Matthias Wuttig and Team (RWTH Aachen)

Tailoring Phase Change Materials for different Applications: The Role of Metavalent Bonding

Physics of Novel Materials, RWTH Aachen University
& JARA Institute Green-IT, Jülich, Germany

Mission and Vision of our Team:

Our Mission: Design of novel opto-electronic materials

For information technology (neuromorphic computing) and energy conversion

Phase Change Materials, Thermoelectrics, Topological Insulators, Photovoltaics,….

Our Vision: Devising and employing (treasure) maps,

i.e. relationship between chemical bonding and material properties

Google Map for Materials

(opto-electronic materials)

by 2028 (2029)

Example: How can we understand, explain and tailor PCMs?

Step 1: Understand and Explain

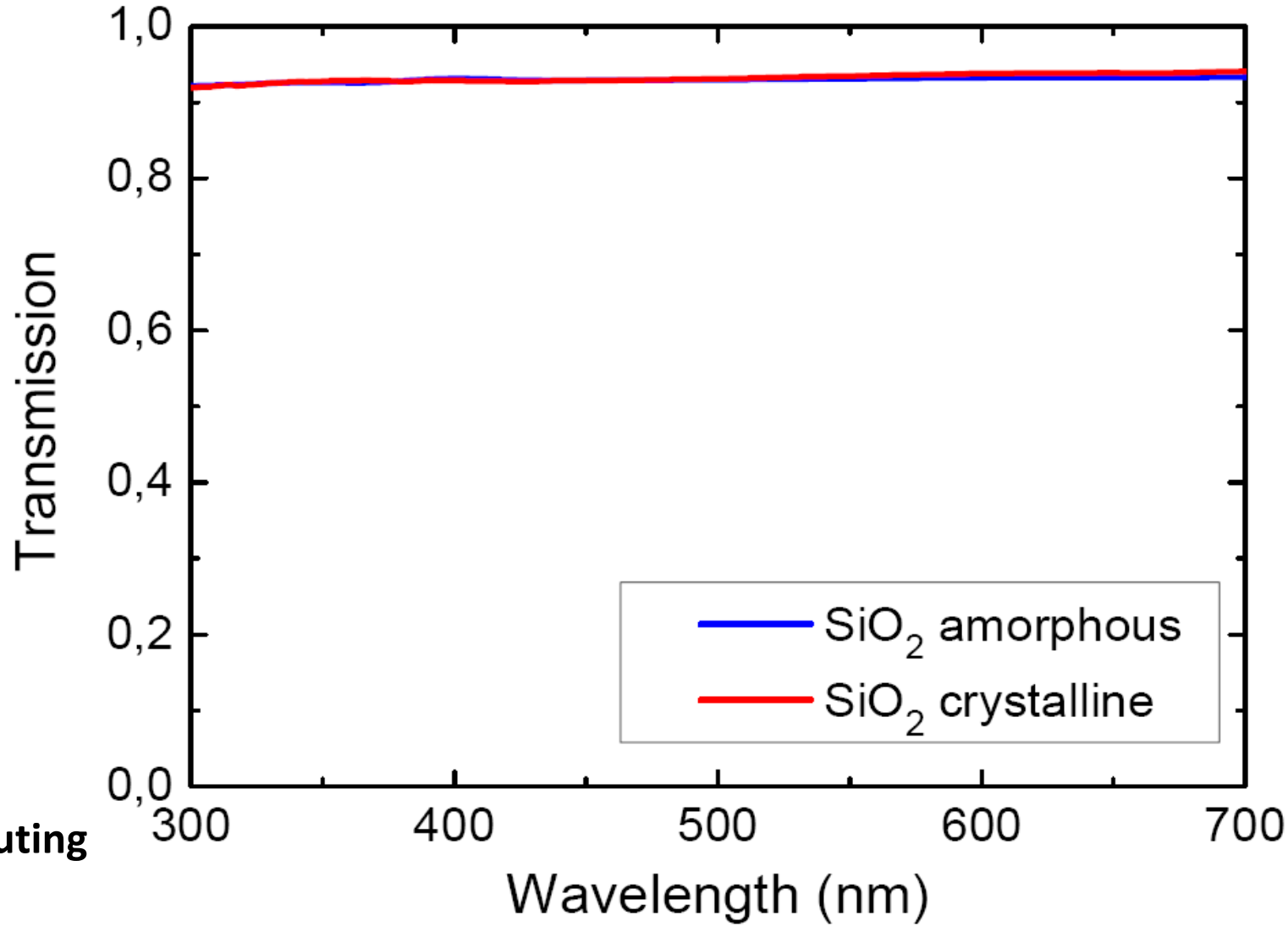
What is unique about PCMs?

