A Systematic Investigation of Defects in Quantum Materials

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August 14th, 2025

Johns Hopkins University

Outline

1. Quantum Materials
2. Quantum Computing
3. Quantum Computing ↔ Quantum Materials
   $Yb_{2-x}Lu_{2x}Ti_2O_7$
4. Quantum Materials ⟶ Quantum Computing
   $Sr_2/Ba_2CaWO_{6-\delta}$

Quantum materials

Quantum materials

Quantum materials have properties which rely on quantum mechanical phenomena such as:

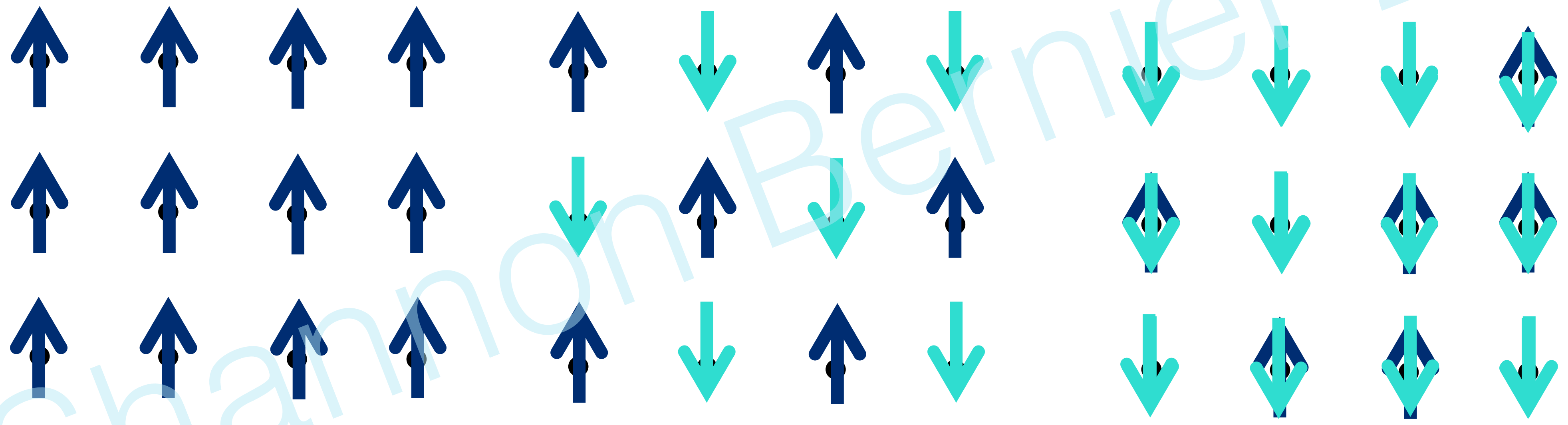
- Quantization
- Entanglement
- Superposition & collapse

The lifetime of a quantum mechanical state is measured by its *coherence time*.

Conventional magnetic order

Spin Alignment Axis

Ferromagnetism | **Antiferromagnetism** | **Paramagnetism**



Strong magnetic field

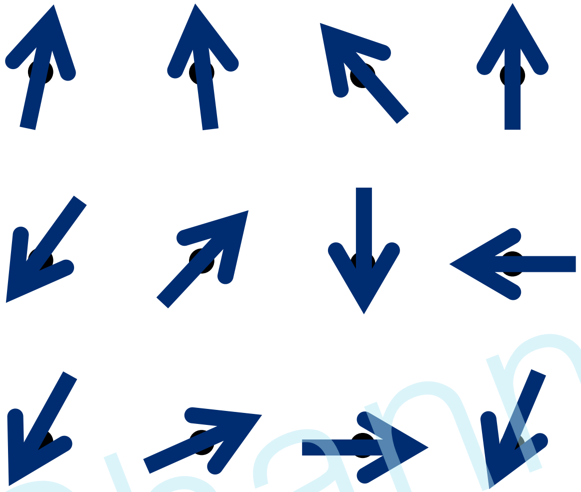
Big magnetic moment
ΣS >> 0

Zero magnetic moment
ΣS = 0

Small magnetic moment, if any
***can order with a magnetic field*

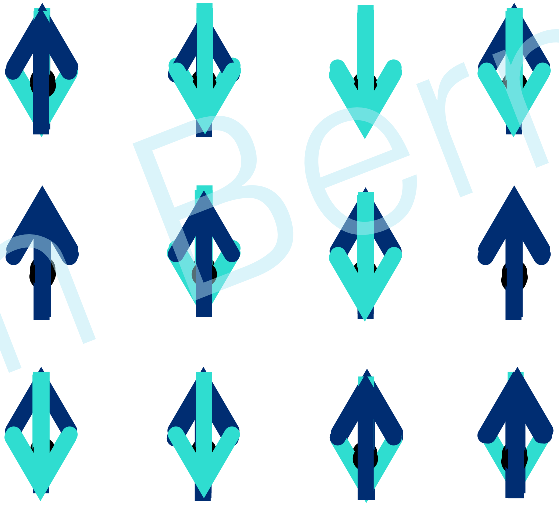
Unconventional dis-order

Spin glass

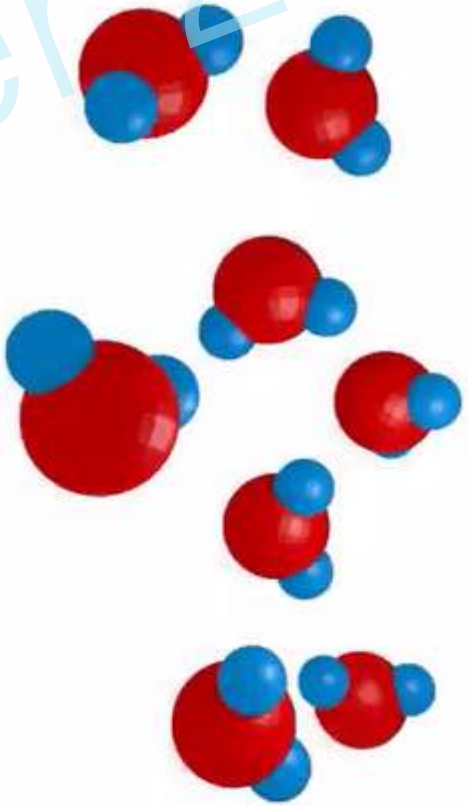


Random spin orientations from frustrated interactions

Spin liquid



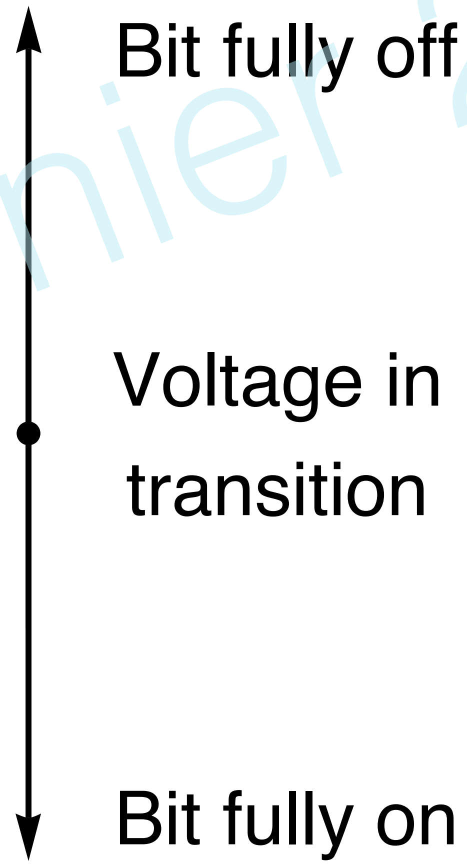
Like a paramagnet, but not "frozen"



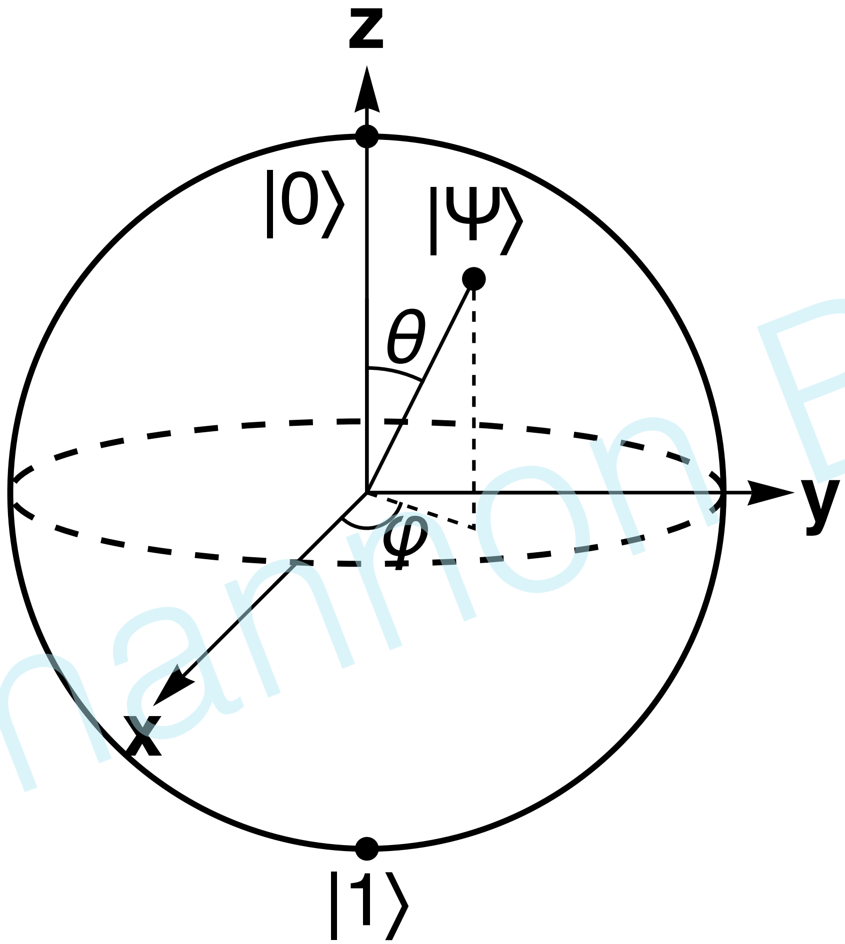
Quantum computing

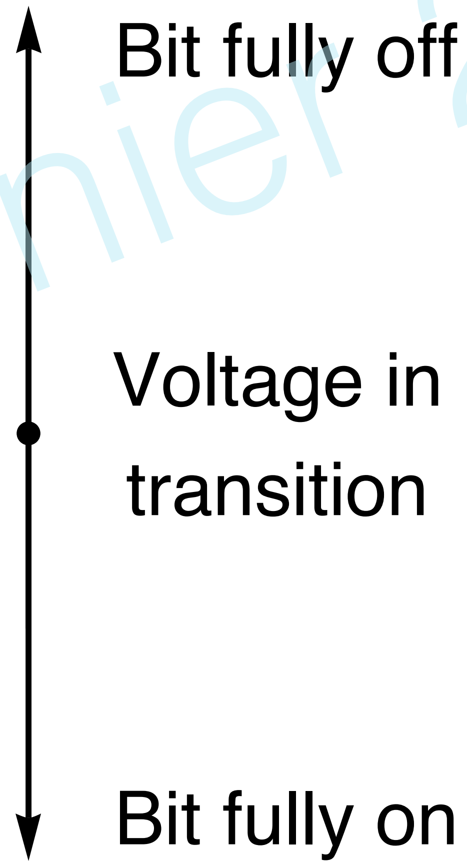
[Quantum] Computers

- Computers perform series of logical or arithmetic operations to solve problems.
- Classical computers do so with logical bits – binary digits – with 2 total states.



[Quantum] Computers

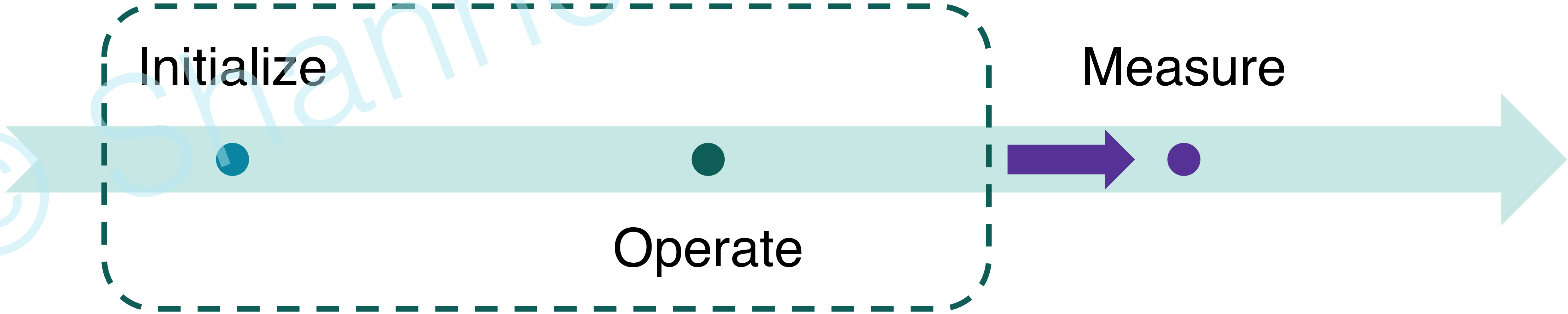




Quantum computers (cont.)

Quantum computers consist of:

- Qubits
- Couplers
- Operators



Other benefits

- May be faster than conventional computation for certain types of problems.
- Uses less memory than classical computers when describing entangled systems.
  - Can offer similar improvement for other quantum mechanical behaviors, such as tunneling.

Modern computer modalities

Digital (aka "Universal")

- A combination of gates (operators) is used to evolve the system.
- Any algorithm input with gates will work.
- The system studied is represented as variables.

Smartphones, Laptops

Analog

- Uses changes in continuous variables to compute.
- The computer can only model systems which obey the same physics as itself.

Slide rules, wristwatches

Quantum Computing ↔ Quantum Materials

D-Wave simulation of $Yb_{2-x}Lu_{2x}Ti_2O_7$

Project design

Goal: Use near-term quantum computing hardware to explore a quantum materials problem.

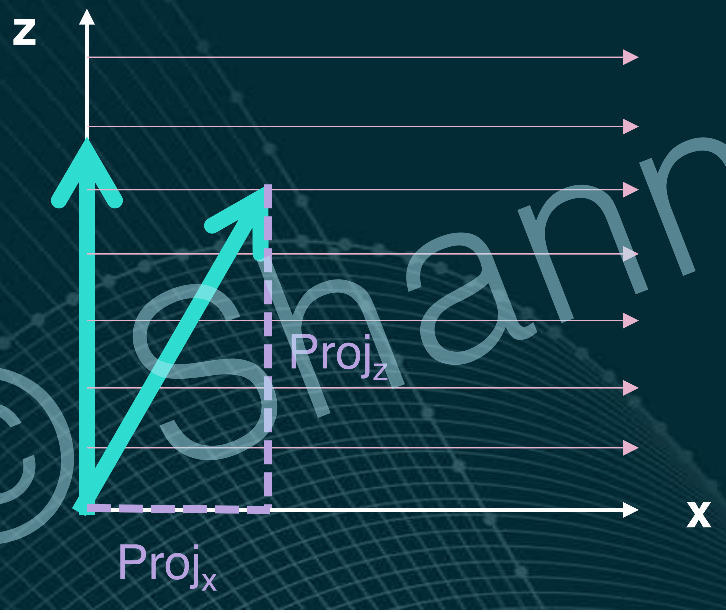
1. Simulate an interesting lattice on a quantum computer.
2. Make a real material with the same lattice [and physics].
3. Measure the same observable on both.

