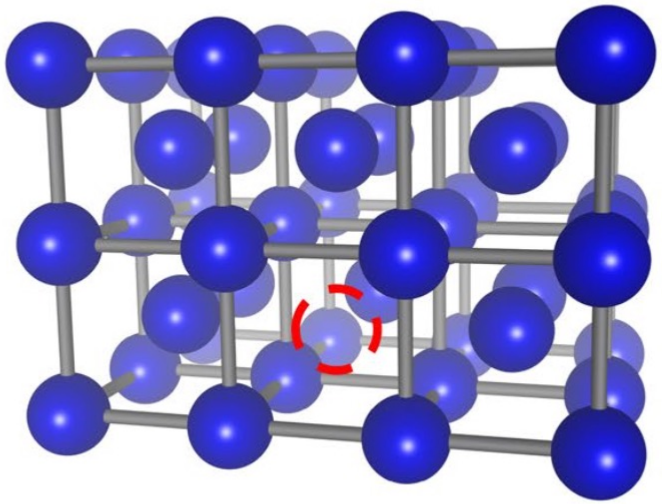
Possible loss for the phonon sensor...
(at least for some time...)

Our observable in phonon-mediated cryodetector

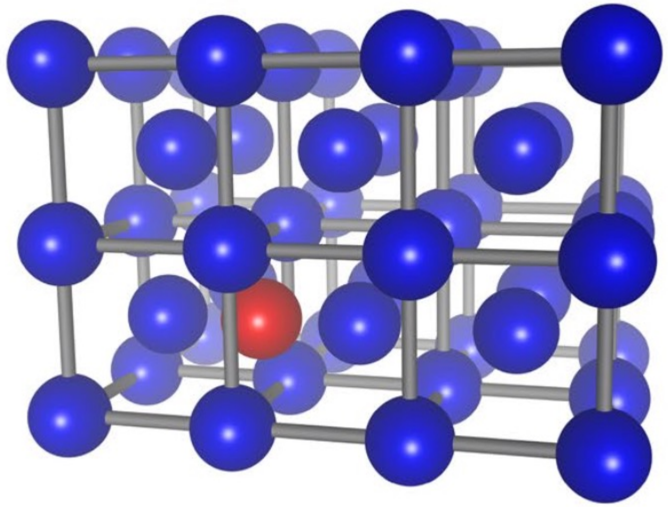
- After a few recombinations (100's ps or so), « part of the incident energy » is used for the creation of defects, defect clusters, dislocations... This is also referred to as the Wigner energy (δE_W)

Example of crystal defects

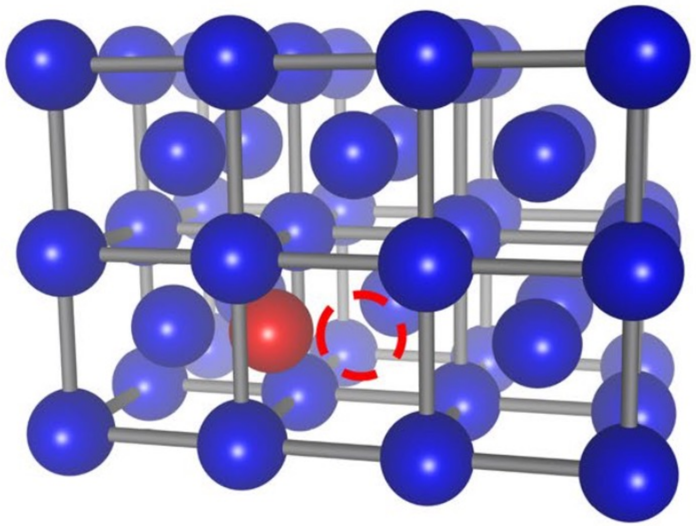
Vacancy



interstitial



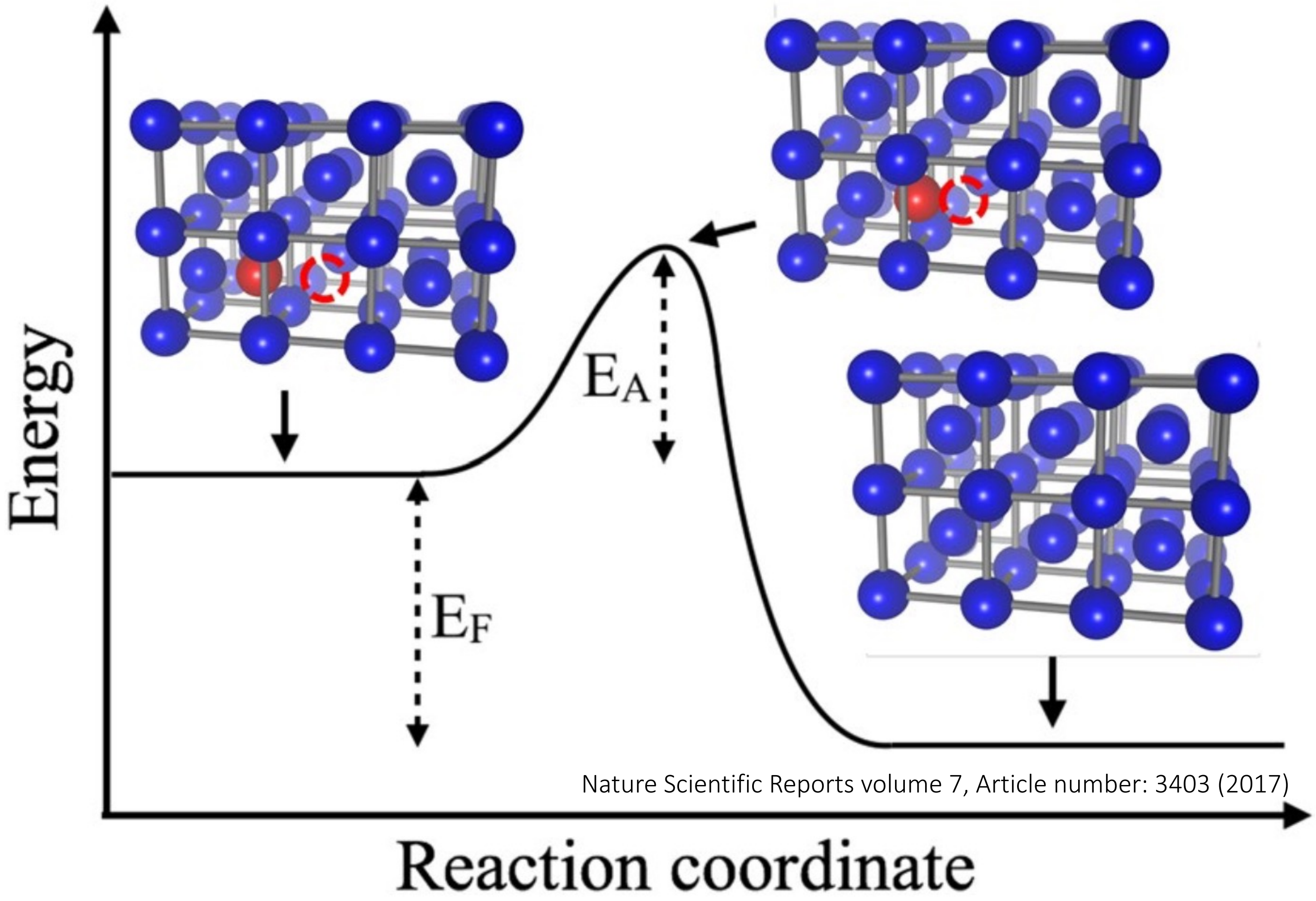
Frenkel pair



- Other defects: Schottky pairs, dislocation (aligned set of defects), cluster of defects, … not considered here …
- The defect tagging itself may be used to detect rare radiation events (DM) and potentially their direction!

Nature Scientific Reports volume 7, Article number: 3403 (2017)

Frenkel pair: formation / recombination



Nature Scientific Reports volume 7, Article number: 3403 (2017)

- Frenkel Pair (FP)

- Creation

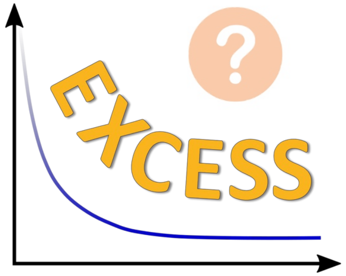
  Need E > E_F + E_A

  E_F: Formation Energy

- Temperature-dependent recombination

  Need E > E_A

  E_A: Activation Energy



Radiation induced CLDQ

- Penetrating radiations of all forms induce lasting defects in most of materials

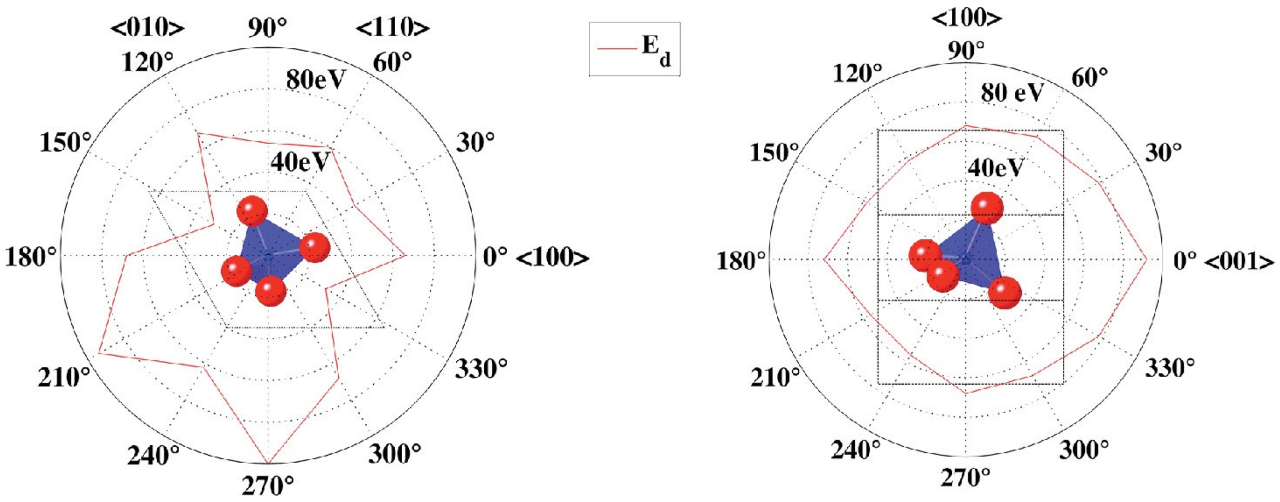
- In solid-state detectors:
  - **High-energy electromagnetic radiation** interacts primarily with the electron system
    - Indirect induction of crystal defects (for high energy radiation of 100's of keV, $E \gg E_F$)
  - **Neutrons (Nuclei)** interact primarily with lattice nuclei
    - Can create long-lived defects for E_{rec} > 10's eV

- Coherent Neutrino Scattering off nuclei inside a crystal
  - Induce a 10-500 eV nuclear recoil: $\boldsymbol{E_{recoil}}$ → **corresponds to the energy scale of defect formation**
  - Part of the energy may create a long-lived defects: $\boldsymbol{\delta E_W(E_{recoil}(\theta), \text{particle ID}}, \text{material})$
  - Possible bias in energy reconstruction:

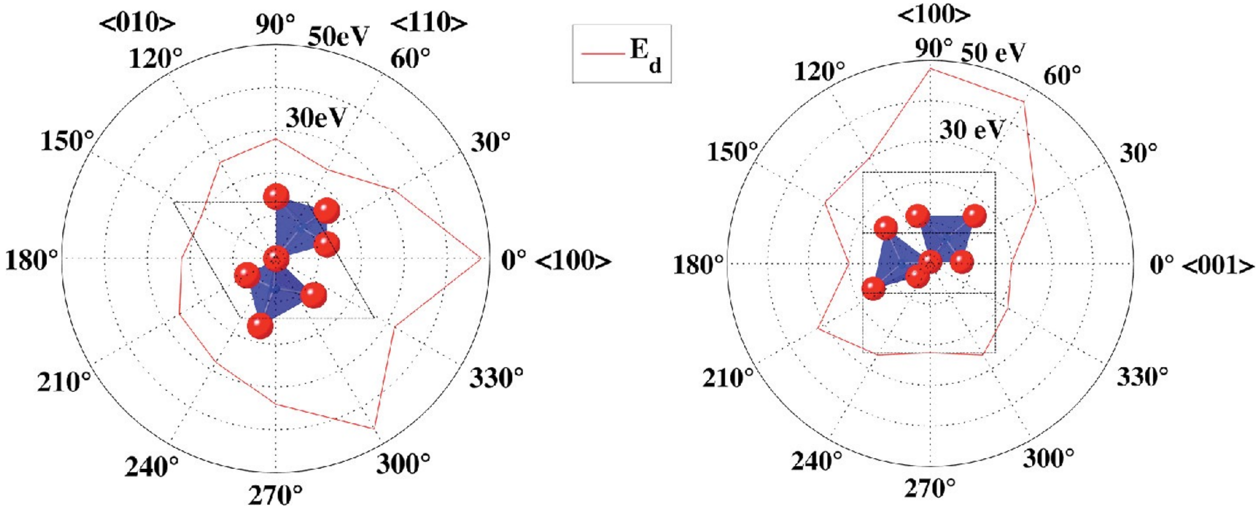
$$\underbrace{\boldsymbol{E_{visible}}} = \underbrace{\boldsymbol{E_{recoil}}} - \underbrace{\boldsymbol{\delta E_W(E_{recoil}(\theta), \text{particle ID}, \text{material})}}$$