Pronounced change of opto-electronic properties upon crystallization,

which happens very rapidly!

Understanding the intellectual challenge:
Explaining strong property contrast and high speed of crystallization

The ability to switch optical properties significantly: is rare

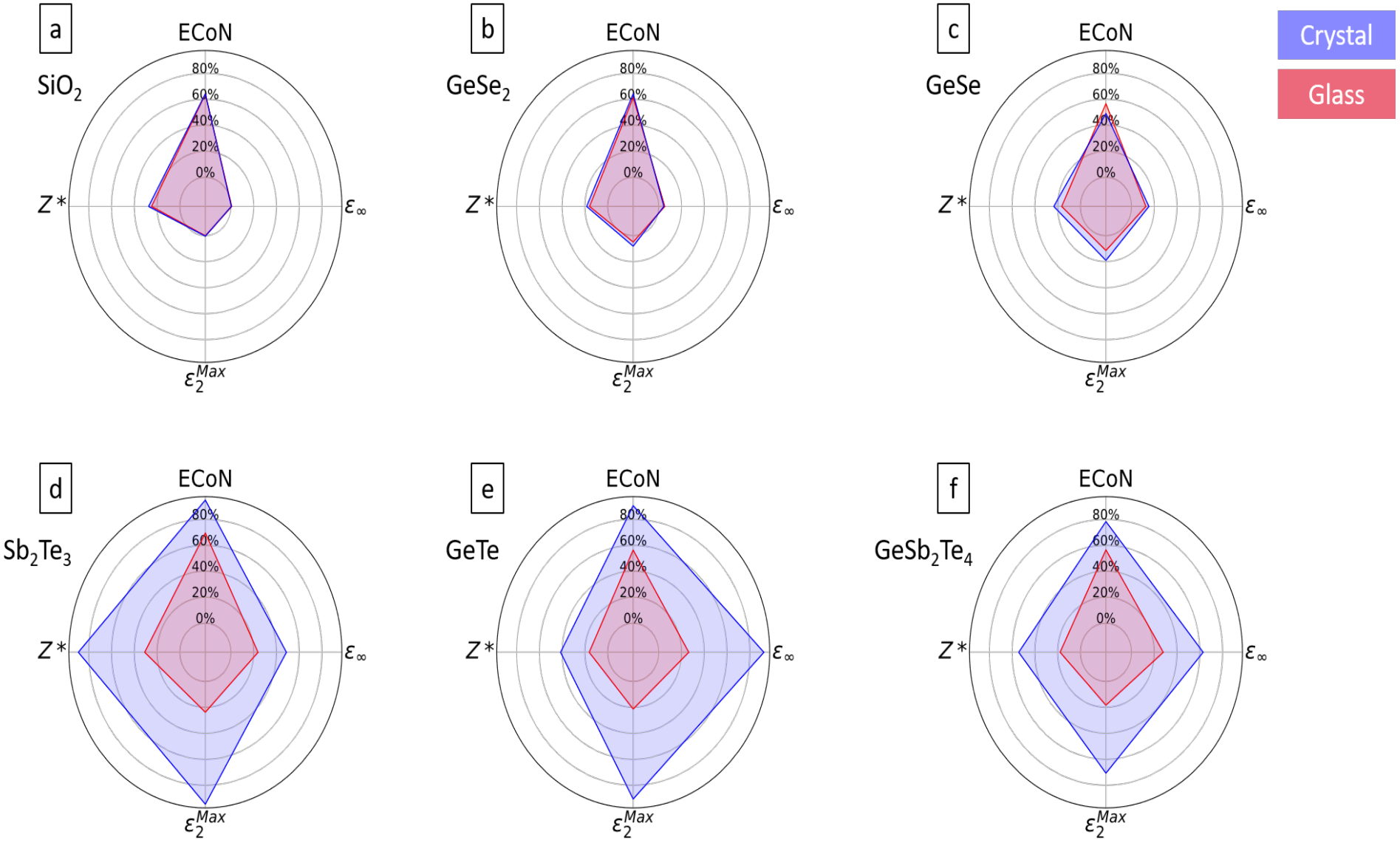


Applications:

- **Optical data storage**
- **Photonic switches**
- **neuromorphic computing**
- **Active metasurfaces**

No significant difference in optical properties!