Feedback for both computers and materials researchers.

D-Wave quantum annealers

$$H_{IM} = -\sum_i \sum_j J_{ij}\sigma_i{}^z \sigma_j{}^z - \sum_i h_i\sigma_i{}^z$$

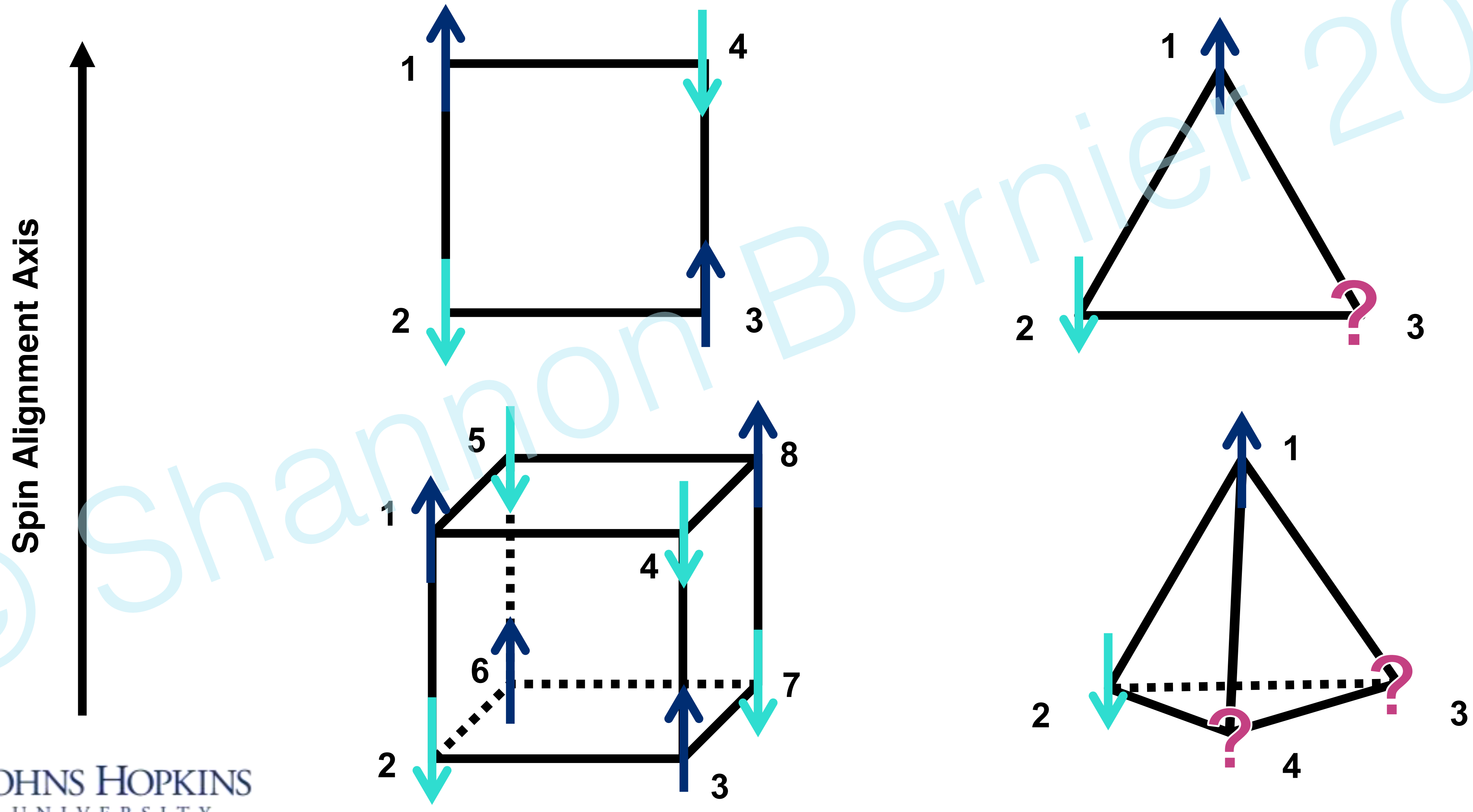
$$H_{TFIM} = -\sum_i \sum_j J_{ij}\sigma_i{}^z \sigma_j{}^z - \sum_i h_i\sigma_i{}^z - \sum_i a_i\sigma_i{}^x$$



Annealer physics

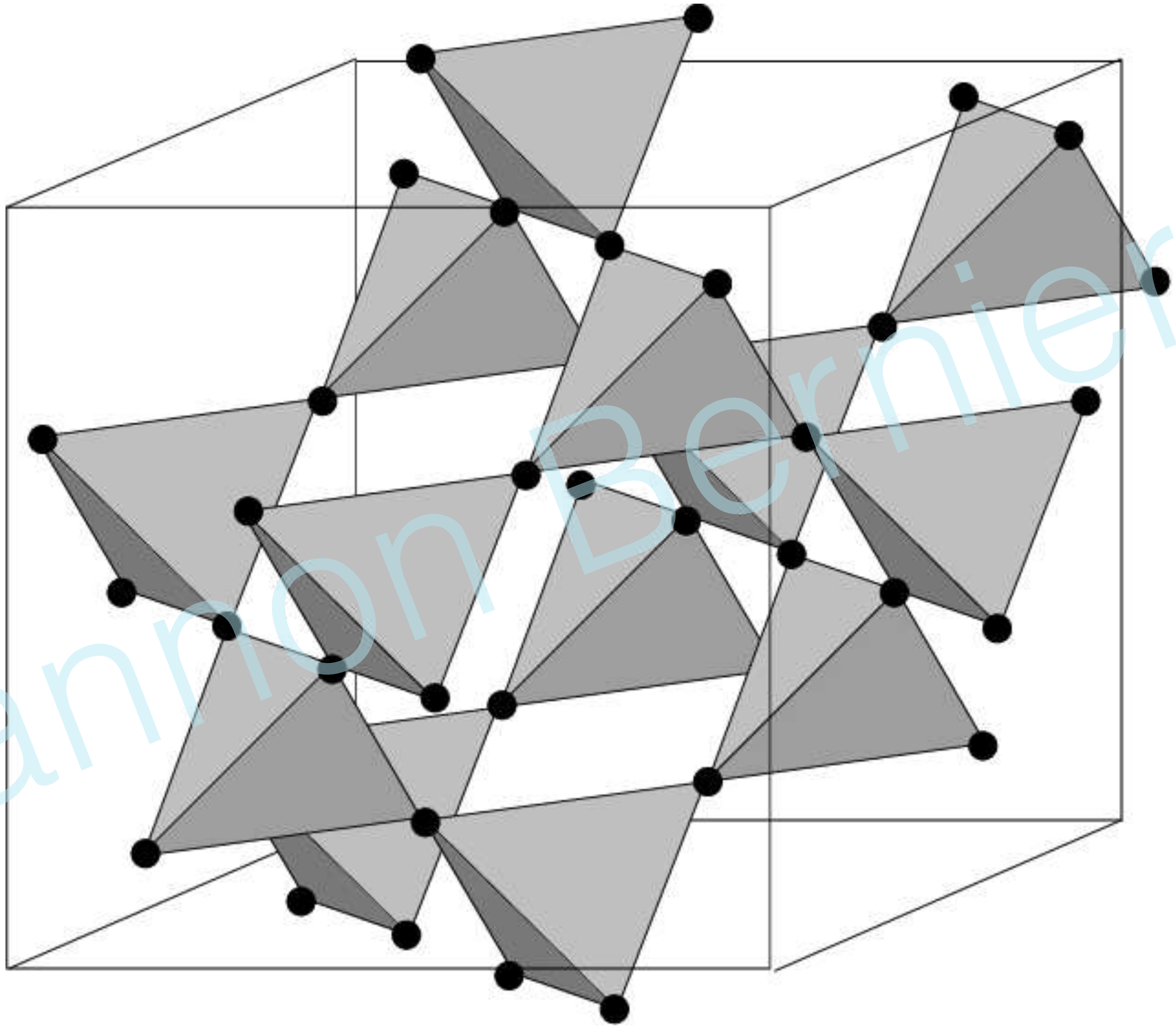
- Ising model only: one spin alignment axis and only one unpaired spin per site
- Every spin has the same susceptibility to the field
  - "Interesting" here thus means geometrical frustration, rather than competing interactions

Aside: Geometric frustration



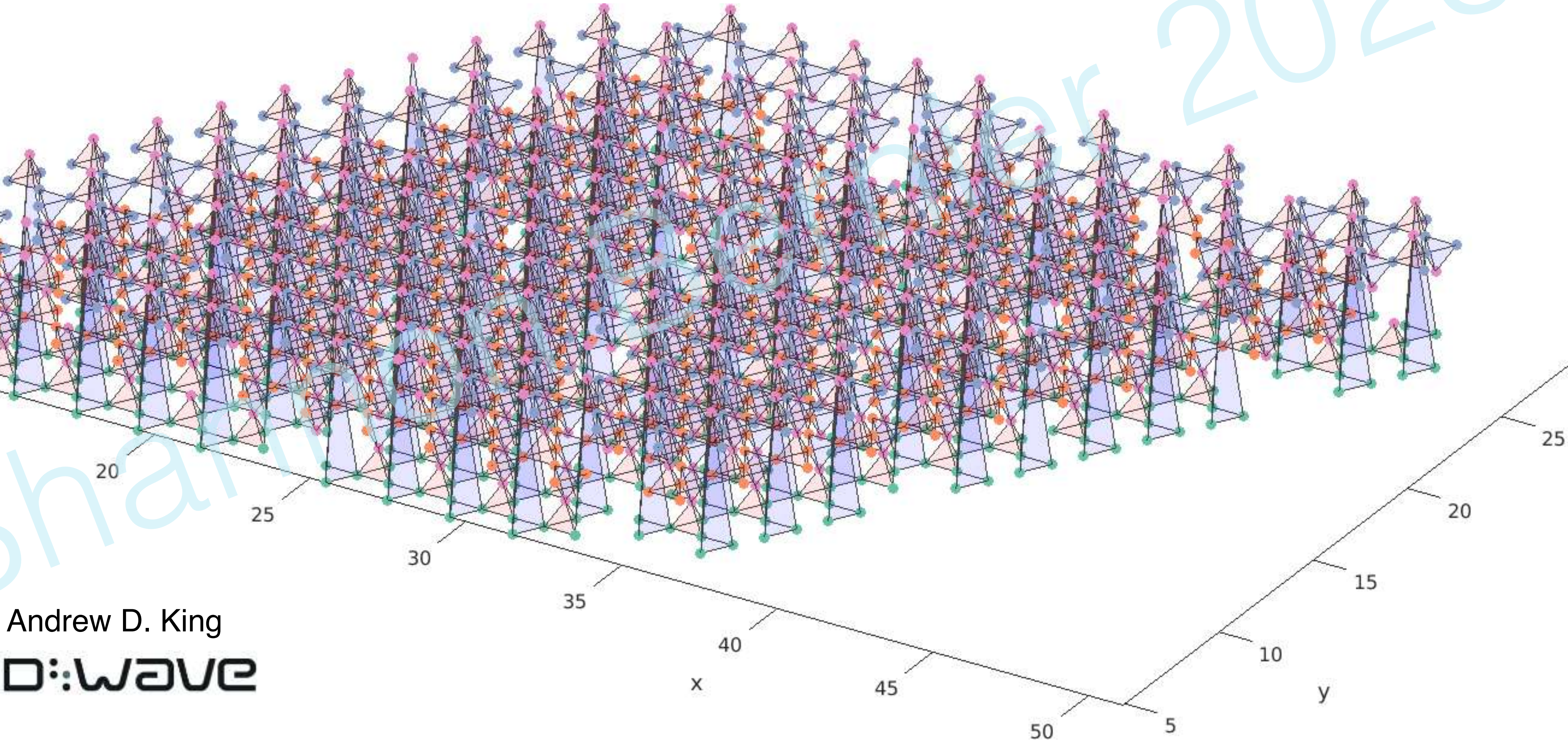
Annealer physics (cont.)

- Ising model <u>only</u>: one spin alignment axis and only one unpaired spin per site
- Every spin has the same susceptibility to the field
  - "Interesting" here thus means geometrical frustration, rather than competing interactions
- The annealer's temperature must be ~12 mK
- h_i and J_{ij} are unitless
- Broken physical qubits → missing logical qubits → gaps in the magnetic lattice



Elhajal et. al. PRB **71** 094420 (2005)

Pyrochlore embedding



Andrew D. King

D:WAVE

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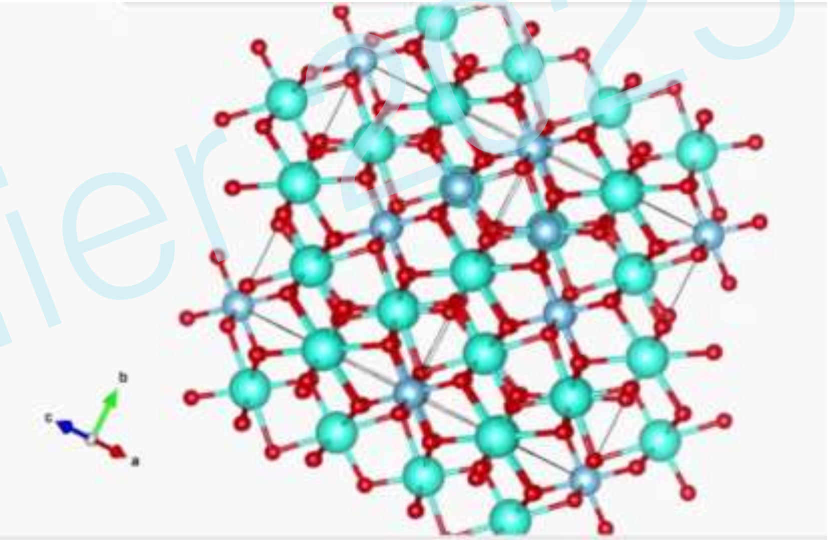
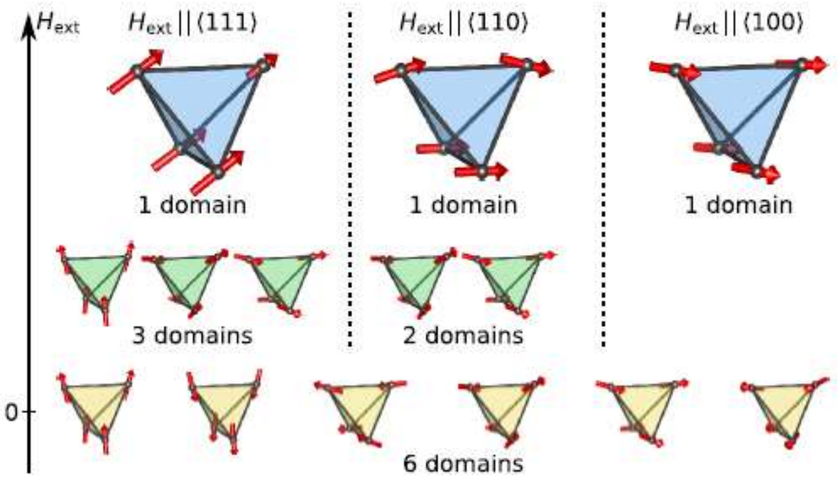
Candidate material: $Yb_{2-x}Lu_{2x}Ti_2O_7$

