UNIVERSITÉ DE GENÈVE
FACULTÉ DES SCIENCES
Section de physique

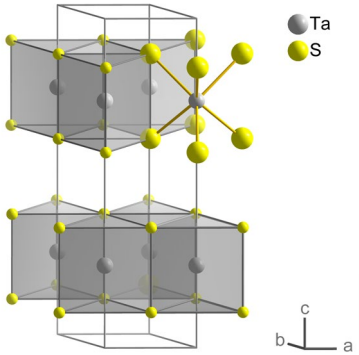
DQMP Department of Quantum Matter Physics
von Rohr Lab

Flat Club Winter Semester 2026
02.10.2026

Controlling the Magnetic Properties of Layered van der Waals Crystals Through Targeted Growth

Maria A. Herz
Department of Quantum Matter Physics (DQMP)| Université de Genève

Which Compounds? 2D van der Waals Materials



2H TaS_2

Tuneable Bandgaps

Large Structural Variability

Supercon-ductivity

Ferro- and piezoelectrics

Asymmetric Structures

Tuneable Functionalisation

Hetero-structure Devices

Chalcogenides

Halides

Mixed chalcogenide-halides

Exotic magnetic properties

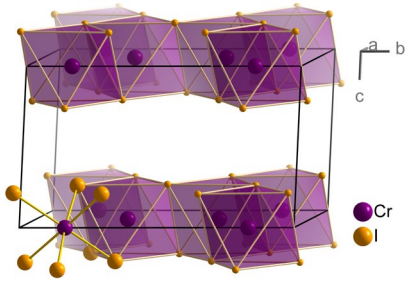
Long range magnetic ordering

Layer-dependent ordering and magnetism

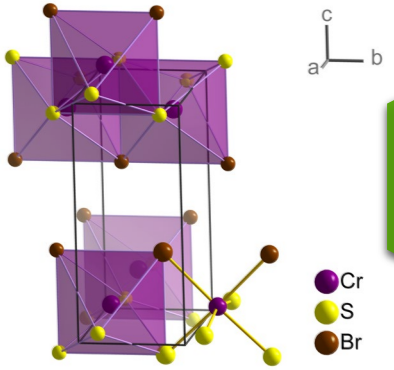
Second Harmonic Generation (SHG)

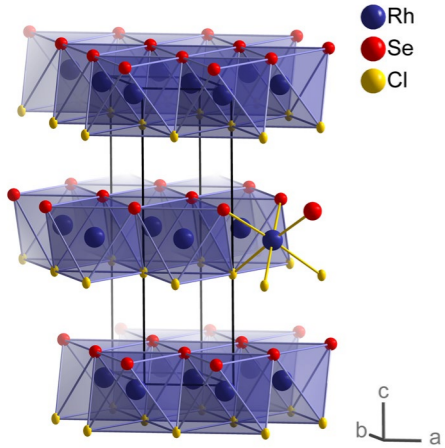
2D Janus Materials

2D Semiconductors



CrI_3



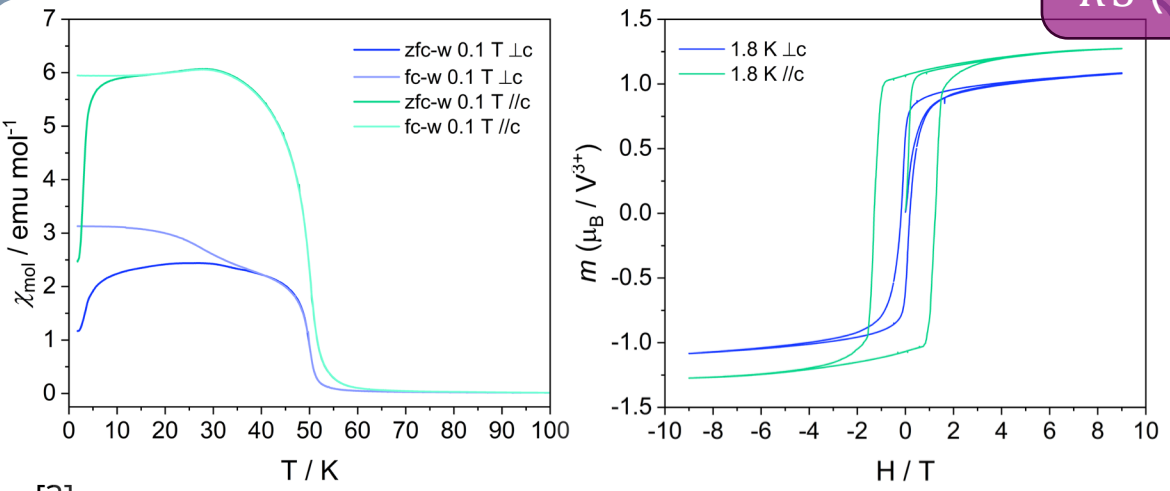


[1] J. Li, S.-S. Han, S.-P. Guo, *Eur. J. Inorg. Chem.* **2022**, e202200419.
[2] U. V. Ghorpade *et al*., *Chem. Rev.* **2023**, *123*, 327.

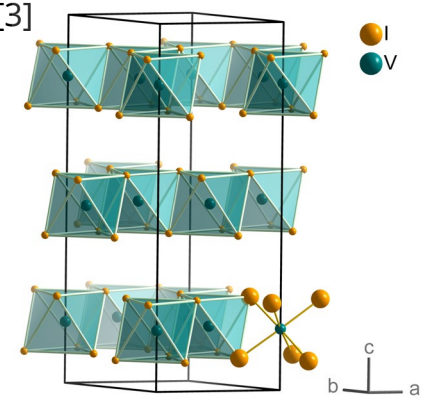
The End Members: VI_3 and CrI_3

CrI_3
$R\bar{3}$ (< 220 K)

CrI_3
$C2/m$ (RT)

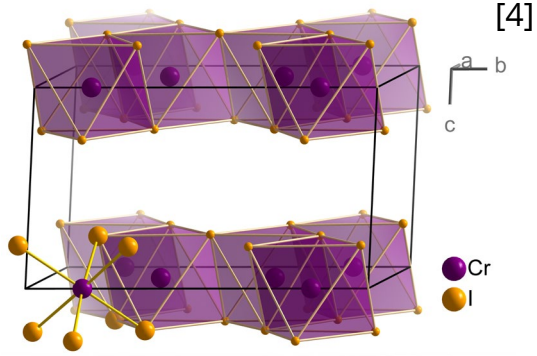


[3]

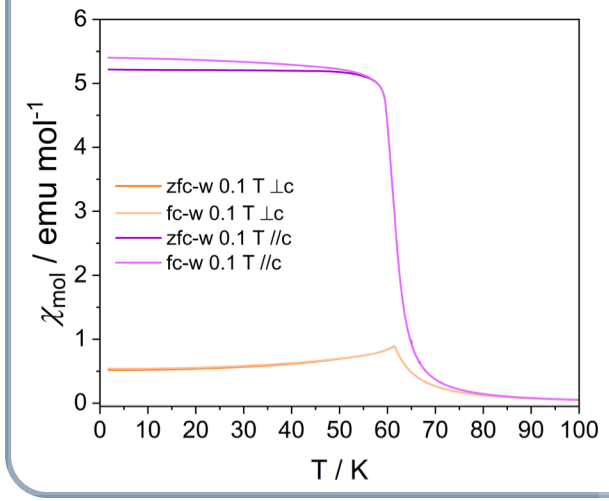


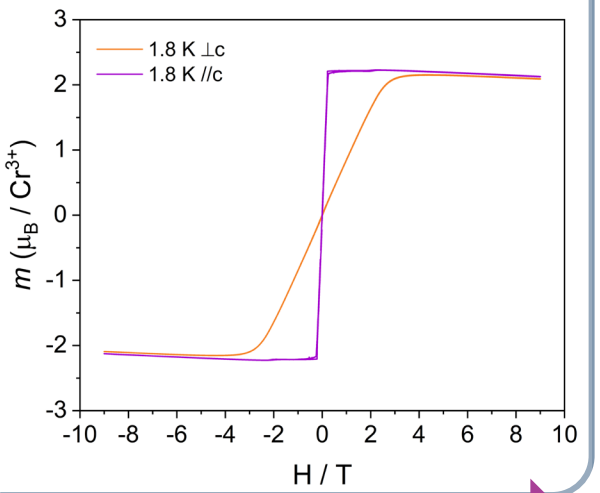
- Bulk ferromagnet
- T_{C1} = 50 K, T_{C2} ≈ 35 K
- Interplay between spin and orbital moment

[4]



- Bulk ferromagnet
  - ➢ Layer-dependent[5]
  - ➢ Odd-even effect
- T_C = 61 K





VI_3
$C2/m$ (< 79 K)

VI_3
$R\bar{3}$ (RT)

[3] S. Tian *et al.*, *J. Am. Chem. Soc.* **2019**, *141*, 5326.
[4] M. McGuire *et al.*, *Chem. Mat.* **2015**, *27*, 612; N.Ubrig *et al.*, *2D Mater.* **2020**, *7*, 015007.
[5] B. Huang *et al.*, *Nature* **2017**, *546*, 270.

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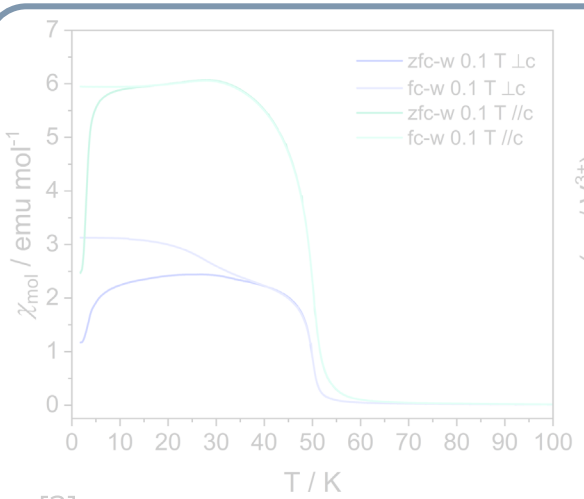

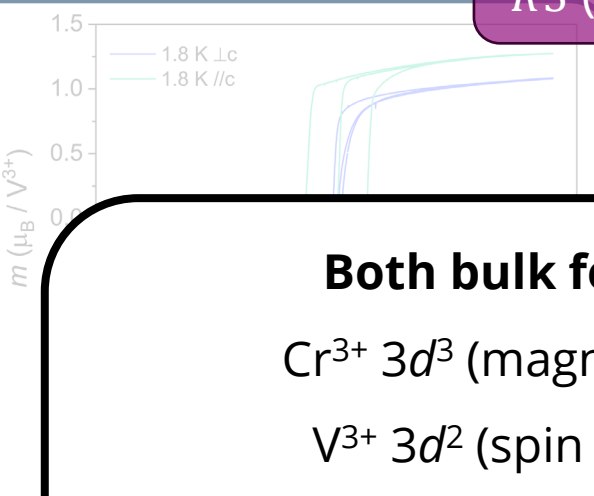

[3]

- B
- T
- Int
  and orbital moment

[4]

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  - ➢ Layer-dependent[5]
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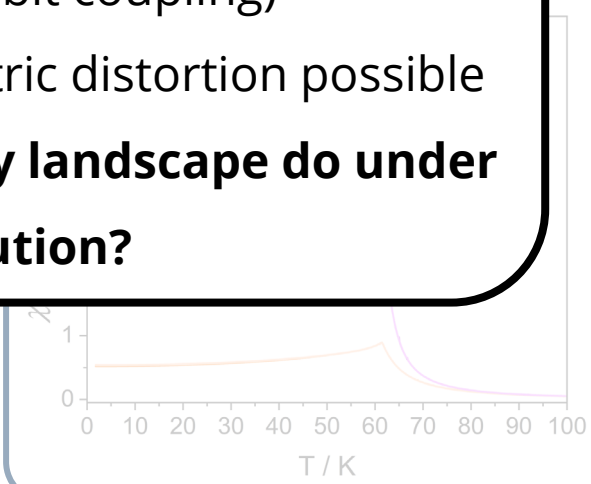

Both bulk ferromagnets:

Cr^{3+} $3d^3$ (magnetic anisotropy)

V^{3+} $3d^2$ (spin orbit coupling)

Structure: Some geometric distortion possible

What does a flat energy landscape do under substitution?

VI_3
$C2/m$ (< 79 K)

VI_3
$R\bar{3}$ (RT)

[3] S. Tian *et al.*, *J. Am. Chem. Soc.* **2019**, *141*, 5326.
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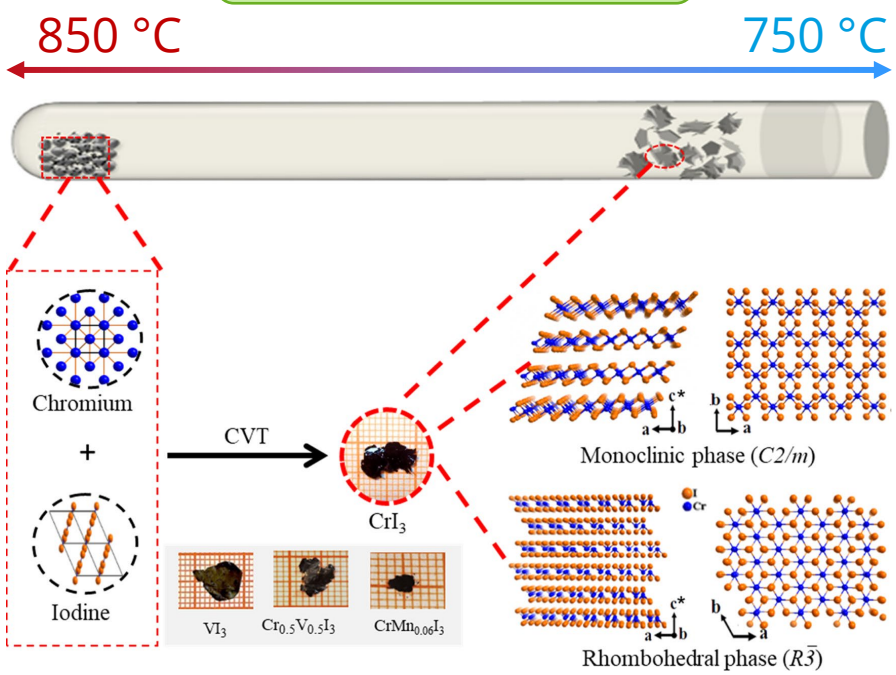
[5] B. Huang *et al.*, *Nature* **2017**, *546*, 270.

The Challenges of $V_xCr_{1-x}I_3$

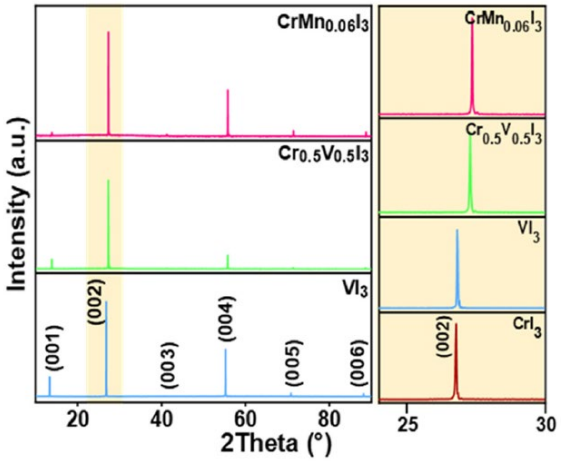
Phase Separation



Chemical vapour transport (CVT) syntheses of $V_{0.5}Cr_{0.5}I_3$[6,7,8]

→ Synthesis method does **not** work

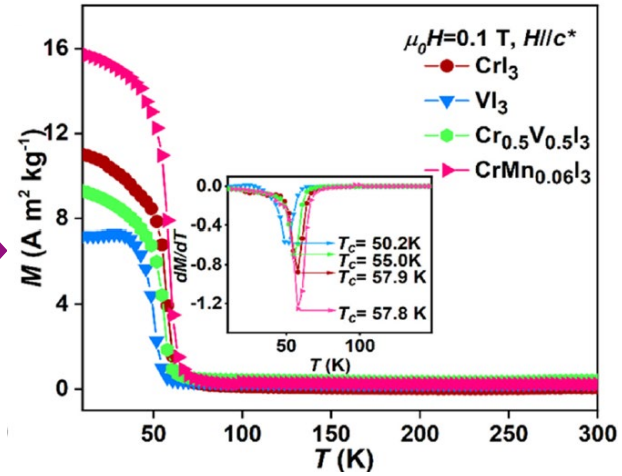
Phase Identification



Powder X-ray Diffraction: Highly oriented samples

→ Uncertain identification

Inconclusive Magnetism



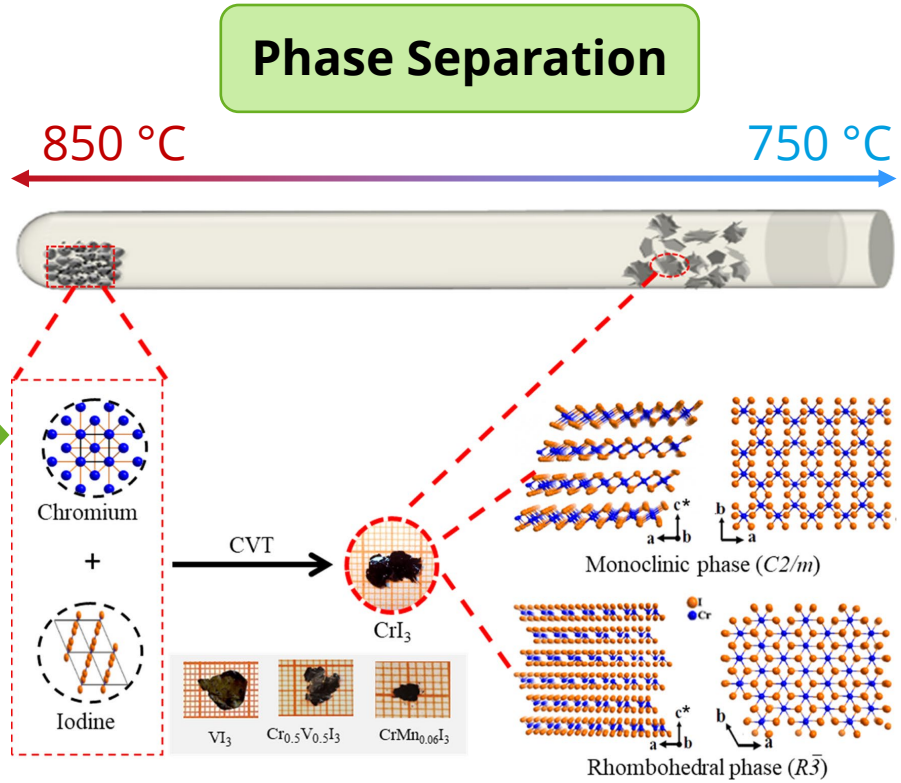
Ferromagnetic behaviour

Inconclusive regarding the exact dynamics

[6] S. Pan *et al.*, *J. Alloys Compd.* **2022**, *908*, 164573.
[7] X. Li *et al.*, *Nature Physics* **2024**, *20*, 1145.
[8] Q. Wang *et al.*, *Appl. Phys. Lett.* **2025**, *127*, 052405.

Synthesis: Why Does It Not Work?

Phase Separation



Chemical vapour transport (CVT) syntheses of $V_{0.5}Cr_{0.5}I_3$[6,7,8]

→ Synthesis method does **not** work

Challenges:

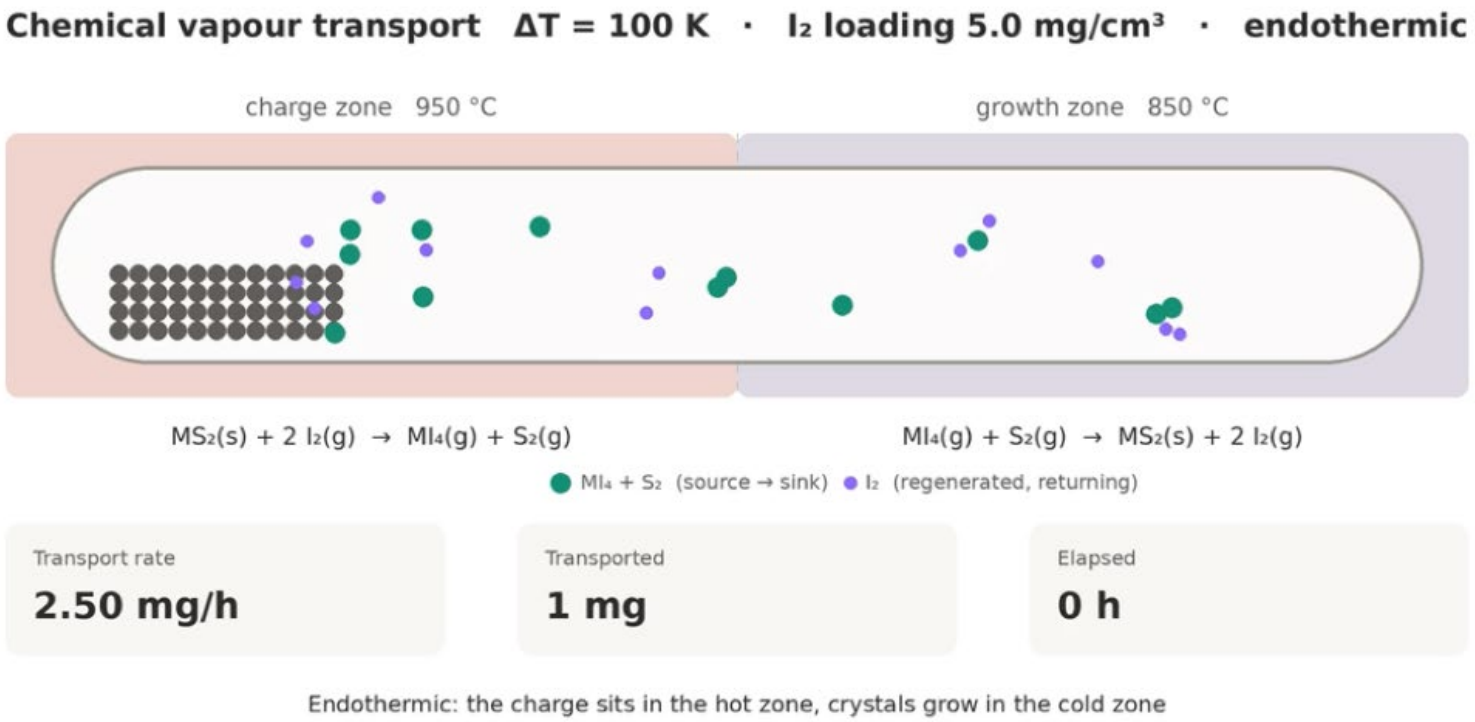
- CVT causes phase separation
  - ➢ Incongruent transport
  - ➢ Competing phase formation

Solution?