Property Change upon Crystallization: Difference between SiO_2 and PCMs



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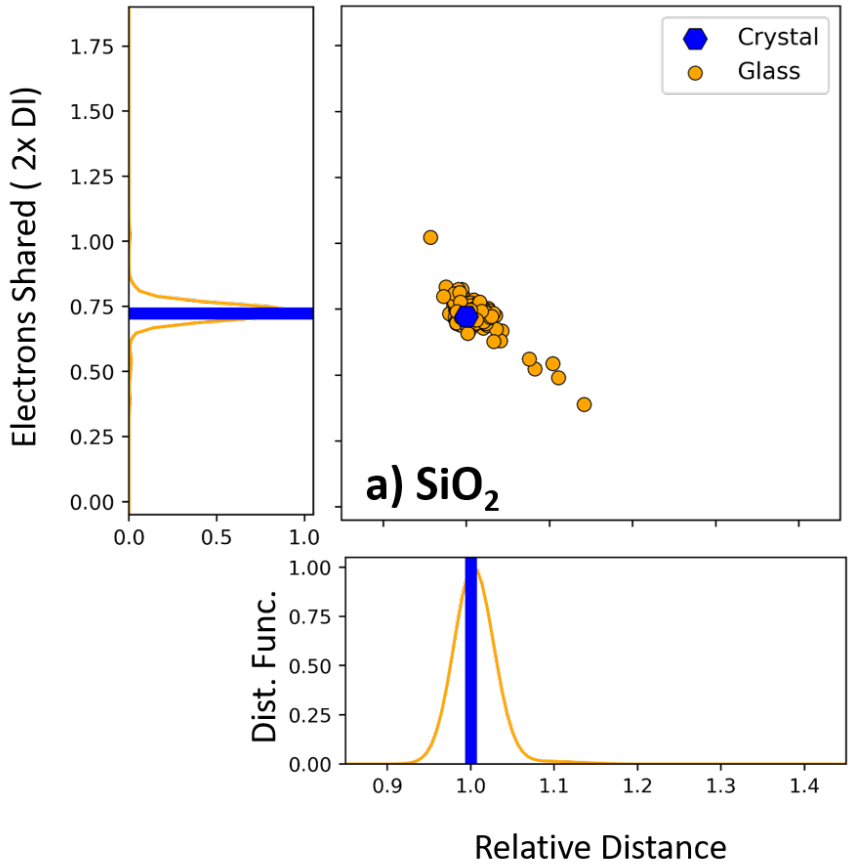
Suspicion concerning origin of difference: differences in bonding



Comparison of chemical bonding and atomic arrangement in glassy and crystalline SiO_2 (left)

with quantum-chemical bonding analysis **(compare Zachariasen conjecture from 1932)**

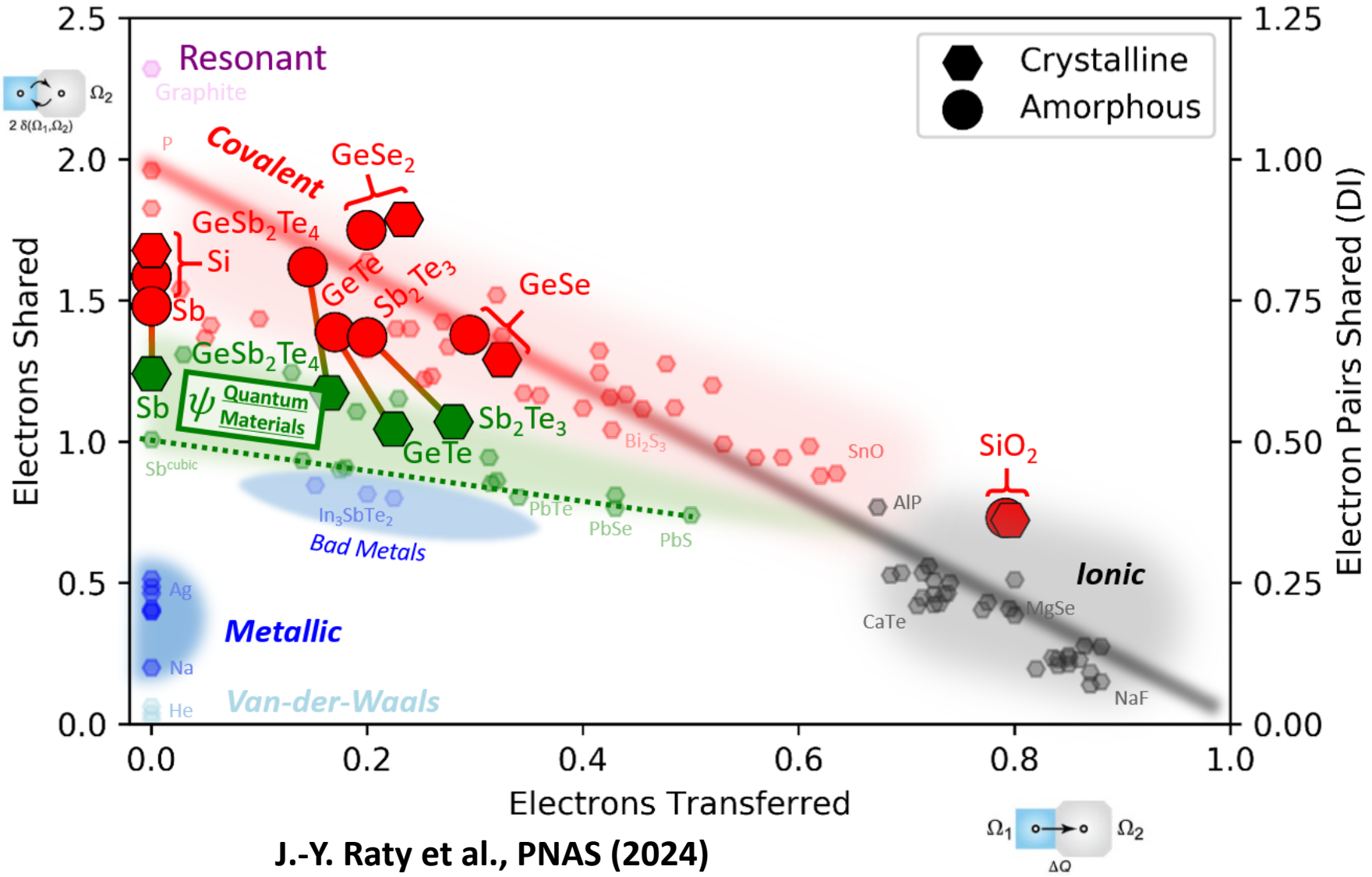
Compare SiO_2 with GeTe (PCM)