Threshold displacement energy (E_d) along different directions in quartz crystal

Si

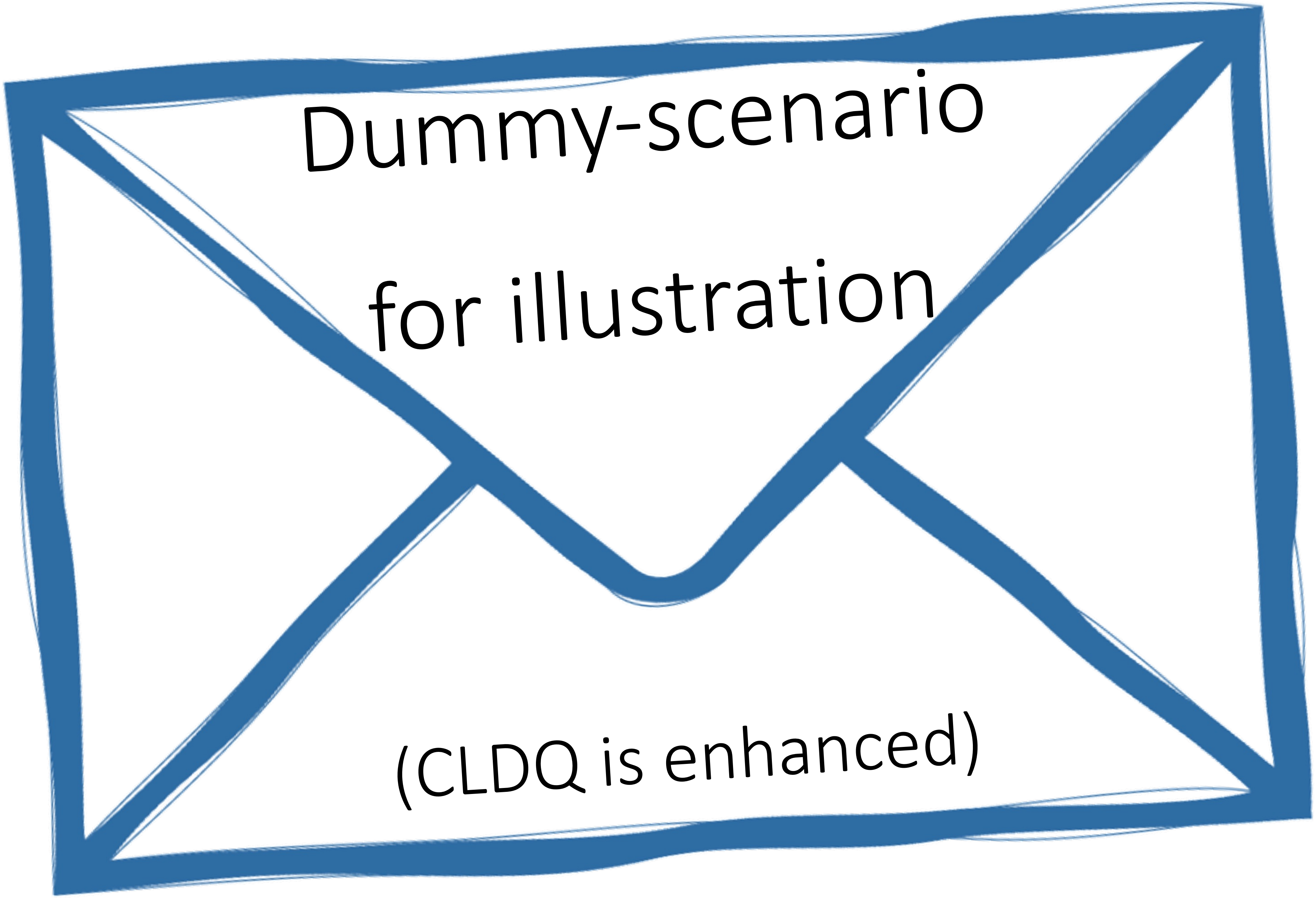


O



Sizable variation with respect the incoming radiation direction

Published in: Bu Wang; Yingtian Yu; Isabella Pignatelli; Gaurav Sant; Mathieu Bauchy; *J. Chem. Phys.* **143,** 024505 (2015) - DOI: 10.1063/1.4926527 Copyright © 2015 AIP Publishing LLC

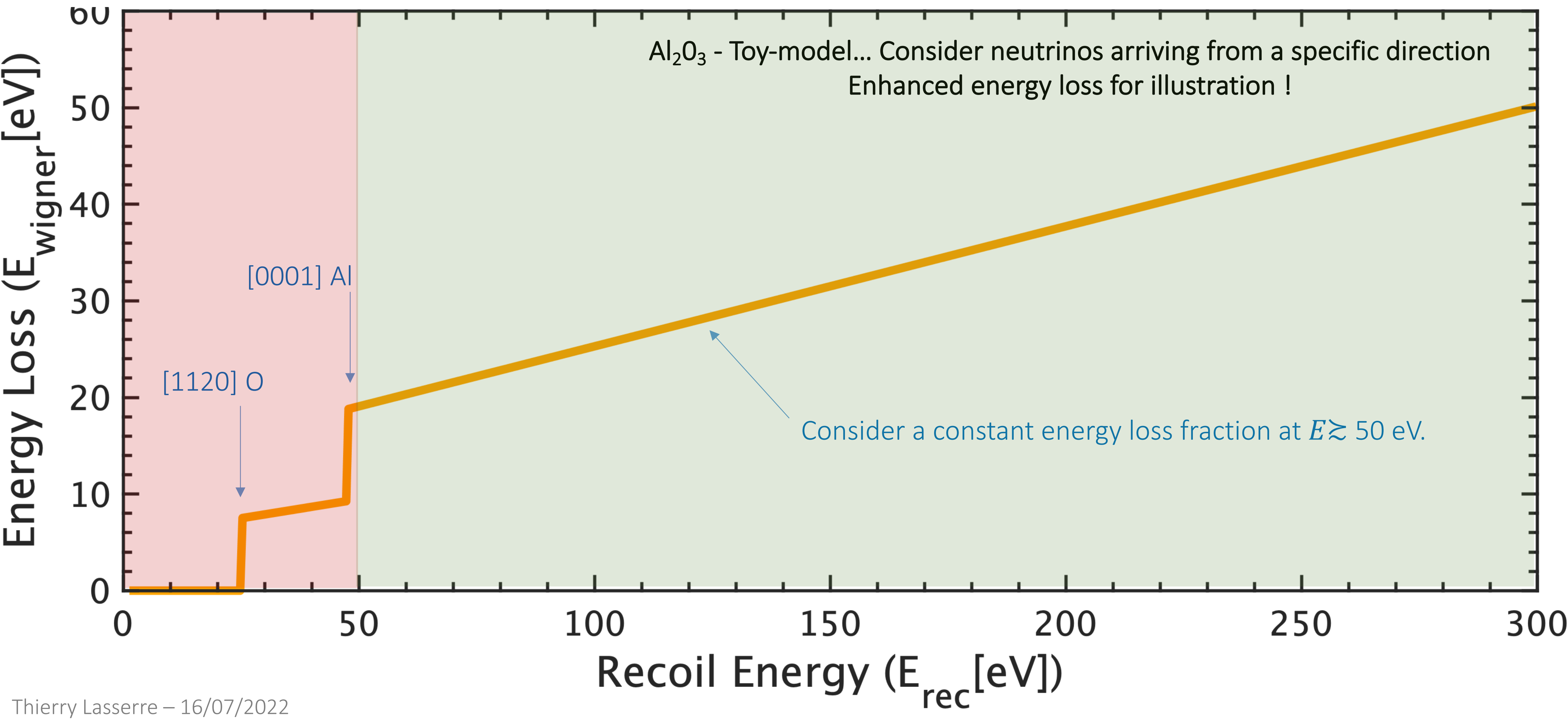


Dummy-scenario

for illustration

(CLDQ is enhanced)

Dummy CLDQ energy loss function

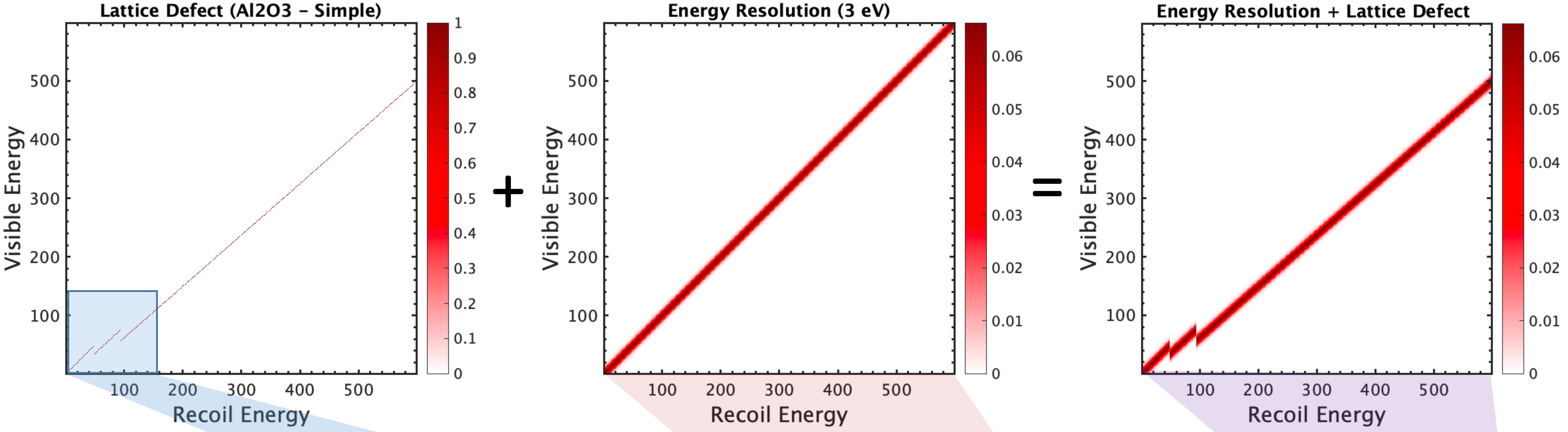


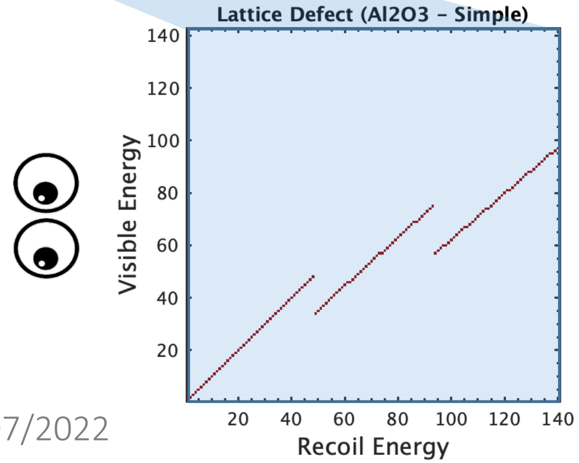
Al_2O_3 - Toy-model… Consider neutrinos arriving from a specific direction

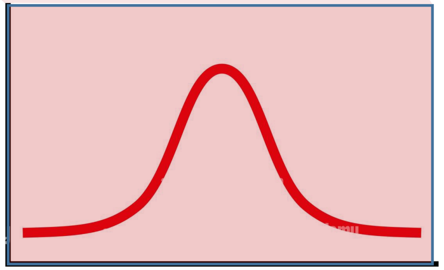
Enhanced energy loss for illustration !

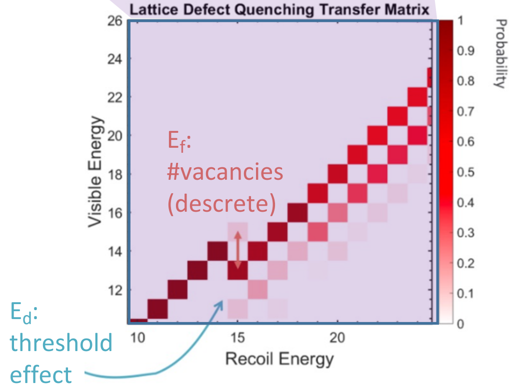
Consider a constant energy loss fraction at $E \gtrsim 50$ eV.

CLDQ + resolution: transfer matrices





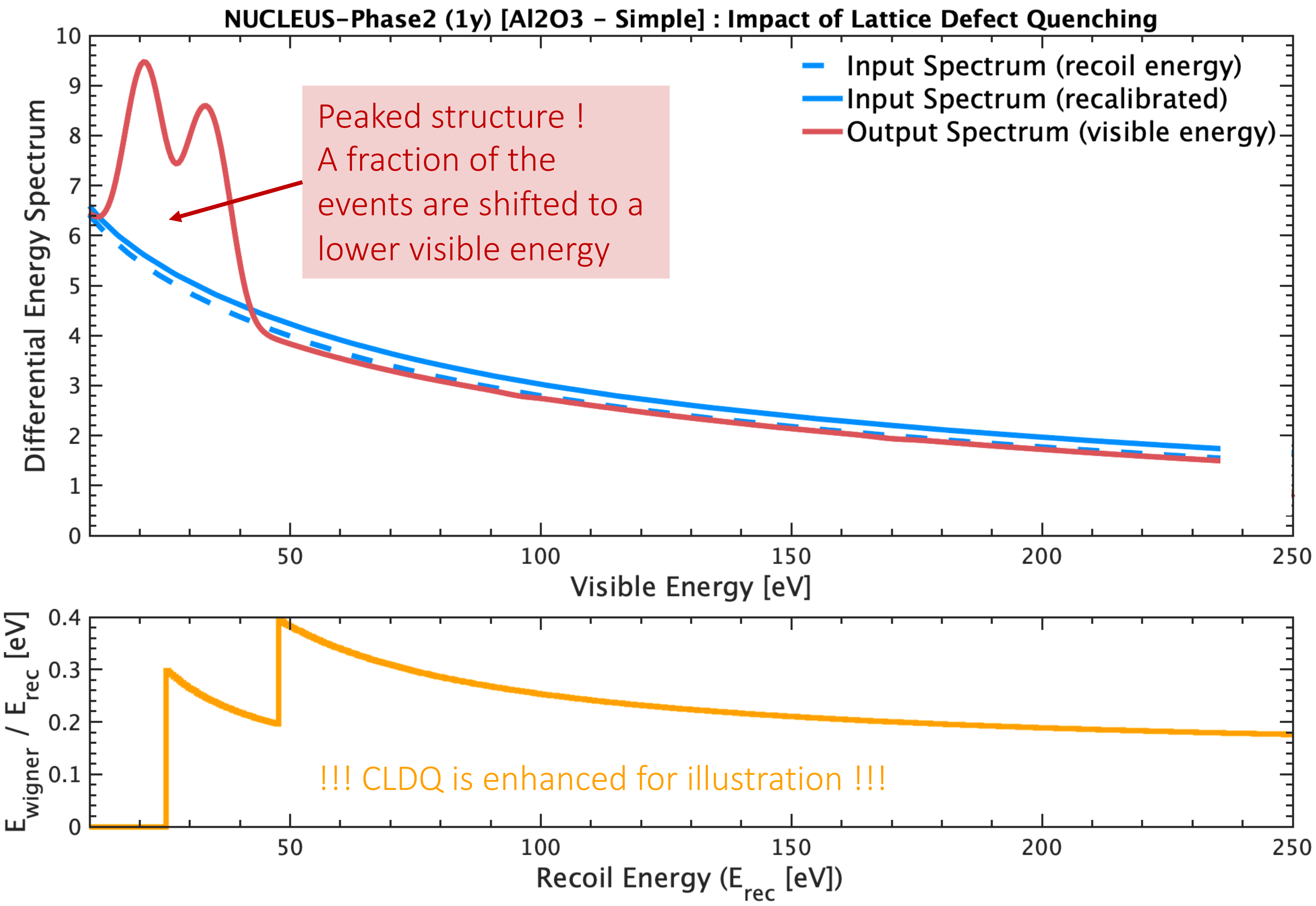




E_f: #vacancies (descrete)