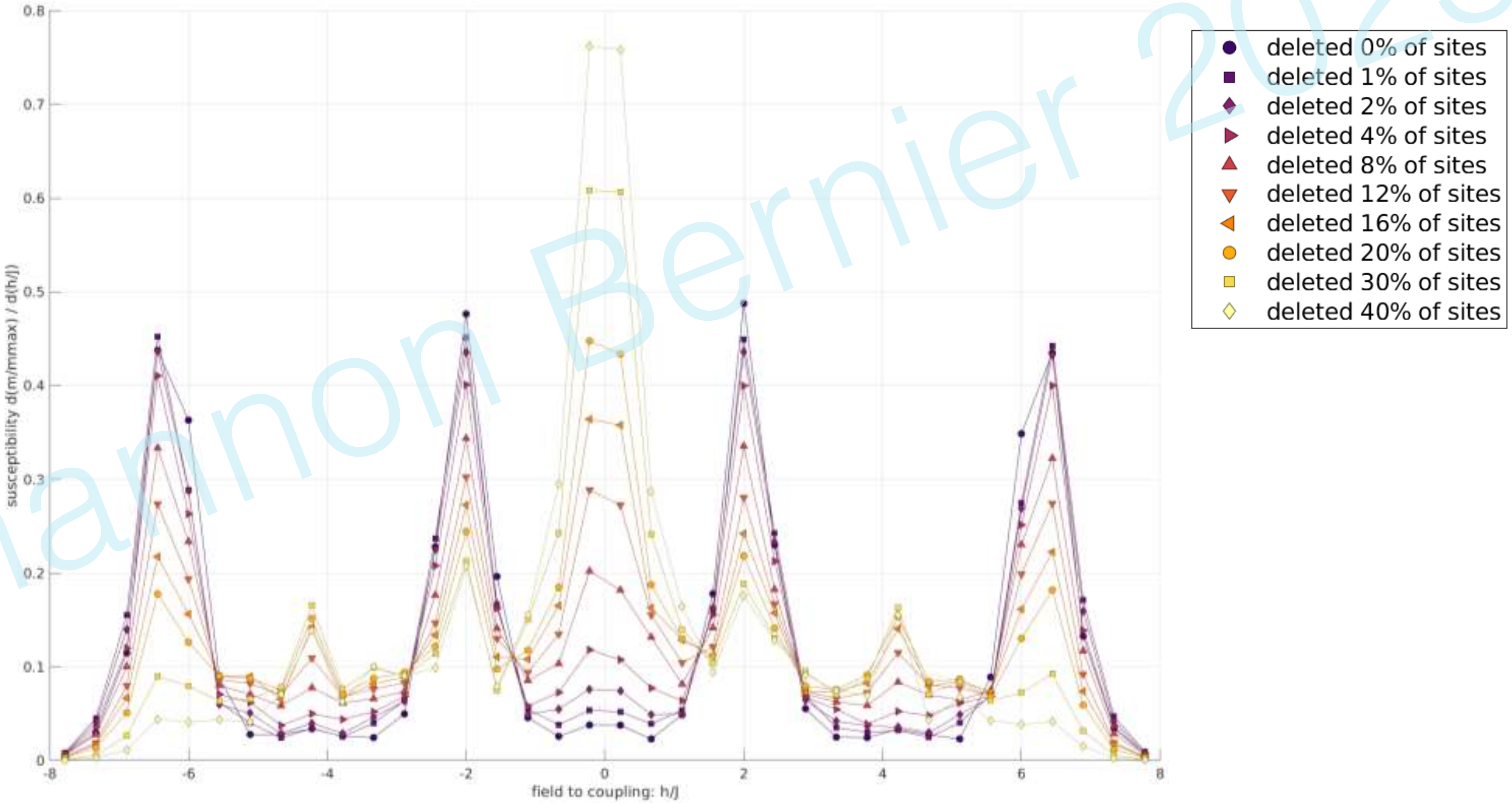
- Yb^{3+} has a single unpaired spin.
  - Lu^{3+} and Ti^{4+} are nonmagnetic.
- Yb^{3+} and Lu^{3+} occupy a geometrically-frustrated pyrochlore lattice.
- The crystal field around the 3+ metal site is the same everywhere.
- Under ⟨100⟩ applied field, there is a single Ising axis.





Säubert et. al. PRB **101** 174434 (2020)