Phase Change Materials change their atomic arrangement and chemical bonding upon crystallization!

GeTe: forms a non-Zachariasen glass; finding builds a link to chemical bonding

PNAS, 121 (2) e2316498121 (2024)

Where do changes of bonding upon crystallization occur?



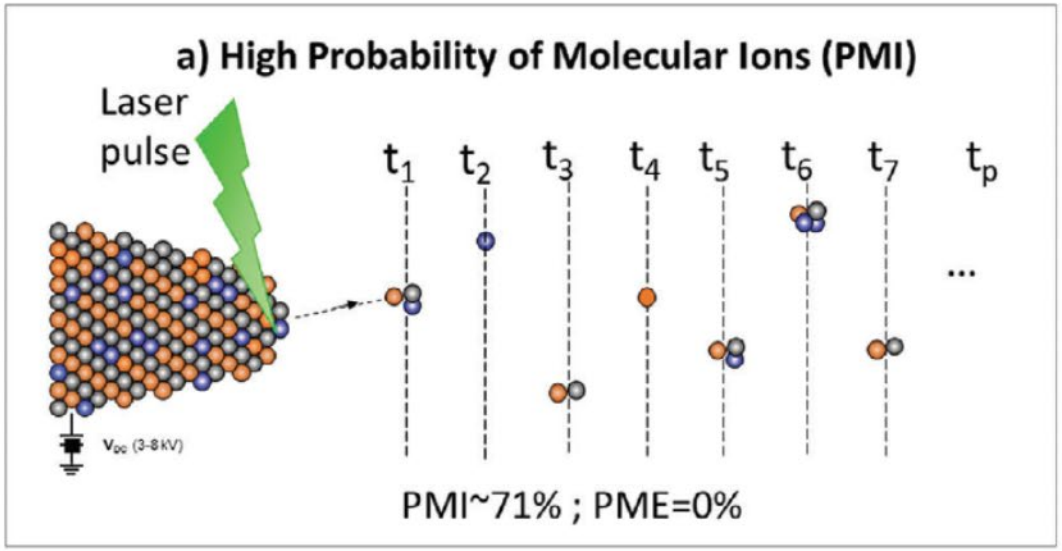
J.-Y. Raty et al., PNAS (2024)