- Different crystal growth route, *i.e.*, physical vapour transport (PVT), self-selecting vapour growth (SSVG), or flux growth
- Consider crystal growth mechanism
- Choice of reagents
- Fine-tuning reaction conditions

Synthesis Pathway: Which One is Best?

Parameter	Chemical Vapour Transport (CVT)
Driving force	ΔT shifts equilibrium
Transport Medium	Reactive gas phase
Purity	High, pot. substitution
Growth Rate	Slow (days – weeks)
Agent / Solvent	Often halogen



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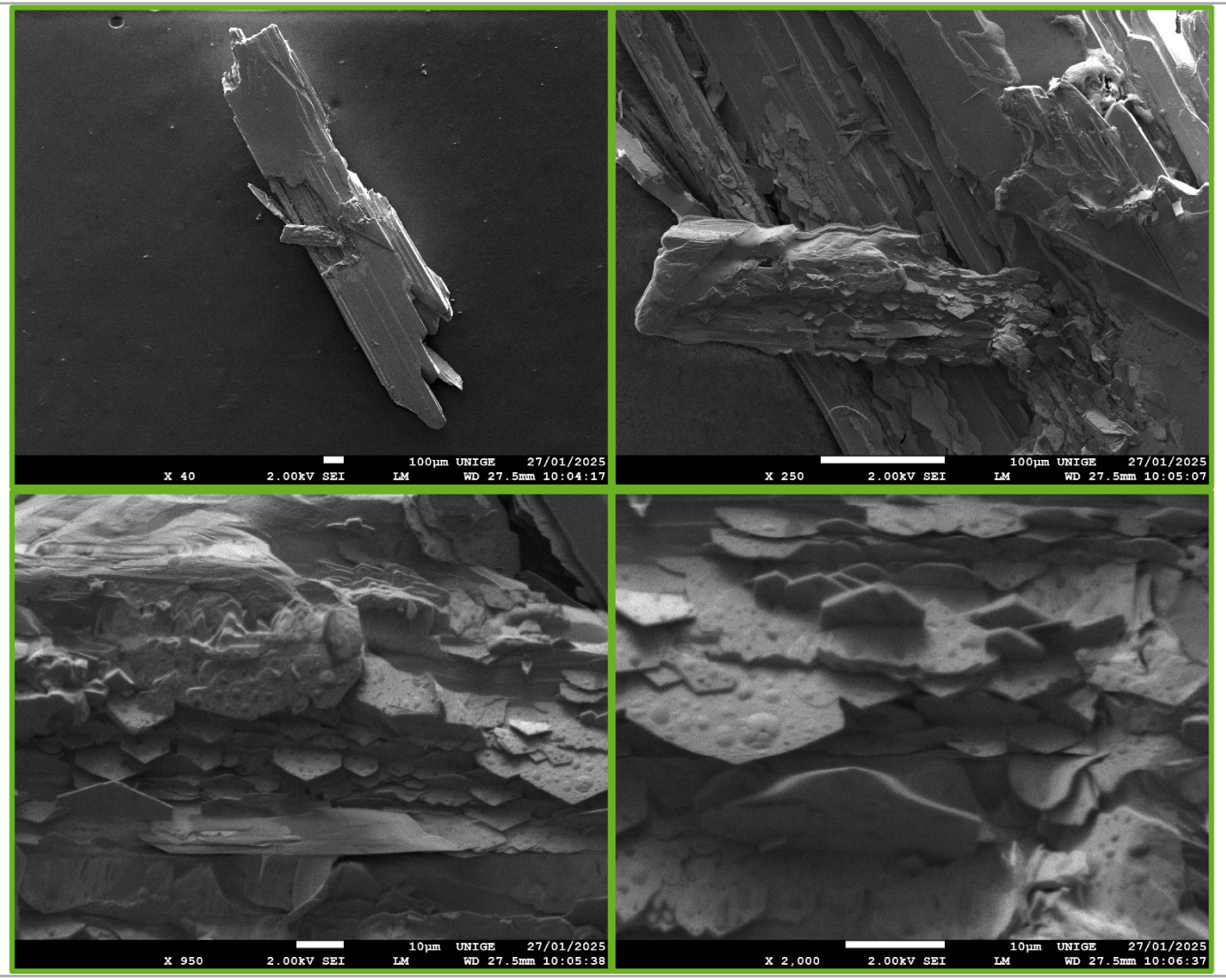
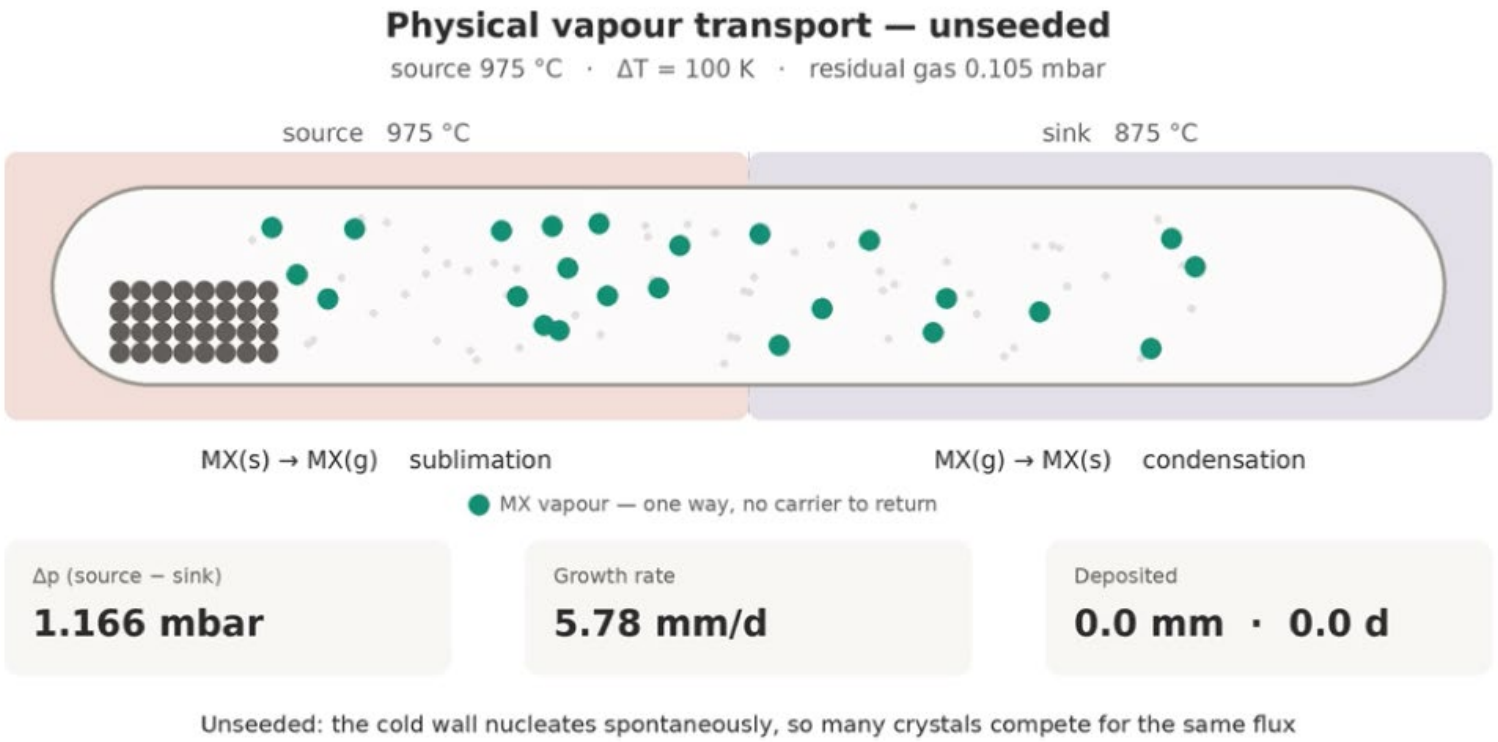


Synthesis Pathway: Which One is Best?

Parameter	Physical Vapour Transport (PVT)
Driving force	ΔT drives sublimation
Transport Medium	Self-transport
Purity	Very high
Growth Rate	Slow – moderate
Agent / Solvent	None



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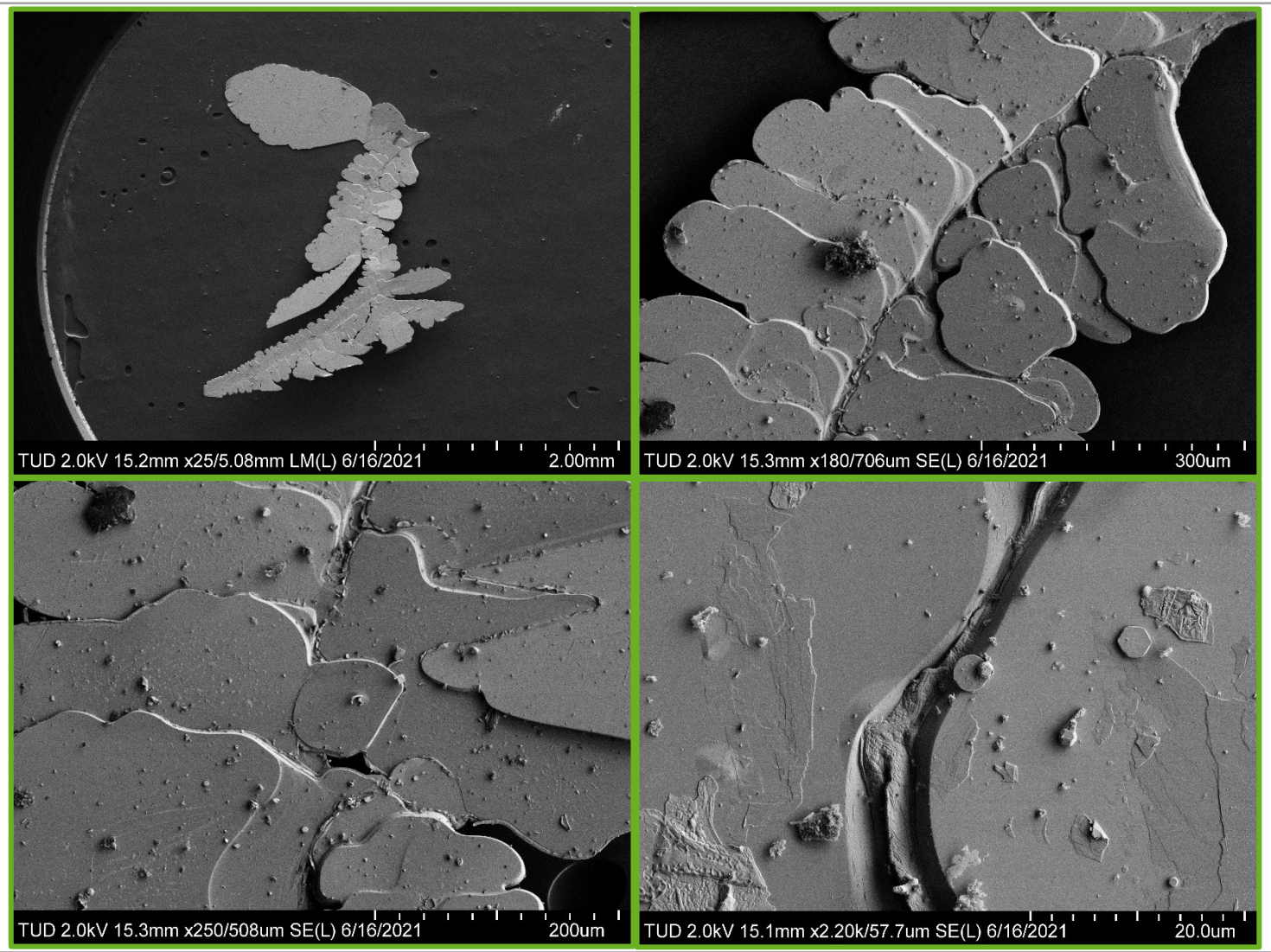
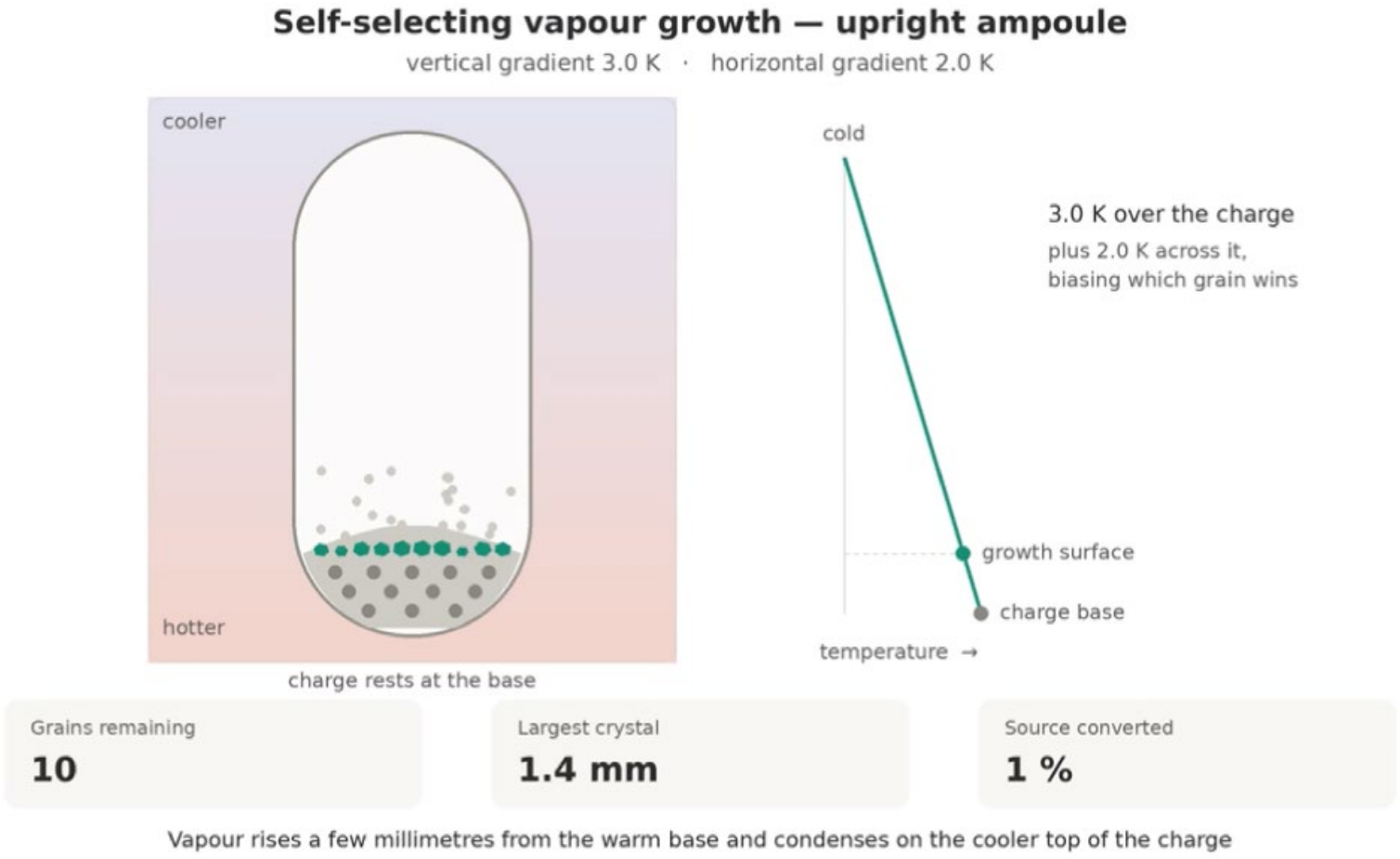


Synthesis Pathway: Which One is Best?

Parameter	Self-Selecting Vapour Growth (SSVG)
Driving force	Near isothermal; $\Delta T \leq 5°C$
Transport Medium	Own vapour pressure
Purity	High; self-selected
Growth Rate	Slow (days – weeks)
Agent / Solvent	None