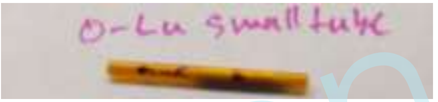
D-Wave results

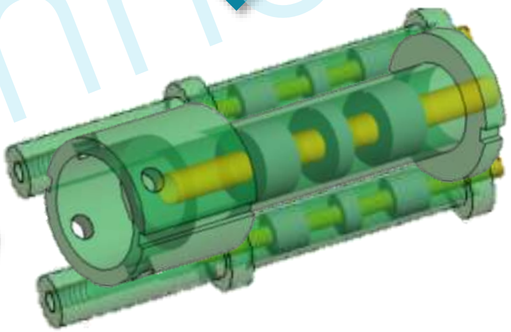


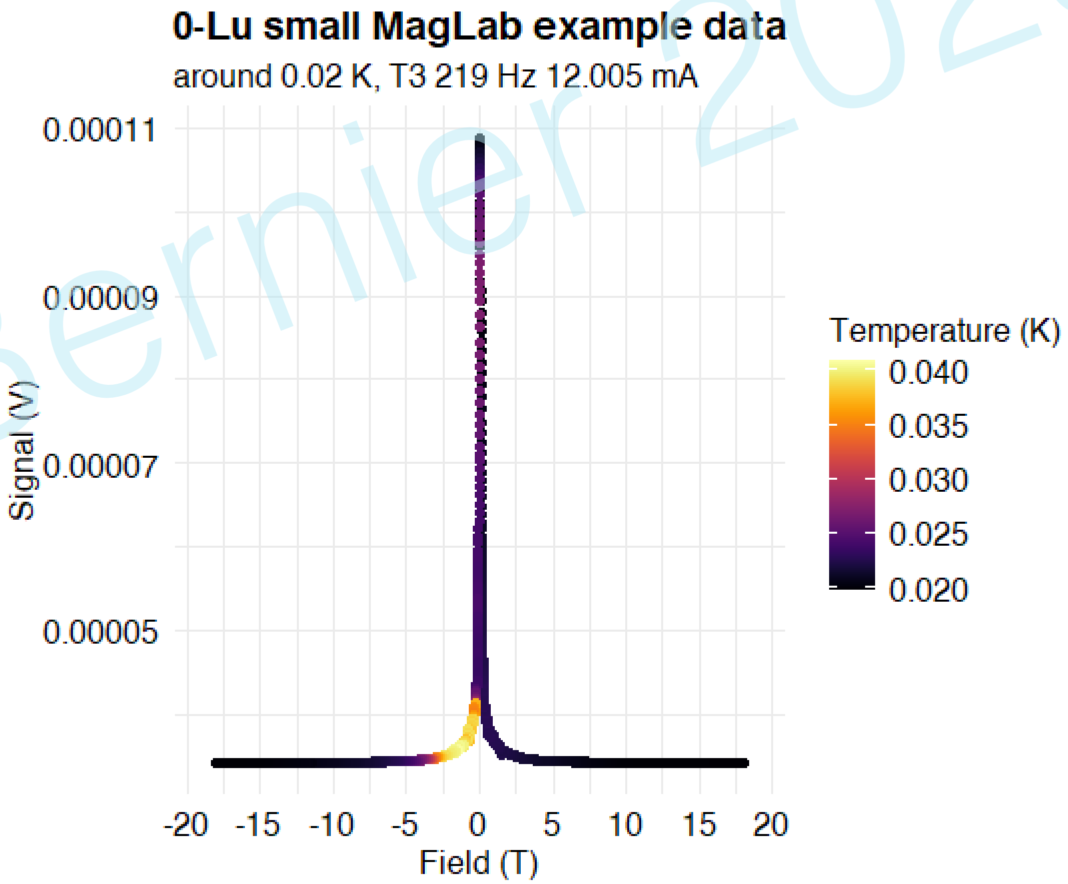
MagLab data

"0-Lu small" sample

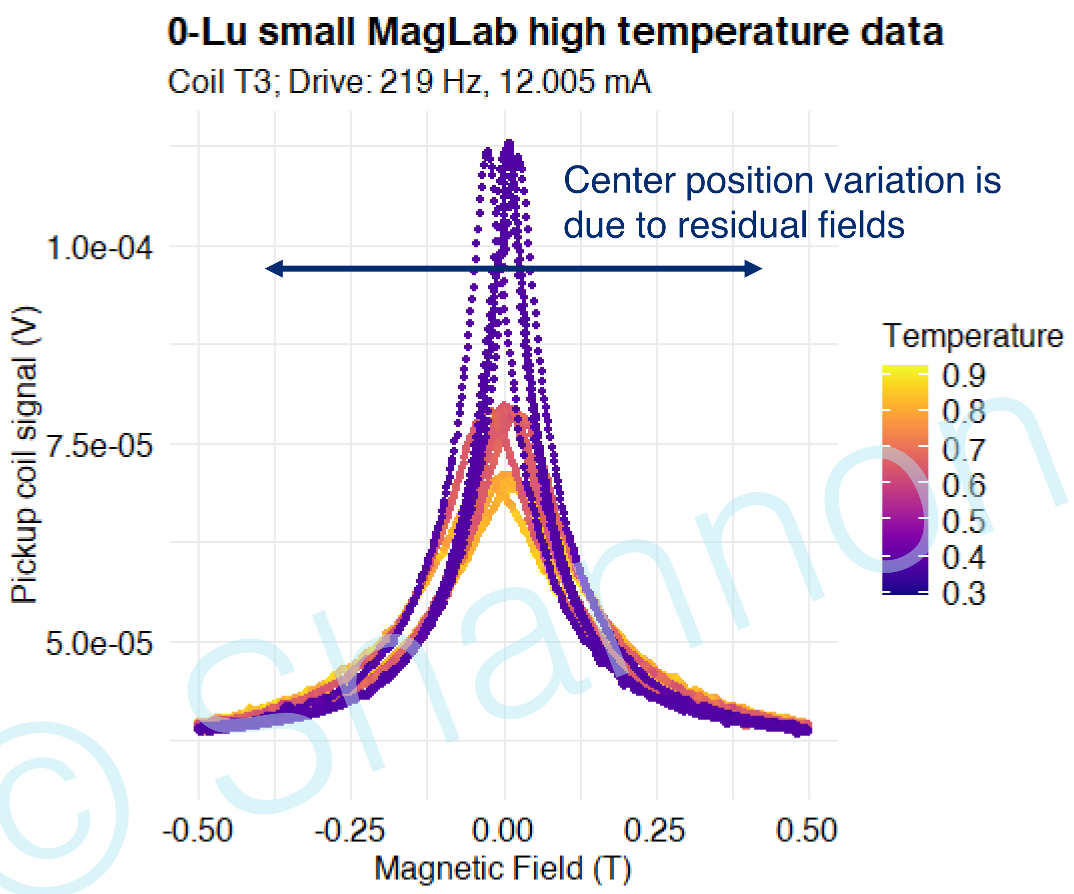
⟨100⟩ direction

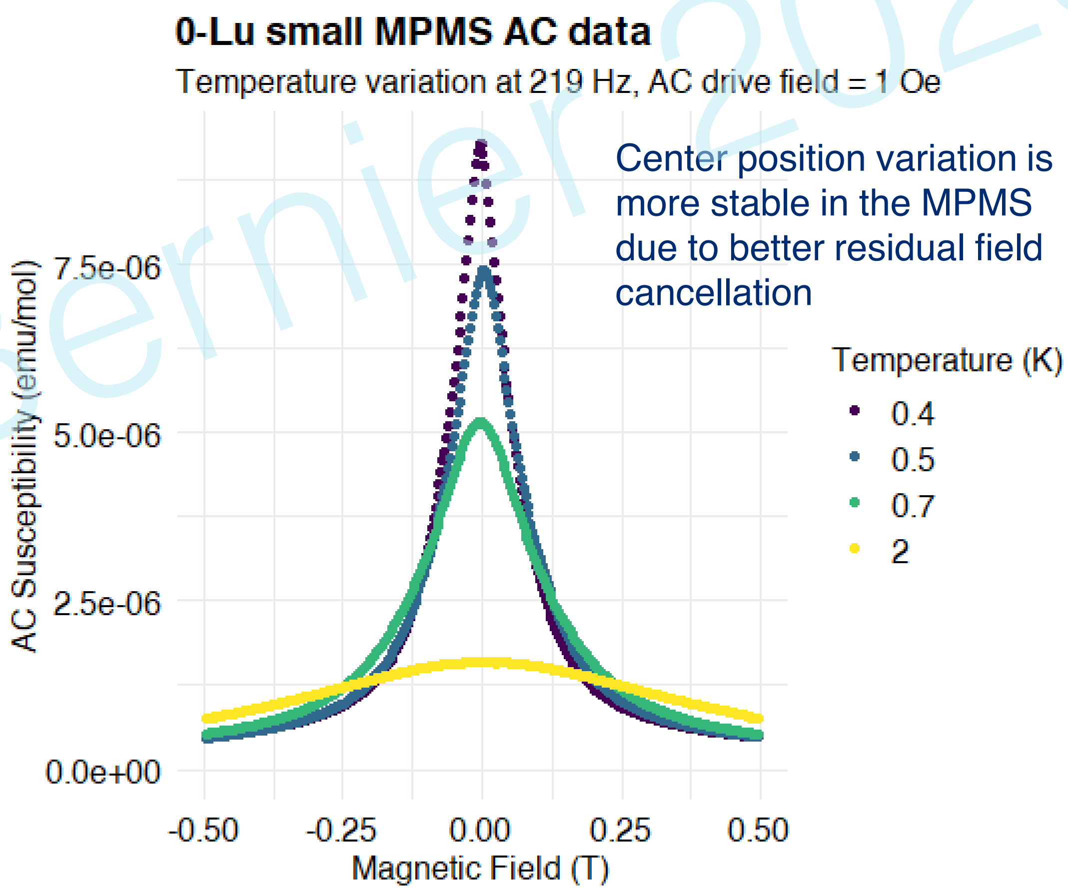






MPMS data as anchor





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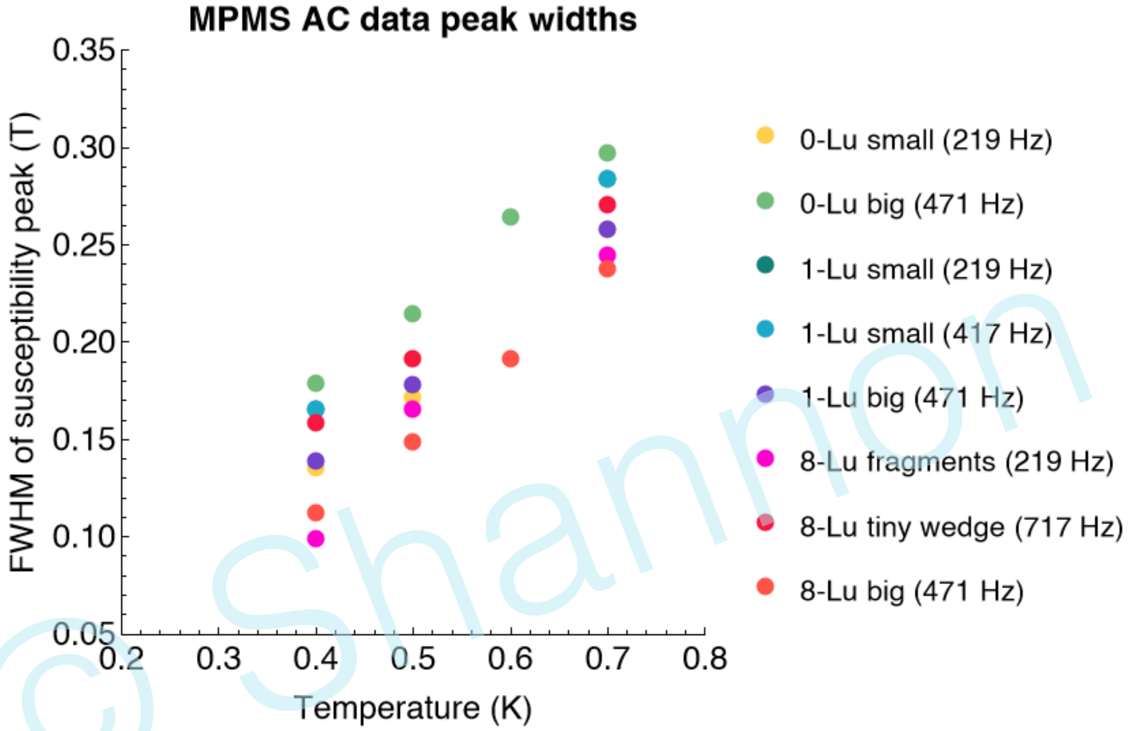
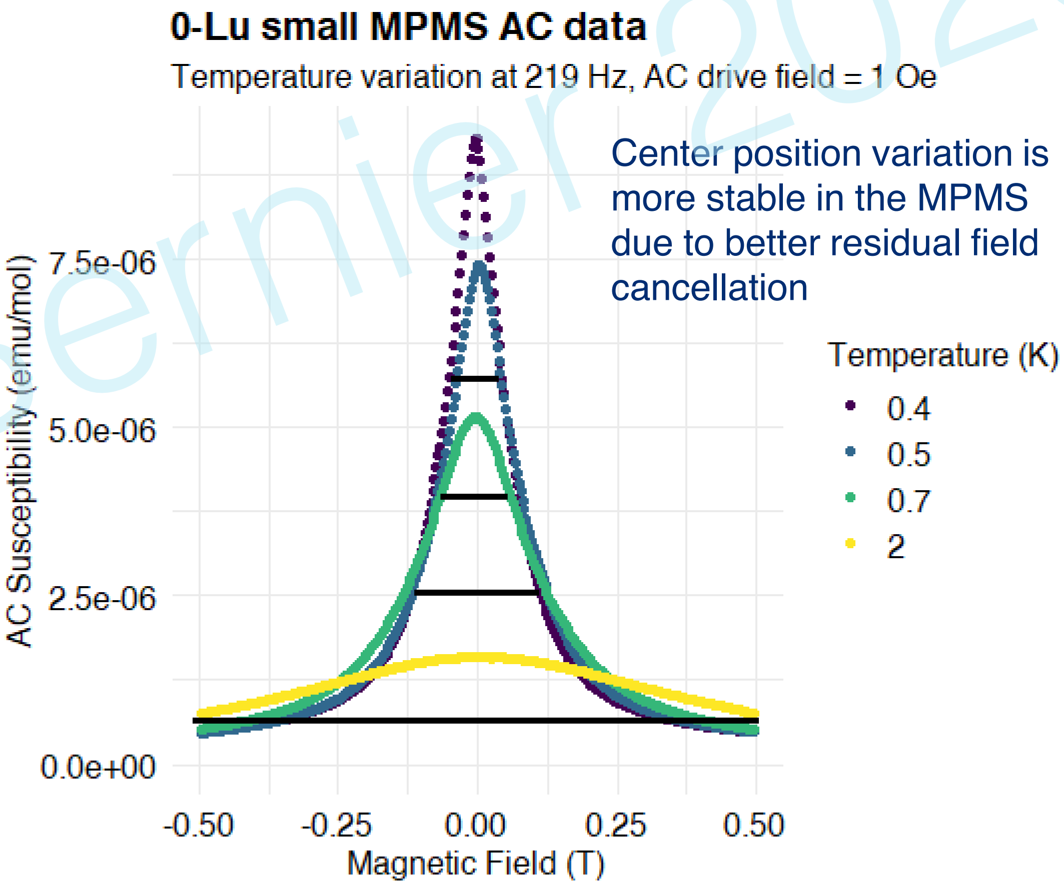
Thermal broadening





Center position variation is more stable in the MPMS due to better residual field cancellation

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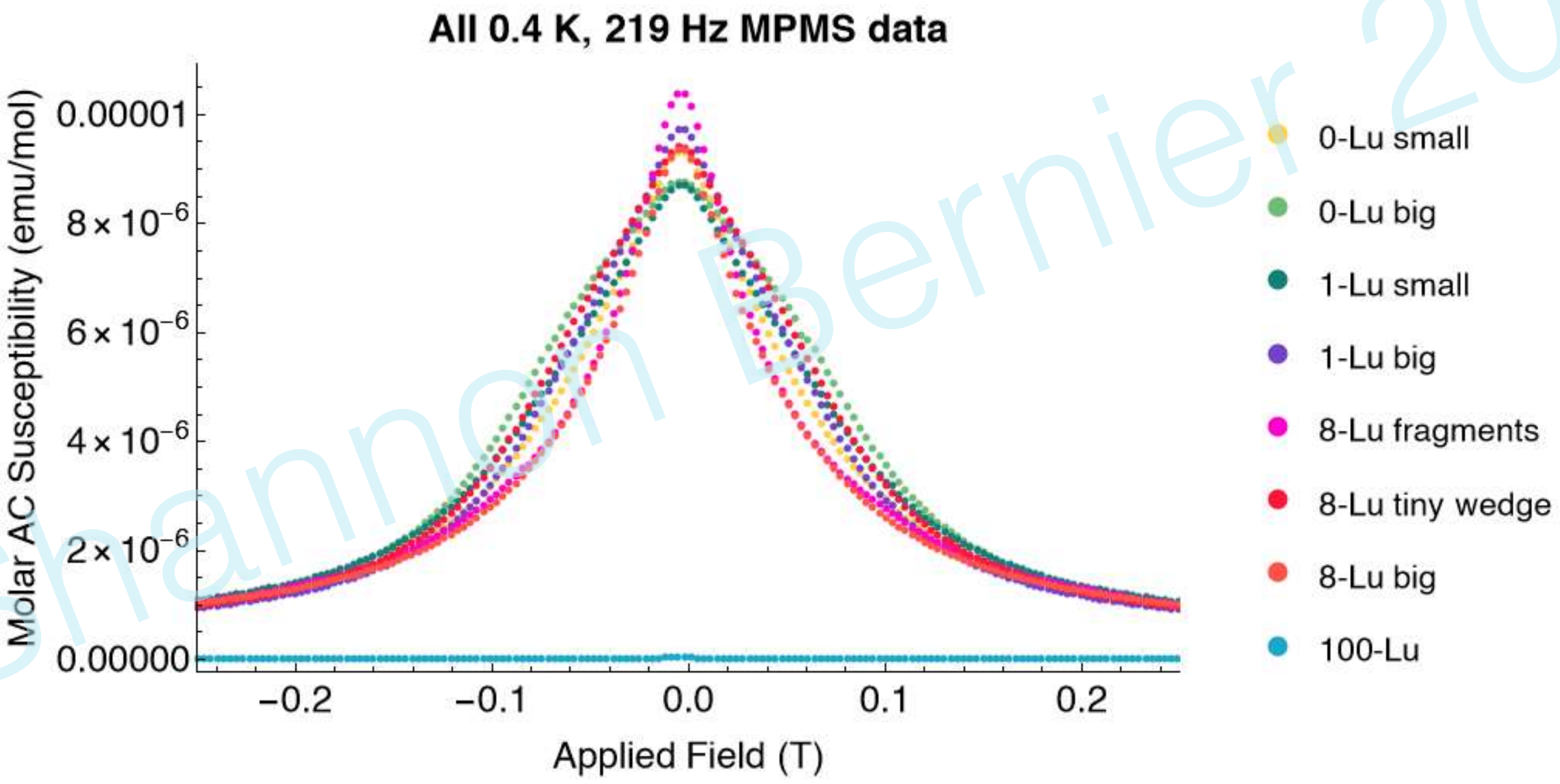
Real material results (cont.)



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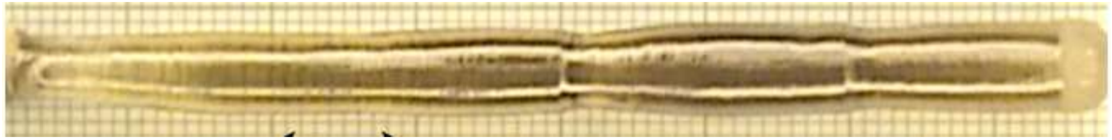
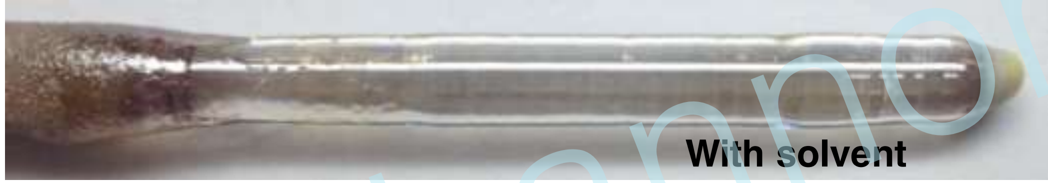
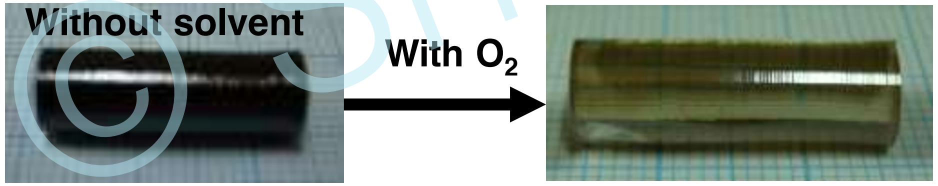
Courtesy of R. D’Ortenzio **(without solvent)**


Courtesy of K. Ross **(without solvent)**



Courtesy of K. Arpino



Courtesy of D. Prabhakaran


Bernier 2023 **(without solvent)**


Bernier 2025 **(with solvent)**


Bernier 2021 **(with solvent)**

Heated in vacuum

With $TeCl_4$

With I_2

$Yb_2Ti_2O_7$

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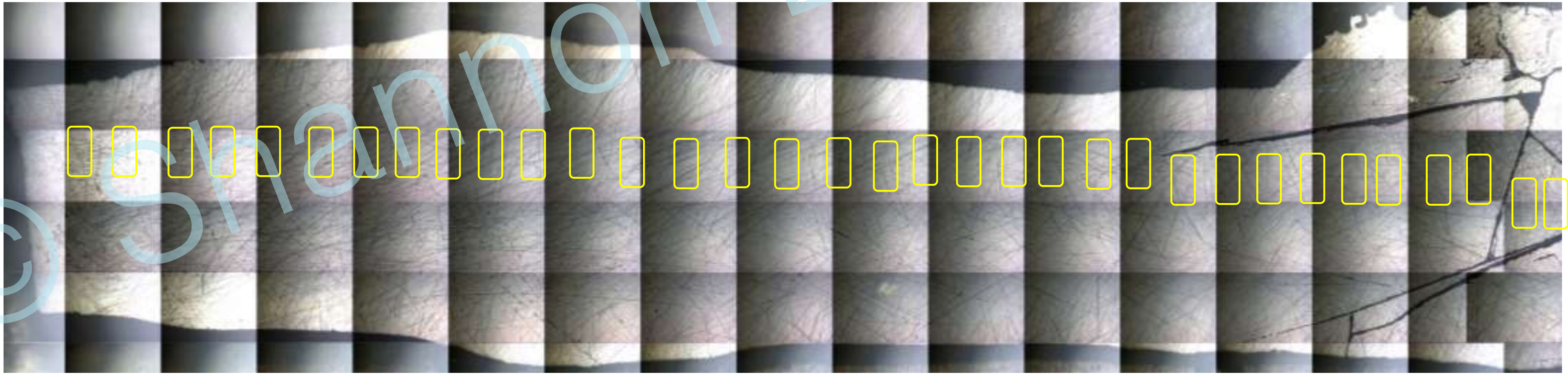
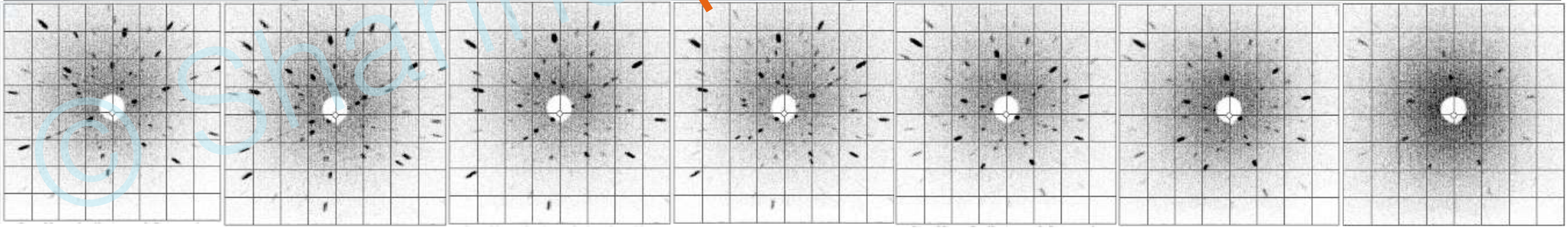
PARADIM
AN NSF MATERIALS INNOVATION PLATFORM