Pronounced bonding changes upon crystallization for several PCMs

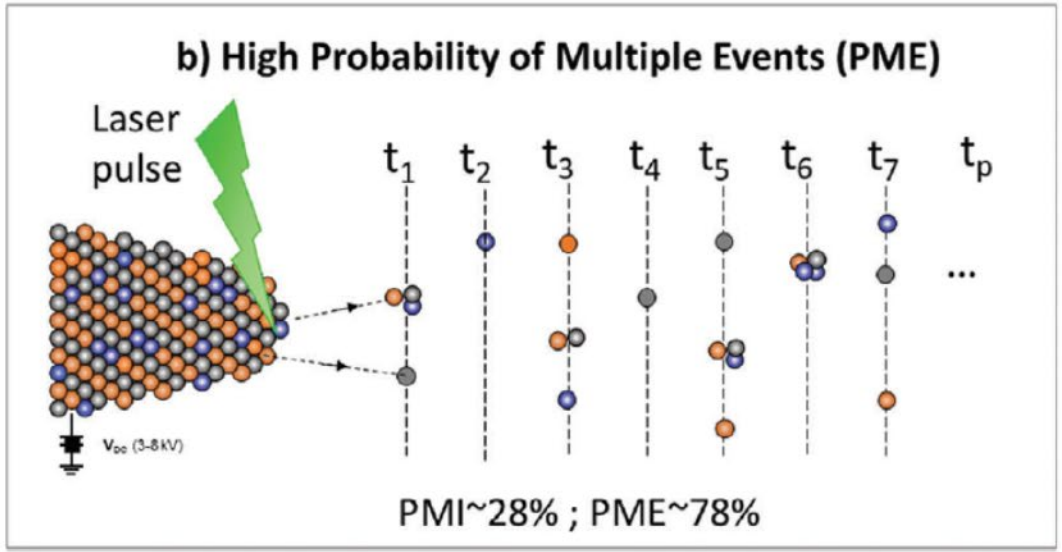
ES and ET are bonding descriptors and property predictors!

Bonding in crystalline PCMs is special!

Can we confirm unconventional bonding in crystalline PCMs? (APT)



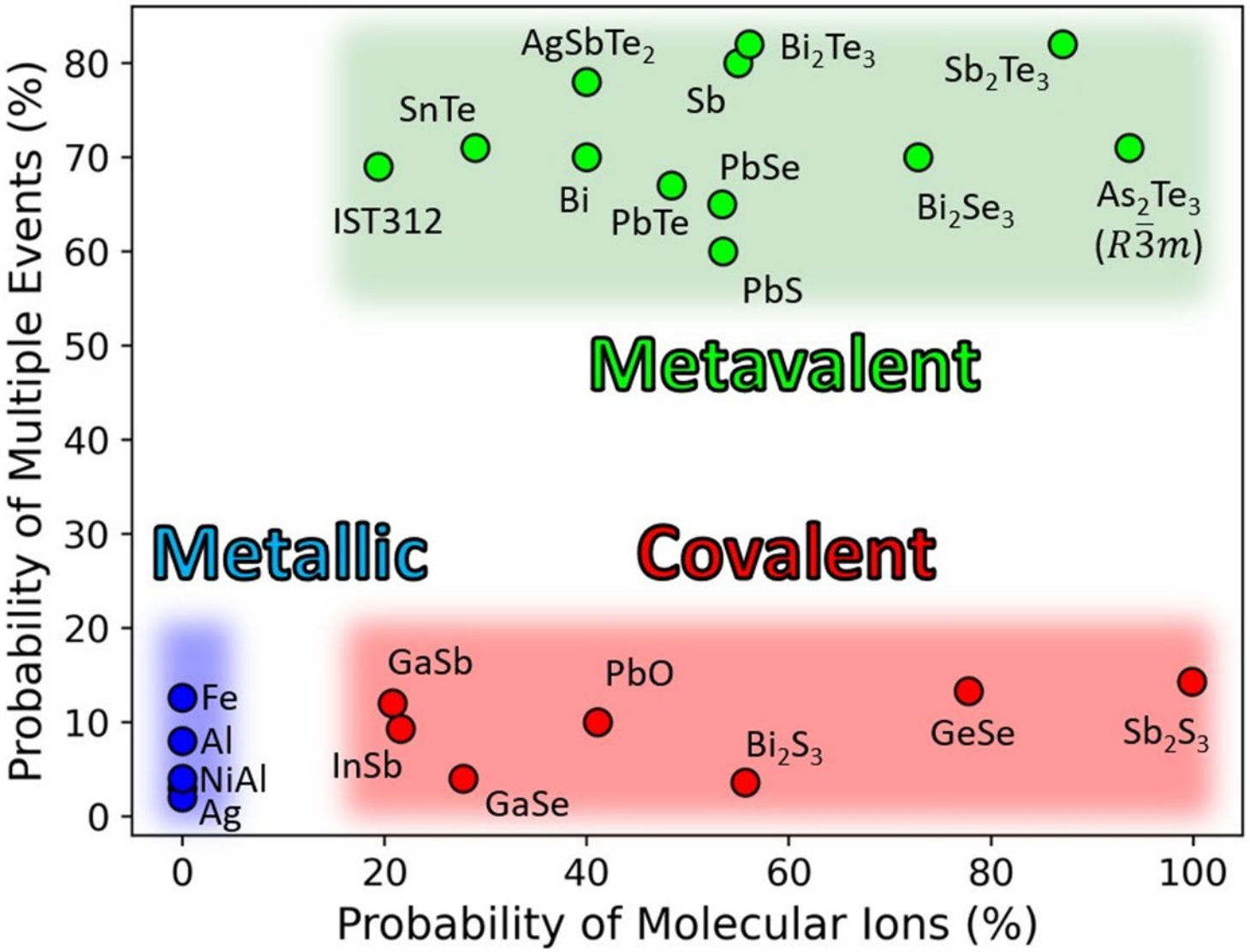
$$\mathbf{PMI} = \frac{\mathbf{X_{MI}}}{(\mathbf{X_{MI}} + \mathbf{X_{SI}})} \times \mathbf{100}$$