E_d: threshold effect

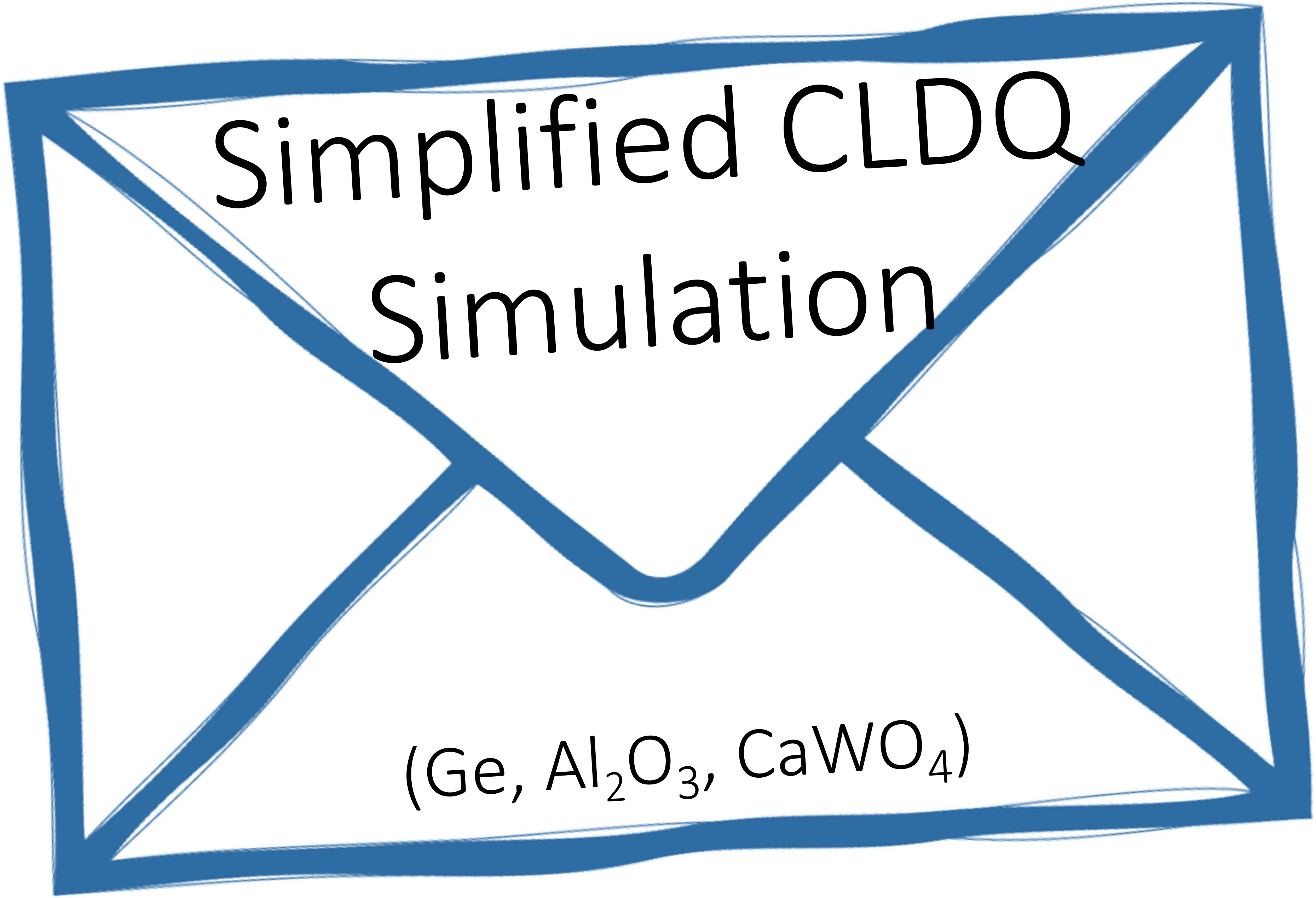
Impact on CEVNS spectrum (Al$_2$O$_3$ – analytical evaluation)



Peaked structure !
A fraction of the
events are shifted to a
lower visible energy

!!! CLDQ is enhanced for illustration !!!

- Blue Curve:
  Input reactor
  CEVNS spectrum
  versus true recoil energy
- Red Curve:
  CEνNS Spectrum including
  lattice defects energy loss
  versus the visible energy
- Gold Curve:
  relative energy loss
  (no shape distortion for
  E>100eV)



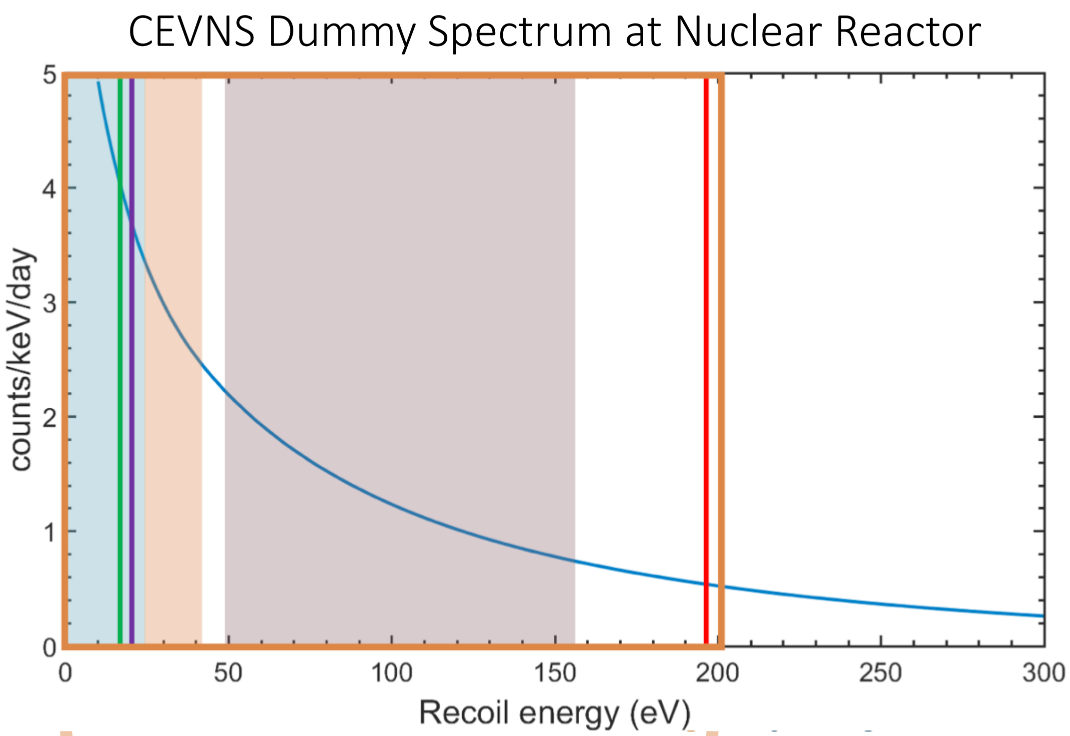
Simplified CLDQ Simulation

(Ge, Al_2O_3, $CaWO_4$)

Displacement energies & Formation energies

Crystal	Atom	E_d	E_f
Germanium	Ge	from 9.5 eV to 28.5 eV	from 2 eV to 4 eV
Al_2O_3	Al	from 45.5 eV to > 150 eV	4.95 eV
	O	from 25 eV to 40.5 eV	6.52 eV
$CaWO_4$	Ca	24.02 eV	4.04 eV
	W	195.83 eV	7.02 eV
	O	20.30 eV	1.23 eV

Jiang et al., A Theoretical Simulation of the Radiation Responses of Si, Ge, and Si/Ge Superlattice to Low-Energy Irradiation, 2018
Jiang et al., Ab initio molecular dynamics simulation of low energy radiation responses of α-Al2O3, 2017
Matsunaga et al., First-Principles Calculations of Intrinsic Defects in Al2O3, 2003.
Shao et al, Computer study of intrinsic defects in CaWO4, 2008.



Displacement energies matches the CEVNS@Reactor Regions Of Interest!

One step further: TRIM/SRIM simulation

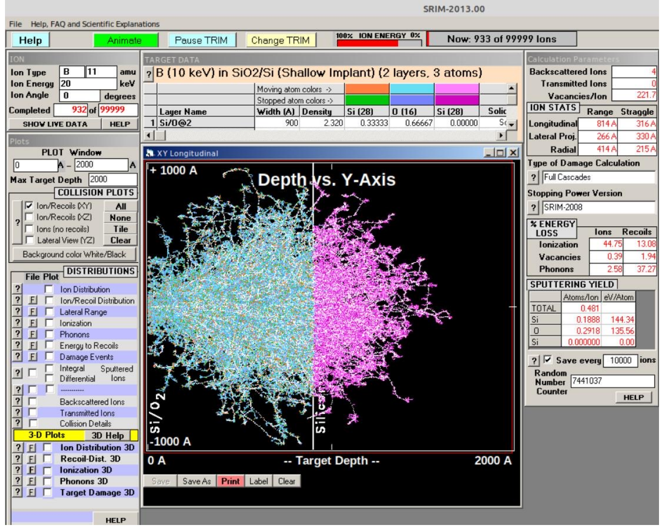
The Stopping and Range of Ions in Matter

J. F. Ziegler
U.S.N.A.
Annapolis, MD, USA

J. P. Biersack
Hahn-Meitner Inst.
Berlin, Germany

SRIM Version: SRIM-2013.00

Significant contributions by Helmut Paul (Linz), Roger Webb (Surrey), Xiao Yu (Beijing)
(c) 1984,1989,1998, 2008, 2012 by J. F. Ziegler, M.D. Ziegler, J. P. Biersack (SRIM.com)



SRIM-2013.00

Now: 933 of 99999 Ions

ION
Ion Type: B 11 amu
Ion Energy: 20 keV
Ion Angle: 0 degrees
Completed: 932 of 99999

TARGET DATA
B (10 keV) in SiO2/Si (Shallow Implant) (2 layers, 3 atoms)

	Layer Name	Width (A)	Density	Si (28)	O (16)	Si (28)	Solic
1	Si/O@2	900	2.320	0.33333	0.66667	0.00000	Sc

PLOT Window: 0 A – 2000 A
Max Target Depth: 2000

Calculation Parameters
Backscattered Ions: 4
Transmitted Ions: 0
Vacancies/Ion: 221.7

ION STATS	Range	Straggle
Longitudinal	814 A	316 A
Lateral Proj.	266 A	330 A
Radial	414 A	215 A

Type of Damage Calculation: Full Cascades
Stopping Power Version: SRIM-2008

% ENERGY LOSS	Ions	Recoils
Ionization	44.75	13.08
Vacancies	0.39	1.94
Phonons	2.58	37.27

SPUTTERING YIELD

	Atoms/Ion	eV/Atom
TOTAL	0.481	
Si	0.1888	144.34
O	0.2918	135.56
Si	0.000000	0.00

☑ Save every 10000 ions
Random Number Counter: 7441037

SRIM - Calculate the stopping and range of ions into matter

- A moving atom is considered as an **"ion"** and all target atoms as **"atoms"**
- Uses a quantum mechanical treatment

TRIM - (the Transport of Ions in Matter) - Monte Carlo

- 2D Target
- Anamorphous
- Specific recoils generated at a specific target location/orientation

Radiation Damage MC simulation

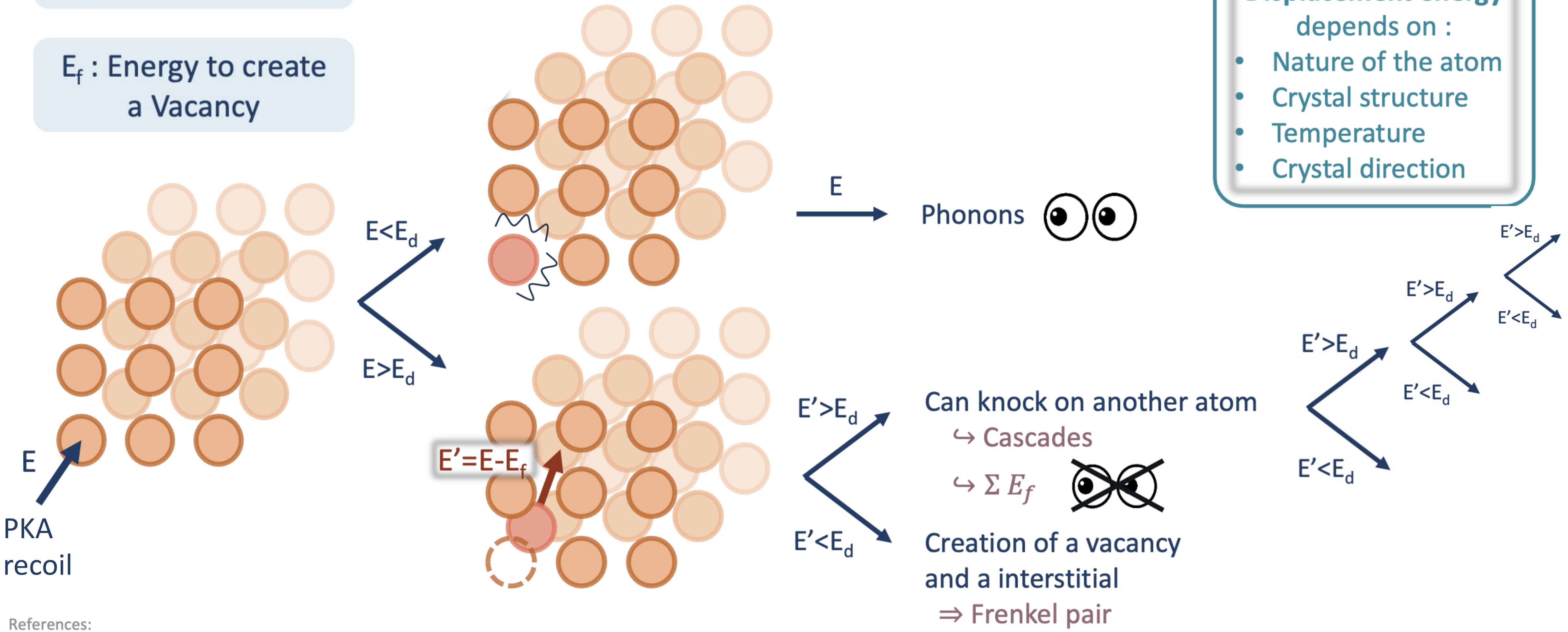
- Emulation of cascades in crystal – fair estimate
- Input parameters
  - E_{disp} : Threshold displacement energy
  - E_{latt} : Defect formation energy
  - Material density

"SRIM - The Stopping and Range of Ions in Solids", by J. F. Ziegler and J. P. Biersack in 1985 (a new edition in 2009)

Kinchin-Pease recoil cascade’s simulation

E_d : Threshold displacement energy

E_f : Energy to create a Vacancy



E

PKA recoil

$E<E_d$

$E>E_d$

$E'=E-E_f$

E → Phonons

$E'>E_d$

Can knock on another atom

↪ Cascades

↪ ΣE_f

$E'<E_d$

Creation of a vacancy and a interstitial

⇒ Frenkel pair

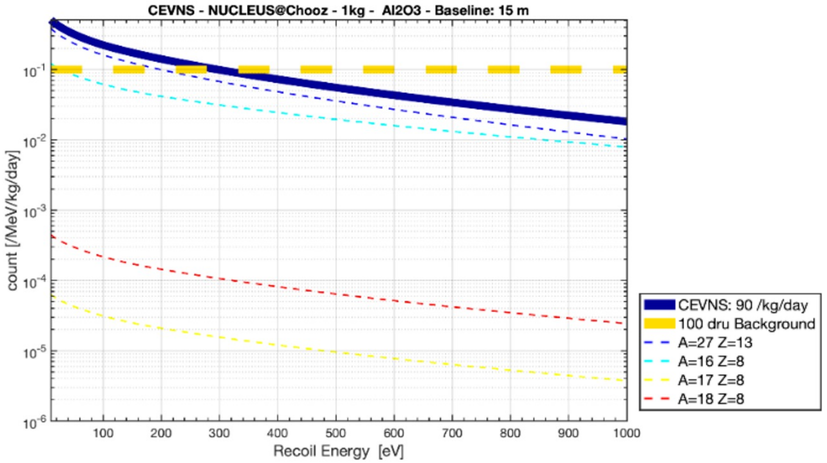
Displacement energy depends on :