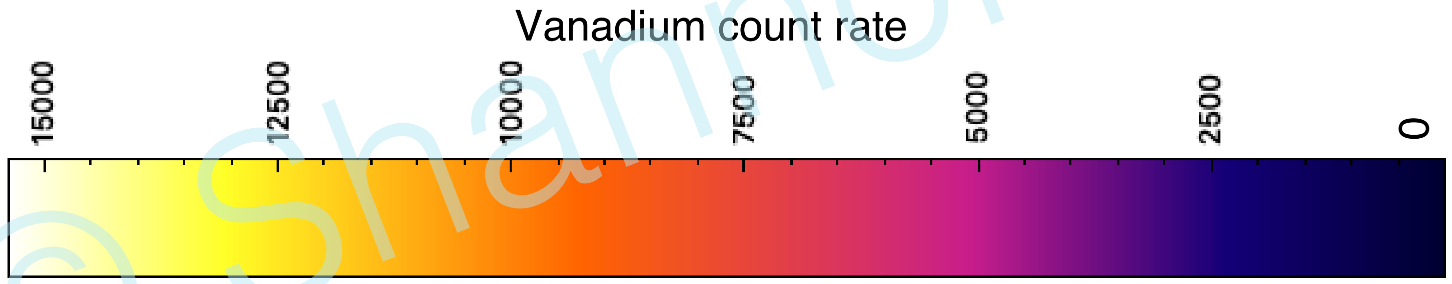
$Lu_2Ti_2O_7$

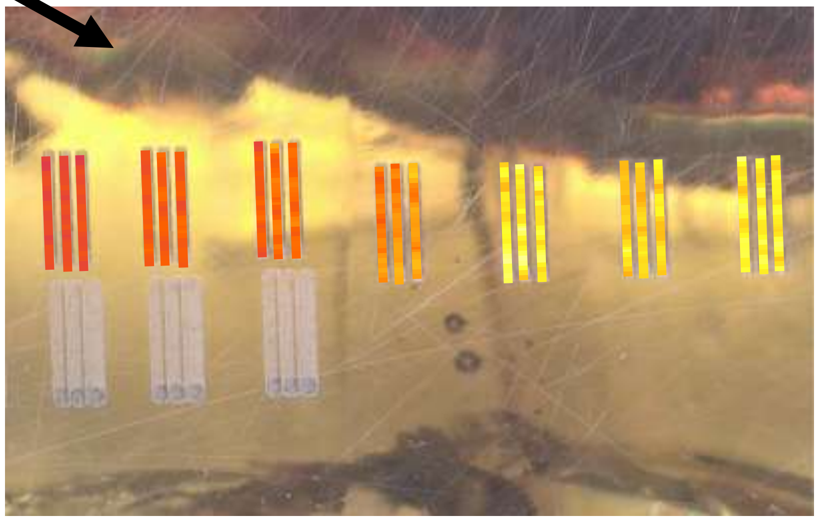


$Lu_2Ti_2O_7$



$Lu_2Ti_2O_7$





$Lu_2Ti_2O_7$

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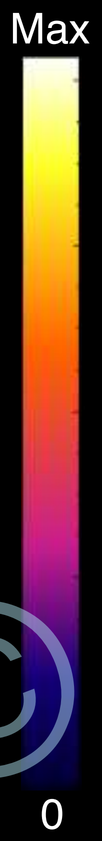
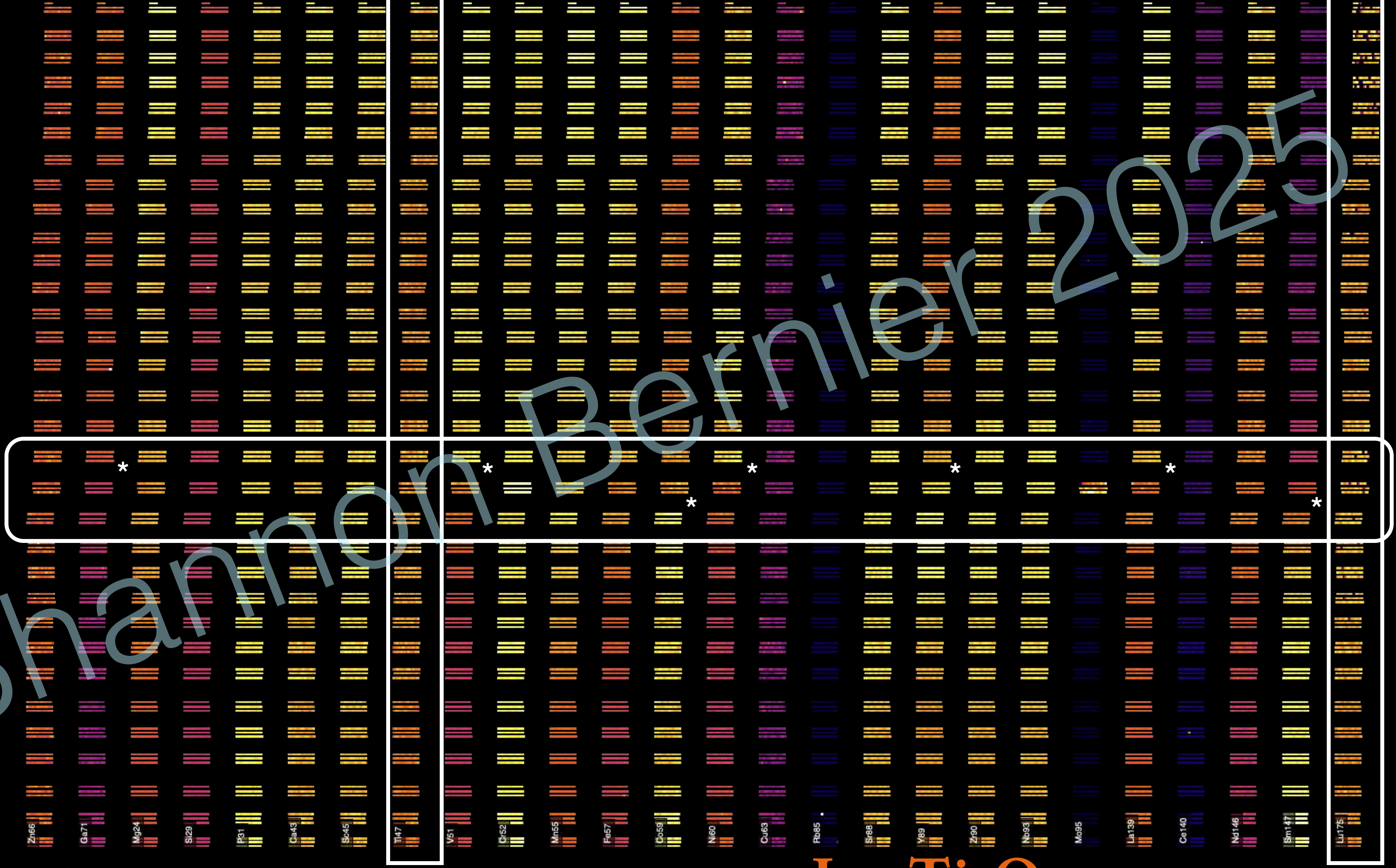
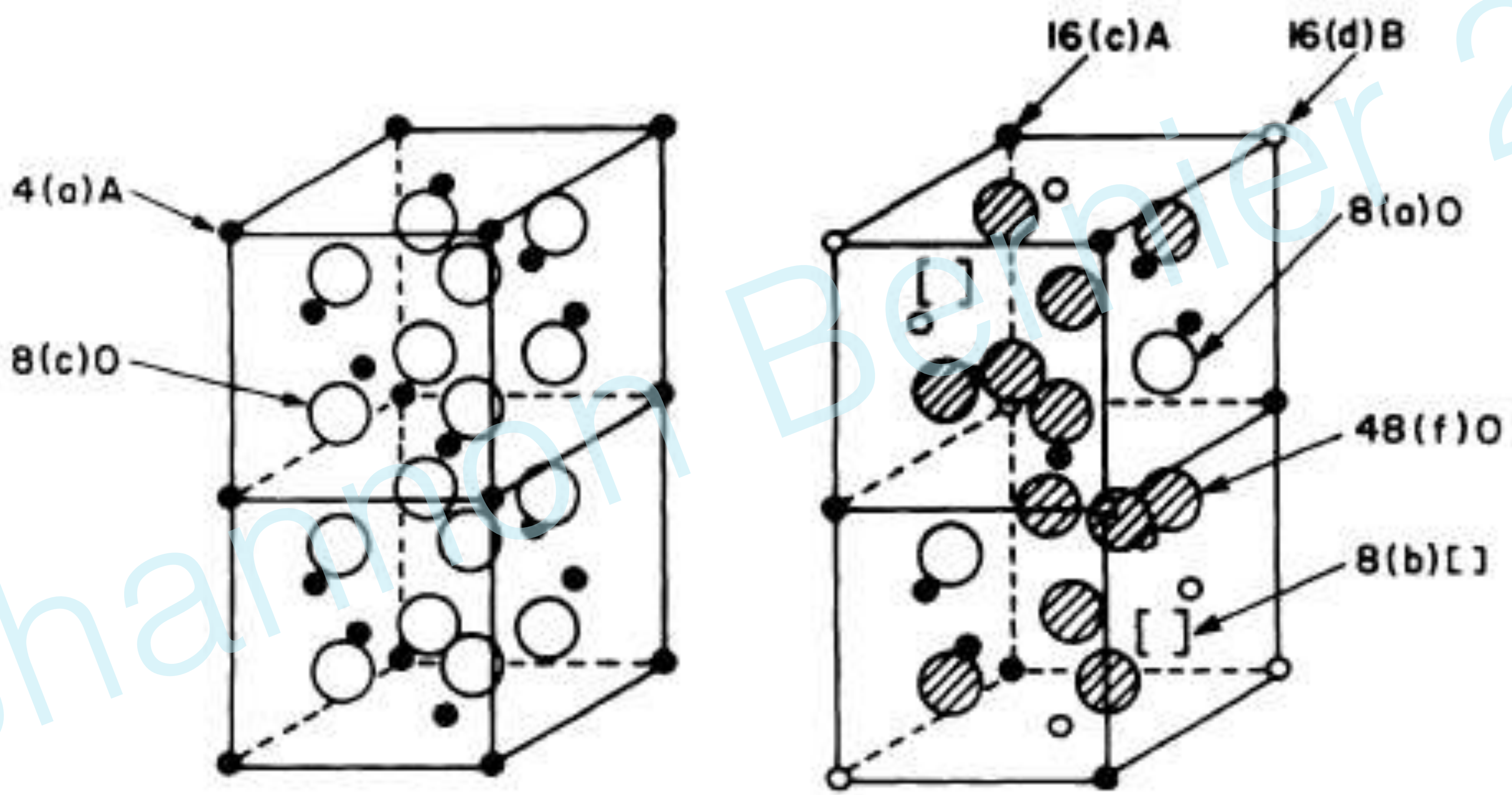
There is a noticeable discontinuity in: Ga, V, Co, Ni, Y, La, and Sm



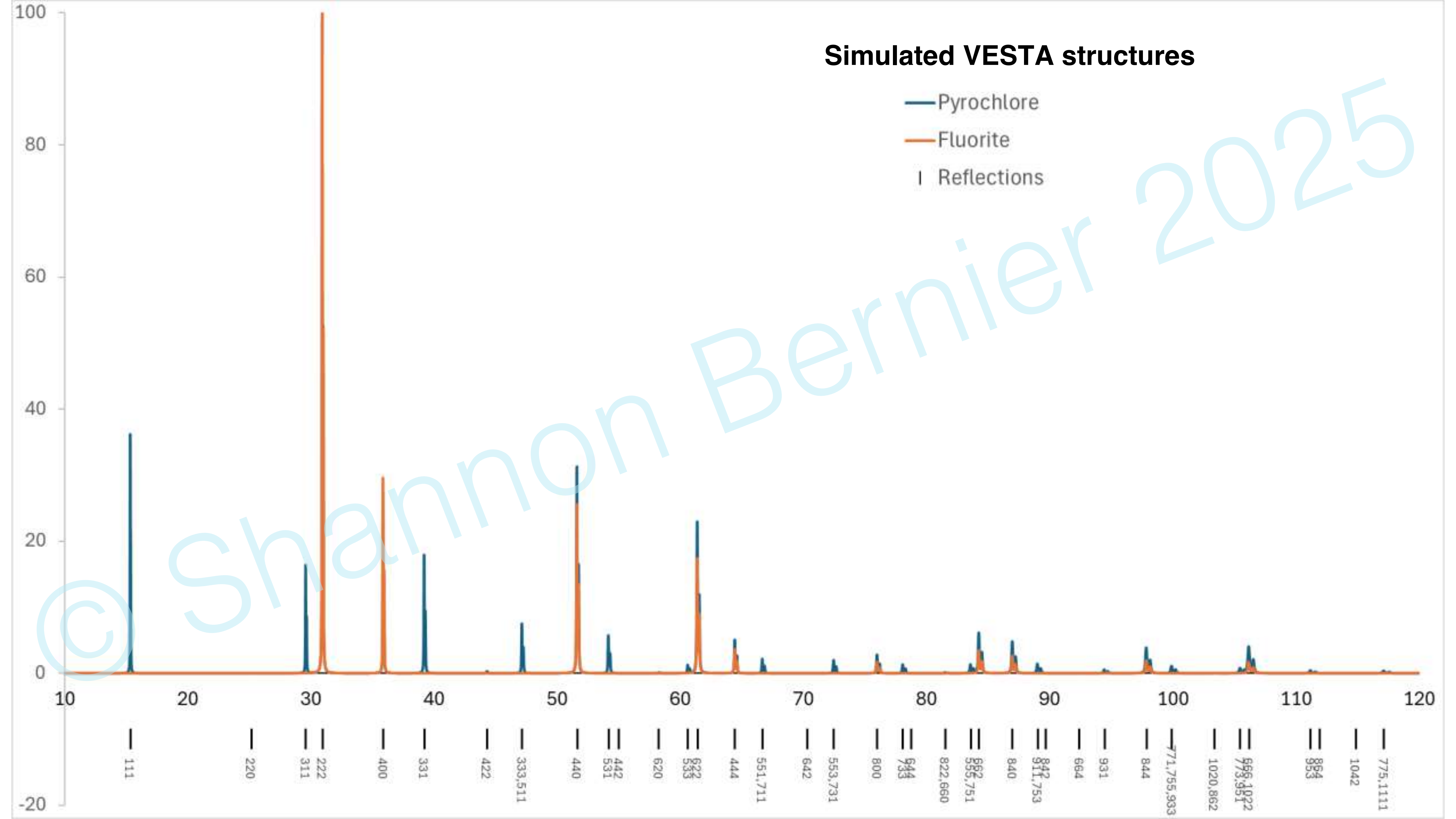


$Lu_2Ti_2O_7$

Disordering transformations?