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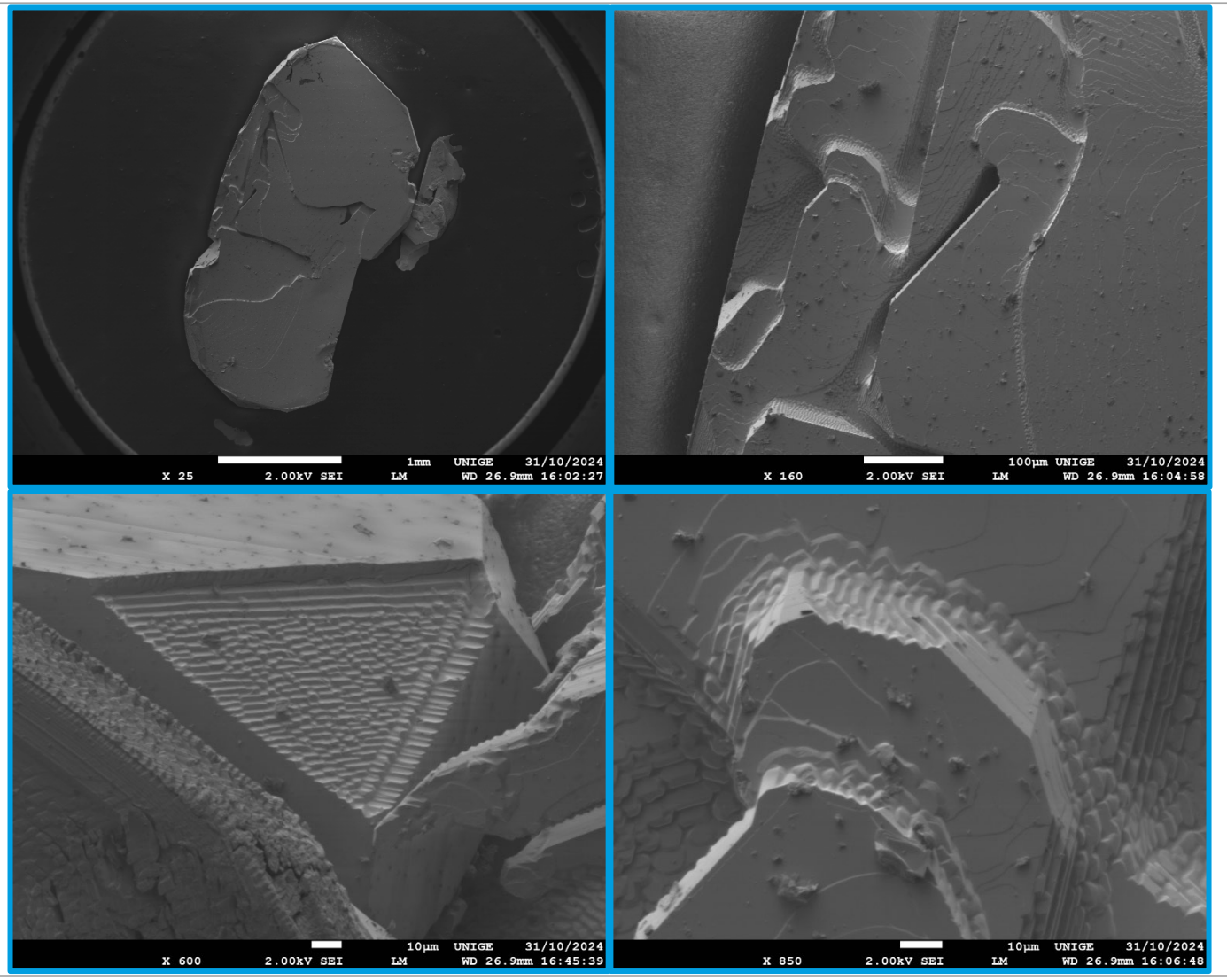
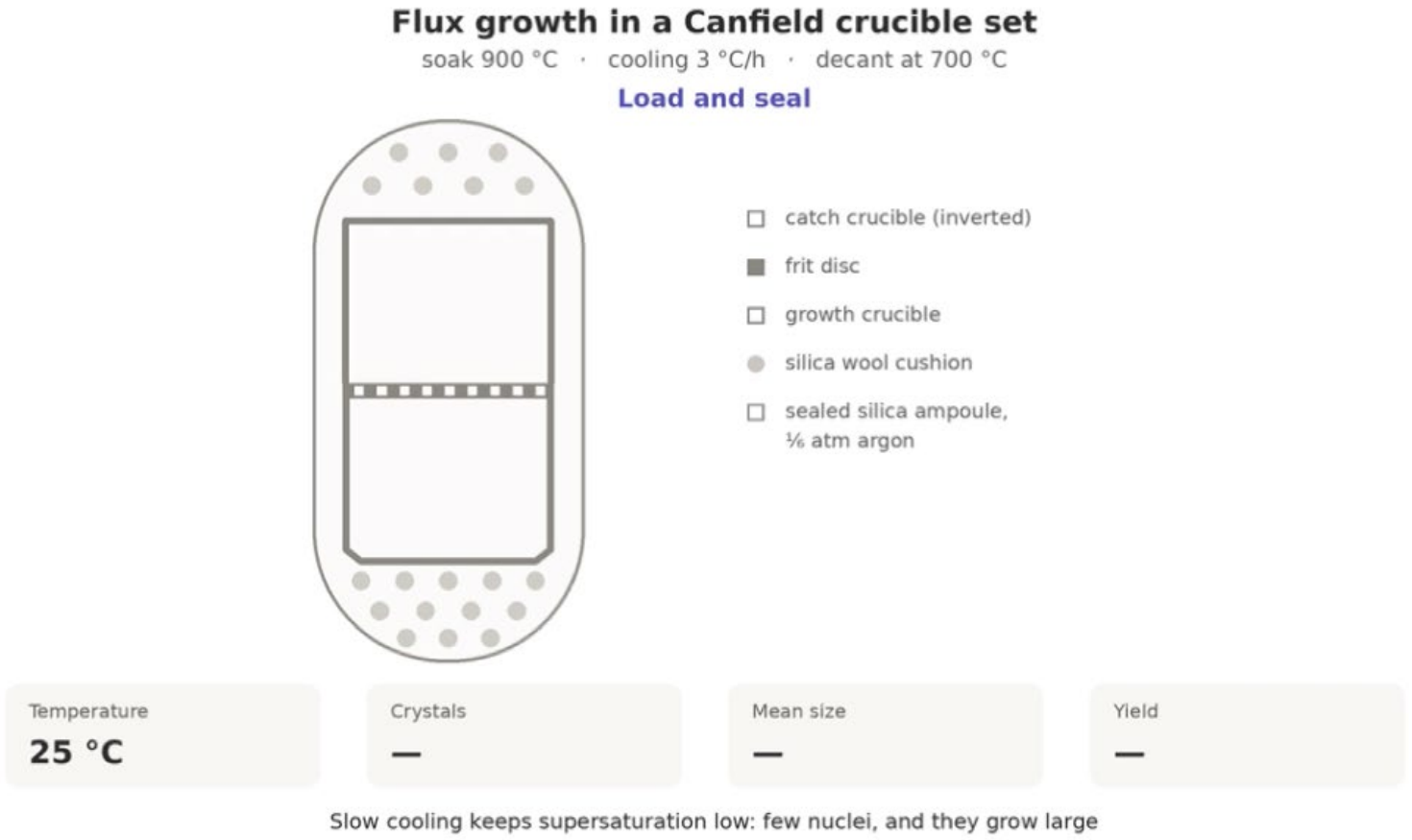


Synthesis Pathway: Which One is Best?

Parameter	Flux Growth
Driving force	Slow cooling → supersaturation
Transport Medium	Molten flux
Purity	Moderate, flux inclusion
Growth Rate	Moderate (days)
Agent / Solvent	Oxide, halide, metal



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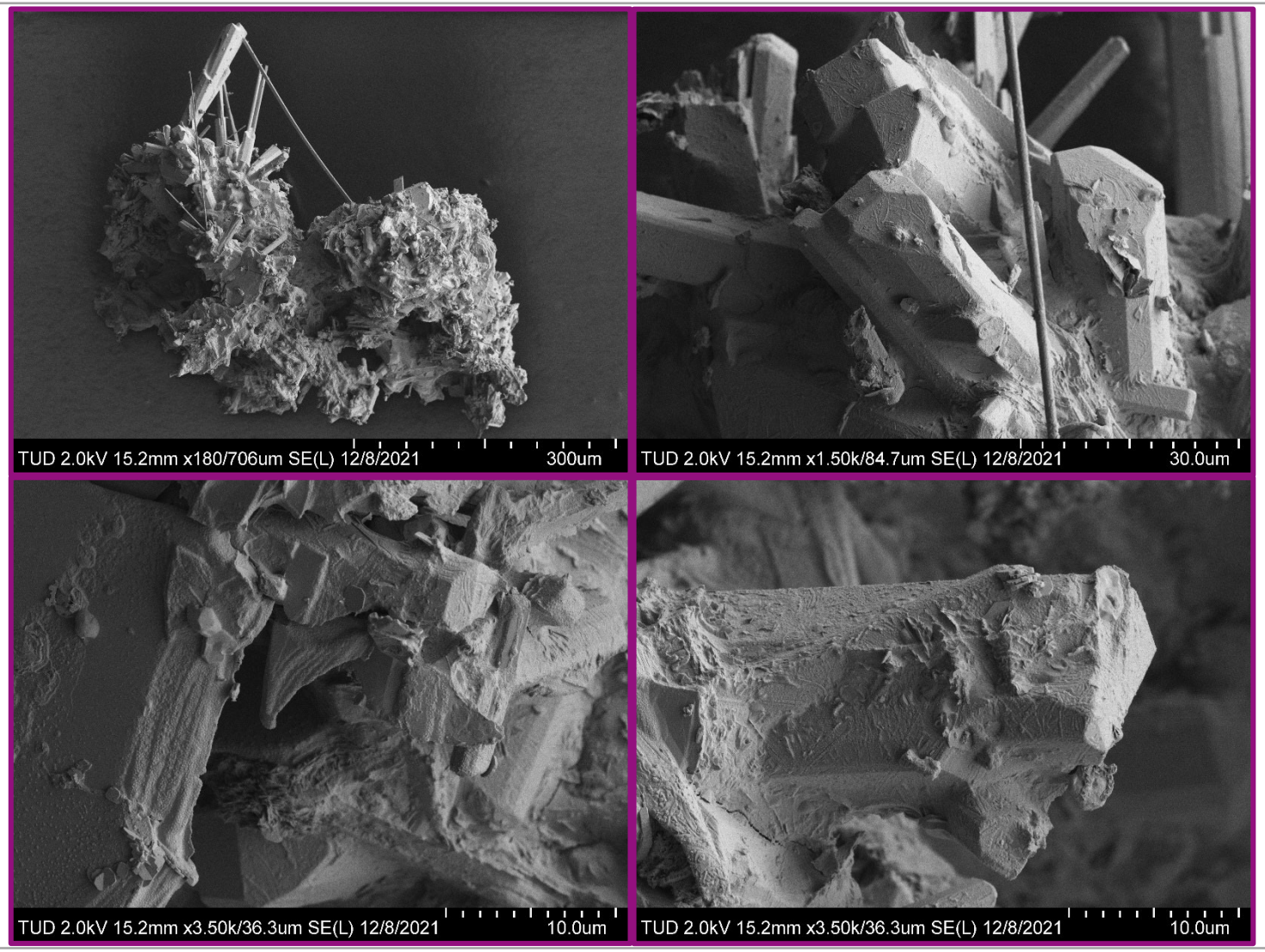


Animation generated with Claude (Anthropic)

Synthesis Pathway: Which One is Best?

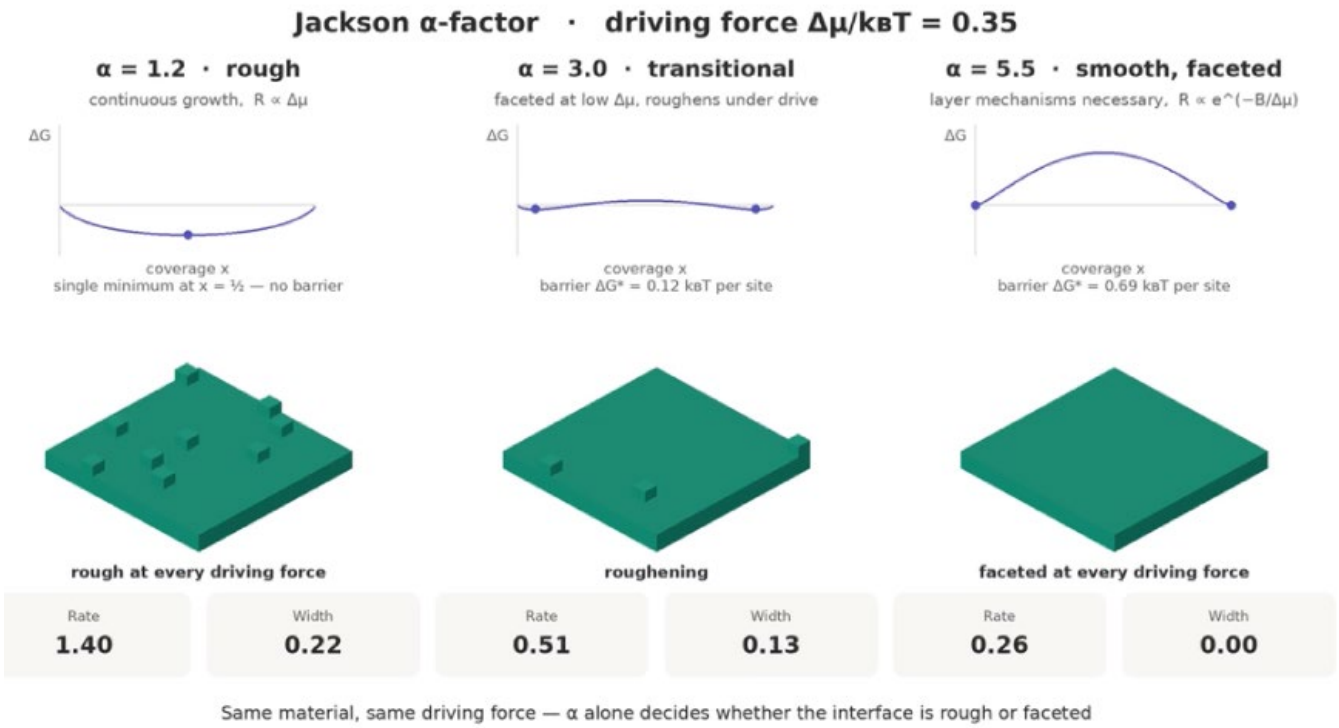
Parameter	Chemical Vapour Transport (CVT)	Physical Vapour Transport (PVT)	Self-Selecting Vapour Growth (SSVG)	Flux Growth
Driving force	ΔT shifts equilibrium	ΔT drives sublimation	Near isothermal; $\Delta T \leq 5°C$	Slow cooling → supersaturation
Transport Medium	Reactive gas phase	Self-transport	Own vapour pressure	Molten flux
Purity	High, pot. substitution	Very high	High; self-selected	Moderate, flux inclusion
Growth Rate	Slow (days – weeks)	Slow – moderate	Slow (days – weeks)	Moderate (days)
Agent / Solvent	Often halogen	None	None	Oxide, halide, metal

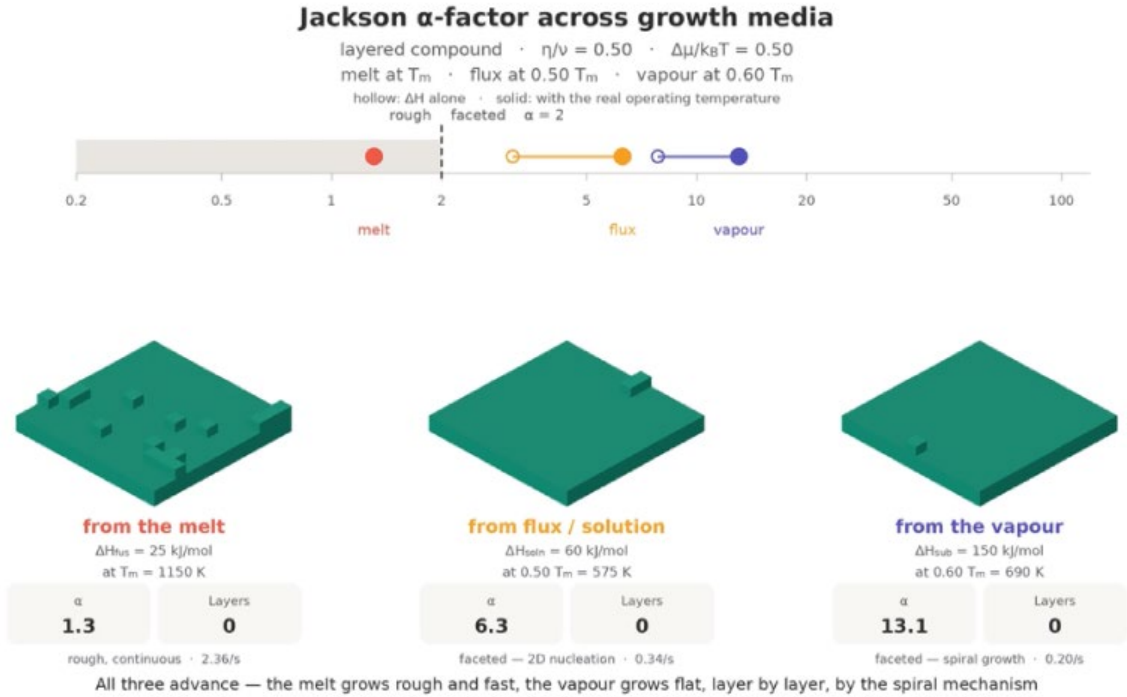
Large, high-purity crystals
No 'foreign' transport agents
Compromise on growth rate

Crystal Growth Mechanisms: Are They Important?

Two factors to consider for the growth mechanisms:

1. Jackson α factor:





$$\alpha = \frac{\Delta H_{trans}}{\kappa_B T} \xi$$

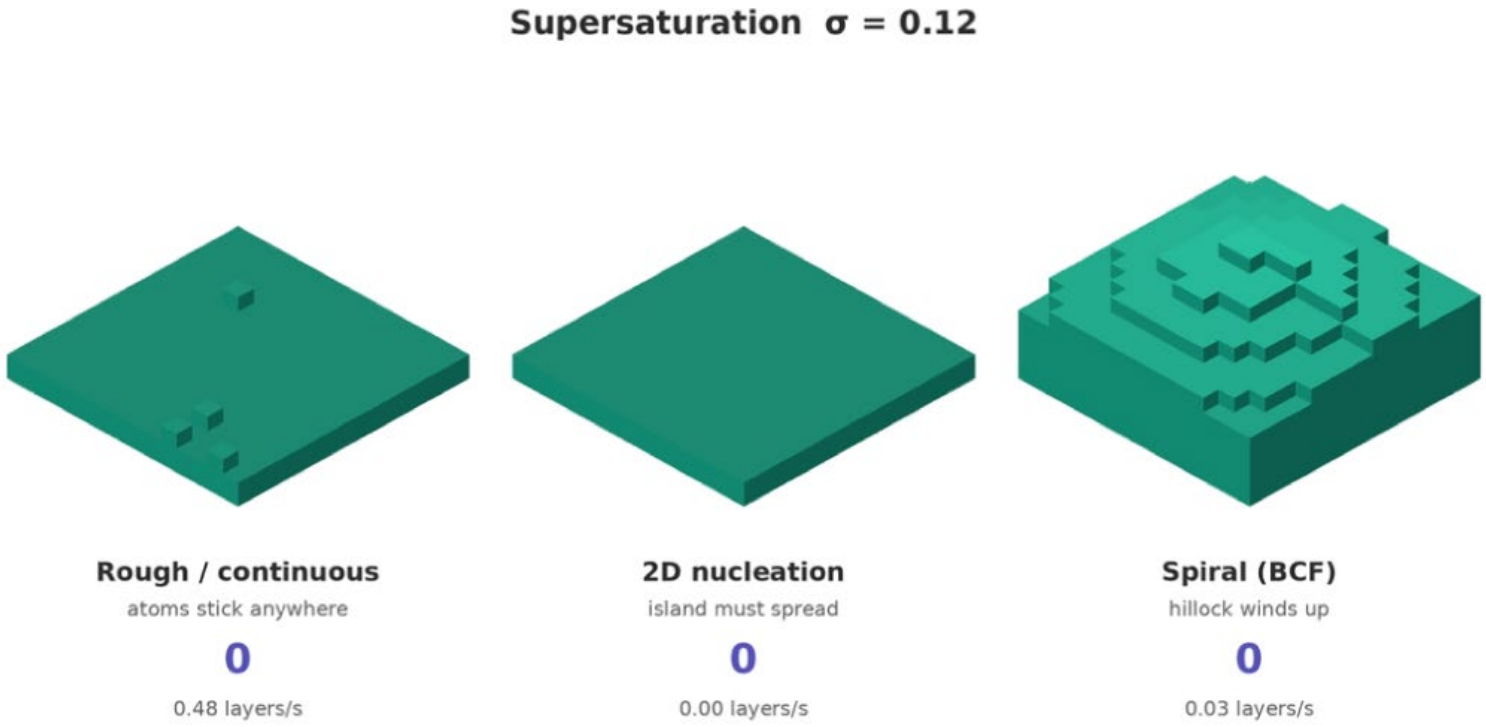
$\alpha \leq 2$: rough interface, continuous growth
$\alpha \geq 2$: smooth, faceted interface,
layer mechanisms necessary

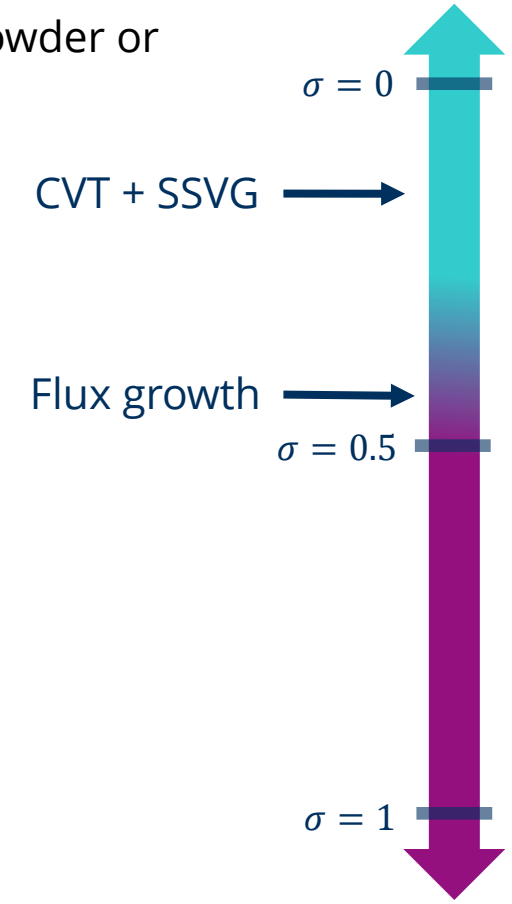
Crystal Growth Mechanisms: Are They Important?

Two factors to consider for the growth mechanisms:

Supersaturation σ: $\sigma = \frac{C - C_{sat}}{C_{sat}} = S - 1$

- **Low supersaturation:** small number of larger, high-quality crystals
- **High supersaturation:** fine powder or small crystals



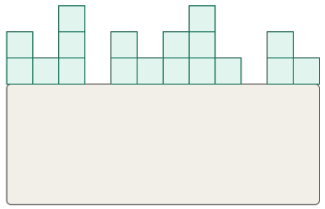


Crystal Growth Mechanisms: Are They Important?