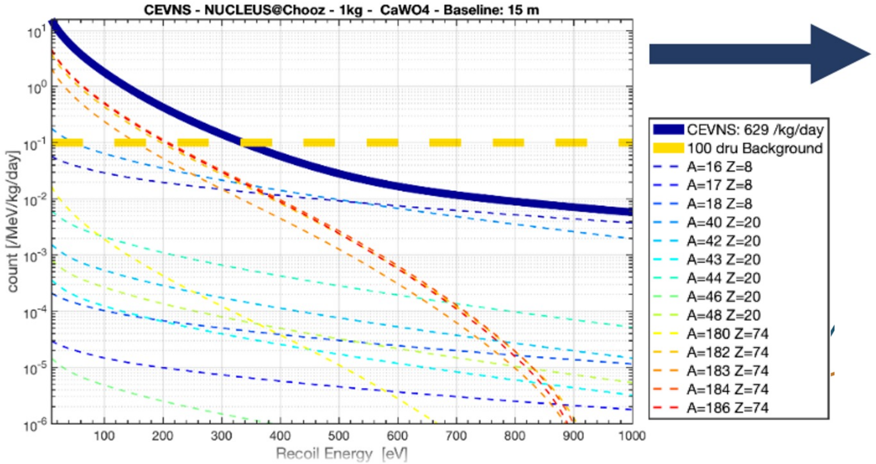
- Nature of the atom
- Crystal structure
- Temperature
- Crystal direction

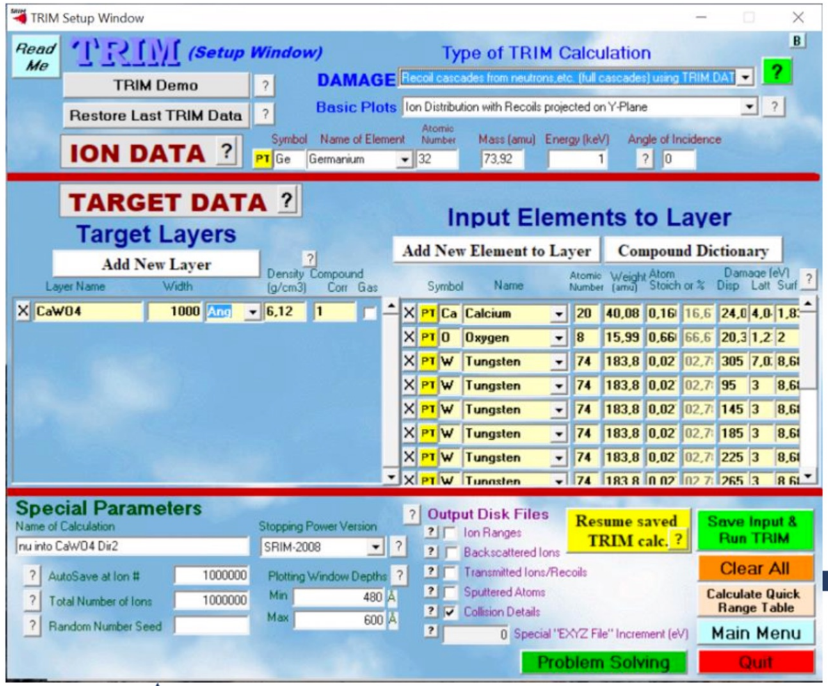
References:
[5] H. Kinchin and R. S. Pease. The displacement of atoms in solids by radiation, jan 1955.

TRIM inputs and outputs

Input: CEνNS @Reactor Recoil Spectrum







Type of TRIM Calculation

DAMAGE: Recoil cascades from neutrons,etc. (full cascades) using TRIM.DAT

Basic Plots: Ion Distribution with Recoils projected on Y-Plane

Symbol	Name of Element	Atomic Number	Mass (amu)	Energy (keV)	Angle of Incidence
Ge	Germanium	32	73,92	1	0

Layer Name	Width		Density (g/cm3)	Compound Corr	Gas
CaW04	1000	Ang	6,12	1	☐

Symbol	Name	Atomic Number	Weight (amu)	Atom Stoich	or %	Damage (eV) Disp	Latt	Surf
Ca	Calcium	20	40,08	0,16	16,6	24,0	4,0	1,8
O	Oxygen	8	15,99	0,66	66,6	20,3	1,2	2
W	Tungsten	74	183,8	0,02	02,7	305	7,0	8,6
W	Tungsten	74	183,8	0,02	02,7	95	3	8,6
W	Tungsten	74	183,8	0,02	02,7	145	3	8,6
W	Tungsten	74	183,8	0,02	02,7	185	3	8,6
W	Tungsten	74	183,8	0,02	02,7	225	3	8,6
W	Tungsten	74	183,8	0,02	02,7	265	3	8,6

Name of Calculation: nu into CaWO4 Dir2

Stopping Power Version: SRIM-2008

AutoSave at Ion #: 1000000

Total Number of Ions: 1000000

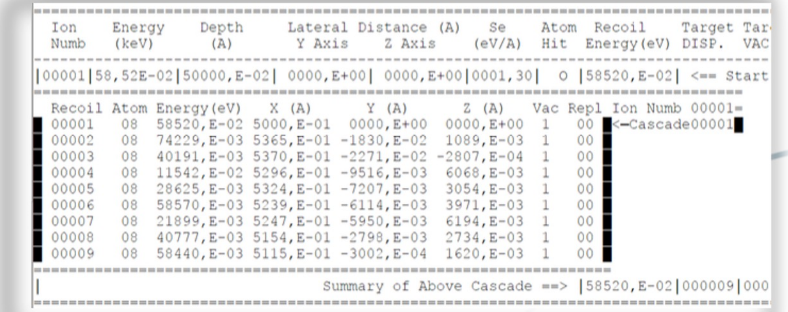
Random Number Seed:

Plotting Window Depths: Min 480 Å, Max 600 Å

- ☐ Ion Ranges
- ☐ Backscattered Ions
- ☐ Transmitted Ions/Recoils
- ☐ Sputtered Atoms
- ☑ Collision Details
- 0 Special "EXYZ File" Increment (eV)

Output :

.txt file with the detail of the collisions and the detail of cascades

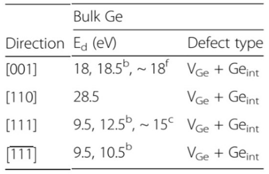


```
=====================================================================================================
 Ion     Energy     Depth      Lateral Distance (A)    Se     Atom  Recoil      Target Tar
 Numb    (keV)       (A)        Y Axis     Z Axis     (eV/A)  Hit  Energy(eV)  DISP.  VAC
-----------------------------------------------------------------------------------------------------
|00001|58,52E-02|50000,E-02| 0000,E+00| 0000,E+00|0001,30|  O |58520,E-02| <== Start
=================================================================================================
 Recoil Atom Energy(eV)   X (A)      Y (A)       Z (A)     Vac Repl Ion Numb 00001=
█00001   08  58520,E-02 5000,E-01  0000,E+00  0000,E+00  1   00 █<—Cascade00001█
█00002   08  74229,E-03 5365,E-01 -1830,E-02  1089,E-03  1   00 █
█00003   08  40191,E-03 5370,E-01 -2271,E-02 -2807,E-04  1   00 █
█00004   08  11542,E-02 5296,E-01 -9516,E-03  6068,E-03  1   00 █
█00005   08  28625,E-03 5324,E-01 -7207,E-03  3054,E-03  1   00 █
█00006   08  58570,E-03 5239,E-01 -6114,E-03  3971,E-03  1   00 █
█00007   08  21899,E-03 5247,E-01 -5950,E-03  6194,E-03  1   00 █
█00008   08  40777,E-03 5154,E-01 -2798,E-03  2734,E-03  1   00 █
█00009   08  58440,E-03 5115,E-01 -3002,E-04  1620,E-03  1   00 █
================================================================================
|                                        Summary of Above Cascade ==> |58520,E-02|000009|000
=====================================================================================================
```

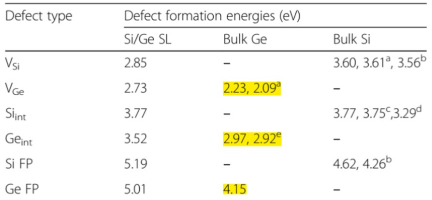
Inpuy: Target simulated

- Germanium
- Al_2O_3
- $CaWO_4$

Specific weighted set of E_d/E_f tc emulated the crystal orientation

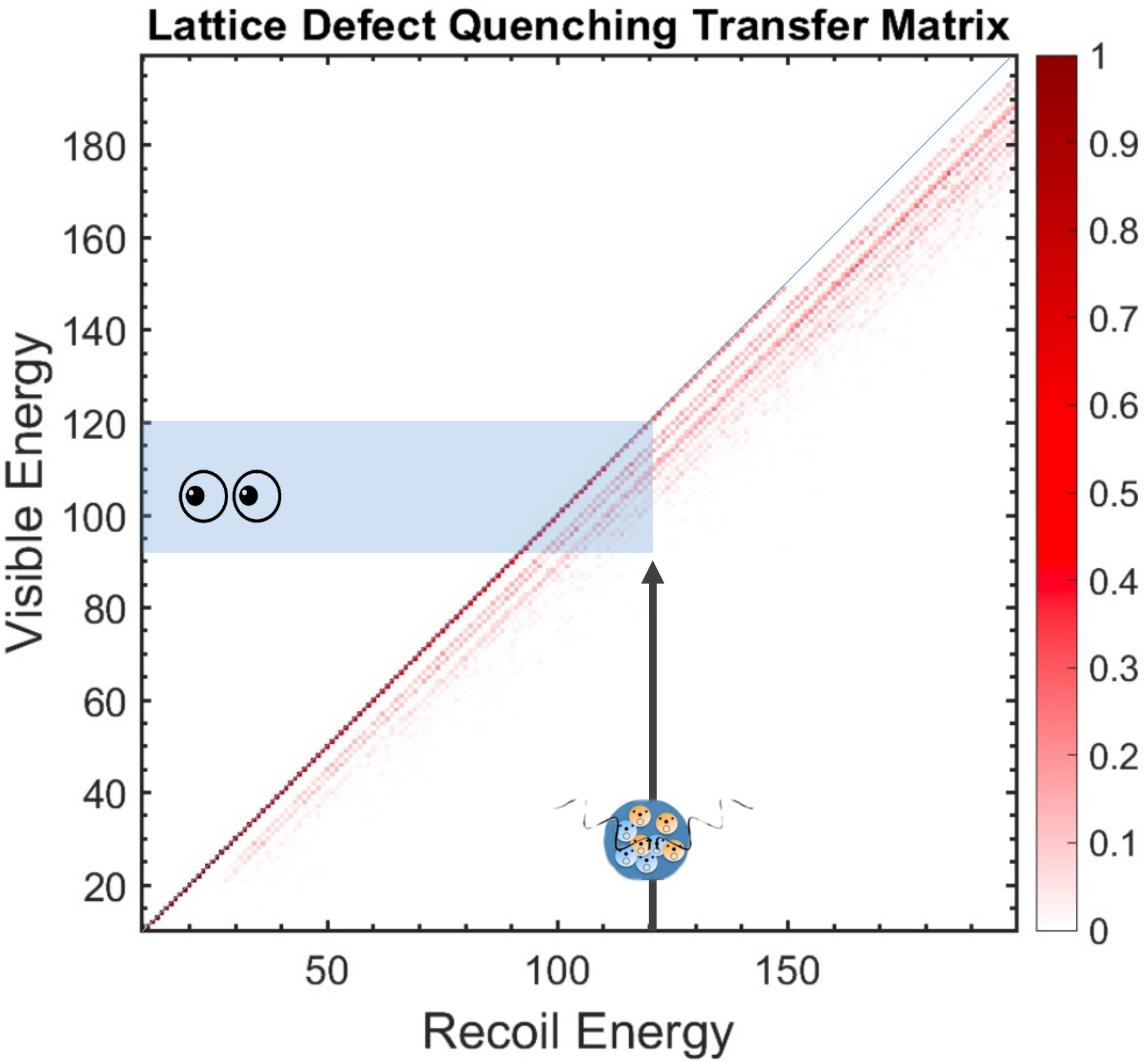


	Bulk Ge	
Direction	E_d (eV)	Defect type
[001]	18, 18.5[b], ~ 18[f]	$V_{Ge} + Ge_{int}$
[110]	28.5	$V_{Ge} + Ge_{int}$
[111]	9.5, 12.5[b], ~ 15[c]	$V_{Ge} + Ge_{int}$
$[\overline{1}\overline{1}\overline{1}]$	9.5, 10.5[b]	$V_{Ge} + Ge_{int}$

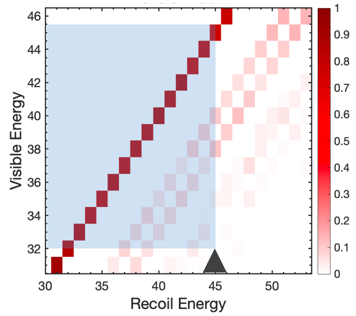


Defect type	Defect formation energies (eV)		
	Si/Ge SL	Bulk Ge	Bulk Si
V_{Si}	2.85	–	3.60, 3.61[a], 3.56[b]
V_{Ge}	2.73	2.23, 2.09[a]	–
Si_{int}	3.77	–	3.77, 3.75[c],3.29[d]
Ge_{int}	3.52	2.97, 2.92[e]	–
Si FP	5.19	–	4.62, 4.26[b]
Ge FP	5.01	4.15	–

Visible/Recoil transfer matrix (here $\alpha - \mathrm{Al_2O_3}$)