$$\mathbf{PME} = \frac{\mathbf{X_{ME}}}{(\mathbf{X_{ME}} + \mathbf{X_{SE}})} \times \mathbf{100}$$

PNAS, 121 (19) e2405294121 (2024)

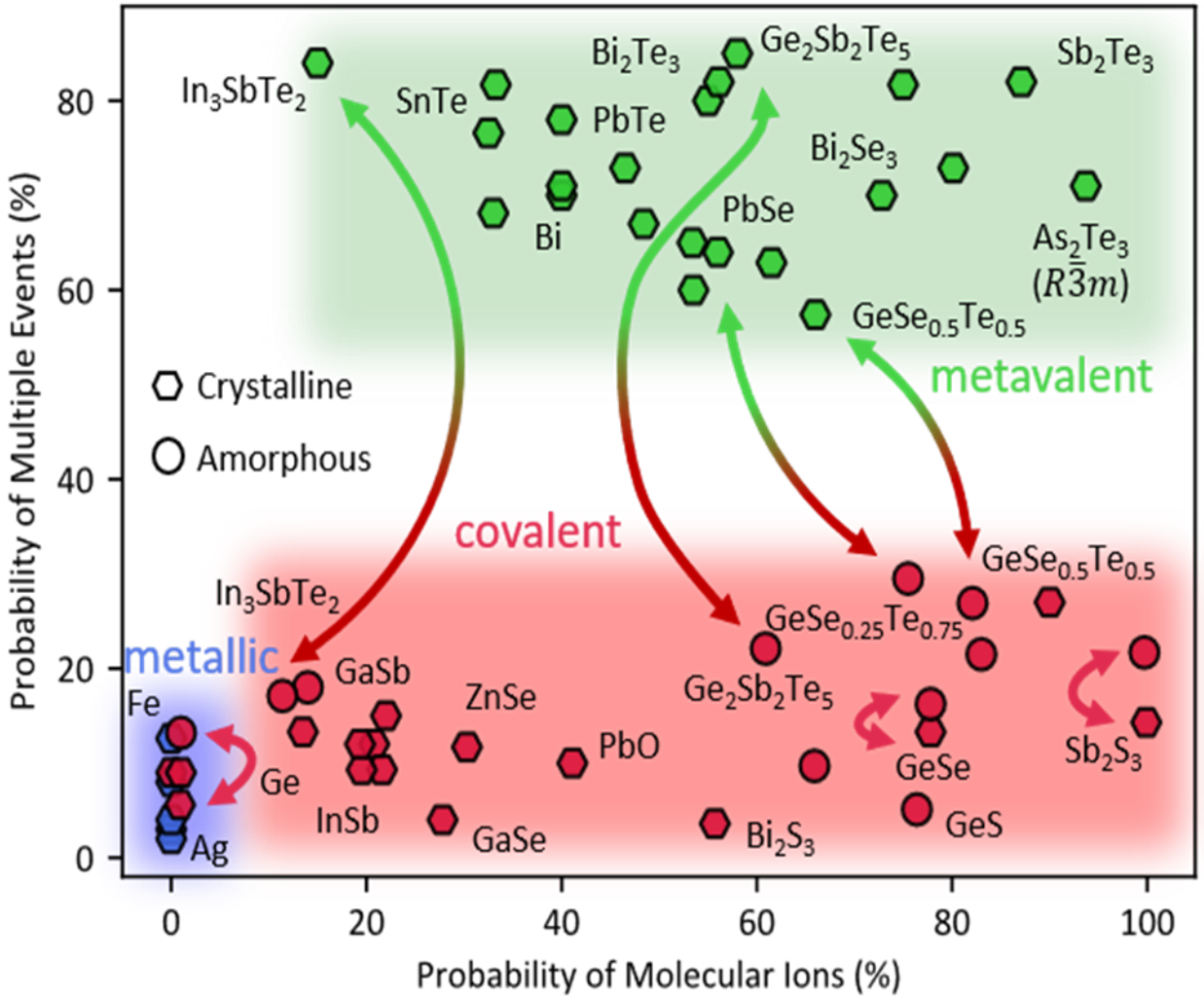
Can we classify bonding by atom probe tomography?