σ = Supersaturation level

$\sigma = 0$ CVT + SSVG Flux growth $\sigma = 0.5$ $\sigma = 1$

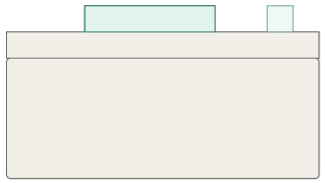
Rough / continuous



Atoms stick anywhere

$R \propto \sigma$

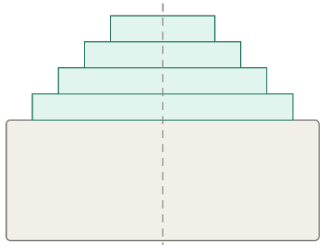
2D nucleation



Needs a new island

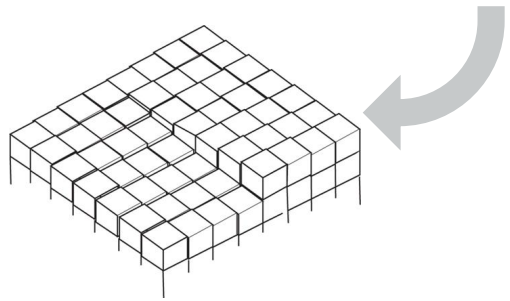
$R \propto \exp(-k/\sigma)$

Spiral (BCF)



Step never runs out

$R \propto \sigma^2$



Method	Phase	Interface	Dominant Mechanism	Typical σ
CVT	Vapour	Strongly faceted	Spiral, 2D nucleation at ↑ σ	Very low
PVT (seeded)	Vapour	Faceted, vicinal seed	Step flow on off-axis seed	Low-moderate
SSVG	Vapour	Strongly faceted	Spiral/layer-by-layer, near equilibrium	Very low
Flux	Solution	Faceted, weaker	Spiral & 2D nucleation common	Low, set by cooling rate

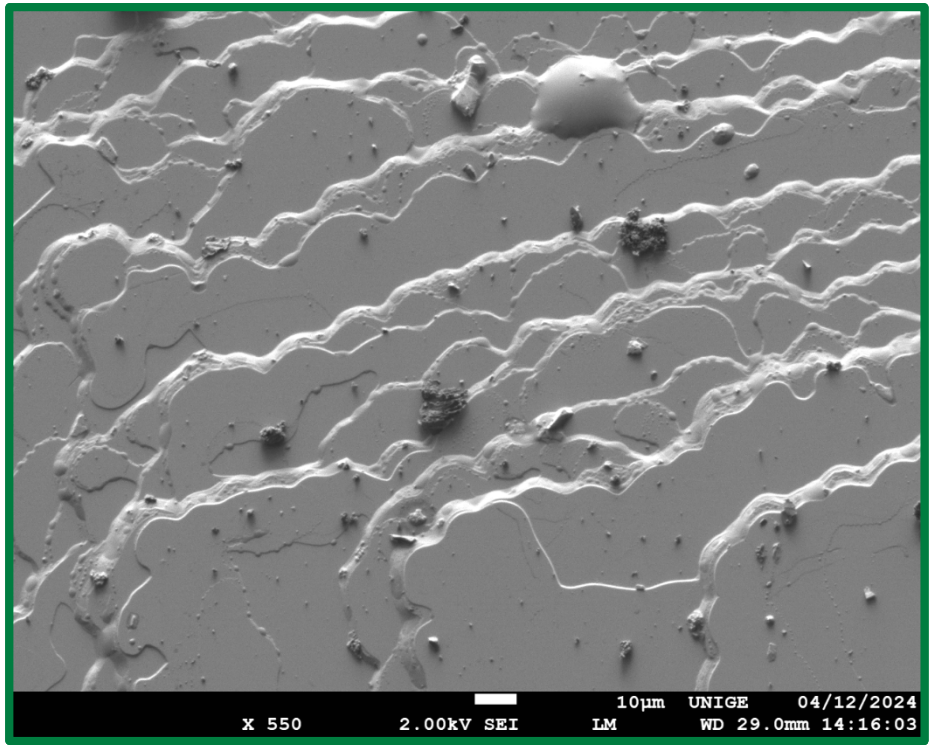
What type of surfaces are required?

Image of growth types generated with Claude (Anthropic)

What About the Reagents?

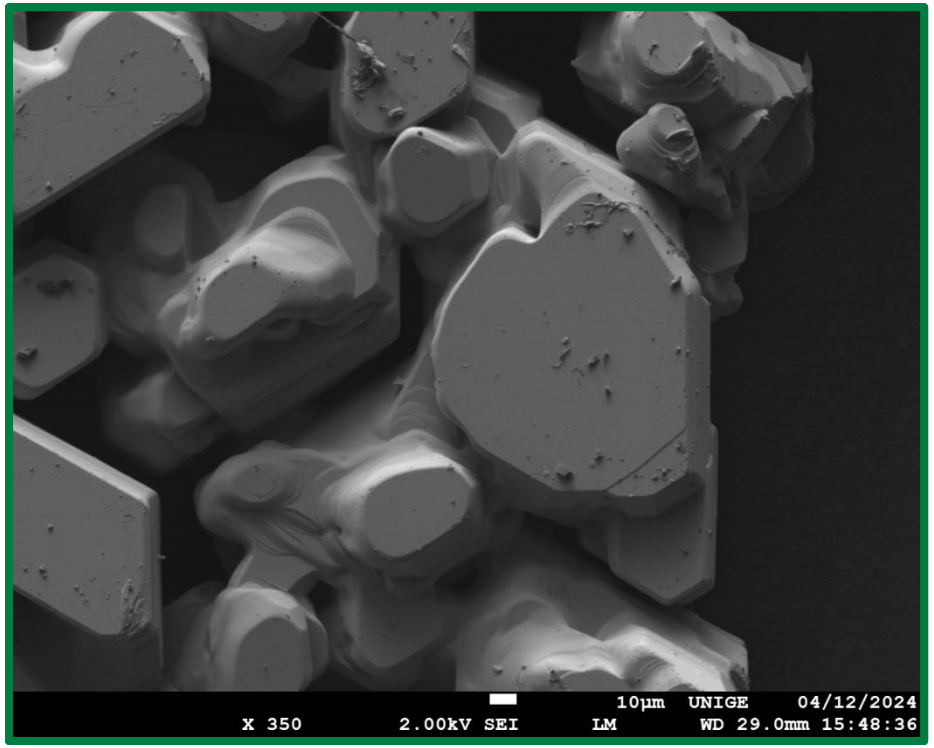
Starting from Elements:

$0.5\ V_{(s)} + 0.5\ Cr_{(s)} + 1.5\ I_{2(s)} \rightarrow V_{0.5}Cr_{0.5}I_{3(s)}$



Starting from Binary Precursors:

$0.5\ VI_{3(s)} + 0.5\ CrI_{3(s)} \rightarrow V_{0.5}Cr_{0.5}I_{3(s)}$



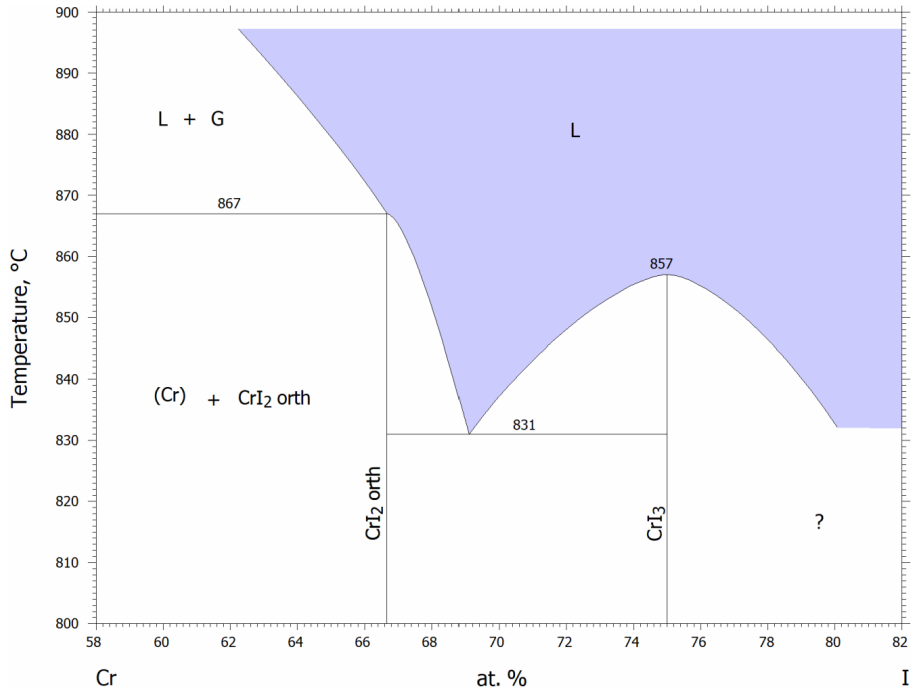
Choice of reagents affects crystal growth

Synthesis Design: Temperature Considerations

Temperature Choice

- No V–I phase diagram exists

Sink solid	*Transport additive*	*Temperature / °C*
VI_2	I_2	580 → 520
	I_2	500 → 400, 800 → 700
VI_3	I_2	400 → 320



→ In literature + decomposition onsets

- CrI_3: 720°C → 640°C; **~700°C:** $2\ CrI_{3(s)} \rightarrow 2CrI_{2(s)} + I_{2(g)}$
- VI_3: 400°C → 350°C; **~270°C:** $2\ VI_{3(s)} \rightleftharpoons VI_{2(s)} + VI_{4(g)}$

Synthesis Design: Temperature Considerations

Temperature Choice

- No V–I phase diagram exists
- Cr–I phase diagram: 857°C and 831°C

In literature + decomposition onsets

- CrI_3: 720°C → 640°C; **~700°C:** 2 $CrI_{3(s)} \rightarrow 2CrI_{2(s)} + I_{2(g)}$
- VI_3: 400°C → 350°C; **~270°C:** 2 $VI_{3(s)} \rightleftharpoons VI_{2(s)} + VI_{4(g)}$

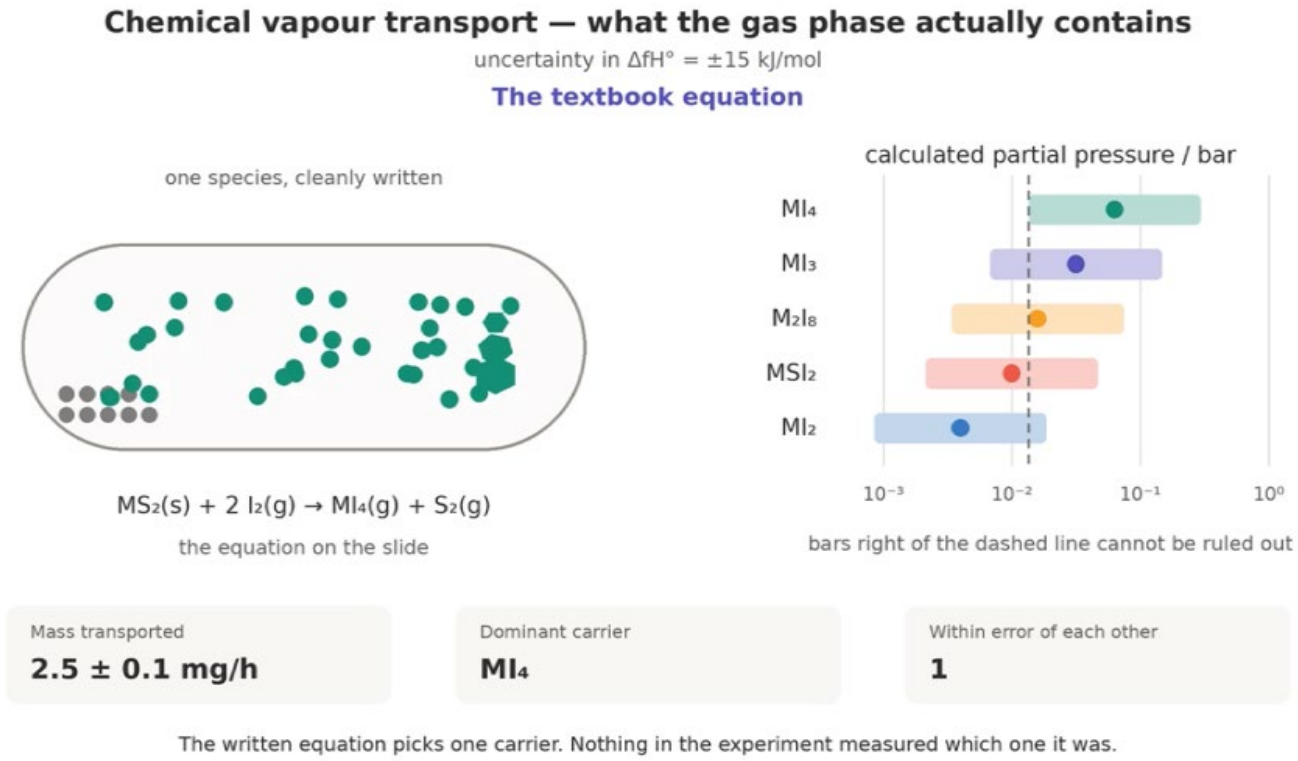
Phase interactions are complicated

- Phase diagrams have an “equilibrium assumption”
  - No heating/cooling rates
  - No information regarding time
- As soon as another component is introduced the formation and decomposition conditions change

Synthesis Design: Further Considerations

Temperature Choice

- No V–I phase diagram exists
- Cr–I phase diagram: 857°C and 831°C

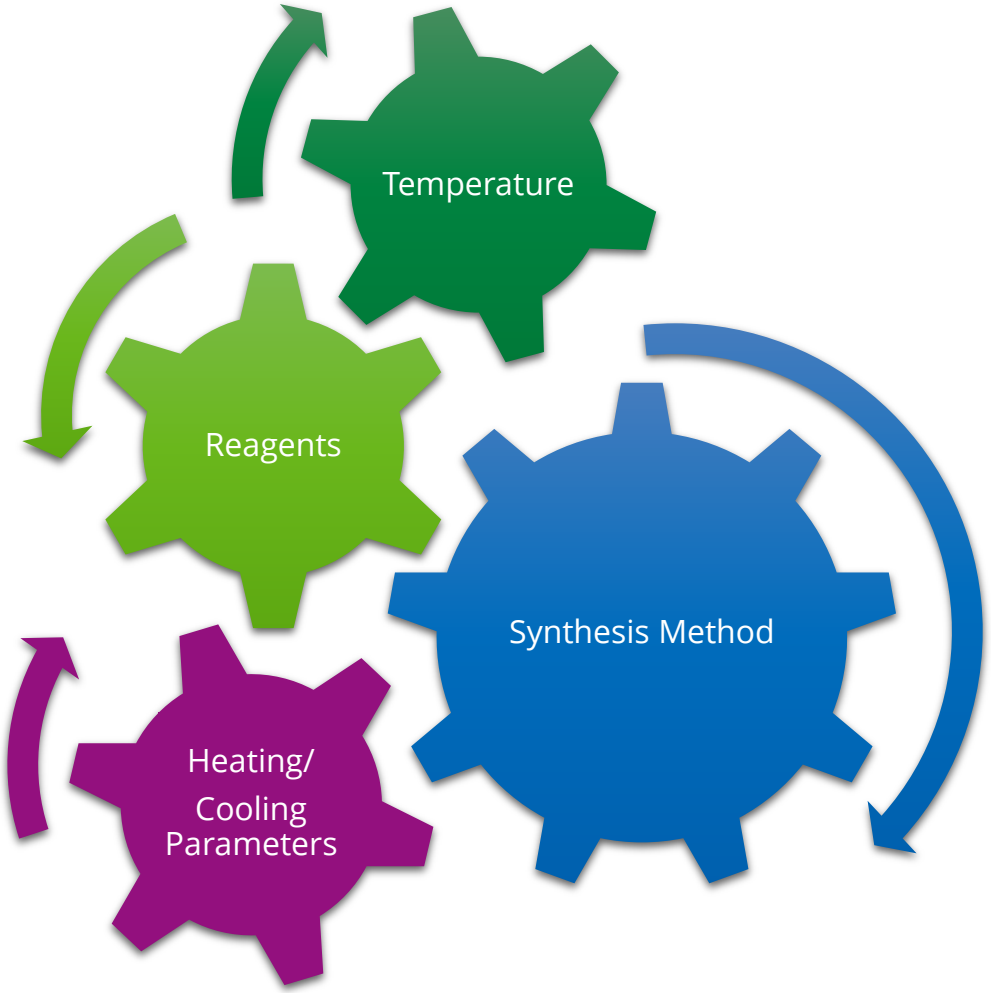


What happens in the transport?

- ➢ Uncertainties in the dominant transport mechanism
- ➢ Uncertainties regarding the solid-state reaction
  - Speciation in the melt
  - Diffusion pathways
  - Intermediates and reaction pathways

The exact reaction pathway is often unknown!

The Roadmap

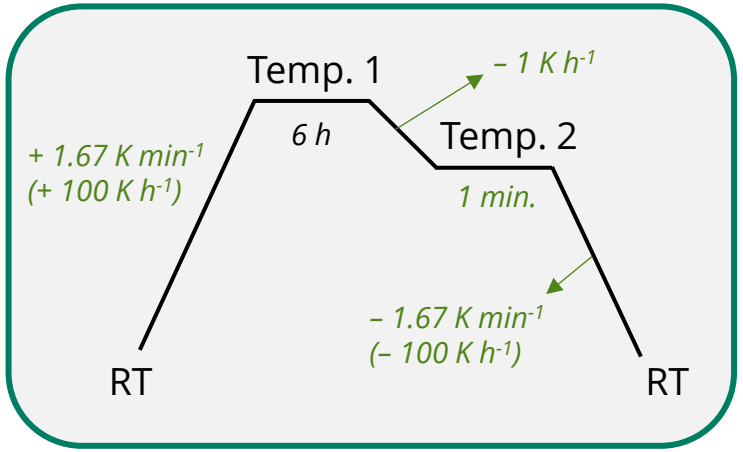


- Choice of synthetic method: self-selecting vapour growth
  - ✓ High-purity, no foreign transport agent
  - ✓ Large crystals possible, layer-by-layer growth
- Choice of reagents: elemental precursors
  - ✓ Favours layer-by-layer growth
- Choice of temperature profile: avoid preferred formation of VI_3 and CrI_3
  - ✓ Remain below 720 °C
  - ✓ Remain above 400 °C
- Heating and cooling rate: Need to be sensible, take vapour pressures into account
- Annealing times: Start small, see if compound forms, then adjust

= An iterative trial-and-error process

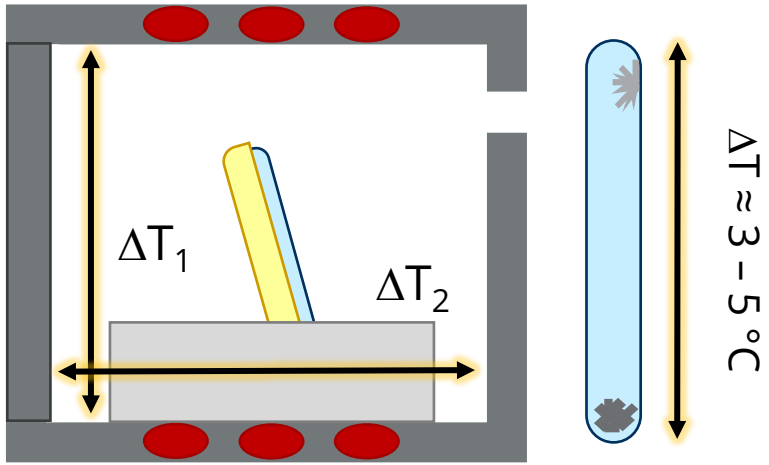
Challenge 1 Solution: Self-Selecting Vapour Growth

- End member syntheses (CVT):
  - CrI_3: 720°C → 640°C
  - VI_3: 400°C → 350°C
- $V_{1-x}Cr_xI_3$ synthesis: self-selecting vapour growth (SSVG)[9]
  - **All substitutions stable across series, $0 \leq x \leq 1$**
  - **Up to mm-sized crystals, homogeneous (per EDX)**



Rotate by 90 °

Inside



[9] A. Lukovkina *et al.*, *Chem. Mat.* **2024**, *12*, 36.