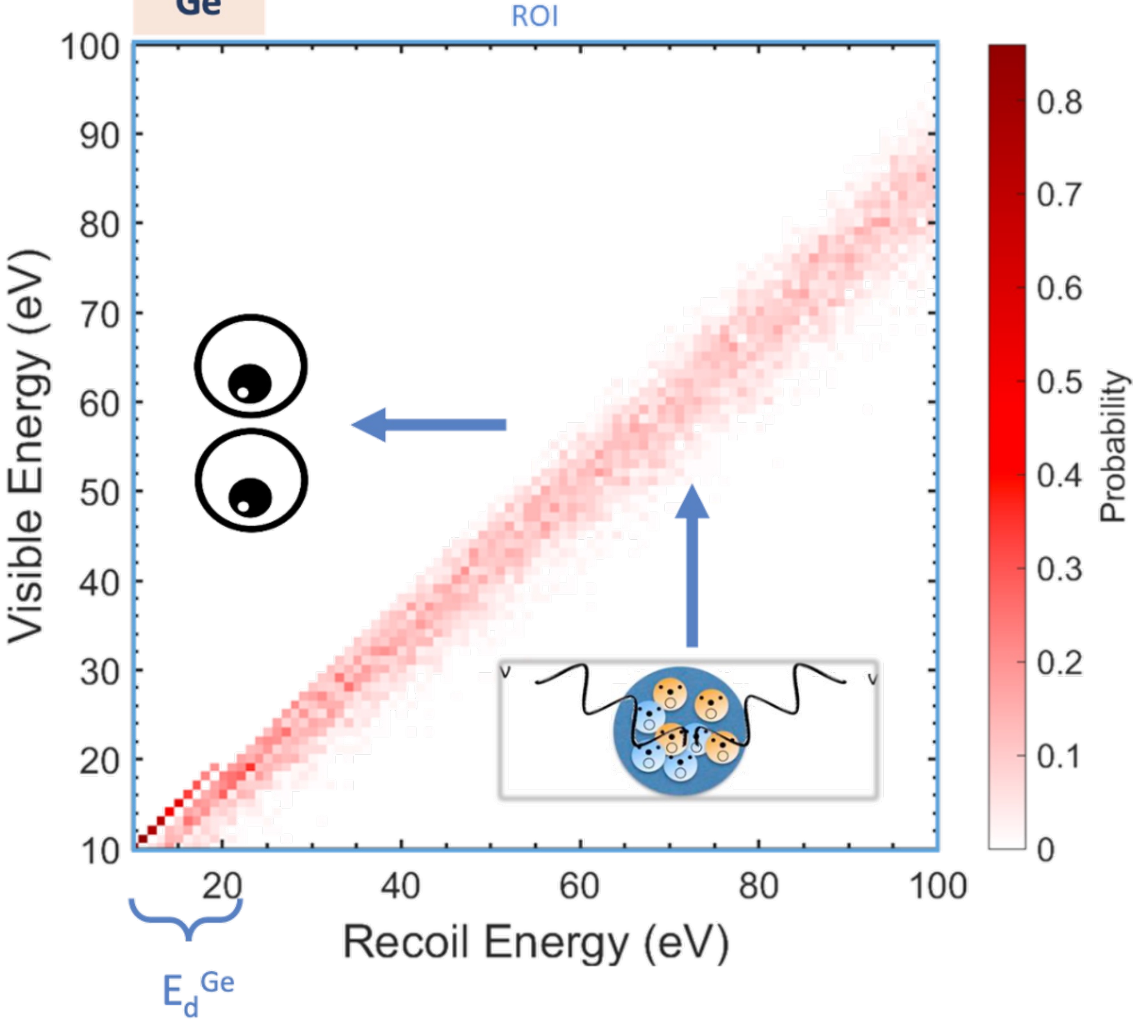


- Transfer Matrix (TRIM):
  Visible energies $\equiv \mathbf{f}(\textbf{Recoil Energy})$
- Visible energy < Recoil Energy
  - CLD Quenching
  - Specific values depend on E_d's, E_f's
  - E_d's, E_f's still not well known
- A range of visible energies can be tied to a single recoil energy!
  - Energy cannot be assigned on an event-by-event basis!
- Energy spectrum reconstruction can be statistically processed

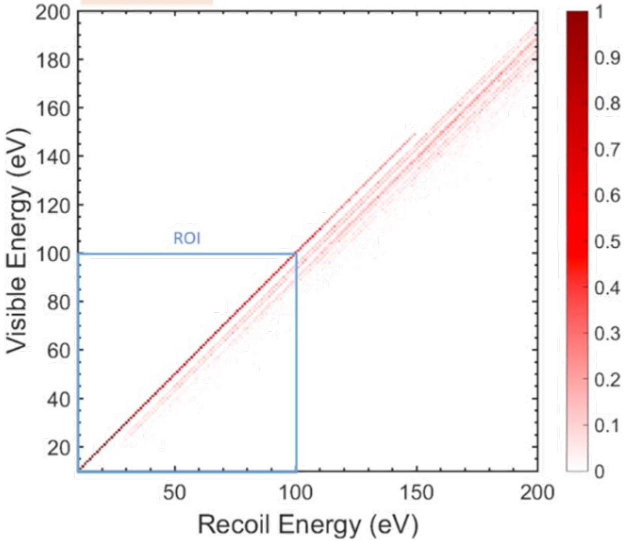


Visible/Recoil transfer matrices

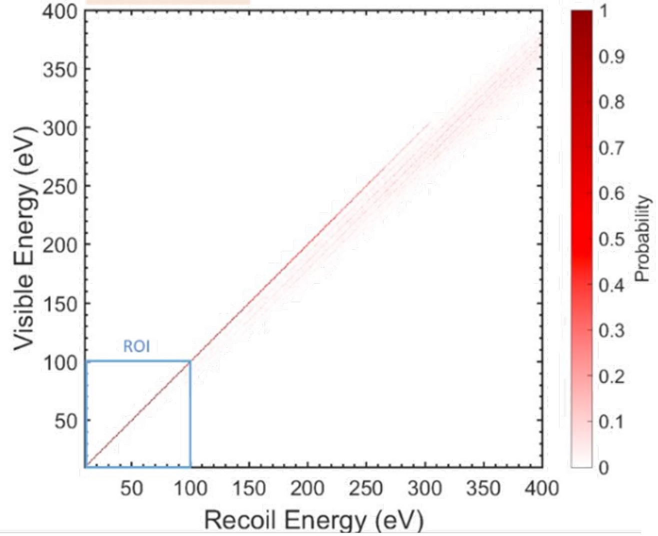
Ge



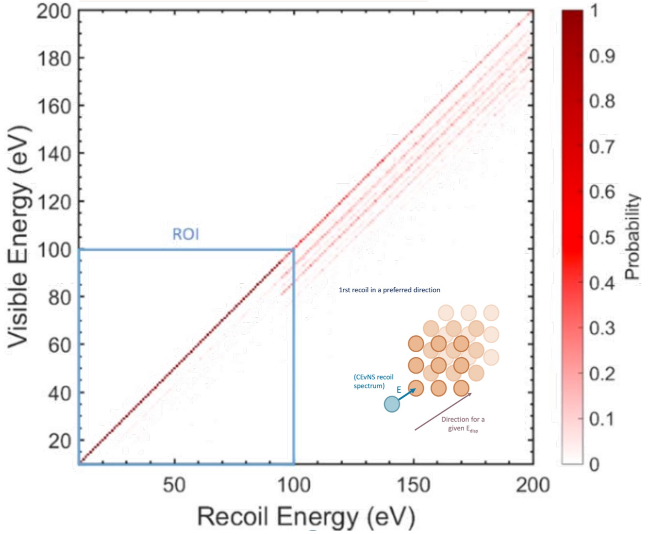
Al_2O_3



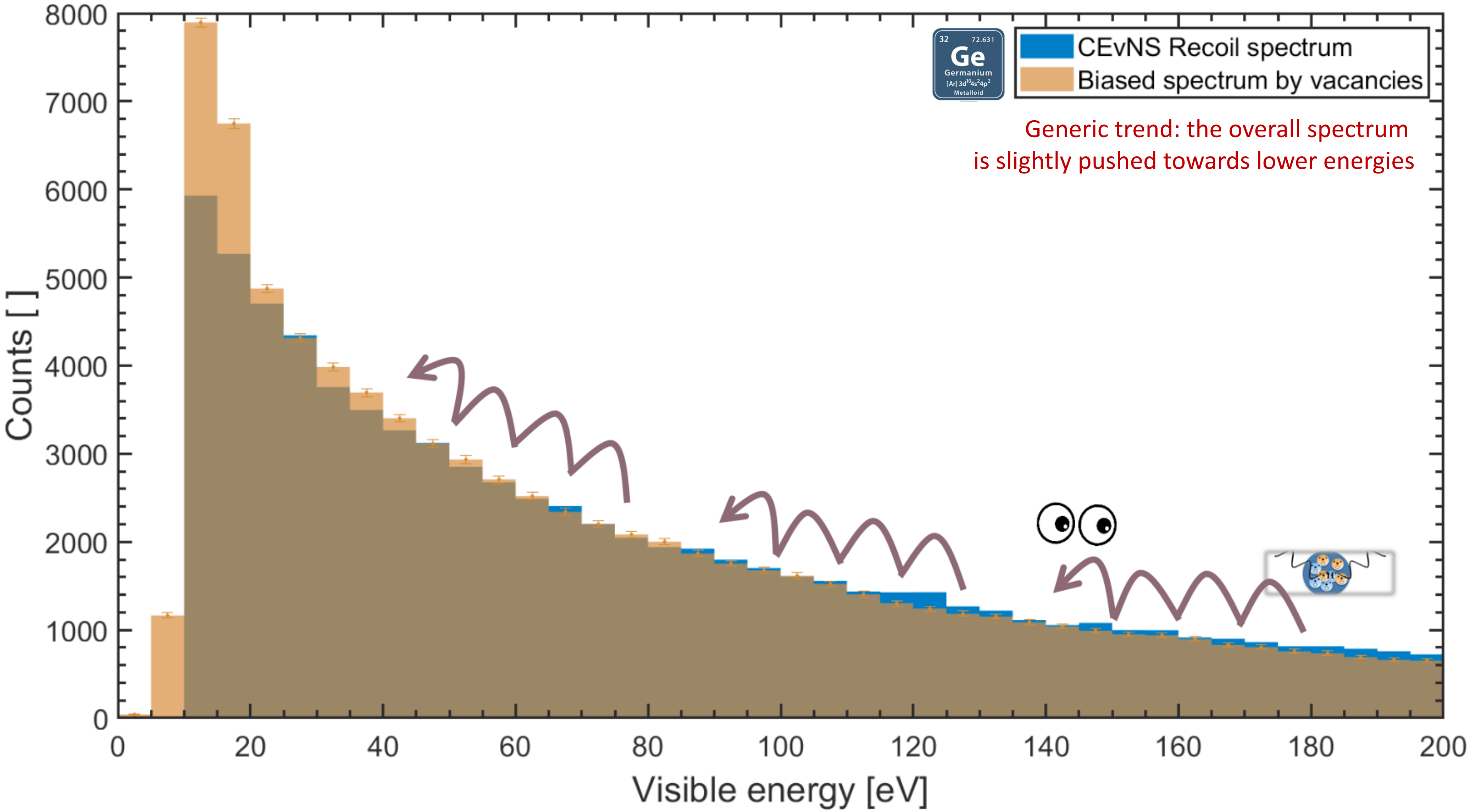
$CaWO_4$



$CaWO_4$ – priv. dir.

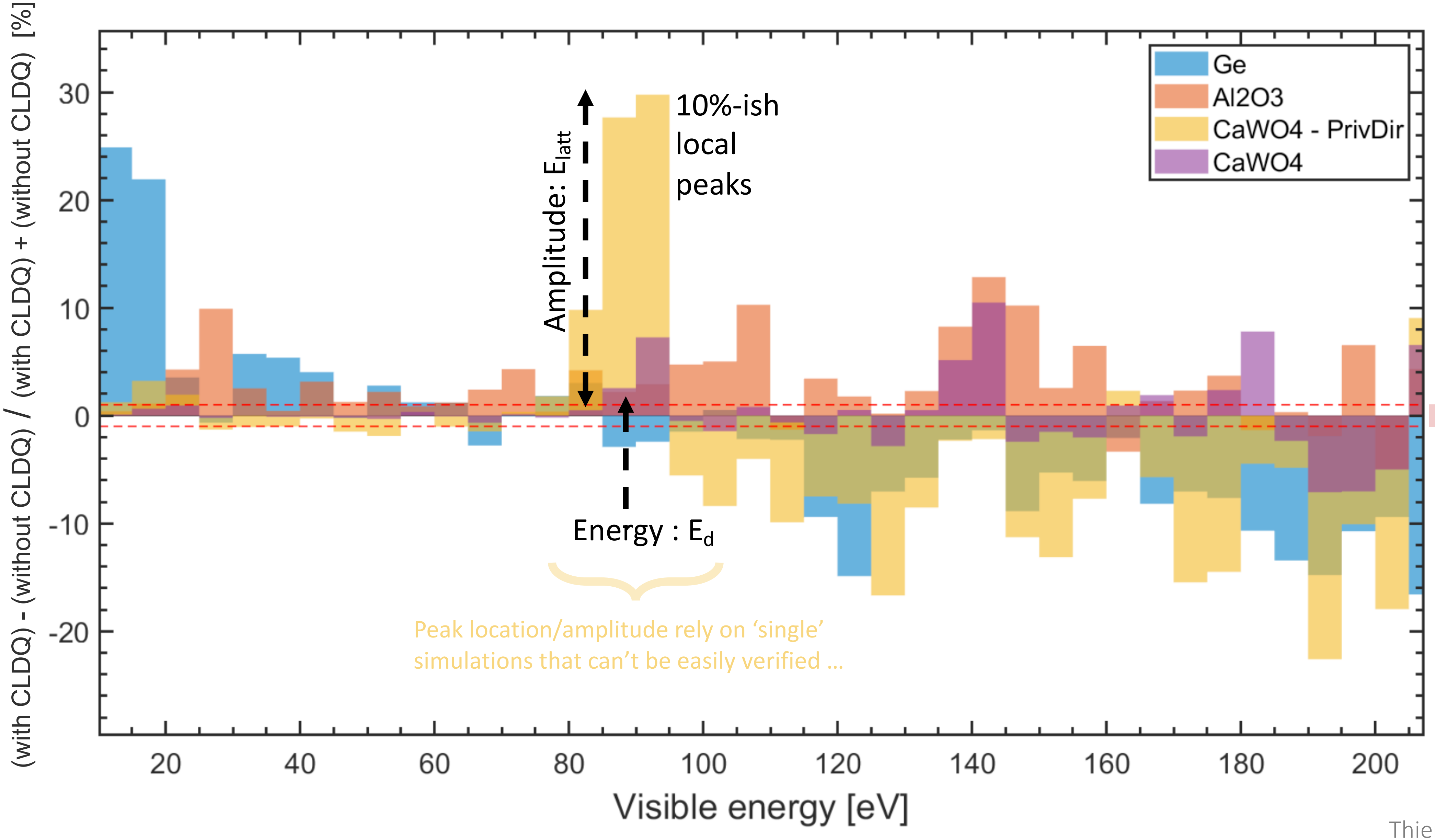


CEVNS@reactor: visible & recoil spectral shapes



Generic trend: the overall spectrum
is slightly pushed towards lower energies

CEVNS@reactor: CLDQ spectral distorsion



10%-ish local peaks

Amplitude: E_{latt}

Energy : E_d

Peak location/amplitude rely on 'single' simulations that can't be easily verified …

±1%

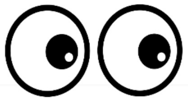
Robust global percent-ish effect

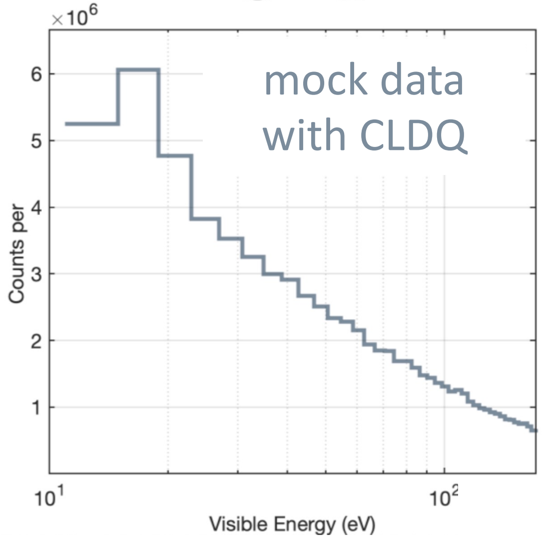
Impact on CEνNS cross-section measurement