Yes, we can!

Can we confirm unconventional bonding in crystalline PCMs (APT)

Classify those solids, which change bond rupture upon crystallization!



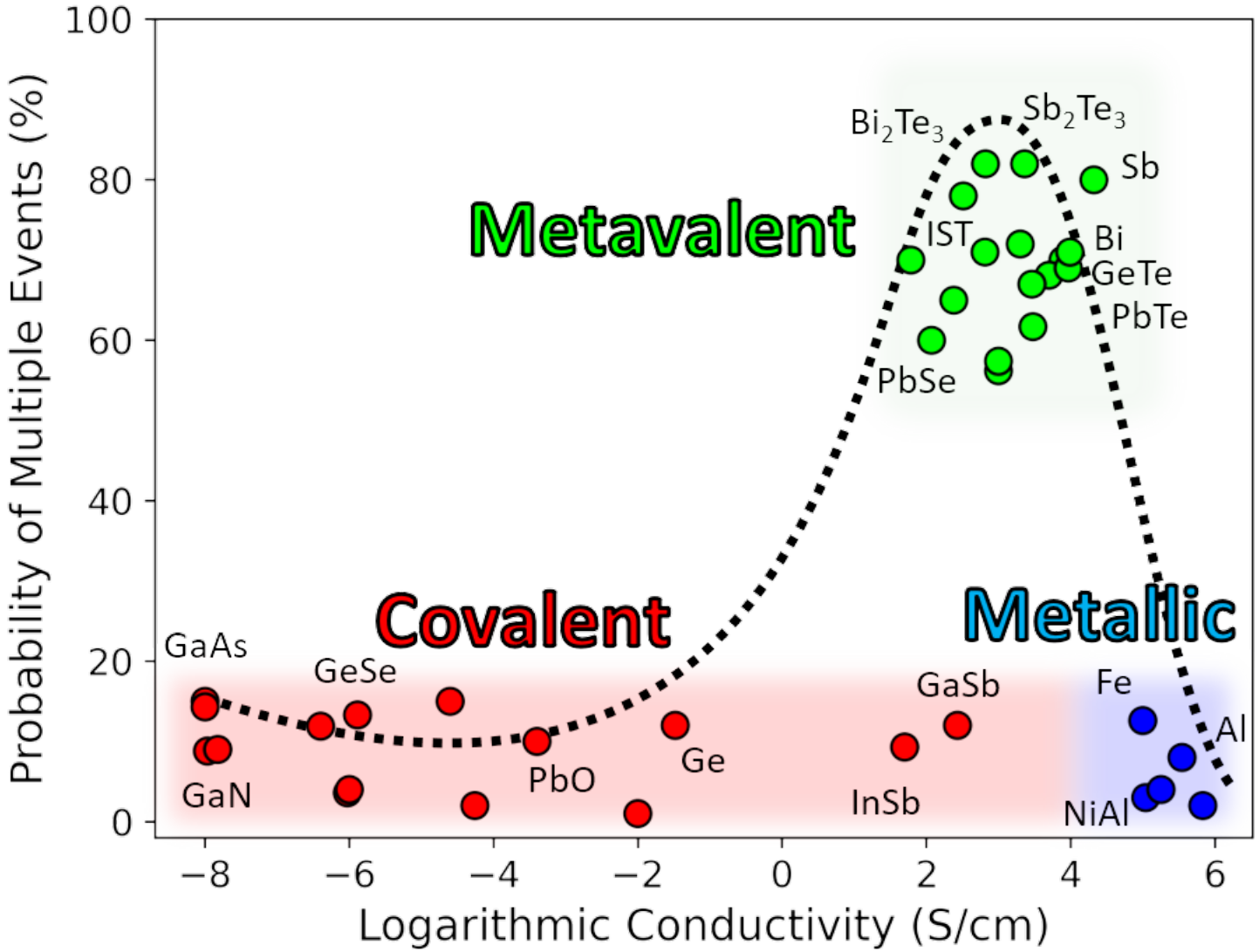
Crystall

Glass

PNAS, 121 (19) e2405294121 (2024)

Bonding in crystalline PCMs is unconventional!