Challenge 1 Solution: Self-Selecting Vapour Growth

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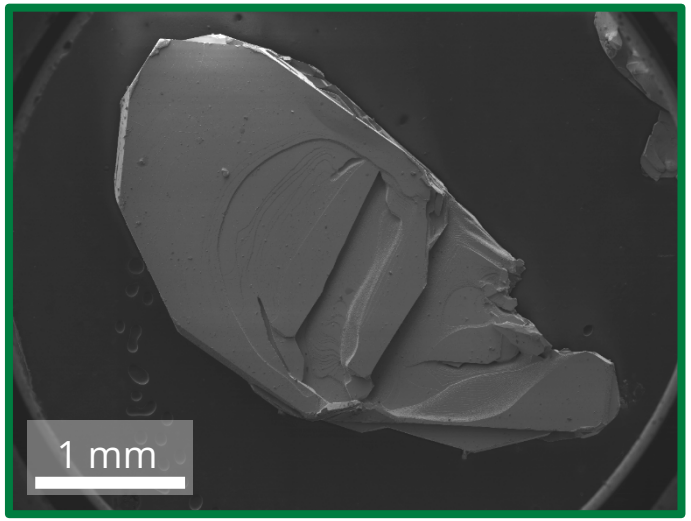
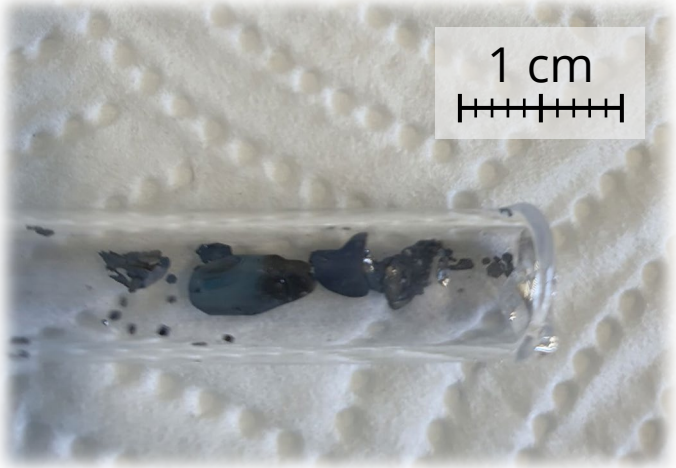
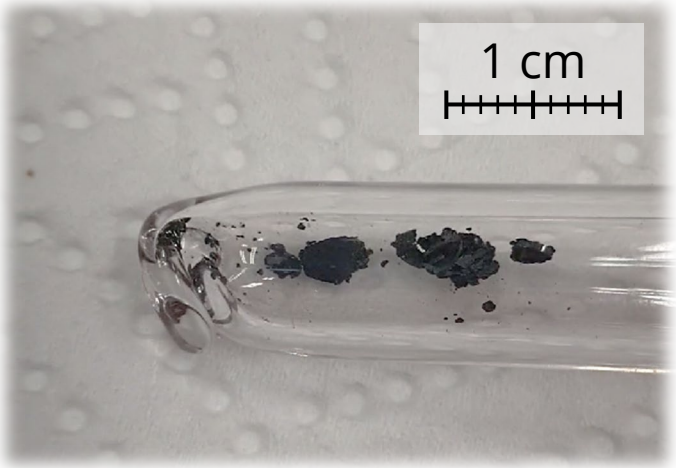
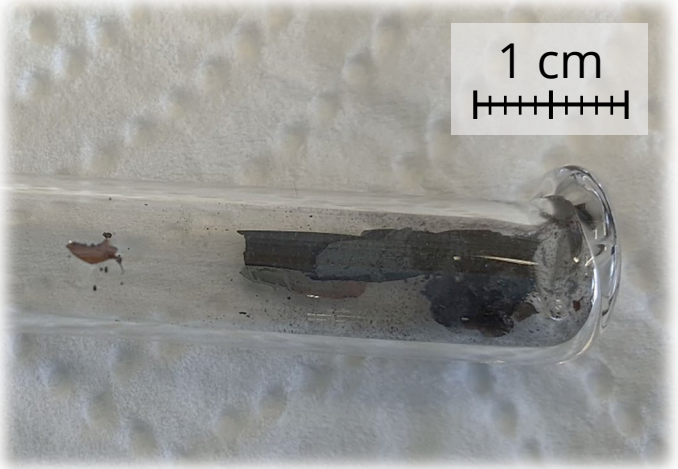




$V_{0.2}Cr_{0.8}I_3$



$V_{0.5}Cr_{0.5}I_3$

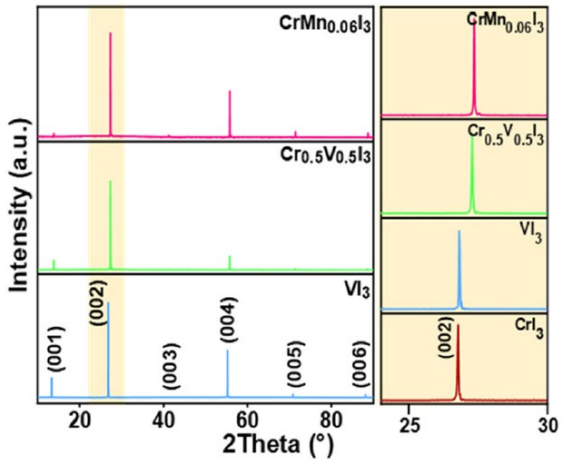


$V_{0.9}Cr_{0.1}I_3$

[9] A. Lukovkina *et al.*, *Chem. Mat.* **2024**, *12*, 36.

Challenge 2: Phase Identification

Phase Identification



Powder X-ray Diffraction:
Highly oriented samples

→ Uncertain identification

Challenges:

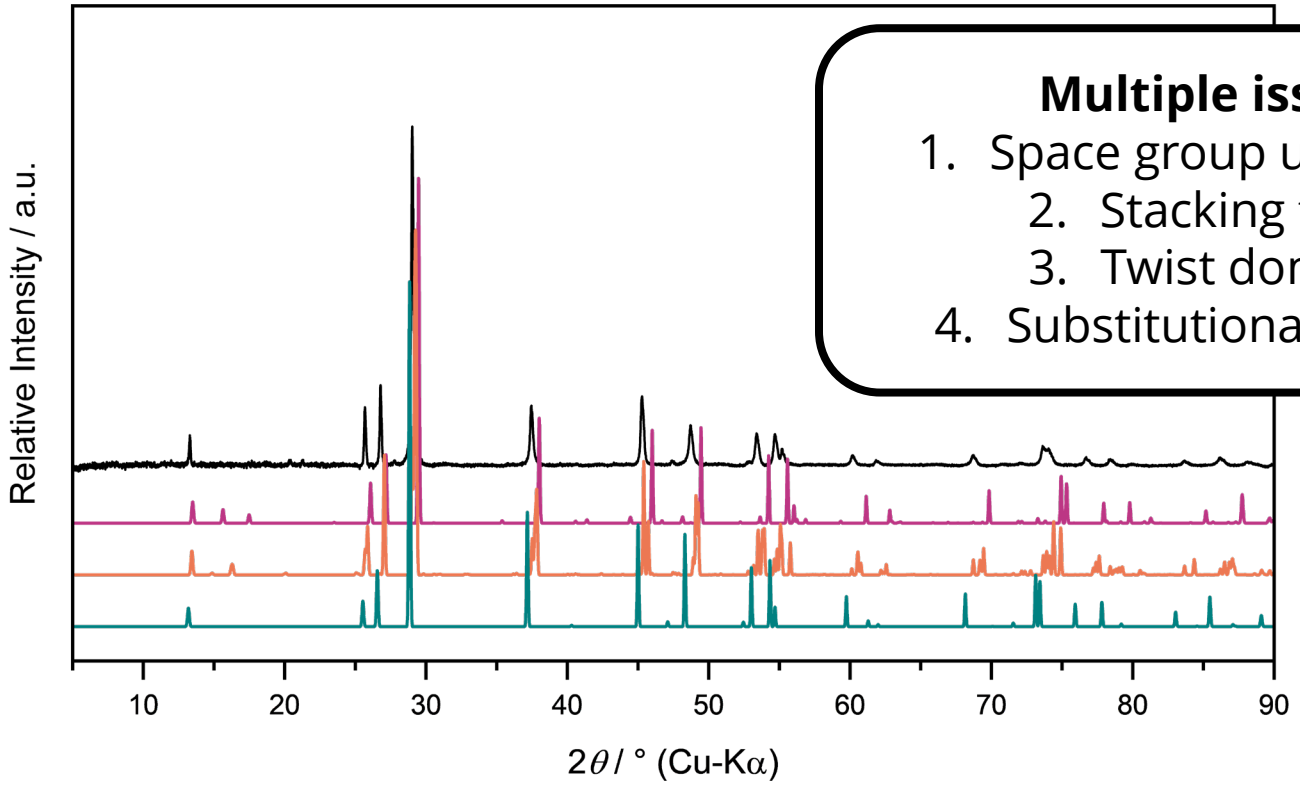
- Only highly-oriented patterns are shown in other study:
  Finer differences cannot be ascertained
  - ➢ Disorder (stacking and otherwise)
  - ➢ Polytypes – this information can also get lost
- Complementary elemental analysis techniques often missing
- No single-crystal data to confirm the structure

Solution?

- Sample identification not hampered by orientation
  - ➢ Type of powder X-ray diffraction (PXRD) measurement:
    Debye-Scherrer in capillary set-up
- Use of complementary techniques

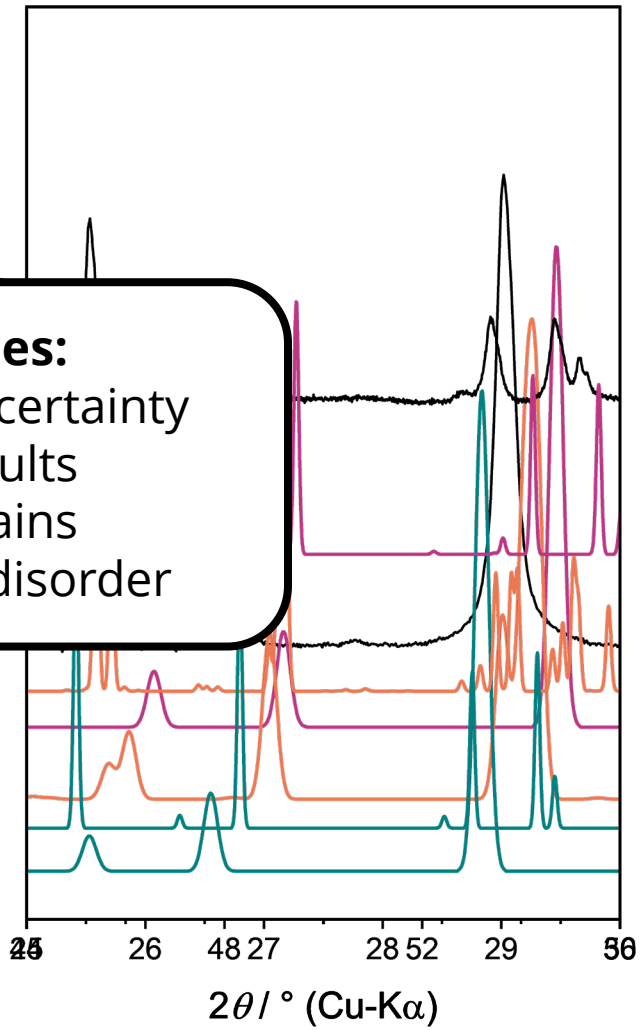
Challenges All Around: Which Phase Is It?

- Disagreement in the literature for VI_3, which space group?
  $R\bar{3} : C2/c : P\bar{3}m1 = 5 : 1 : 1$
- CrI_3 issue: Stacking faults and 120° twist domains[10]
  → broadening of, and shift in, identifiable peaks

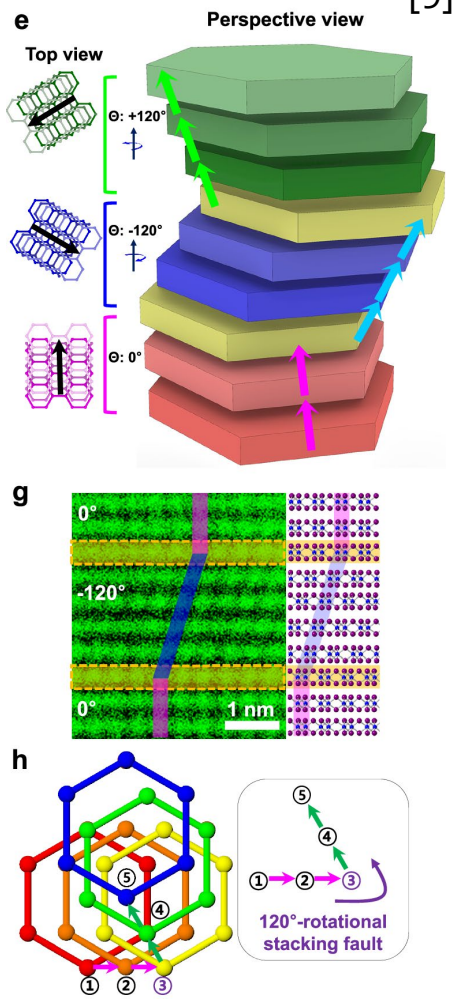


Multiple issues:

1. Space group uncertainty
2. Stacking faults
3. Twist domains
4. Substitutional disorder

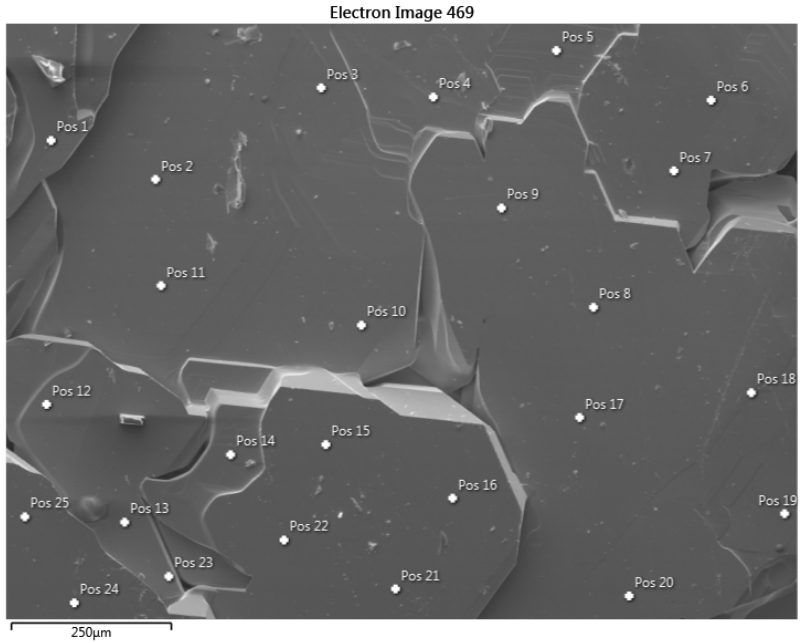


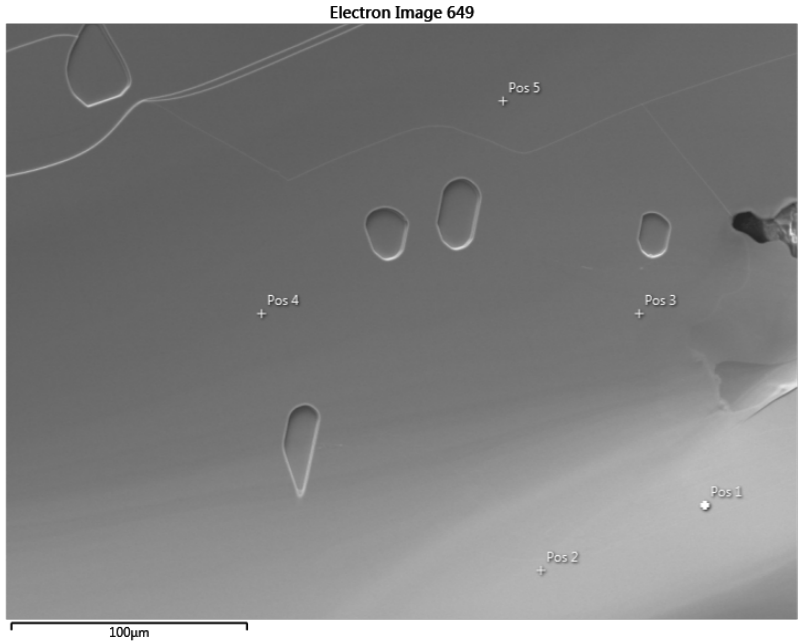
[9]

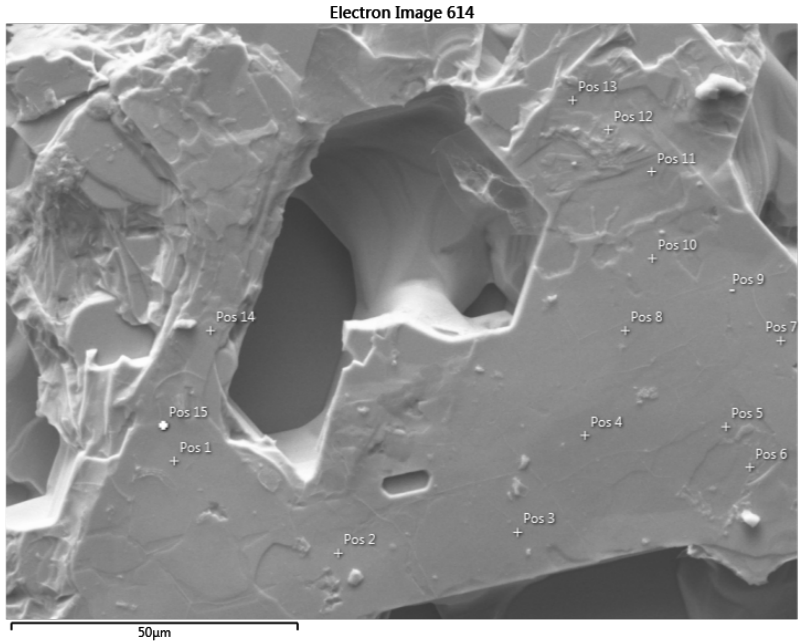


[10] M. Jang *et al.*, *Nat Commun* **15**, 5925 (2024).

Confirming Composition with EDX







Statistics	V	Cr	I
Max	3.54	22.59	74.87
Min	2.90	21.80	74.12
Avg.	3.20	22.23	74.54
σ	0.15	0.22	0.19

$\approx V_{0.1}Cr_{0.9}I_3$

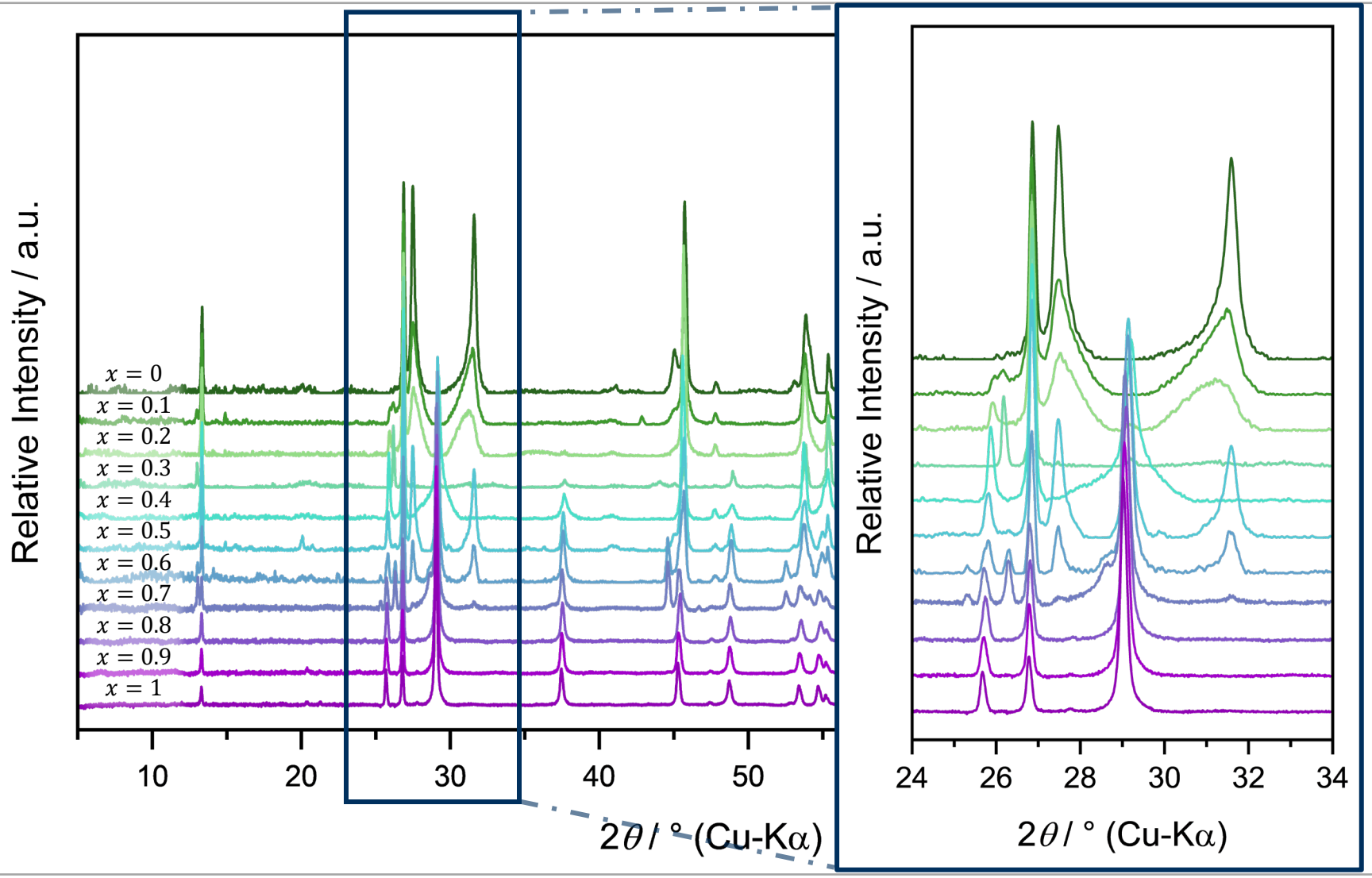
Statistics	V	Cr	I
Max	12.28	15.13	74.34
Min	10.70	14.04	73.56
Avg.	11.33	14.73	73.94
σ	0.61	0.42	0.33

$\approx V_{0.45}Cr_{0.55}I_3$

Statistics	V	Cr	I
Max	17.28	8.95	74.70
Min	16.59	8.38	73.96
Avg.	16.96	8.54	74.46
σ	0.19	0.17	0.23

$\approx V_{0.7}Cr_{0.3}I_3$

Stacking Changes Across the Series



No phase separation

Lattice parameters follow Vegard's law

Structure scheme:

$x \leq 0.2/0.3 : C2/m$

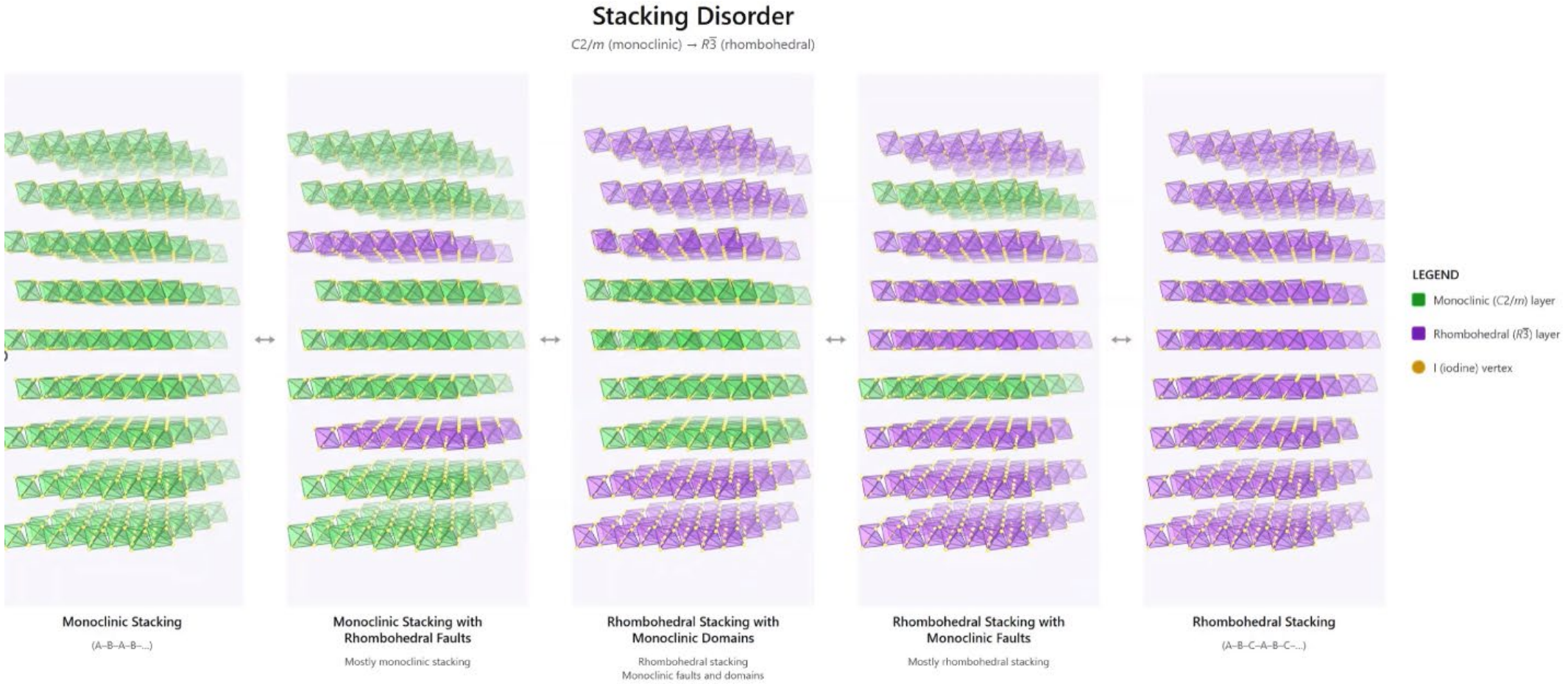
$x \geq 0.3 : R\bar{3}$

Issues with stacking/ disorder:

1. Broader and asymmetric peaks
2. Shifting peaks

What Is Stacking Disorder?

What does this disorder actually mean with respect to the structure?



Animation generated with Claude (Anthropic)

Are There Further Issues?

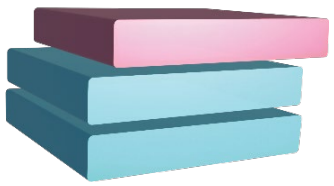
To ensure that there are no further issues – check single crystals and generate the precession images.

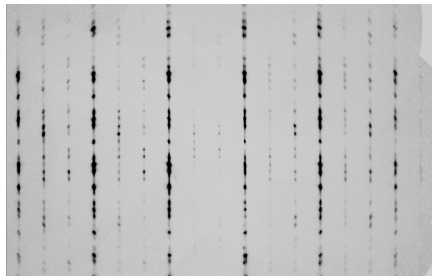
➢ What are the possible issues?

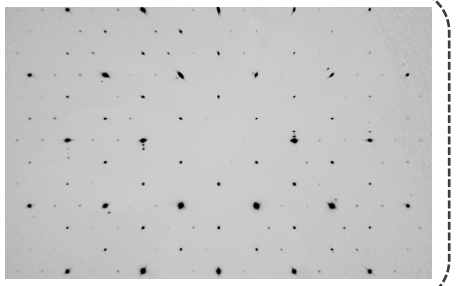
Viewing direction

a*/b* [*hnl*] **c*** [*hk*0]

Stacking Disorder



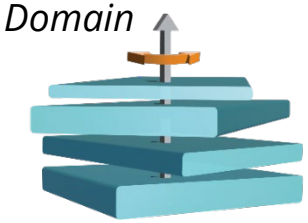


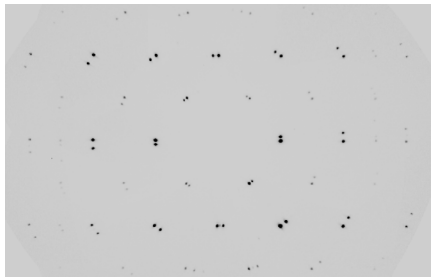


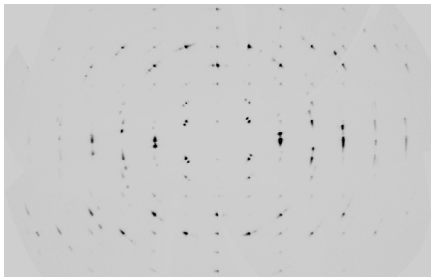
Viewing direction

a*/b* [*hnl*] **c*** [*hk*0]

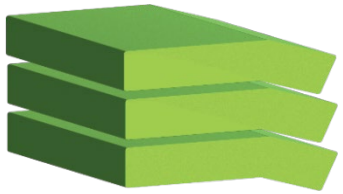
'Multi'-Crystal/ Domain

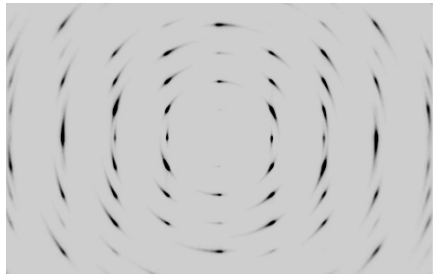


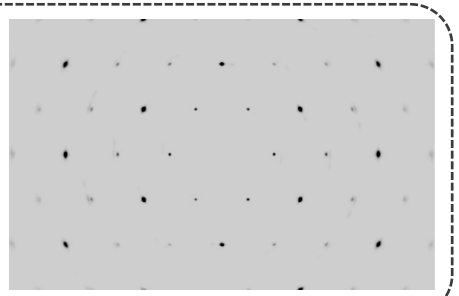




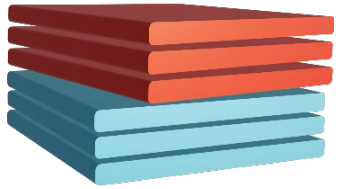
'Bent' Crystals

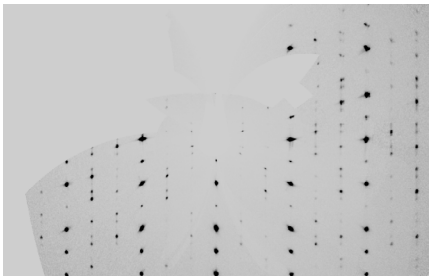


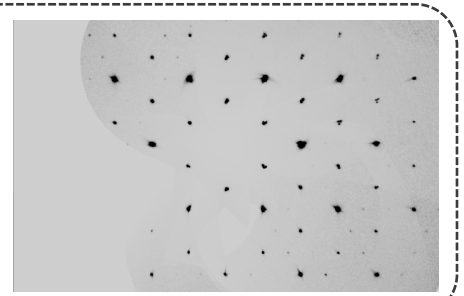




Reverse/Obverse Twinning





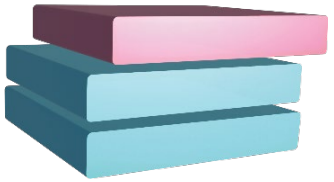


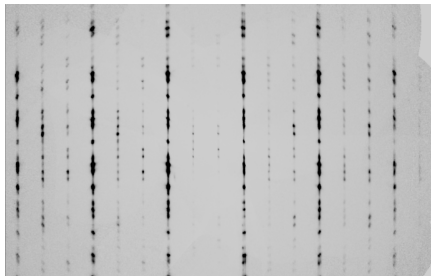
Confirmation of Disorder

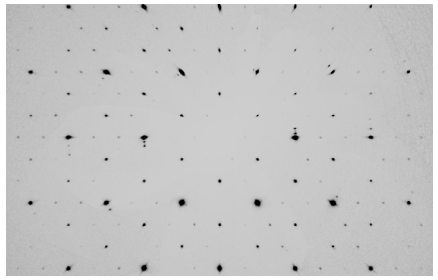
Viewing direction

a*/b* [*hnl*] **c*** [*hk*0]

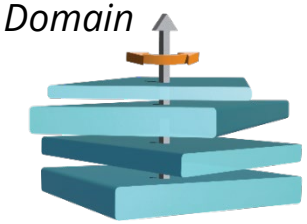
Stacking Disorder

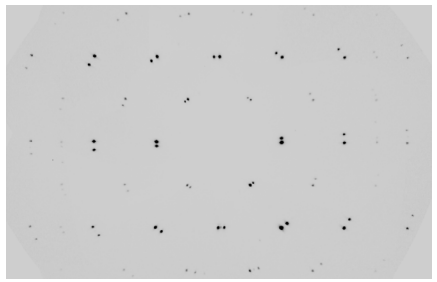


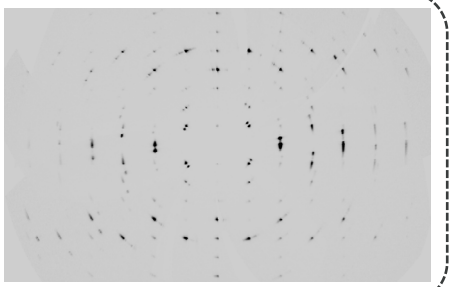




'Multi'-Crystal/ Domain

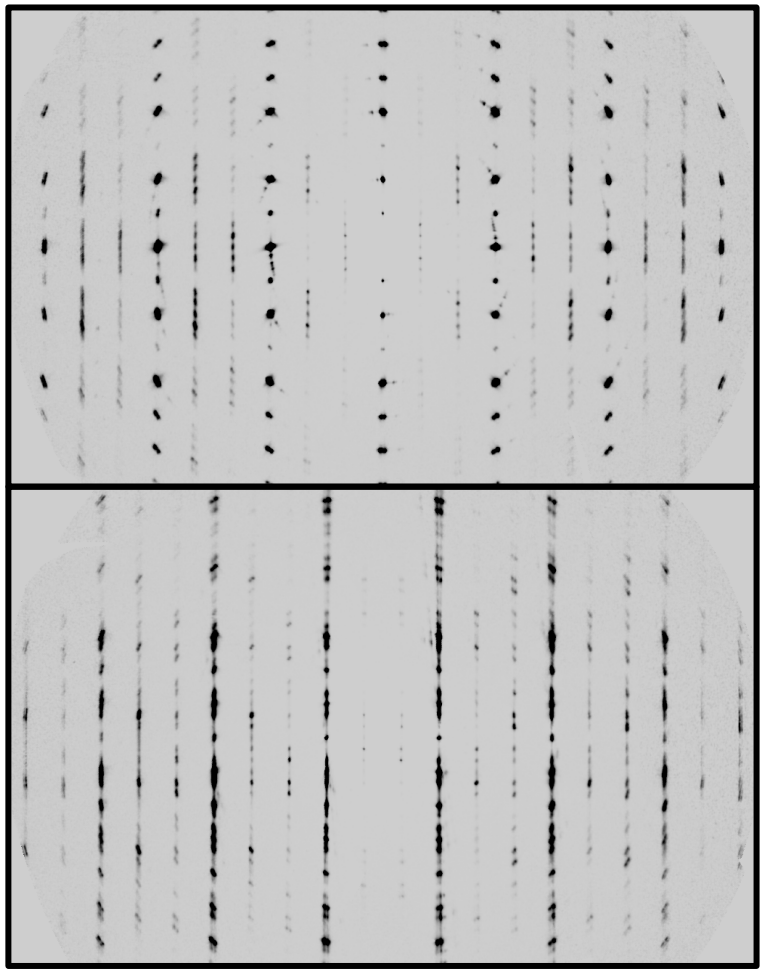






Stacking and domain disorder present

$V_{0.1}Cr_{0.9}I_3$



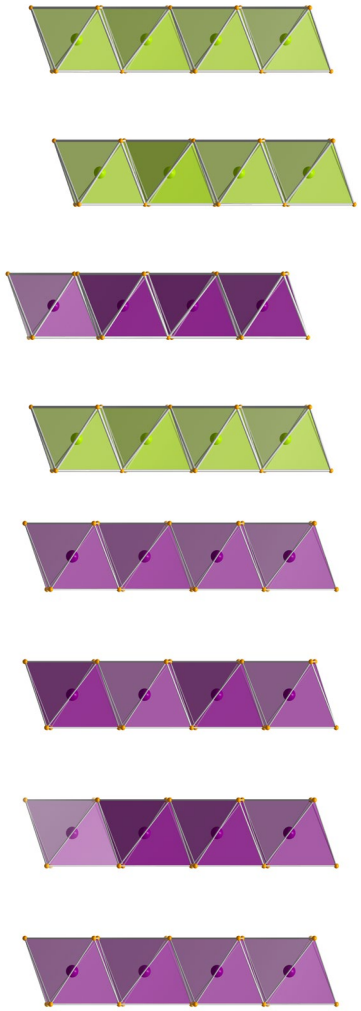
Challenge 2 Solution: Stacking Faults Solved with DIFFaX

Composition	*x*	Dominant Stacking	Mono./Rh. Stacking Disorder	Coherent Mono./Rh. Domains
CrI_3	0	Monoclinic	Rh.: 3.6 %	–
$V_{0.1}Cr_{0.9}I_3$	0.1	Monoclinic	Rh.: 8 %	–
$V_{0.2}Cr_{0.8}I_3$	0.2	Monoclinic	Rh.: 12 %	Rh.: 6 %
$V_{0.3}Cr_{0.7}I_3$*	0.3	Rhombohedral	0	–
$V_{0.4}Cr_{0.6}I_3$	0.4	Rhombohedral	Mono.: 12 %	–
$V_{0.5}Cr_{0.5}I_3$	0.5	Rhombohedral	Mono.: 4%	Mono.: 17%, Rh.: 79%
$V_{0.6}Cr_{0.4}I_3$	0.6	Rhombohedral	Mono.: ~2%	Mono.: 15%, Rh.: 36%
$V_{0.7}Cr_{0.3}I_3$	0.7	Rhombohedral	Mono.: ~2% Obv./Rev.: ~2%	Mono.: 15%, Rh.: 37%
$V_{0.8}Cr_{0.2}I_3$	0.8	Rhombohedral	Obv./Rev.: 1-2%	–
$V_{0.9}Cr_{0.1}I_3$	0.9	Rhombohedral	Obv./Rev.: 1-2%	–
VI_3	1	Rhombohedral	0	–

Monoclinic with R-faults

Rhombohedral with M-faults

Rhombohedral



* Strong preferred orientation in measurements.
Mono. = monoclinic, Rh. = rhombohedral

Challenge 2 Solution: Stacking Faults Solved with DIFFaX

Composition	*x*	Dominant Stacking	Mono./Rh. Stacking Disorder	Coherent Mono./Rh. Domains
CrI_3	0	Monoclinic	Rh.: 3.6 %	–
$V_{0.1}Cr_{0.9}I_3$	0.1	Monoclinic	Rh.: 8 %	–
$V_{0.2}Cr_{0.8}I_3$	0.2	Monoclinic		
$V_{0.3}Cr_{0.7}I_3$*	0.3	Rhombohedral		
$V_{0.4}Cr_{0.6}I_3$	0.4	Rhombohedral		
$V_{0.5}Cr_{0.5}I_3$	0.5	Rhombohedral		
$V_{0.6}Cr_{0.4}I_3$	0.6	Rhombohedral		
$V_{0.7}Cr_{0.3}I_3$	0.7	Rhombohedral	Obv./Rev.: ~2%	
$V_{0.8}Cr_{0.2}I_3$	0.8	Rhombohedral	Obv./Rev.: 1-2%	–
$V_{0.9}Cr_{0.1}I_3$	0.9	Rhombohedral	Obv./Rev.: 1-2%	–
VI_3	1	Rhombohedral	0	–

Three structural regimes exist

Dominant stackings:

$x \leq 0.2/0.3 : C2/m$

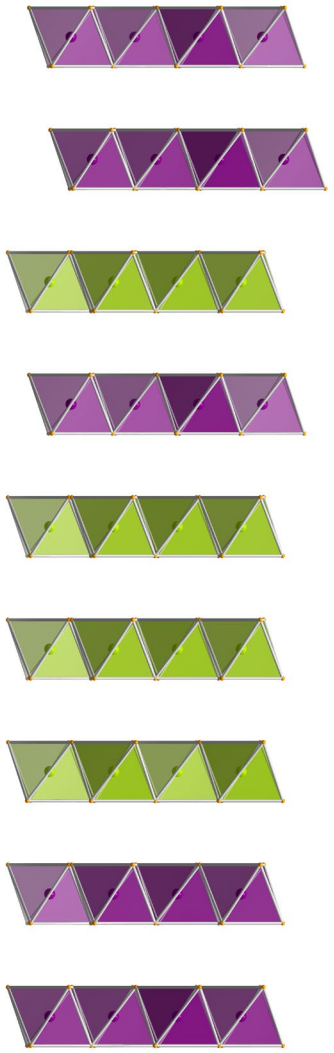
$x \geq 0.3 : R\bar{3}$

→ **Do the structural disorder and magnetic properties correlate?**

Monoclinic with R-faults

ohedral -faults

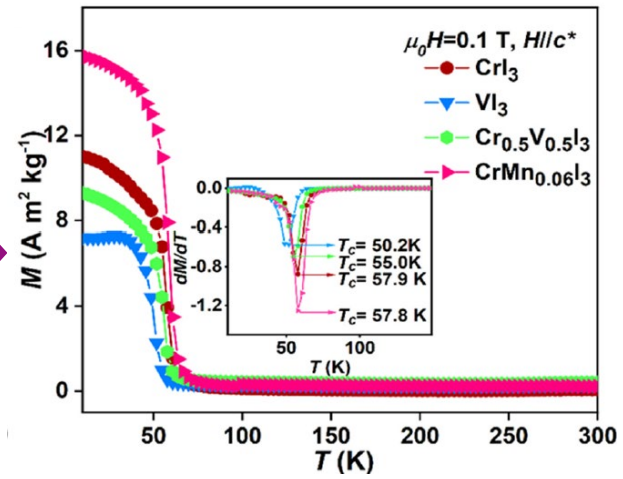
Rhombohedral



* Strong preferred orientation in measurements.
Mono. = monoclinic, Rh. = rhombohedral

Challenge Number 3: Inconclusive Magnetic Analysis

Inconclusive Magnetism



Ferromagnetic behaviour

Inconclusive regarding the exact dynamics

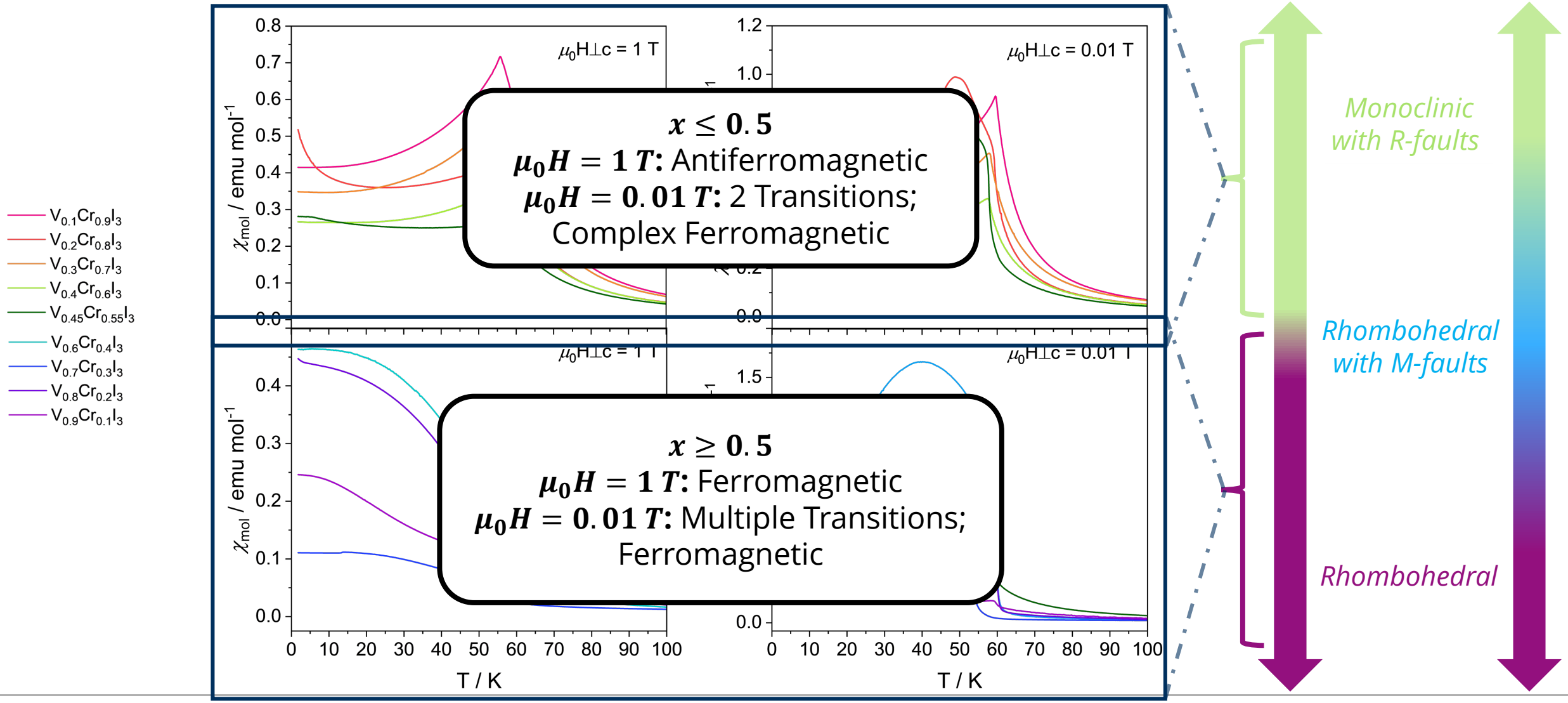
Challenges:

- Uncertain structure determination = wrong assignment
- Limited magnitude of externally applied fields shown/tested:
  - Specifically low-field data is often missing → Finer details lost
  - Global vs. local behaviour: no insights
- Measurements only showcase static behaviour → What about dynamics?