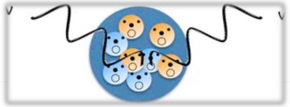
- **Goal: estimate the impact of the CLDQ on the CEνNS cross section measurement**

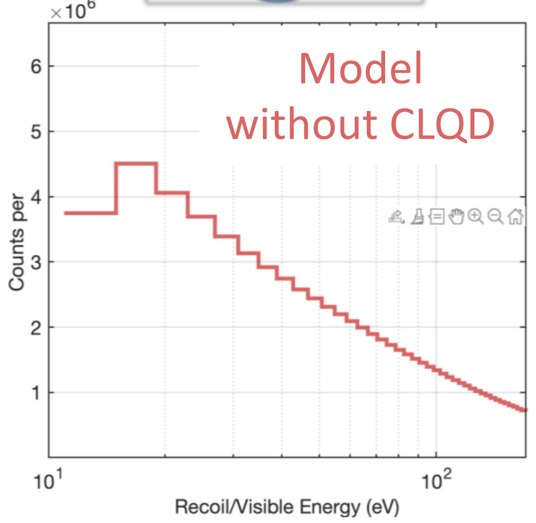
- Method:
  - Use SRIM simulations to evaluate CLDQ impact on CEνNS spectrum (conversion from recoil energy to visible energy)
  - Compare (fit) mock data with CLDQ to the CEνNS model not including CLDQ
  - Consider infinite statistics – 100 millions CEνNS int. –

- **Normalization bias = estimator of the CLDQ systematics**



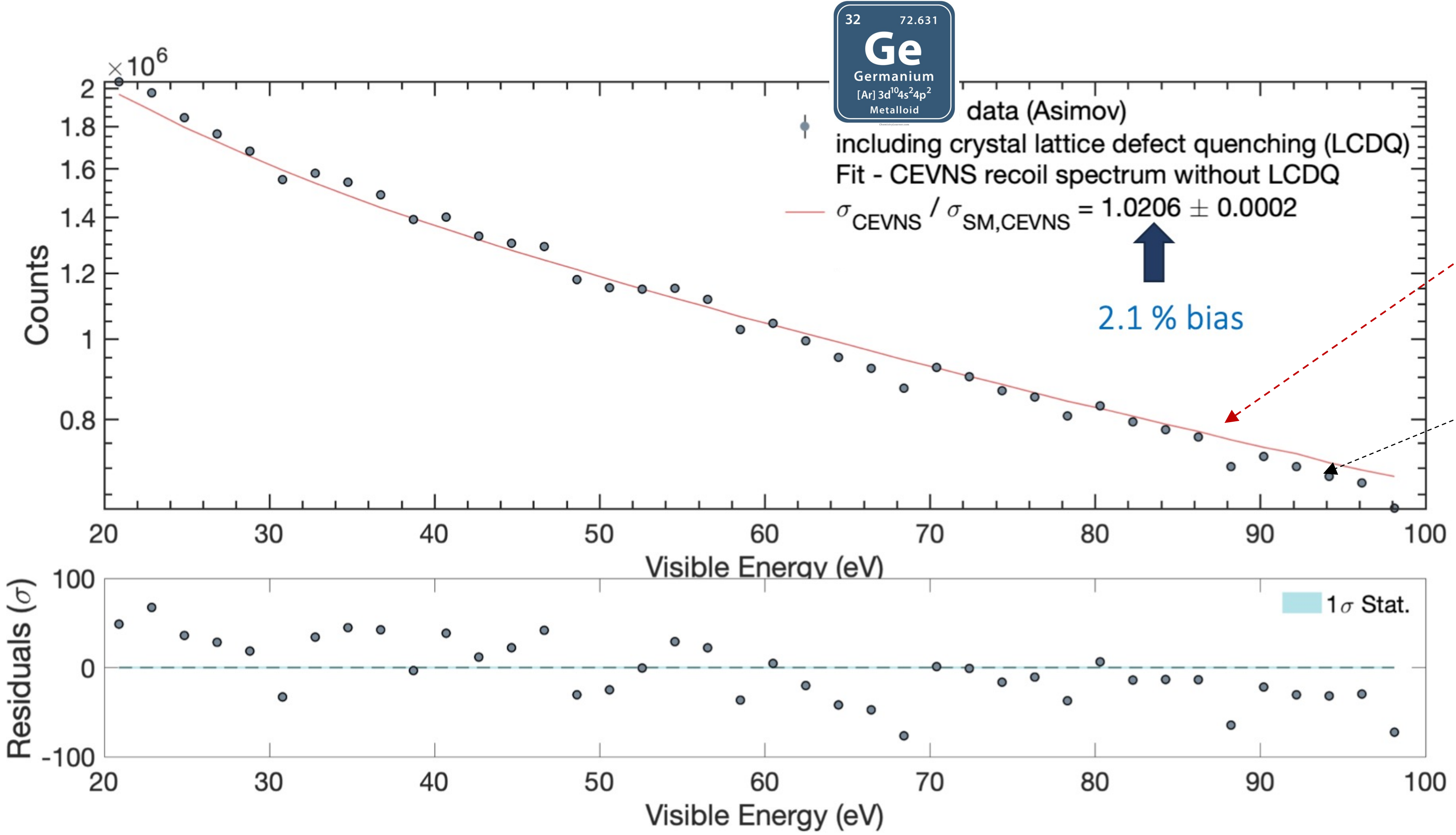






Impact on CEνNS cross-section measurement

Fit Ge CEνNS mock data with model without CLDQ
ROI between 20 eV and 100 eV

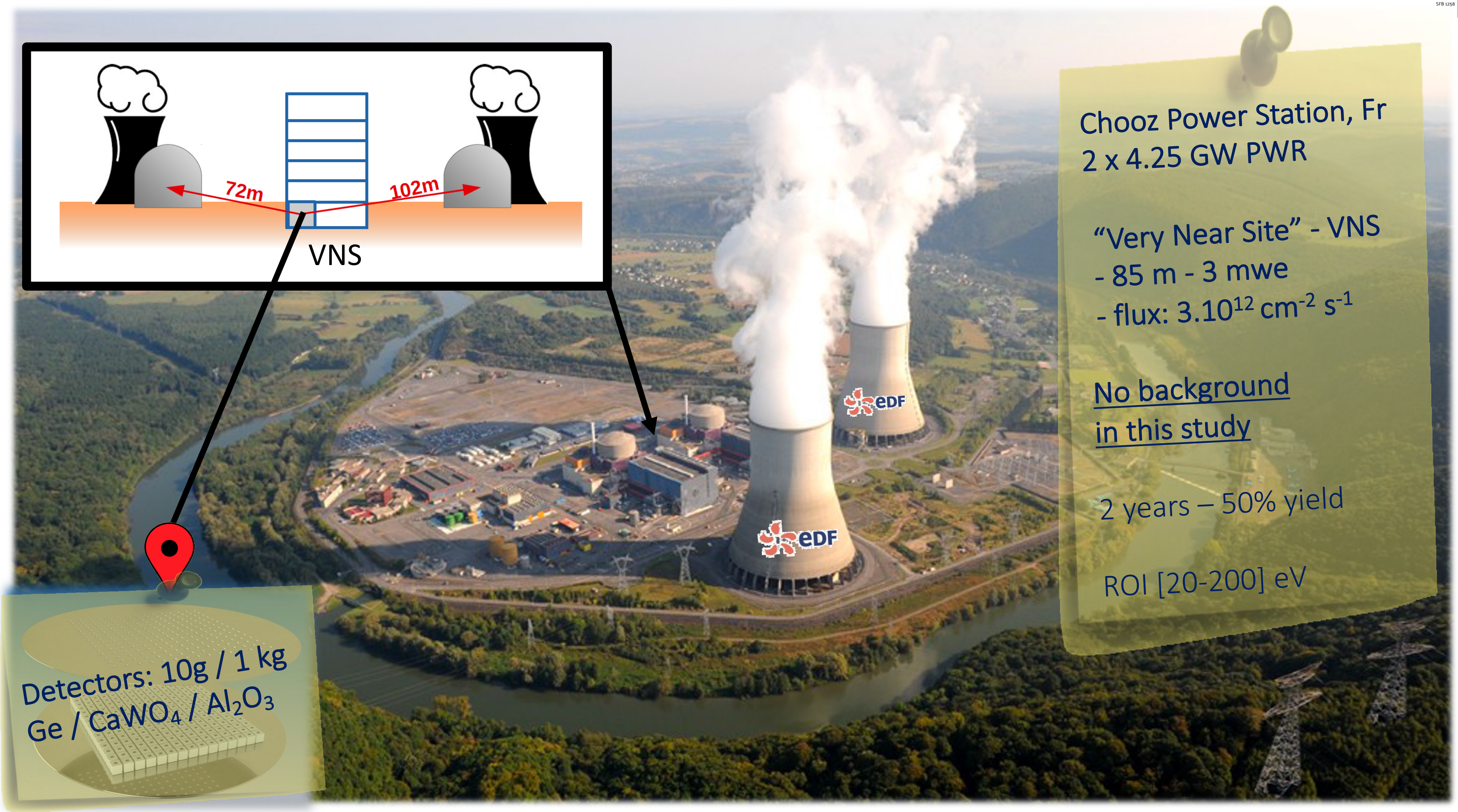


Best fit
CEνNS model
no CLDQ

CEνNS mock data
with sim. CLDQ

CLDQ systematics:
2.1%

Proxy-setup : NUCLEUS@Chooz – Phase I & II



Chooz Power Station, Fr
2 x 4.25 GW PWR

“Very Near Site” - VNS
- 85 m - 3 mwe
- flux: 3.10^{12} cm^{-2} s^{-1}

<u>No background in this study</u>

2 years – 50% yield

ROI [20-200] eV

Detectors: 10g / 1 kg
Ge / $CaWO_4$ / Al_2O_3

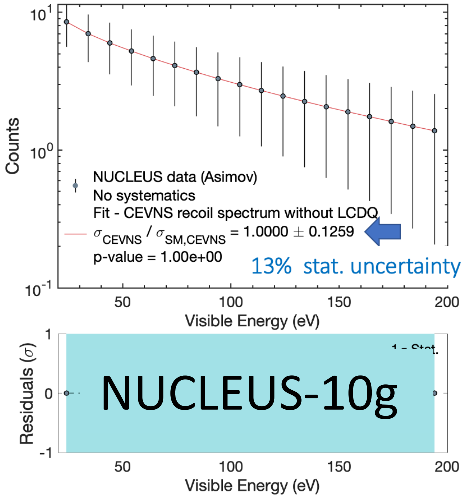
CLDQ Systematics for CEνNS cross-section

$\sigma_{\mathrm{stat}} \gg \sigma_{\mathrm{CLDQ}}$

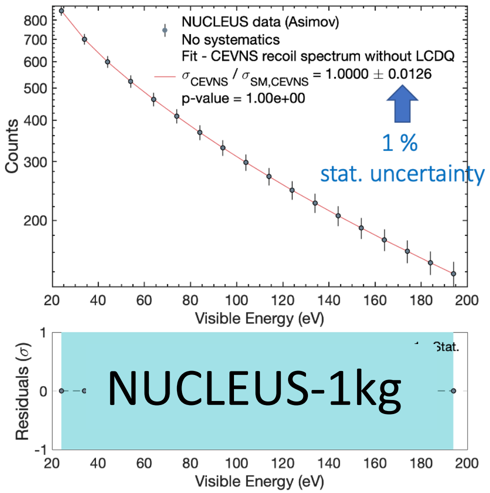
$\sigma_{\mathrm{stat}} \lesssim \sigma_{\mathrm{CLDQ}}$

	NUCLEUS-10g (2 years)		**NUCLEUS-1kg (2 years)**		**CLDQ**
Target	CEvNS events	Stat. Error	CEvNS events	Stat. Error	Sys. Error
Ge	75	13%	7500	1.3%	2.1%
Al_2O_3	59	17%	5900	1.7%	3.2%
$CaWO_4$	156	9.1%	15600	0.9%	0.6-1.8%

Simulation of CEνNS cross section measurement →



13% stat. uncertainty



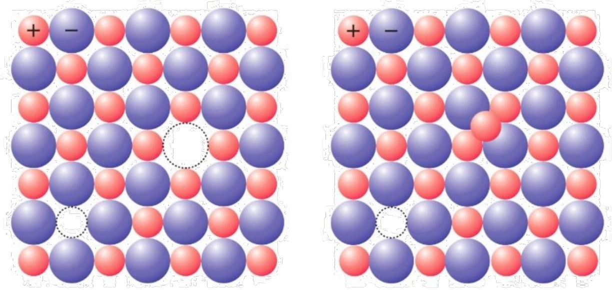
1 %
stat. uncertainty

Limitations of this study

☹ Displacement energies & formation energies are still not yet known

Ge		Al$_2$O$_3$		CaWO$_4$	
E_d 🟩	E_f 🟩	E_d 🟩	E_f 🟥	E_d 🟥	E_f 🟥

☹ Limited type of defects have been considered (others?)



(a) Schottky defect (b) Frenkel defect

☹ TRIM/SRIM : 2D amorphous material

A user-friendly toolkit for a fair estimate

The Stopping and Range of Ions in Matter

J. F. Ziegler
U.S.N.A.
Annapolis, MD, USA

J. P. Biersack
Hahn-Meitner Inst.
Berlin, Germany

SRIM Version
SRIM-2013.00

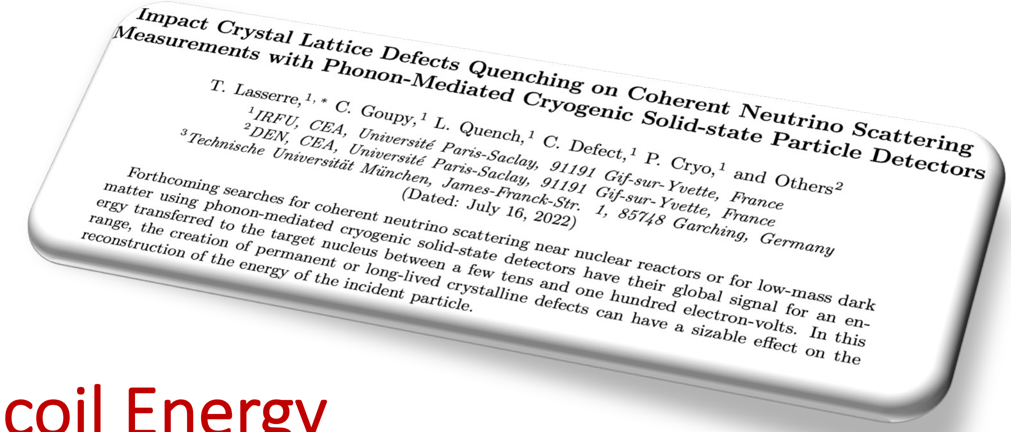
Significant contributions by Helmut Paul (Linz), Roger Webb (Surrey), Xiao Yu (Beijing)

(c) 1984,1989,1998, 2008, 2012 by J. F. Ziegler, M.D. Ziegler, J. P. Biersack (SRIM.com)

Conclusions

A quantitative study of the impact of the **Crystal Lattice Defect Quenching** for the forthcoming **reactor neutrino CEνNS** measurements

- Rely on:
    - Litterature values of E_{dis} / E_f
    - Frenkel defect only
    - TRIM/SRIM cascade simulations



Impact Crystal Lattice Defects Quenching on Coherent Neutrino Scattering Measurements with Phonon-Mediated Cryogenic Solid-state Particle Detectors

T. Lasserre,[1,*] C. Goupy,[1] L. Quench,[1] C. Defect,[1] P. Cryo,[1] and Others[2]

[1] IRFU, CEA, Université Paris-Saclay, 91191 Gif-sur-Yvette, France
[2] DEN, CEA, Université Paris-Saclay, 91191 Gif-sur-Yvette, France
[3] Technische Universität München, James-Franck-Str. 1, 85748 Garching, Germany

(Dated: July 16, 2022)

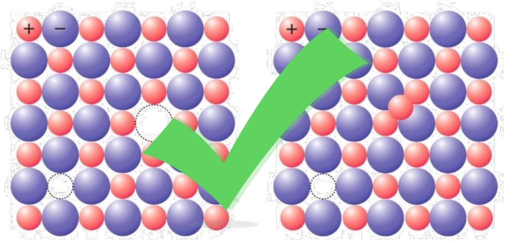
Forthcoming searches for coherent neutrino scattering near nuclear reactors or for low-mass dark matter using phonon-mediated cryogenic solid-state detectors have their global signal for an energy transferred to the target nucleus between a few tens and one hundred electron-volts. In this range, the creation of permanent or long-lived crystalline defects can have a sizable effect on the reconstruction of the energy of the incident particle.