Scheetz & White Opt. Eng. **22** 302-207 (1983)



Simulated VESTA structures
Pyrochlore
Fluorite
Reflections
100
80
60
40
20
0
-20
10
20
30
40
50
60
70
80
90
100
110
120
111
220
311
222
400
331
422
333,511
440
531
442
620
533
622
444
551,711
642
553,731
800
733
644
822,660
555,751
662
840
911,753
842
664
931
844
771,755,933
1020,862
773,951
666,1022
953
864
1042
775,1111
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Potential for spectroscopy

Pyrochlore $Ln_2Ti_2O_7$

Atom	Wycoff position	Site symmetry
Ln	16 c	D_{3d}
Ti	16 d	D_{3d}
O	8a	T_d
O’	48 f	C_{2v}

Fluorite $Ln_2Ti_2O_7$

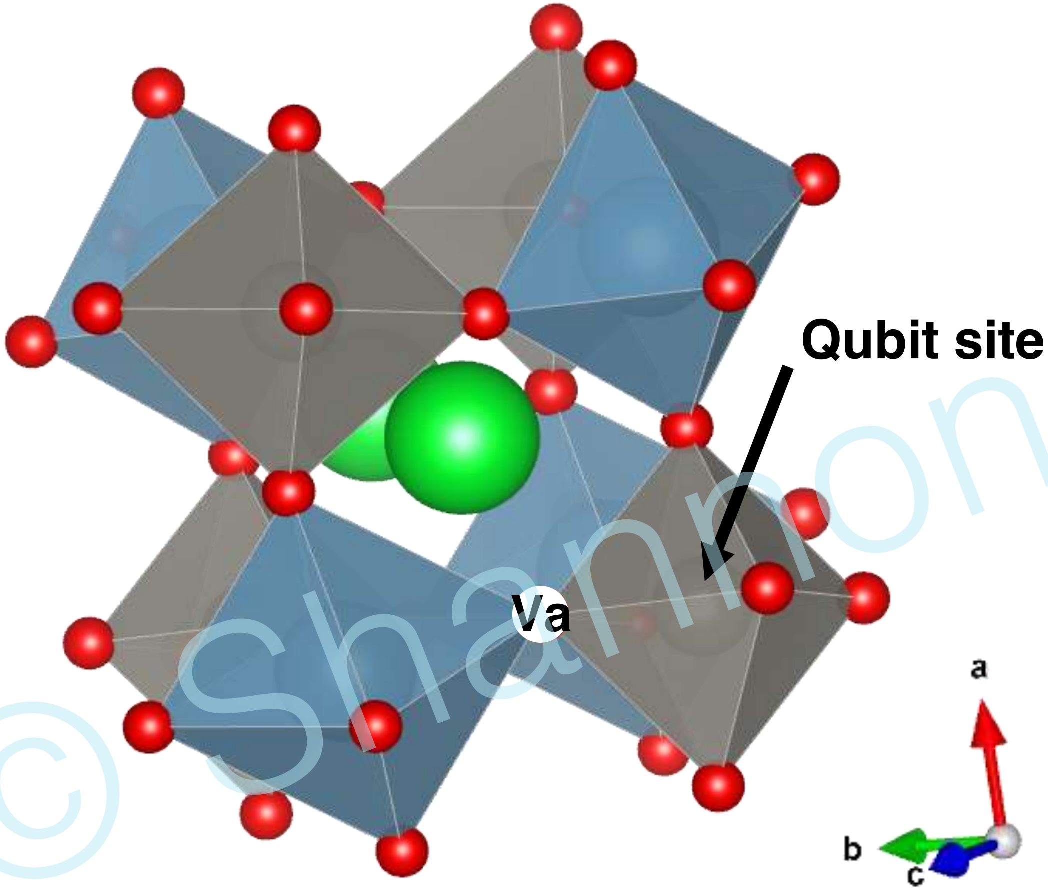
Atom	Wycoff position	Site symmetry
Ln	4 a	O_h
Ti	4 a	O_h
O	8 c	T_d
O’	*N/A*	*N/A*

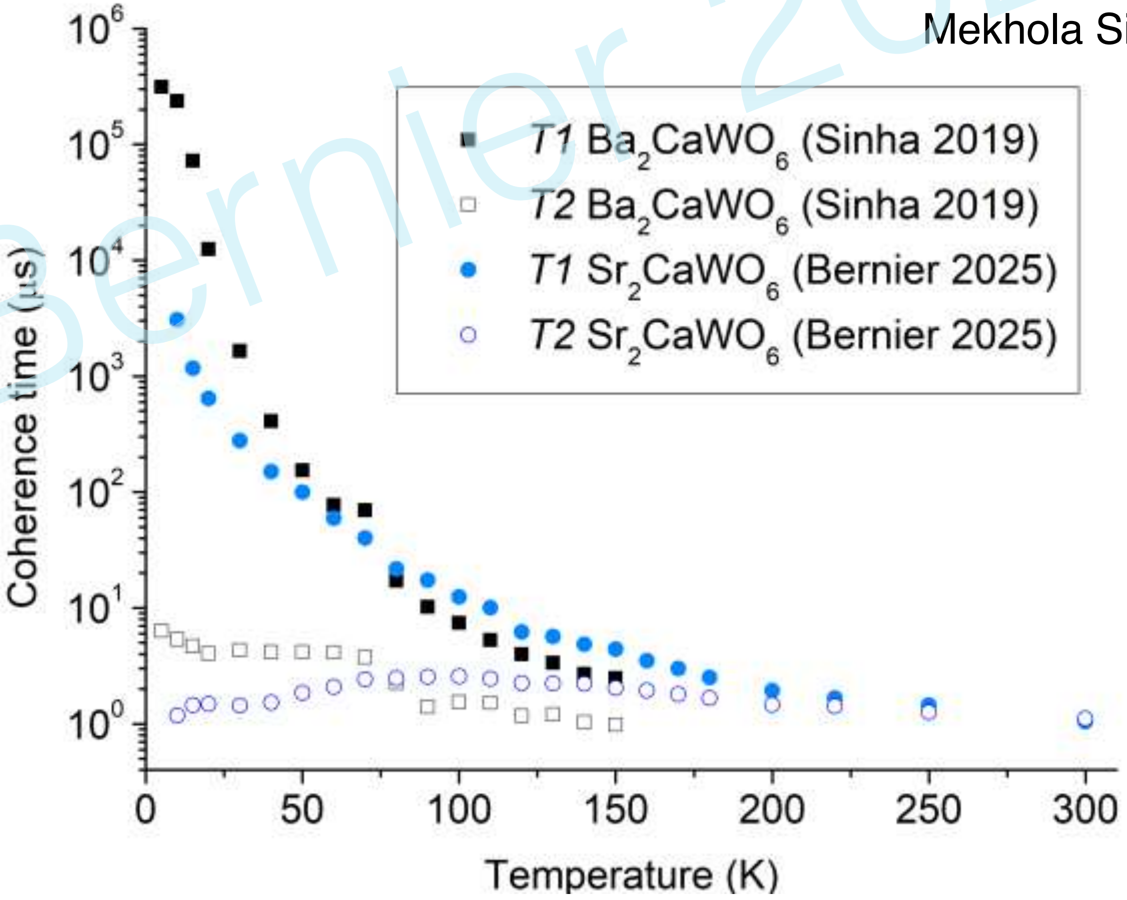
Quantum Materials → Quantum Computing

$Sr_2/Ba_2CaWO_{6-\delta}$ qubit candidates

Electron spin qubit host

Mekhola Sinha

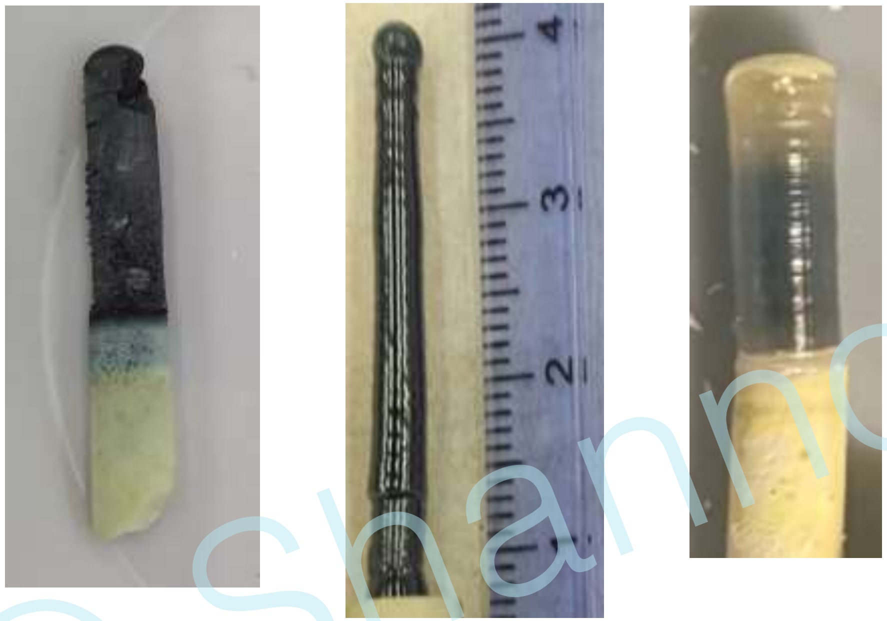


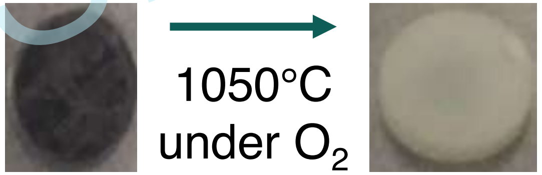


Bernier et. al. npj Quantum Mater. **10** 62 (2025)

Sr_2/Ba_2CaWO_6 oxidation state

Sinha **$Sr_2CaWO_{6-\delta}$** | **Sinha** **$Ba_2CaWO_{6-\delta}$** | **Neill** **$Ba_2CaWO_{6-\delta}$**





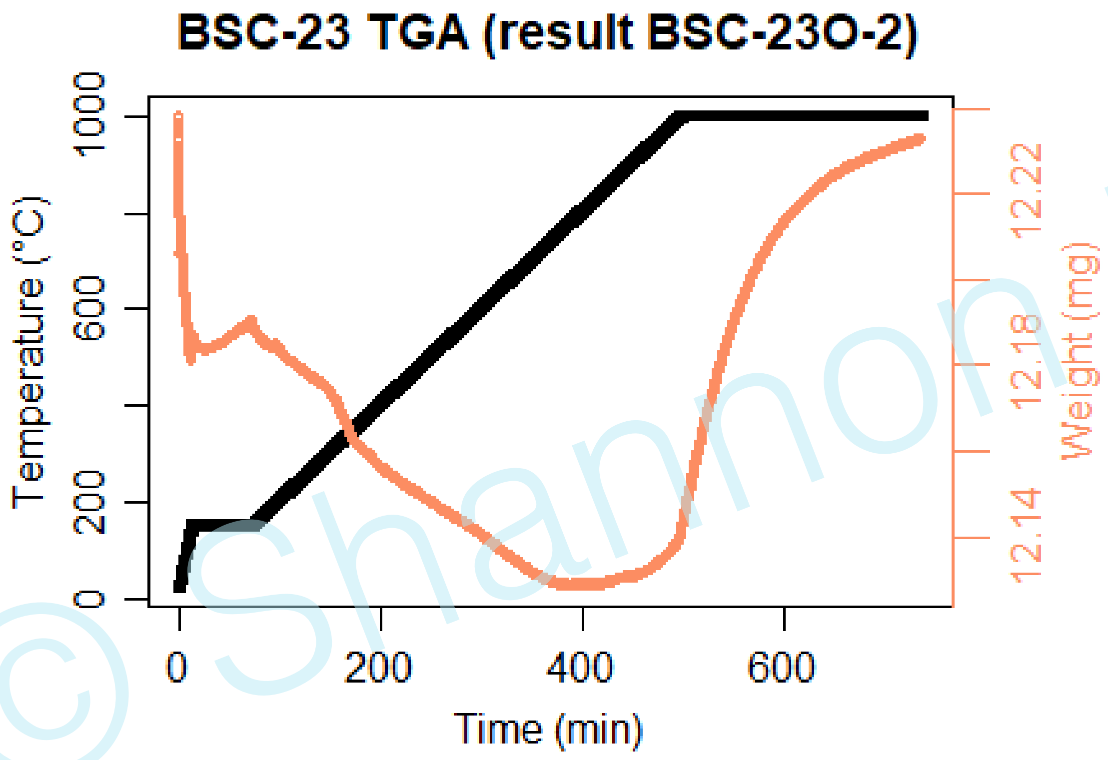
- Color changes under reduction indicate at least some W^{5+} centers.
- The exact concentration is important to interpret T_2 values.

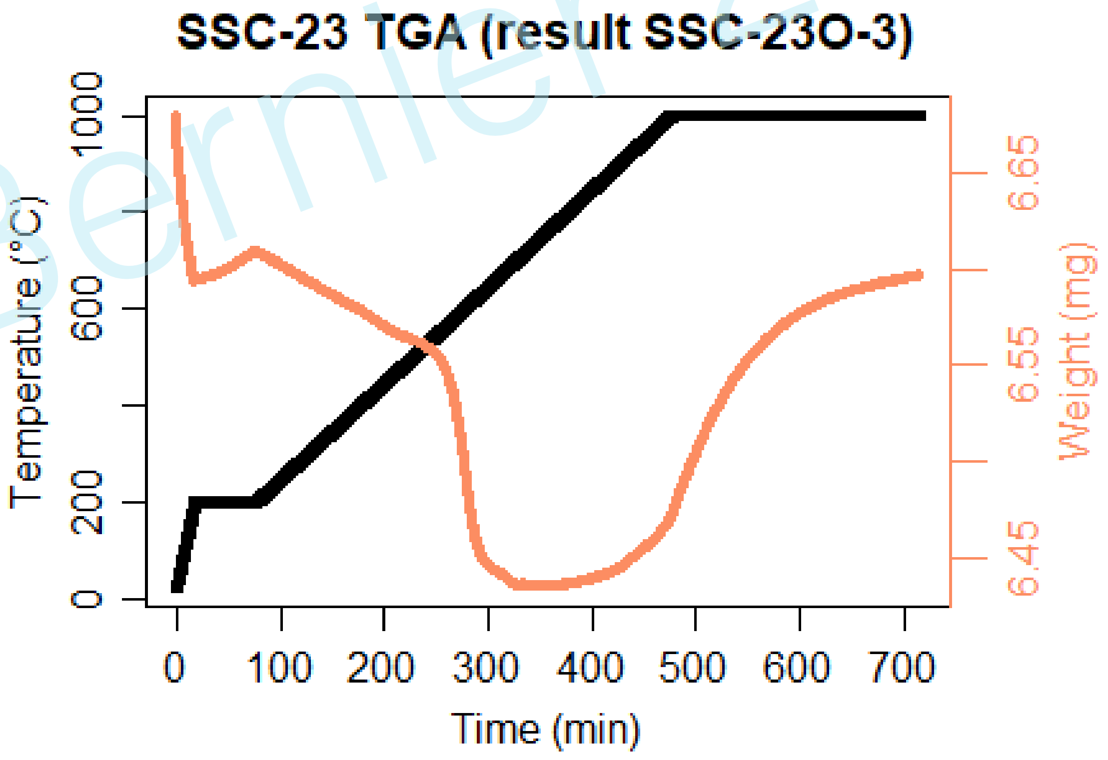
Mekhola Sinha

Abby Neill

Sinha et. al. PRB **3** 125002 (2019)
Neill et. al. Mat. Today Quant. **5** 100029 (2025)