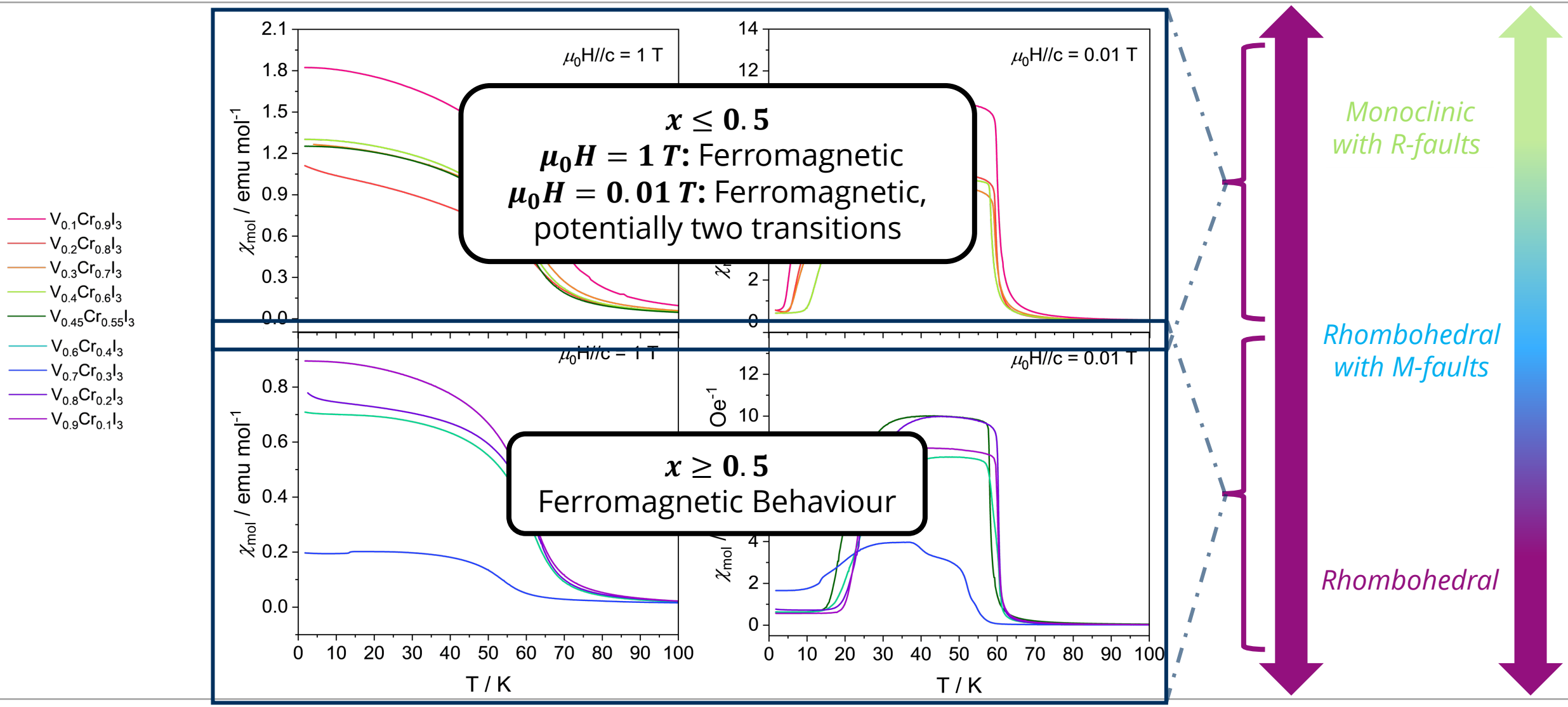
Solution?

- Measure across a range of fields
  - Hopefully see a correlation between structure disorder and properties
- Employ alternating current magnetic susceptibility (ACMS) to gain an insight into spin dynamics

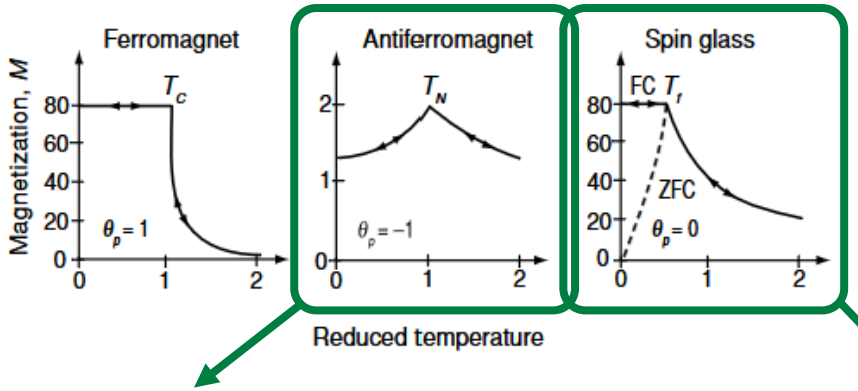
Magnetic Properties $\mu_0 H = 0.1\ T$ and $1\ T \perp c$

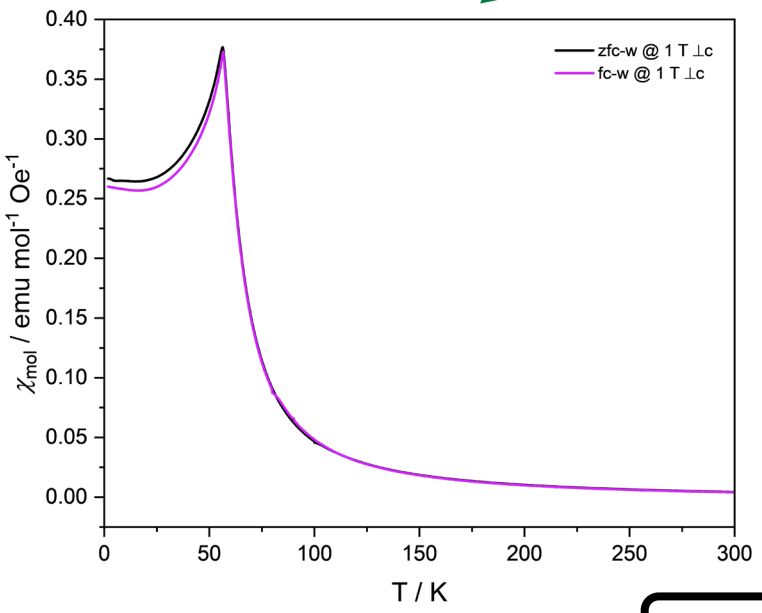


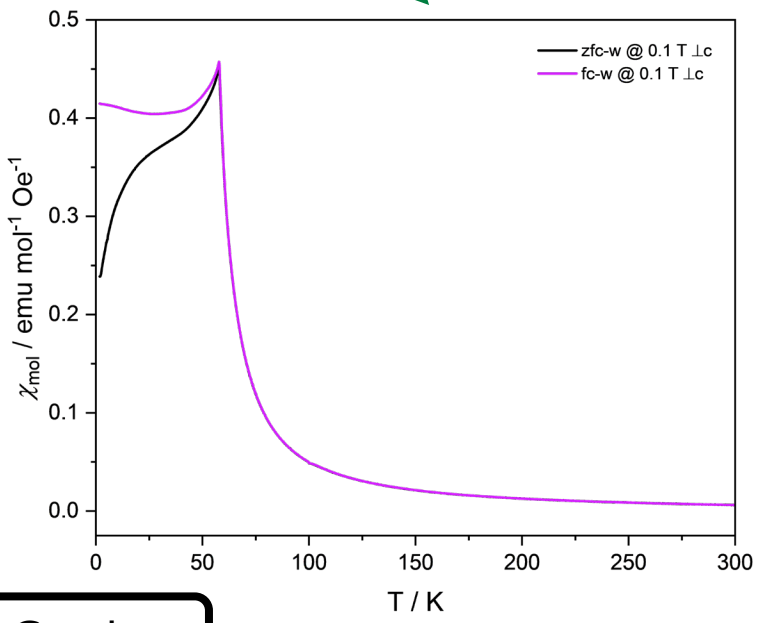
Magnetic Properties $\mu_0 H = 0.1\ T$ and $1\ T \parallel c$



Applied External Field → Unclear Magnetic State







$V_{0.4}Cr_{0.6}I_3$

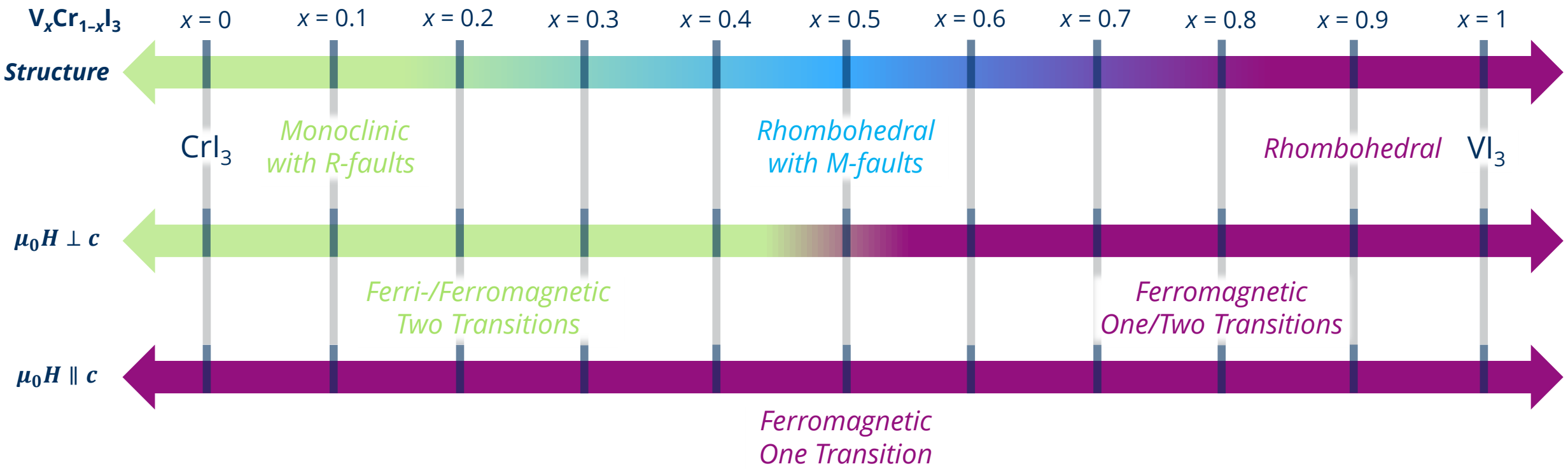
In VSM measurements:

- High field measurements indicate AFM long-range ordering
- Lower field measurements indicate potential spin glass (or similarly complicated) state

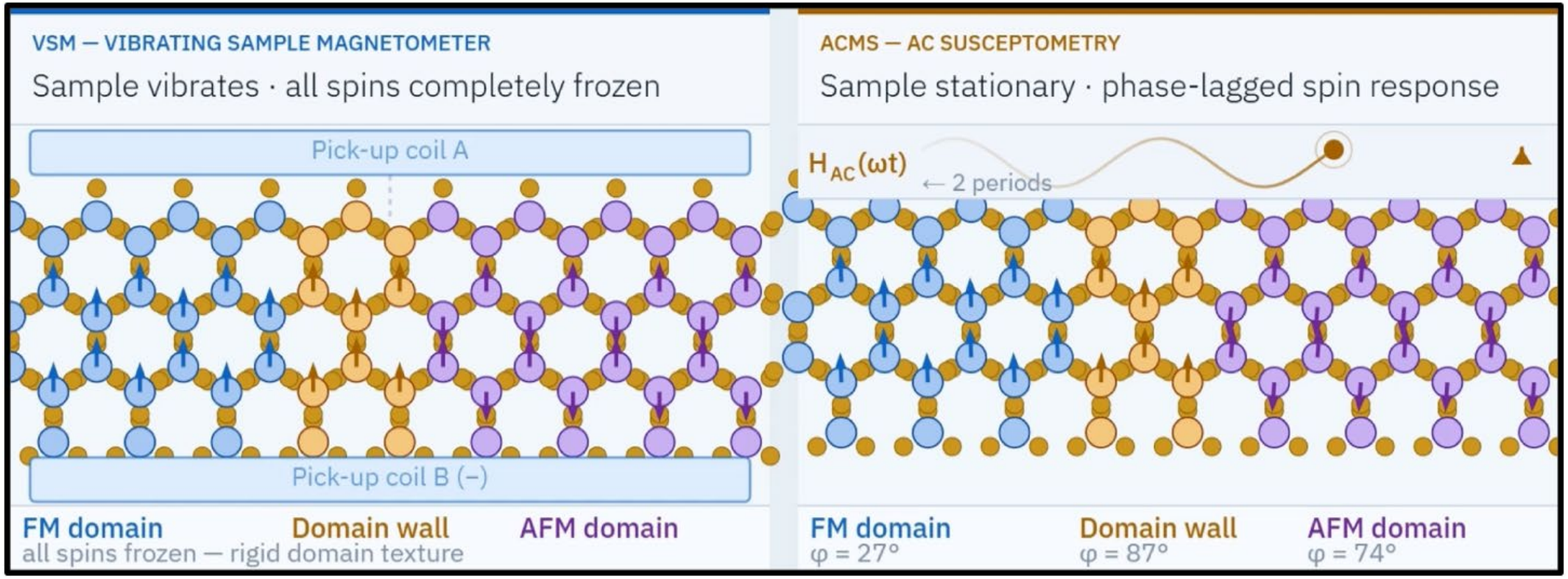
→ Not enough information for a clear determination

Magnetic Properties

- Complex dynamic magnetic behaviour at low fields (< 0.1 T)
- **No correlation with structural regimes:**



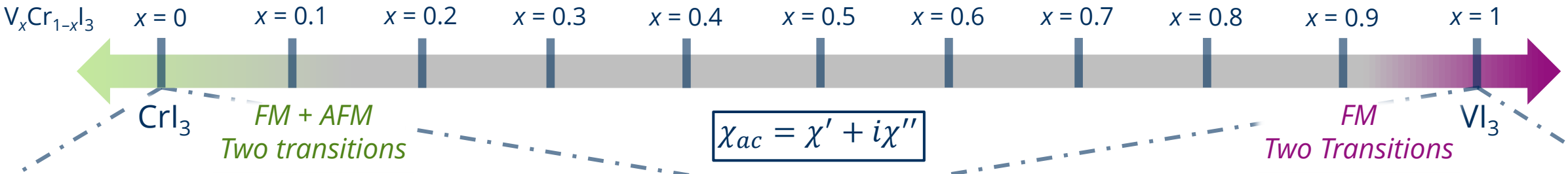
Solution Regarding Dynamics: ACMS



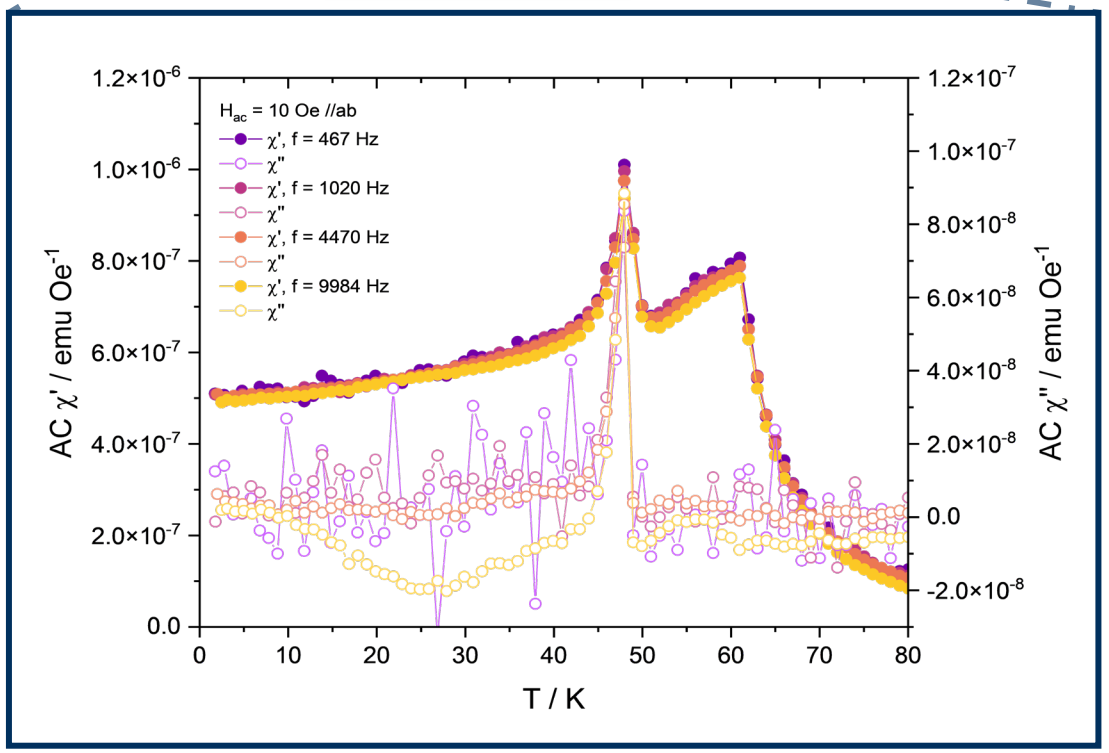
VSM = average spin behaviour

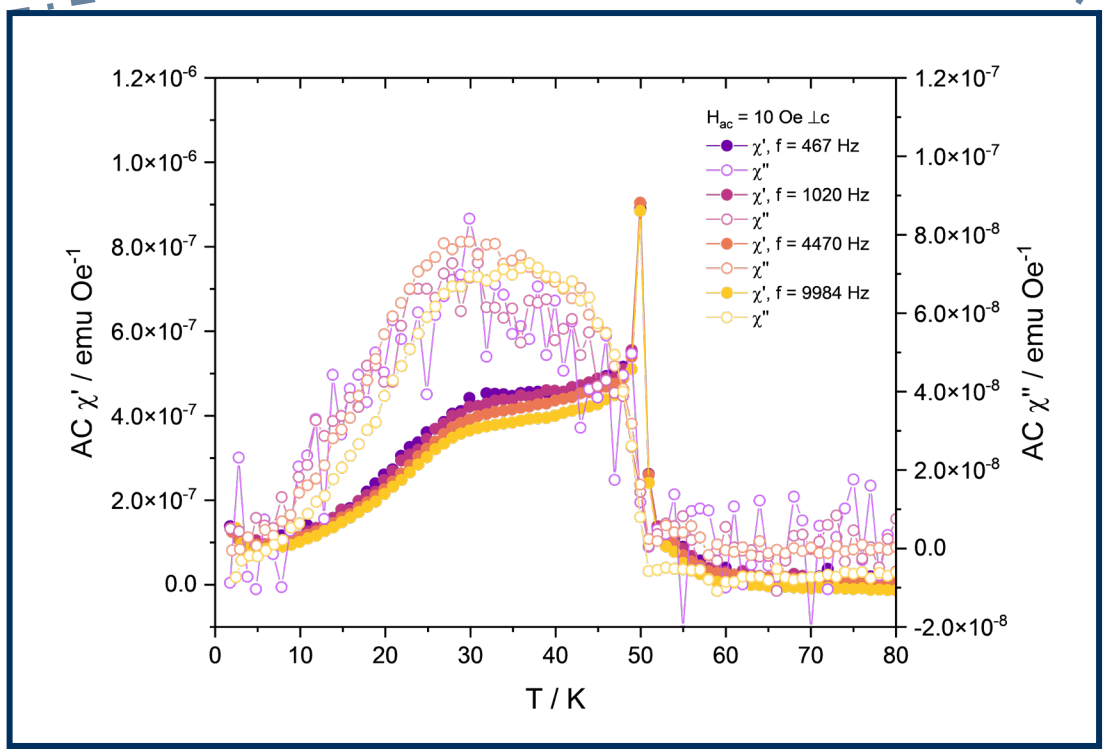
ACMS = speed of change and whether spins move coherently or at a spread of rates