- « Phonon detector »: Visible Energy ≠ Recoil Energy
    - Impact on energy calibration
    - Non-linear effect for $E_{vis} \lesssim \max(E_d)$

A 1-3% systematics effect on the CEνNS cross section (if neglected)

- Could this jeopardize beyond standard model physics searches?

Perspectives by 2024

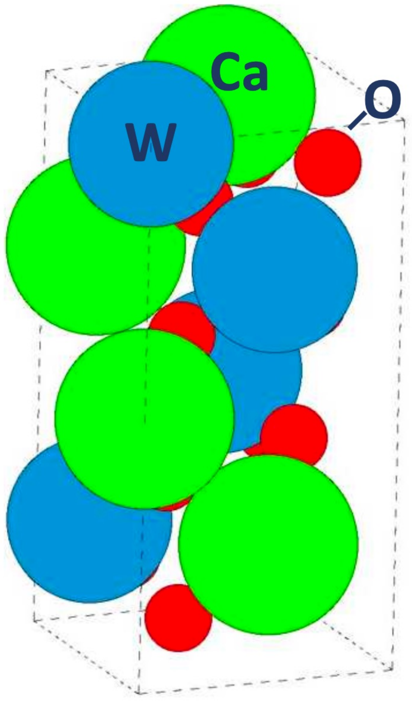


(a) Schottky defect (b) Frenkel defect

Ge		Al_2O_3		$CaWO_4$	
E_d	E_f	E_d	E_f	E_d	E_f

Ongoing collaboration with solid state physicists at CEA – Saclay

- Dedicated VASP simulations
- Identify defects and calculate their formation energies
- Calculate displacement energies



@G. Soum

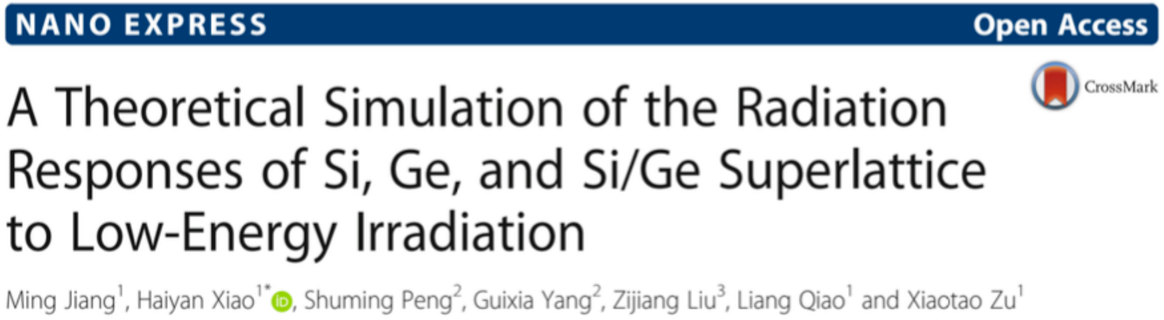
Thank you for your attention

VIII. Appendices

B. Simulated Targets - Ge

Target germanium simulated :

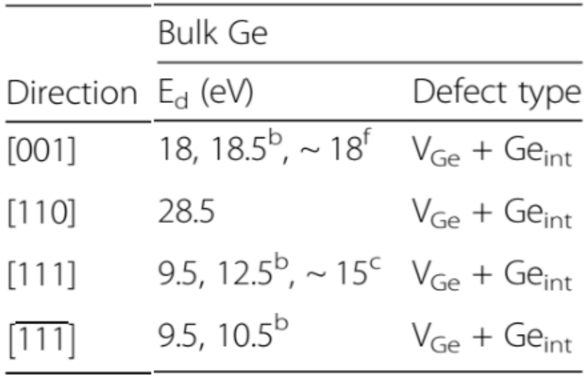
E_{disp} (eV)	E_{latt} (eV)
10	2
	3
	4
12	2
	3
	4
14	2
	3
	4
16	2
	3
	4
18	2
	3
	4
20	2
	3
	4



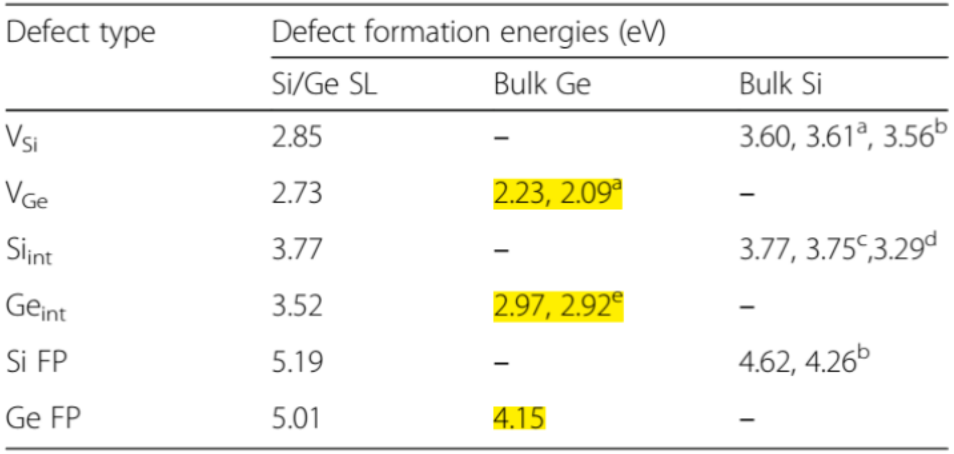
NANO EXPRESS Open Access

A Theoretical Simulation of the Radiation Responses of Si, Ge, and Si/Ge Superlattice to Low-Energy Irradiation

Ming Jiang[1], Haiyan Xiao[1*], Shuming Peng[2], Guixia Yang[2], Zijiang Liu[3], Liang Qiao[1] and Xiaotao Zu[1]



	Bulk Ge	
Direction	E_d (eV)	Defect type
[001]	18, 18.5^b, $\sim 18^f$	$V_{Ge} + Ge_{int}$
[110]	28.5	$V_{Ge} + Ge_{int}$
[111]	9.5, 12.5^b, $\sim 15^c$	$V_{Ge} + Ge_{int}$
$[\bar{1}11]$	9.5, 10.5^b	$V_{Ge} + Ge_{int}$



Defect type	Defect formation energies (eV)		
	Si/Ge SL	Bulk Ge	Bulk Si
V_{Si}	2.85	–	3.60, 3.61^a, 3.56^b
V_{Ge}	2.73	2.23, 2.09^a	–
Si_{int}	3.77	–	3.77, 3.75^c, 3.29^d
Ge_{int}	3.52	2.97, 2.92^e	–
Si FP	5.19	–	4.62, 4.26^b
Ge FP	5.01	4.15	–

VIII. Appendices

B. Simulated Targets – Al_2O_3

Yuan, Y.G., Jiang, M., Zhao, F.A. *et al.* Ab *initio* molecular dynamics simulation of low energy radiation responses of α-Al_2O_3 (2017).

Threshold energy displacement: E_{disp} in Al_2O_3 ?

Direction classes – dir1, dir2, dir3, dir4, dir5, dir6, dir7, dir8, dir9

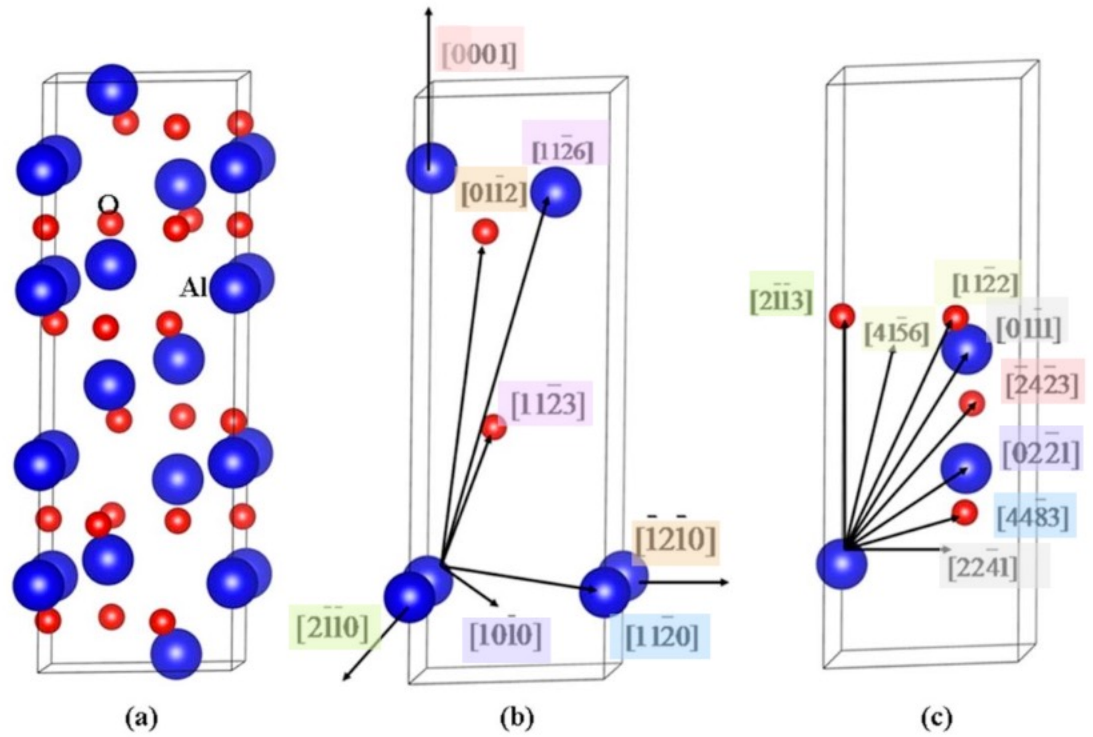
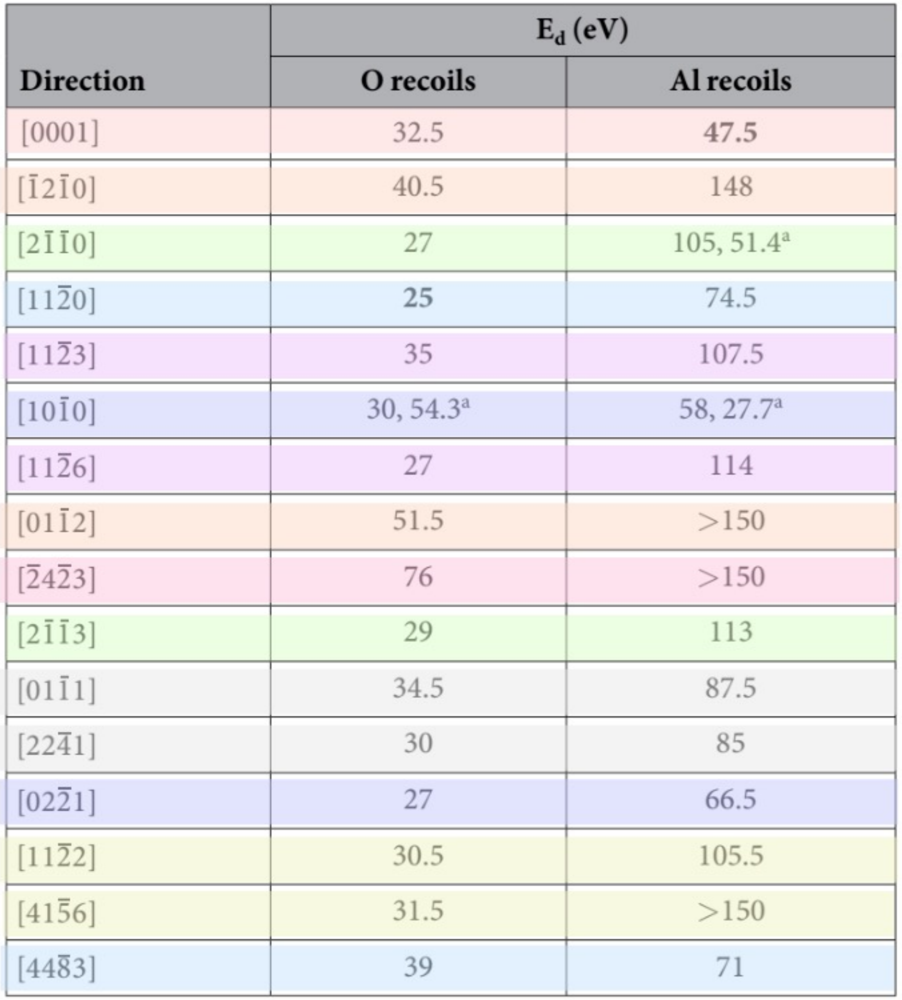