Thermogravimetric analysis





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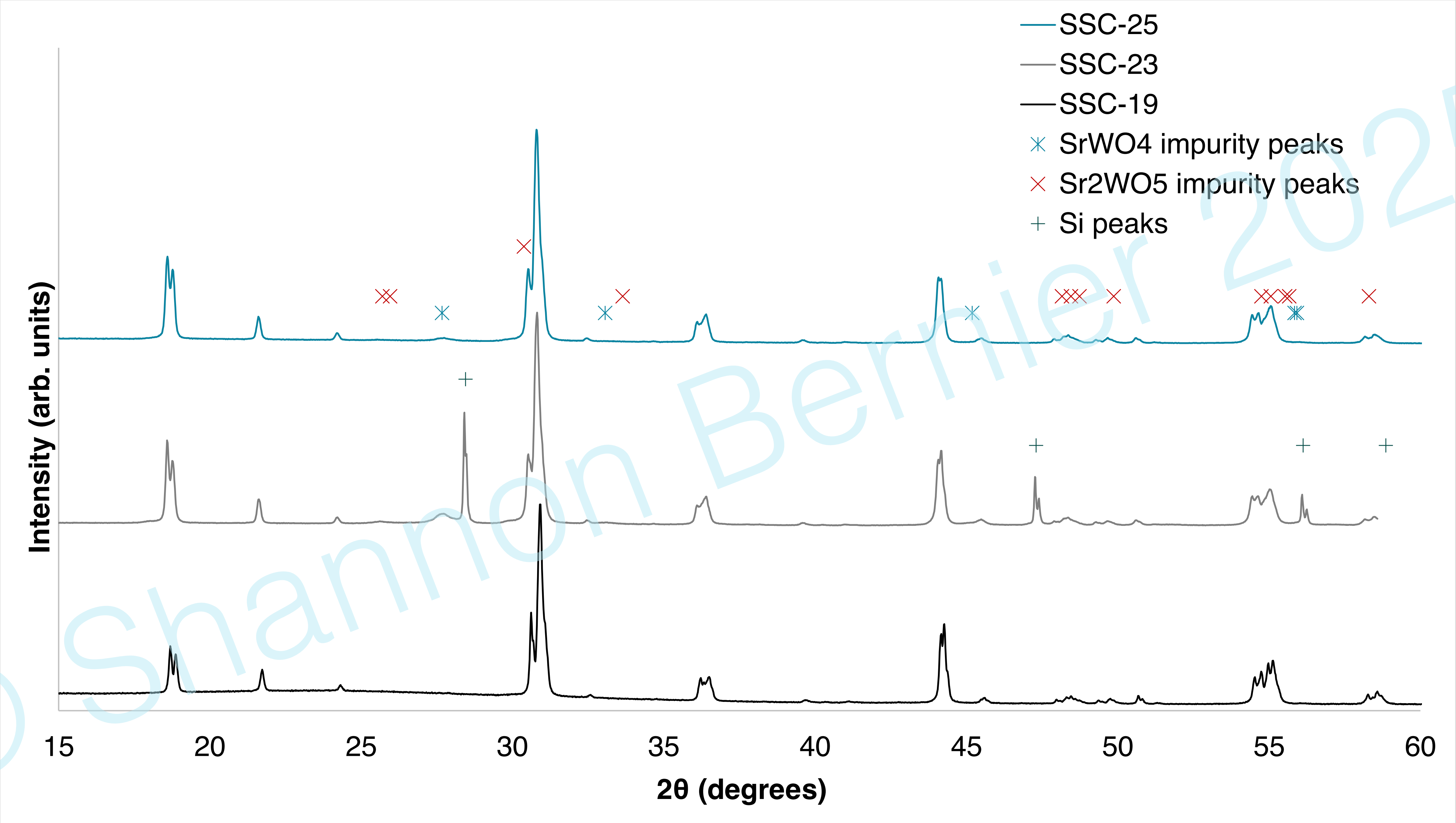

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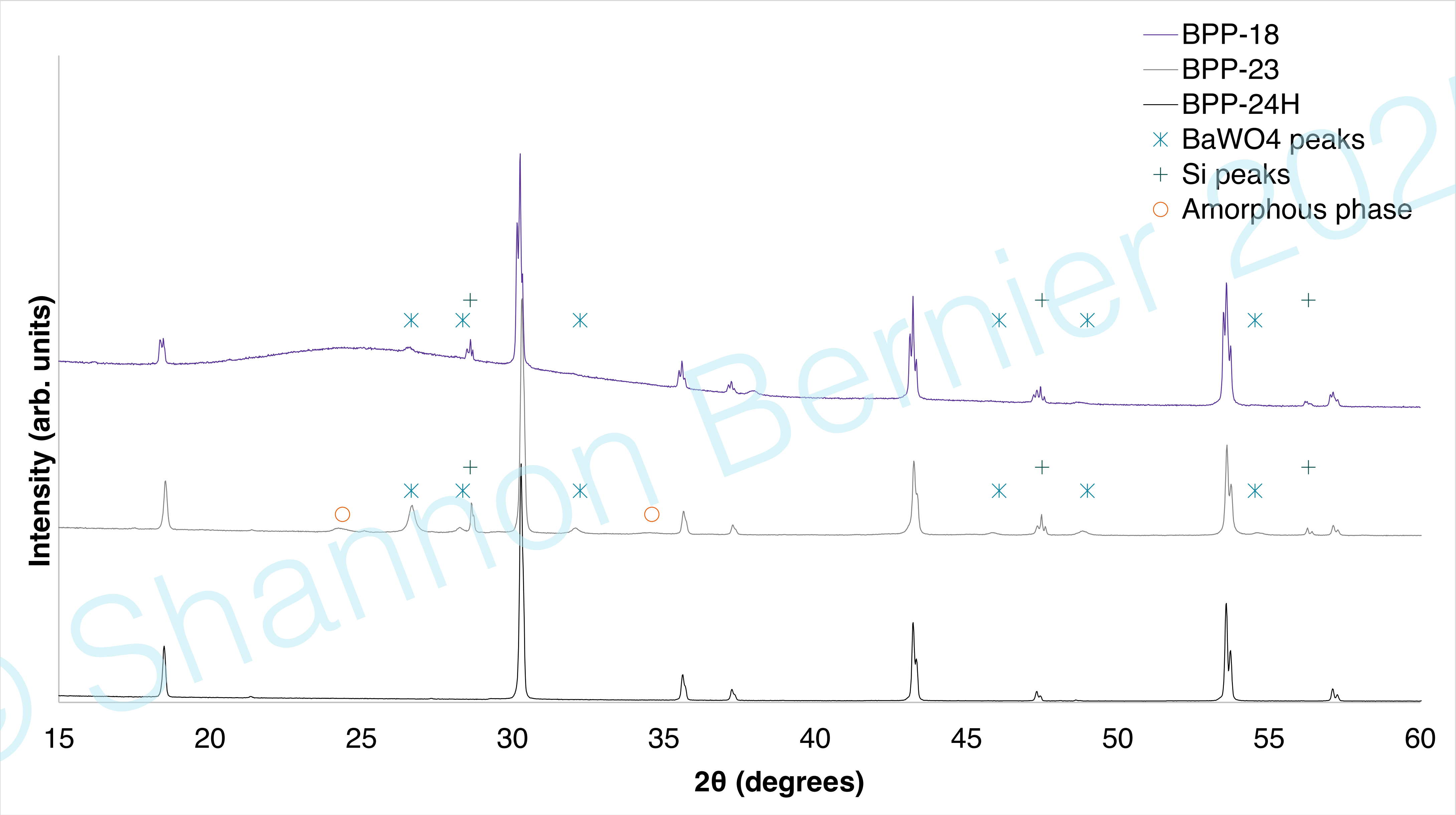
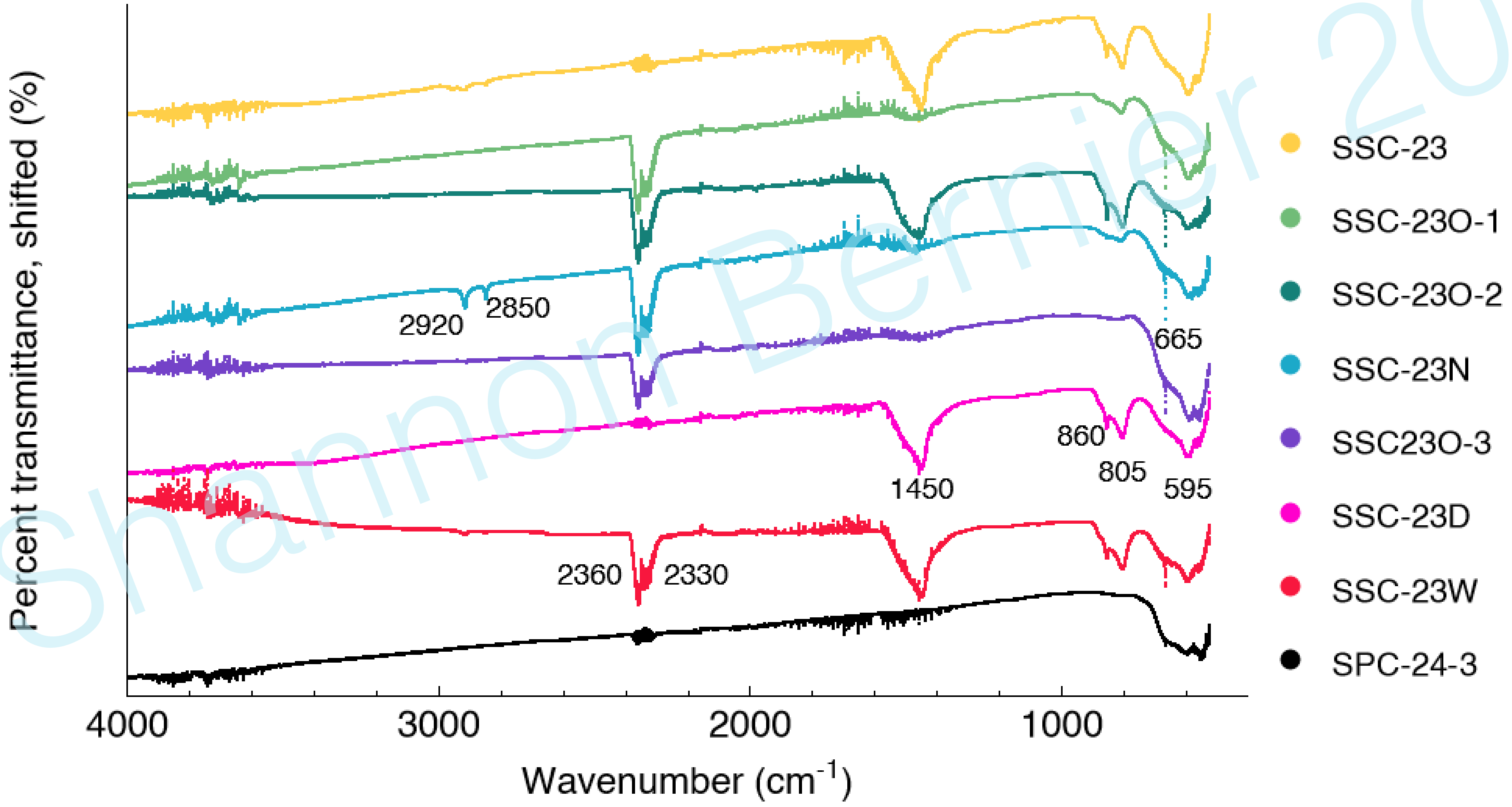


BPP-18
BPP-23
BPP-24H
BaWO4 peaks
Si peaks
Amorphous phase
Intensity (arb. units)
15
20
25
30
35
40
45
50
55
60
2θ (degrees)

Infrared spectroscopy



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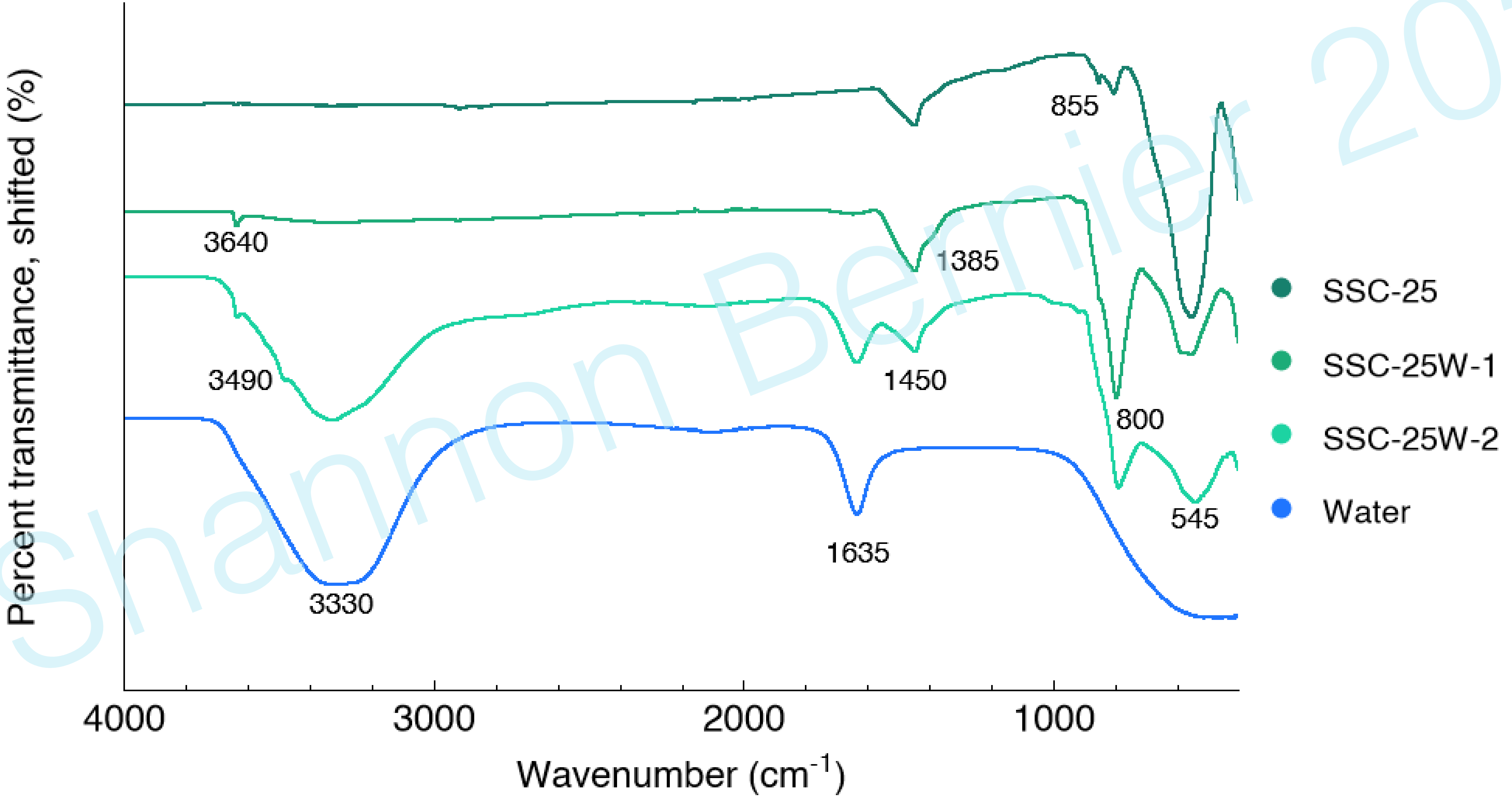
Infrared spectroscopy (cont.)