AC Susceptibility $\perp c$ (In-plane)

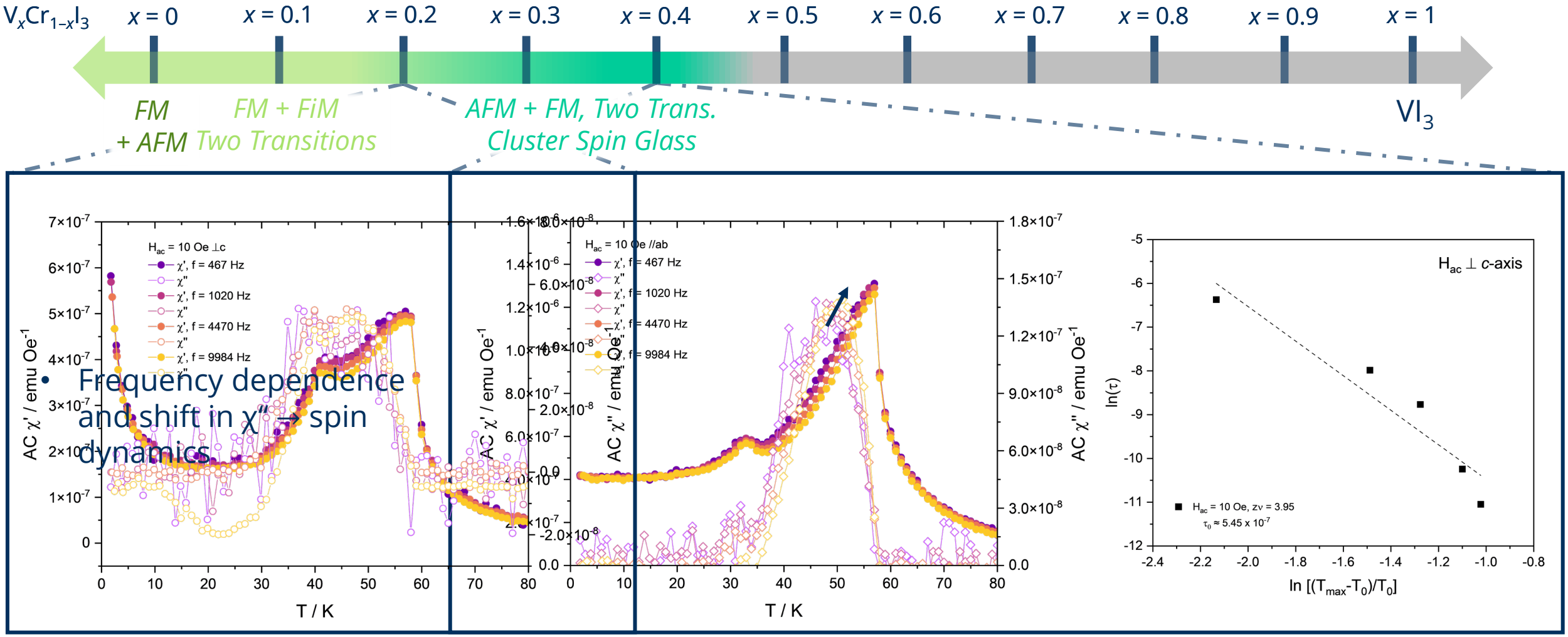


$$\chi_{ac} = \chi' + i\chi''$$

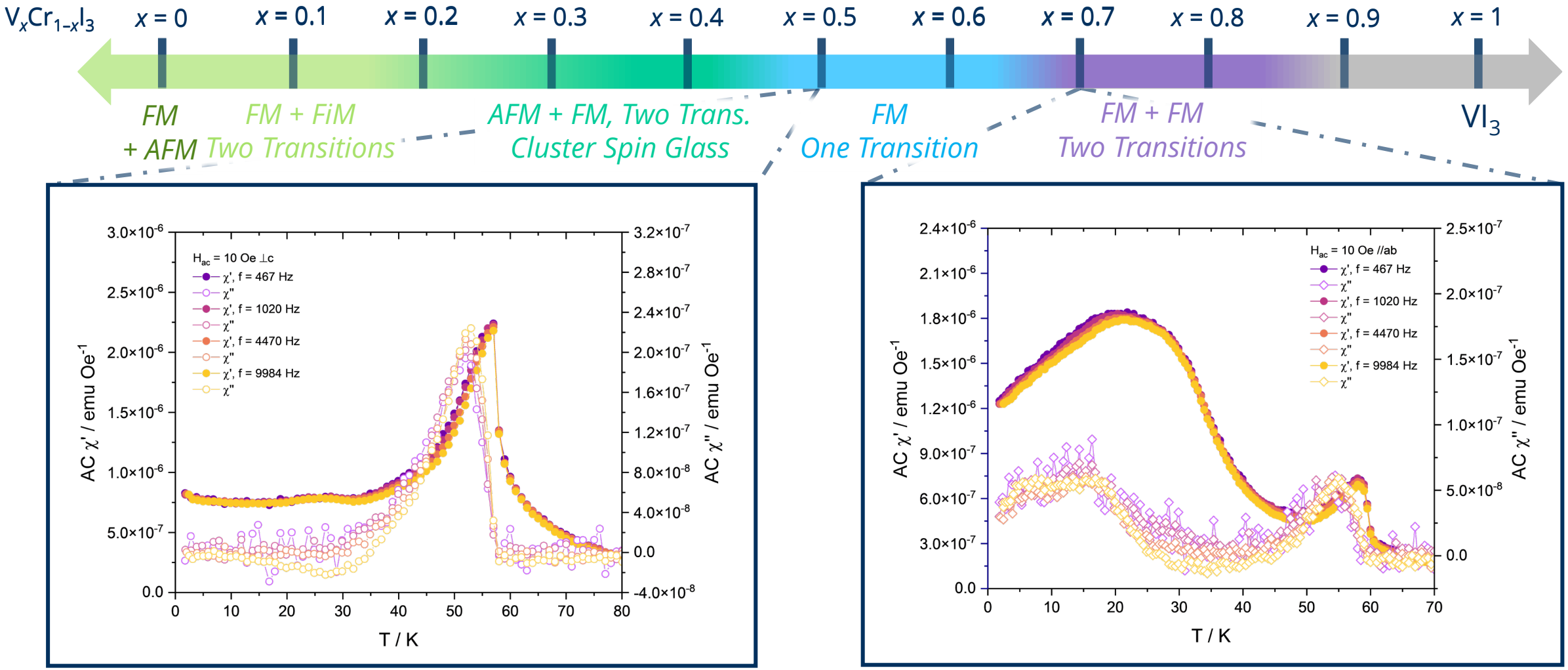




AC Susceptibility ⊥ c (In-plane)



AC Susceptibility ⊥ c (In-plane)



AC Susceptibility ⊥ c (In-plane)

$V_xCr_{1-x}I_3$ $x = 0$ $x = 0.1$ $x = 0.2$ $x = 0.3$ $x = 0.4$ $x = 0.5$ $x = 0.6$ $x = 0.7$ $x = 0.8$ $x = 0.9$ $x = 1$

FM + AFM

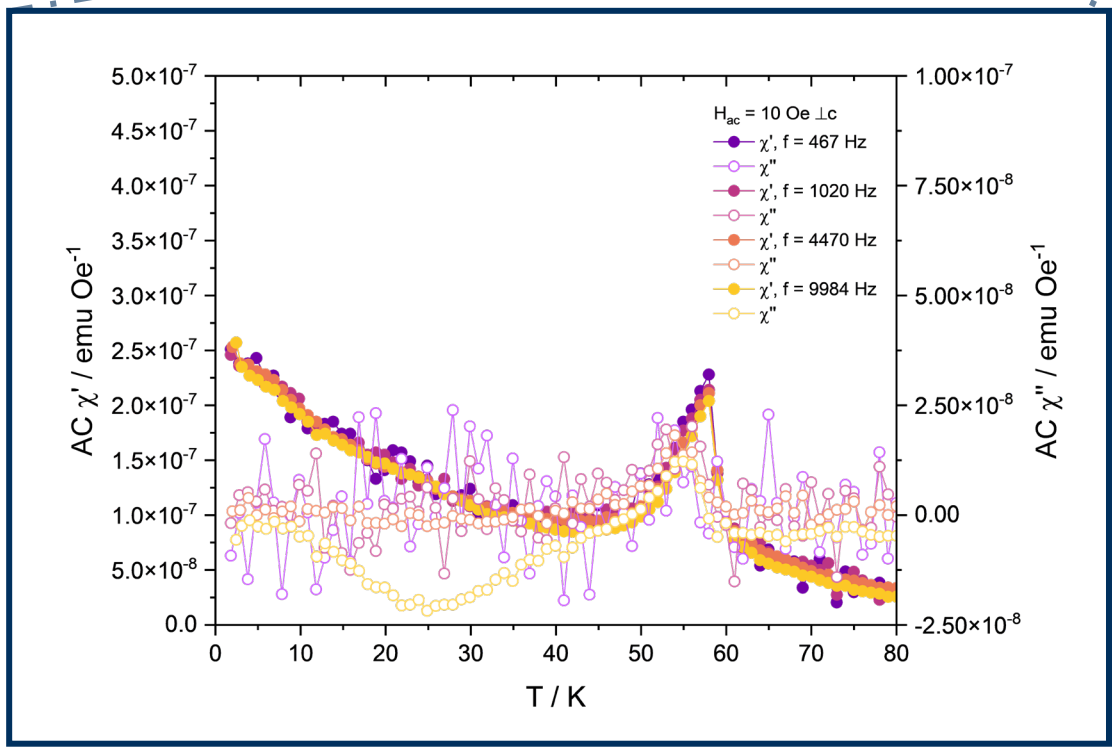
FM + FiM Two Transitions

AFM + FM, Two Trans. Cluster Spin Glass

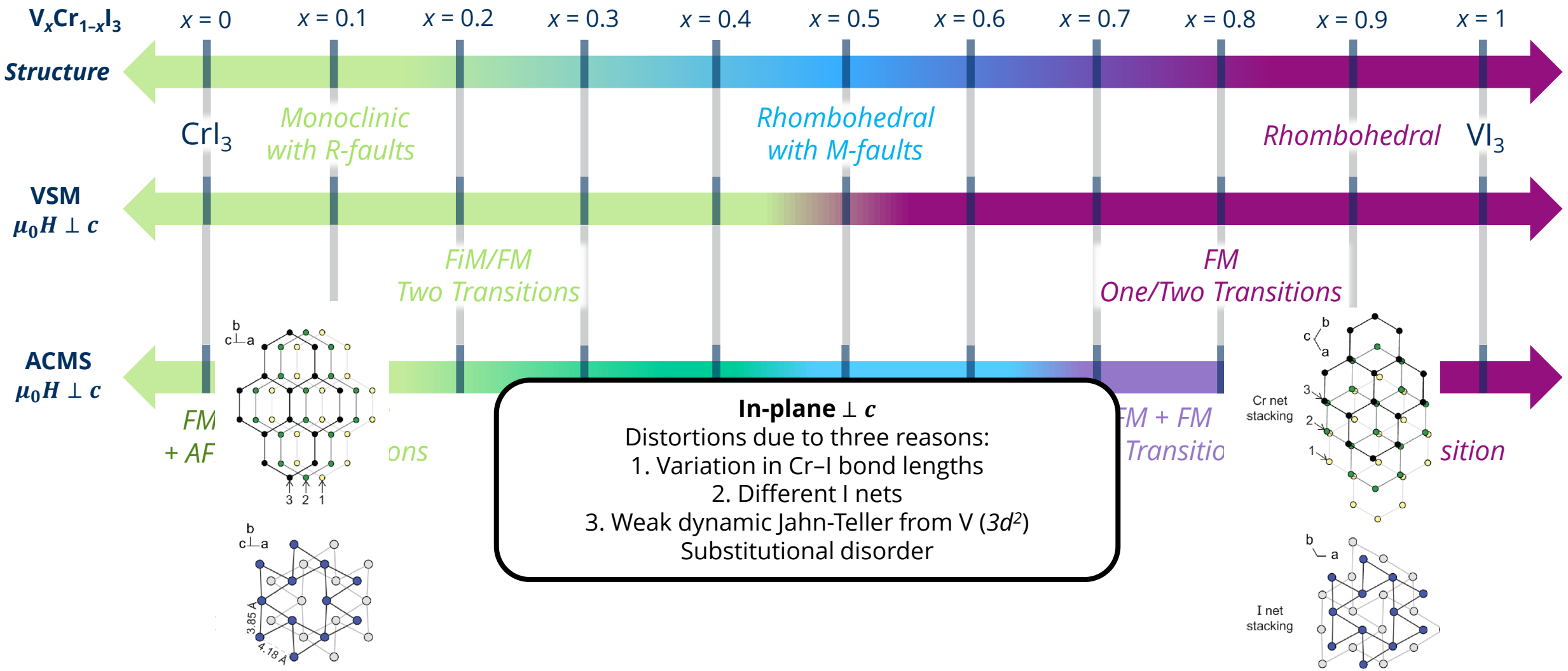
FM One Transition

FM + FM Two Transitions

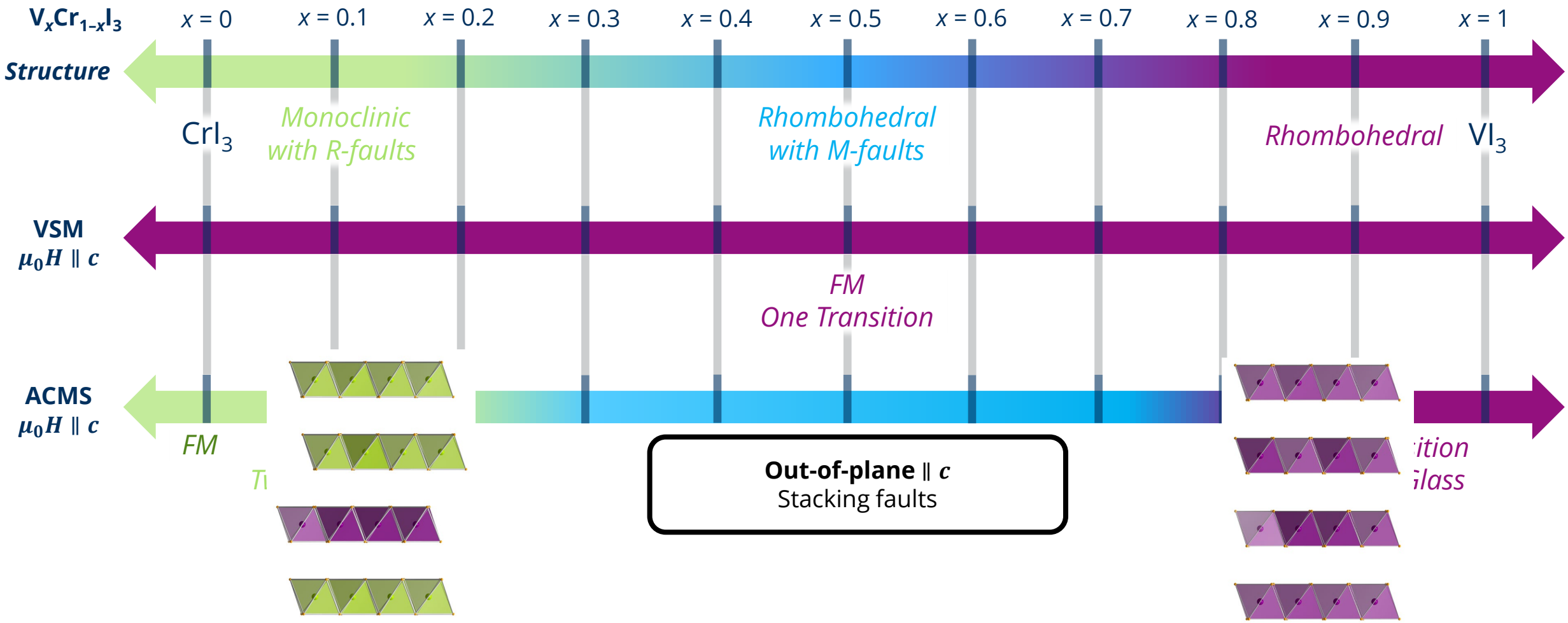
FM One Transition



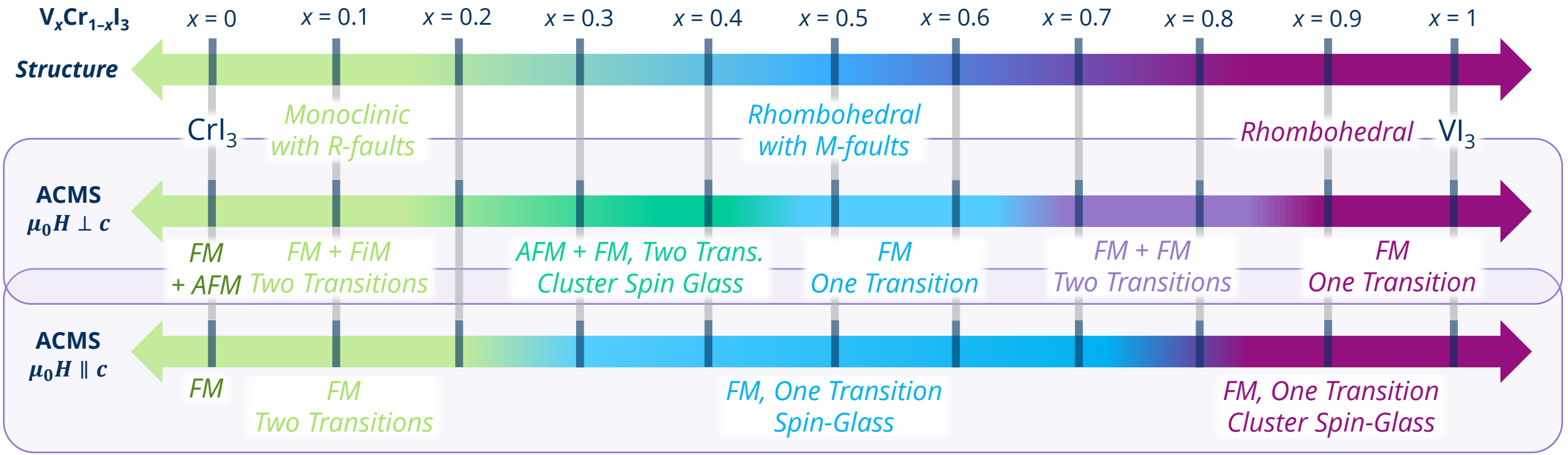
Magnetic Properties $\perp c$



Magnetic Properties $\parallel c$



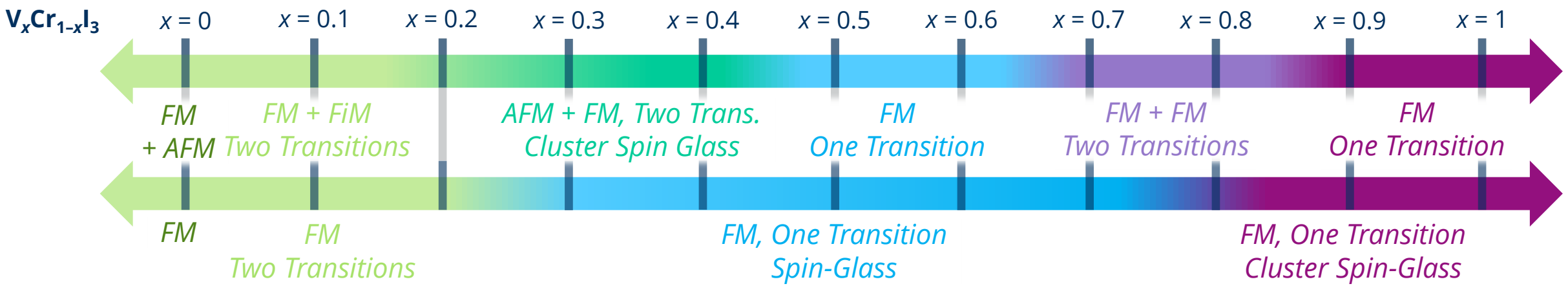
Magnetic Properties of $V_xCr_{1-x}I_3$



In-plane $\perp c$
Geometric distortions
Substitutional disorder

Out-of-plane $\parallel c$
Stacking faults

(Dis)Order Makes a Difference!



- Solid solutions (and structures) containing transition metal elements have multiple challenges
  - ➢ Oxidation number, valency, coordination → structure is affected!
  - ➢ Synthesis pathway plays a significant role
- Understanding the structural peculiarities is key to understanding the physical properties and their origin
- Proper characterisation of crystals/material is key → issues can arise at any step!

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