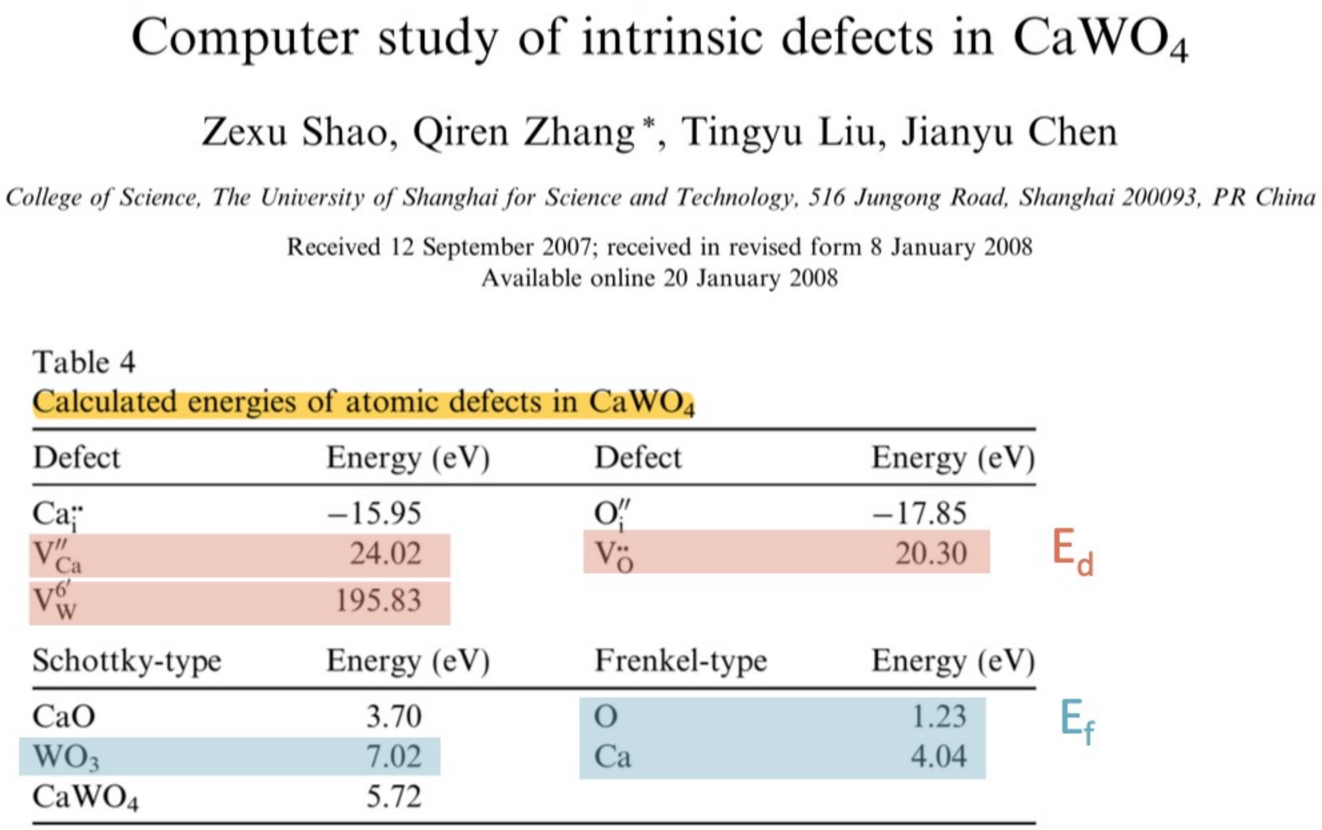
Figure 8. Illustration of schematic view of (**a**) α-Al_2O_3 structure; (**b**,**c**) incident directions in α-Al_2O_3.



	E_d (eV)	
Direction	**O recoils**	**Al recoils**
[0001]	32.5	**47.5**
[$\bar{1}$2$\bar{1}$0]	40.5	148
[2$\bar{1}\bar{1}$0]	27	105, 51.4[a]
[11$\bar{2}$0]	**25**	74.5
[11$\bar{2}$3]	35	107.5
[10$\bar{1}$0]	30, 54.3[a]	58, 27.7[a]
[11$\bar{2}$6]	27	114
[01$\bar{1}$2]	51.5	>150
[$\bar{2}$4$\bar{2}$3]	76	>150
[2$\bar{1}\bar{1}$3]	29	113
[01$\bar{1}$1]	34.5	87.5
[22$\bar{4}$1]	30	85
[02$\bar{2}$1]	27	66.5
[11$\bar{2}$2]	30.5	105.5
[41$\bar{5}$6]	31.5	>150
[44$\bar{8}$3]	39	71

VIII. Appendices

B. Simulated Targets – $CaWO_4$



Computer study of intrinsic defects in $CaWO_4$

Zexu Shao, Qiren Zhang *, Tingyu Liu, Jianyu Chen

College of Science, The University of Shanghai for Science and Technology, 516 Jungong Road, Shanghai 200093, PR China

Received 12 September 2007; received in revised form 8 January 2008
Available online 20 January 2008

Table 4
Calculated energies of atomic defects in $CaWO_4$

Defect	Energy (eV)	Defect	Energy (eV)
$Ca_i^{\cdot\cdot}$	−15.95	O_i''	−17.85
V_{Ca}''	24.02	$V_O^{\cdot\cdot}$	20.30
$V_W^{6'}$	195.83		
Schottky-type	**Energy (eV)**	**Frenkel-type**	**Energy (eV)**
CaO	3.70	O	1.23
WO_3	7.02	Ca	4.04
$CaWO_4$	5.72		

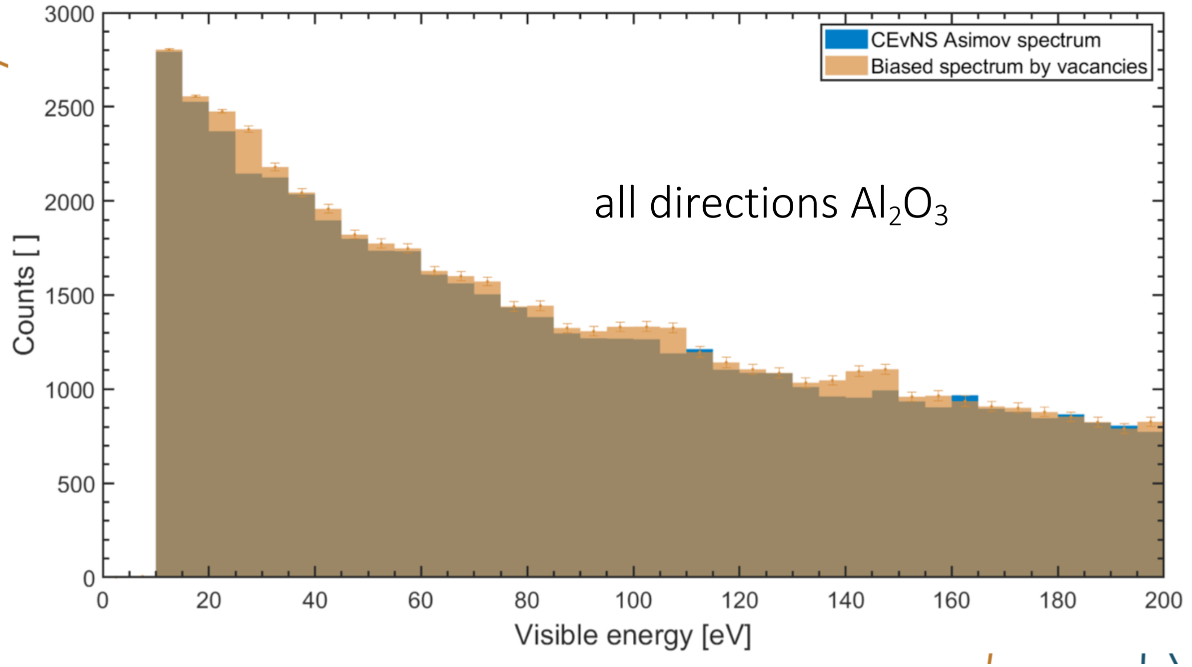
E_d

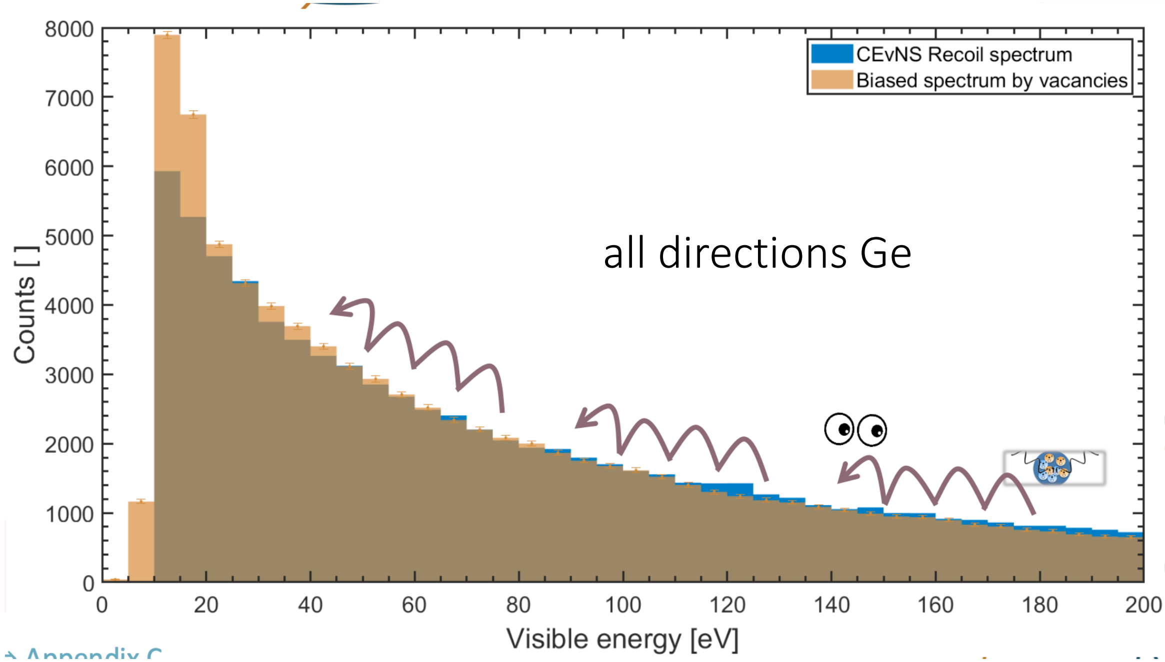
E_f

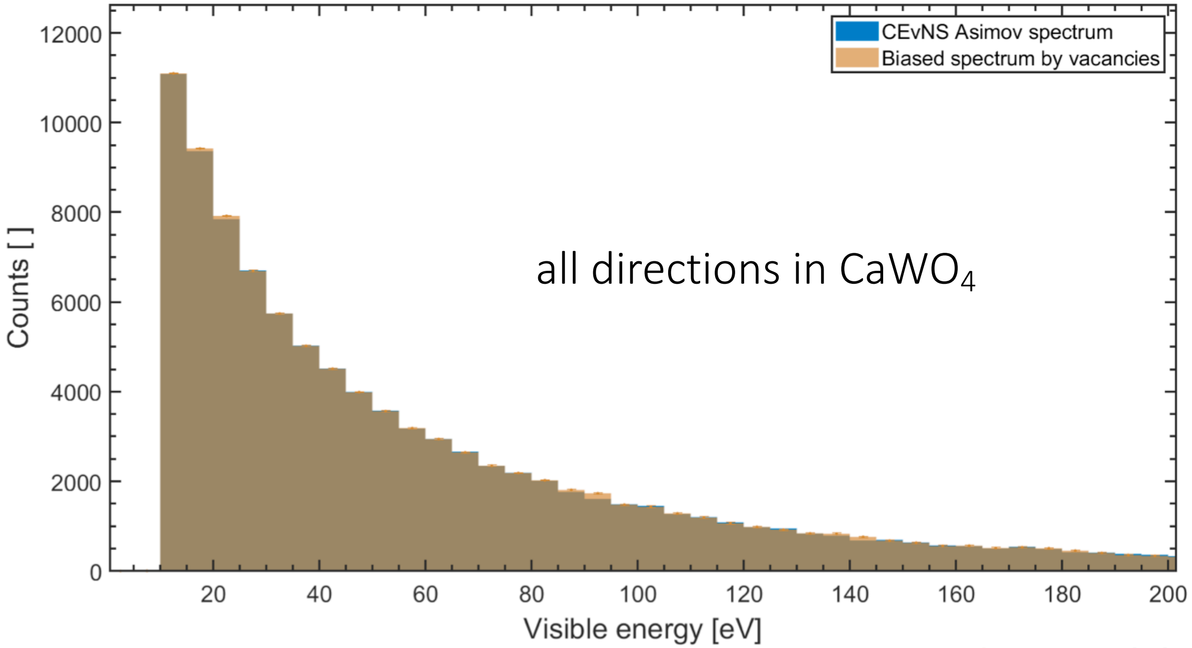
But we want to take into account the variation of the value with the direction in the crystal

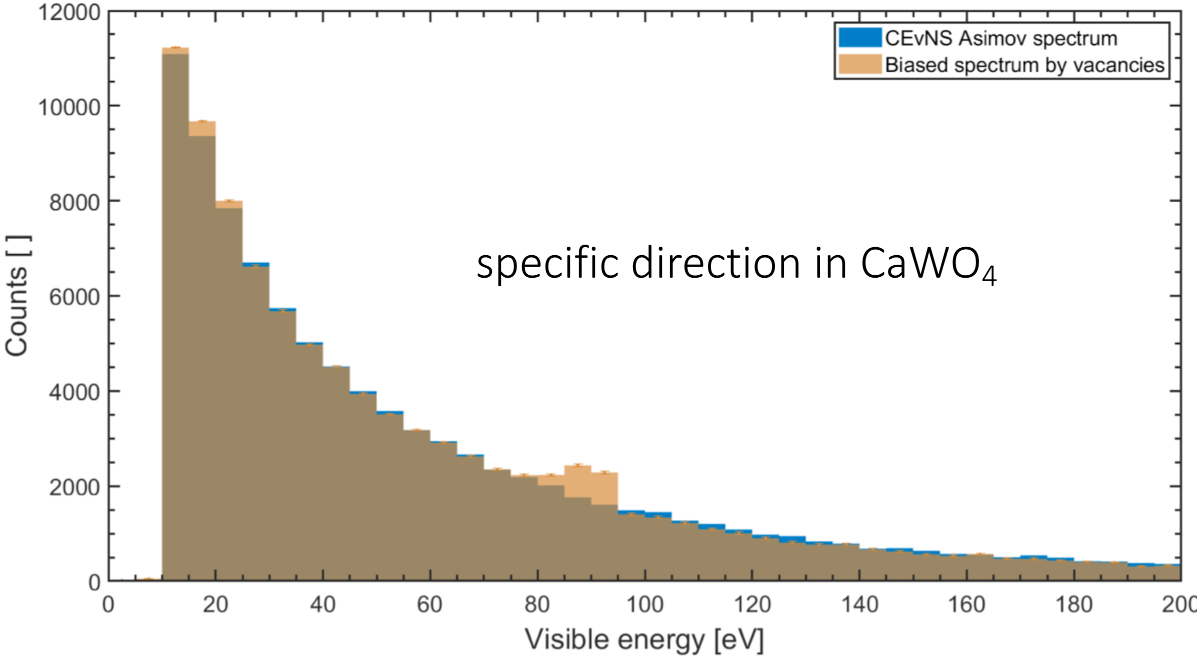
E_d +- 50%
⇒ Applied on Tungsten Only

Atom	E_d (eV)	E_f (eV)
Ca	24.02	4.04
O	20.30	1.23
W	95	7.02
W	145	
W	185	
W	225	
W	265	
W	305	


all directions Al_2O_3


all directions Ge


all directions in $CaWO_4$


specific direction in $CaWO_4$