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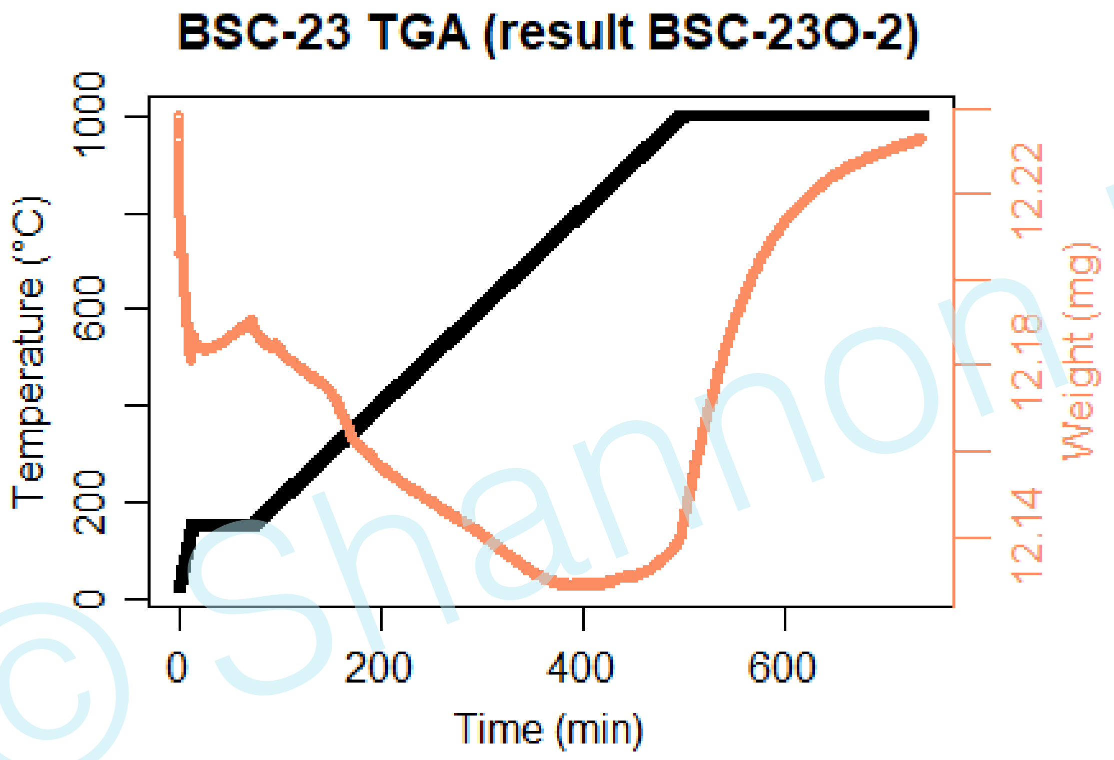
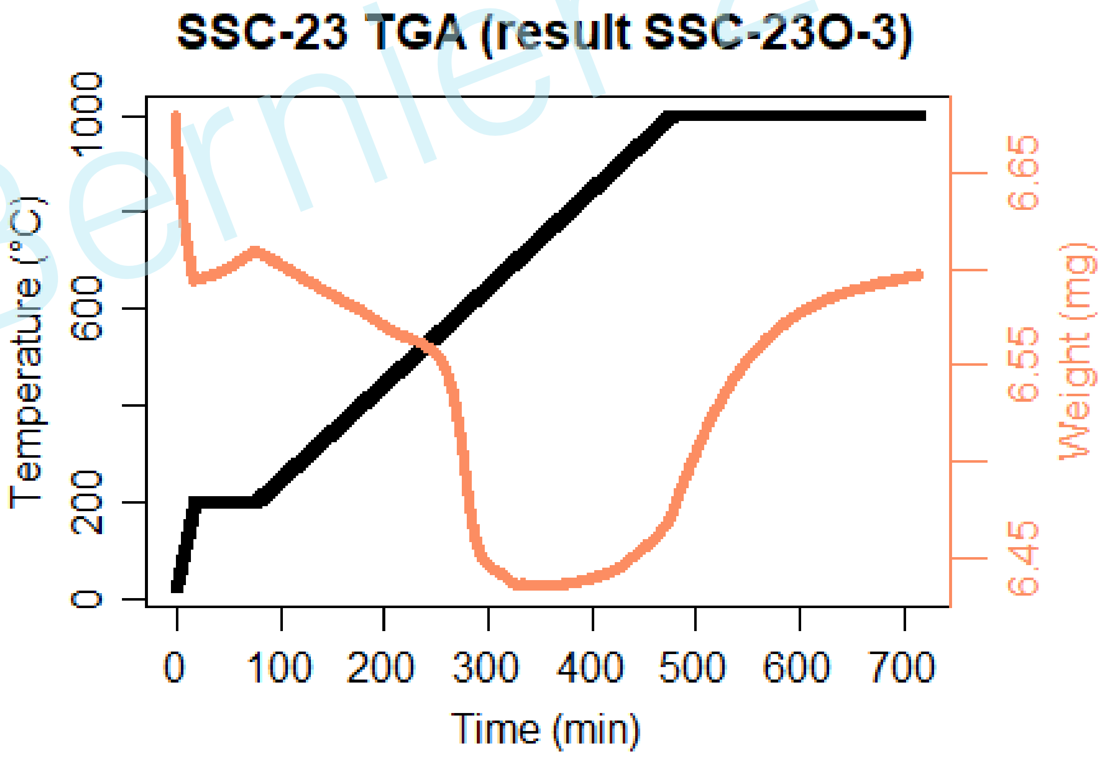
Infrared spectroscopy (cont.)

Peak (cm^{-1})	Assignment
500-800	Metal-oxygen bonds
800, 850	$-CO_3$ out-of-plane bending
1450	C-O bond stretching and M-O-H bending (overlapped)
1635	H-O-H bending of uncoordinated water
2280-2380	CO_2

TGA (reprise)

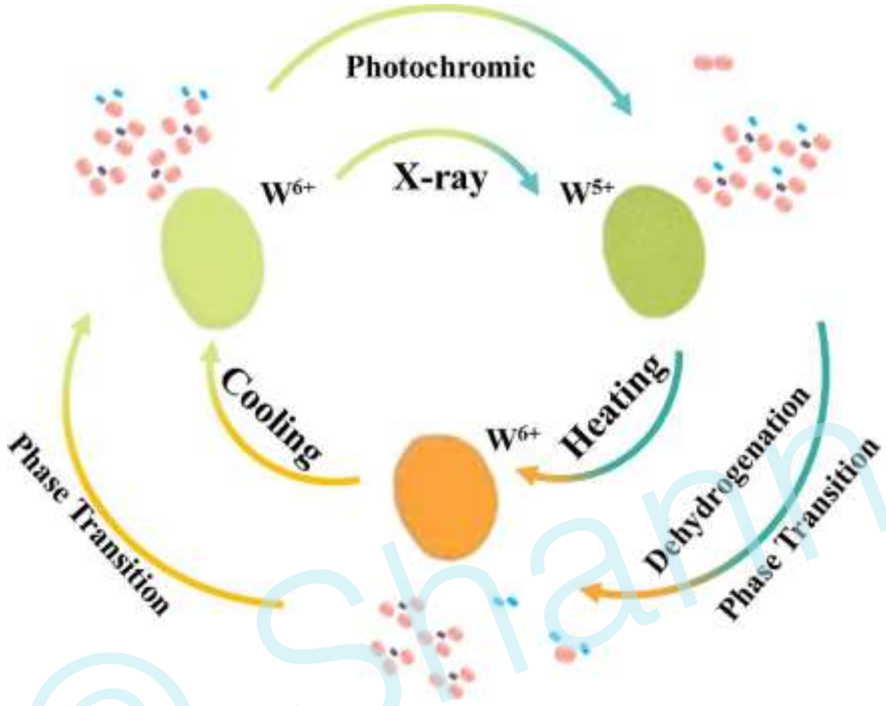


BSC-23 TGA (result BSC-23O-2)
Temperature (°C)
0
200
600
1000
Weight (mg)
12.14
12.18
12.22
0
200
400
600
Time (min)



SSC-23 TGA (result SSC-23O-3)
Temperature (°C)
0
200
600
1000
Weight (mg)
6.45
6.55
6.65
0
100
200
300
400
500
600
700
Time (min)

WO_3 photochromism



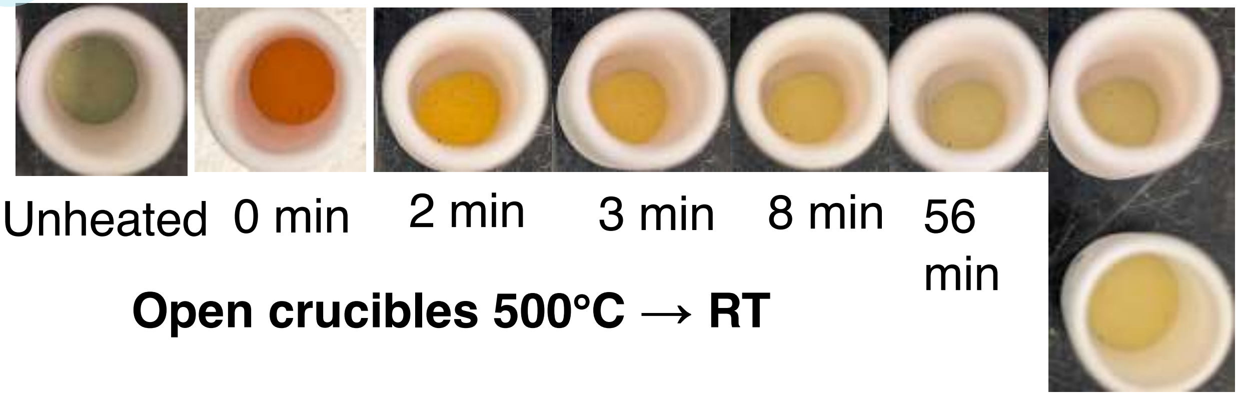
Unheated, 18 hr XRD

Heated, 30 min XRD

Dark-cooled

Light-cooled

Sealed tube 500°C → RT



Unheated | 0 min | 2 min | 3 min | 8 min | 56 min | 57 min

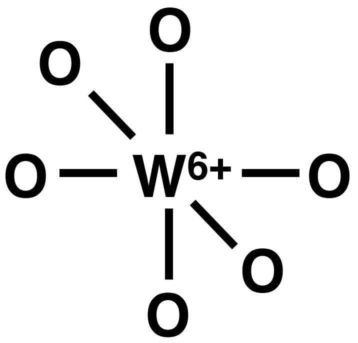
Light-cooled

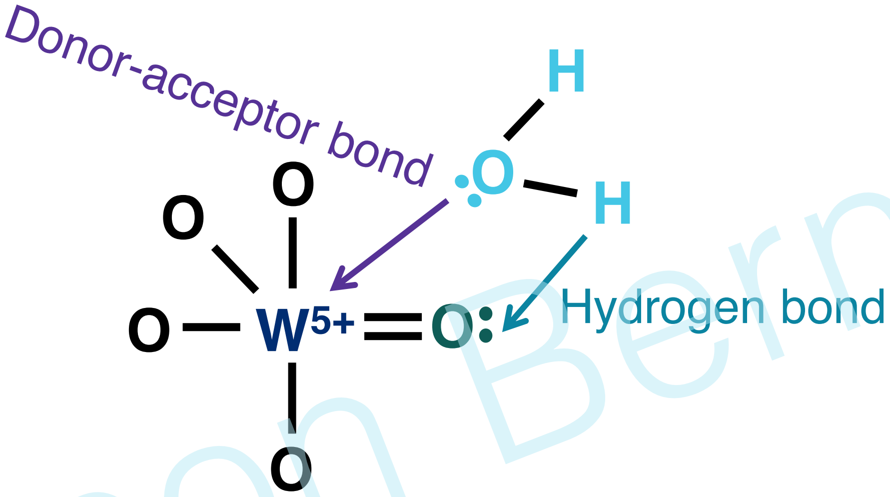
Dark-cooled

Open crucibles 500°C → RT

Yuan et. al. Mat. Lett. **349** 134688 (2023)

WO_3 photochromism (cont.)

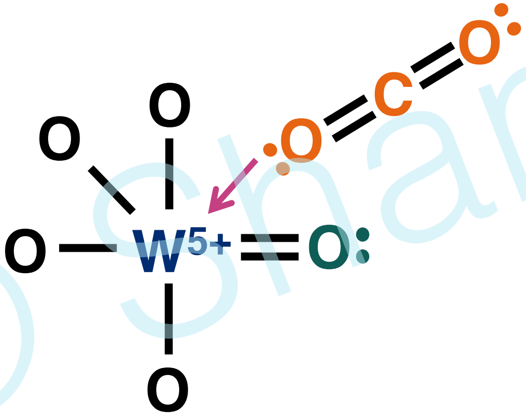




$$WO_3 + h\nu \longrightarrow WO_3^{*} + e^- + \text{hole}^+$$

$$4\text{hole}^+ + 2H_2O \longrightarrow 4H^+ + O_2$$

$$WO_3 + xe^- + xH^+ \longrightarrow H_xWO_3$$



$$\text{Ba/Sr}_2\text{CaWO}_6 + \textit{reductant} \longrightarrow \text{Ba/Sr}_2\text{CaW}^{6+}_{1-\delta}\text{W}^{5+}_{\delta}\text{O}_{6-\delta}$$

$$M_3W^{6+}_{1-\delta}W^{5+}_{\delta}O_{6-\delta} + H_2O \longrightarrow M_3WO_{6-\delta}H_\delta + O^{-2}$$

$$M_3WO_{6-\delta}H_\delta + CO_2 \longrightarrow MWO_{4-\delta}H_\delta + MCO_3$$

or $M_3WO_{6-\delta} + CO_2 \longrightarrow MWO_{4-\delta} + MCO_3$

where M = Sr, Ba, or Ca

Gavrilyuk App. Surf. Sci. **364** 498-504 (2016)

Results

- Breakdown of Ba/Sr_2CaWO_6 crystals into carbonates and $Ba/SrWO_4$ is slow but appears related to light and water exposure.
- This should be accelerated by more oxygen vacancies.
- δ was eventually calculated by measuring the Curie-Weiss constant (C).

Sample	C (emu K/mol)	Calculated formula
BSC-23	0.0005739	$Ba_2CaWO_{5.99}$
SSC-23	0.004451	$Sr_2CaWO_{5.93}$

Funding & collaborators

NATIONAL HIGH MAGNETIC FIELD LABORATORY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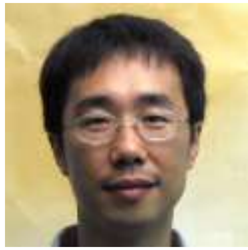
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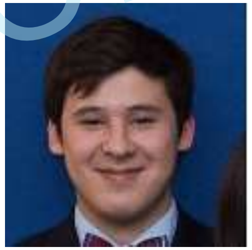
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Summary

1. Quantum Materials
2. Quantum Computing
3. Quantum Computing ↔ Quantum Materials
   $Yb_{2-x}Lu_{2x}Ti_2O_7$
4. Quantum Materials → Quantum Computing
   $Sr_2/Ba_2CaWO_{6-\delta}$