How can we understand the bonding in crystalline PCMs?



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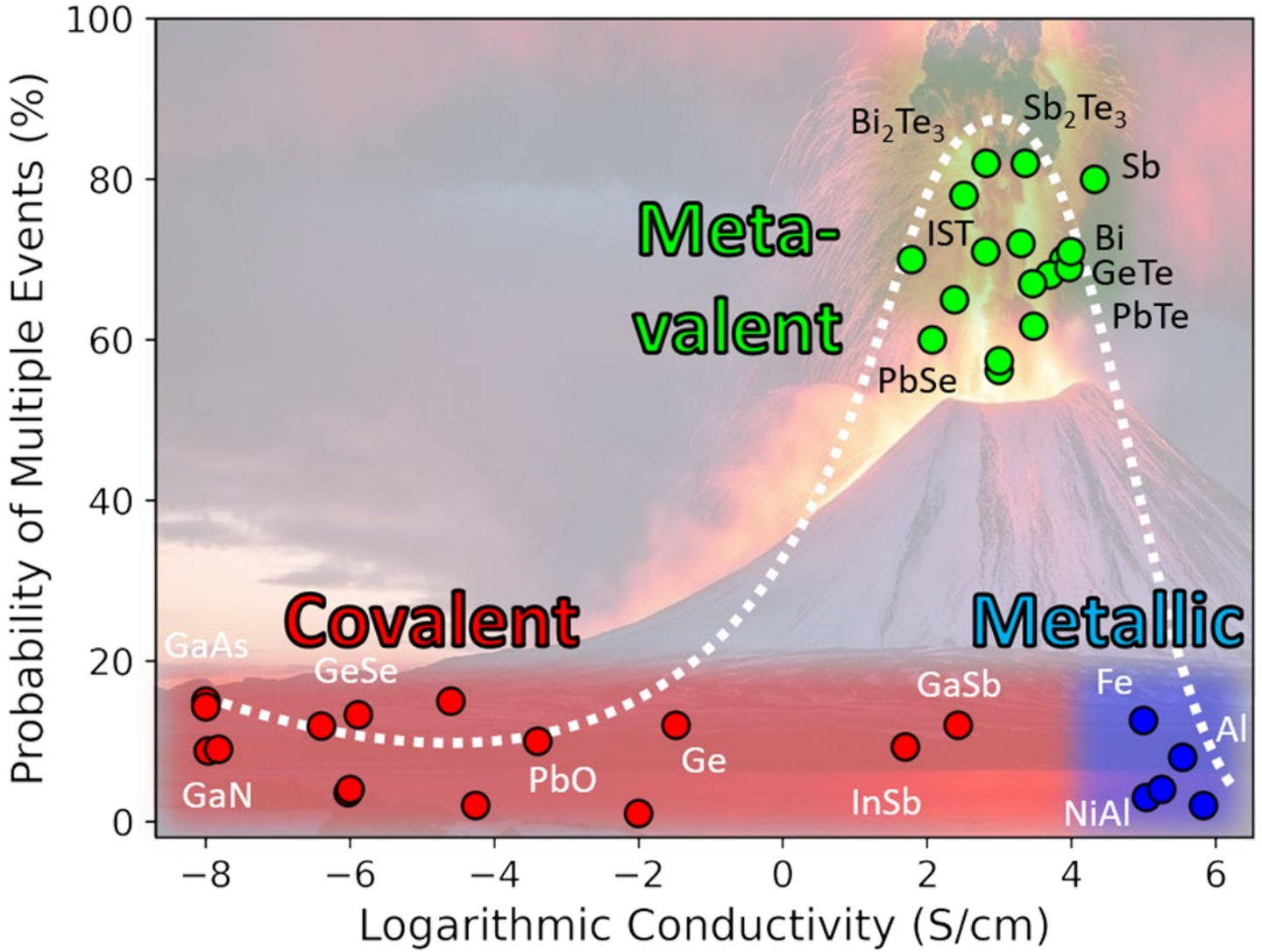


Bonding in crystalline PCMs is unconventional!

Treasure Map helps to design Kinetics in PCMs and zT in thermoelectrics

Study of crystallization kinetics

using 2 chemical bond descriptors

GeTe – GeSe

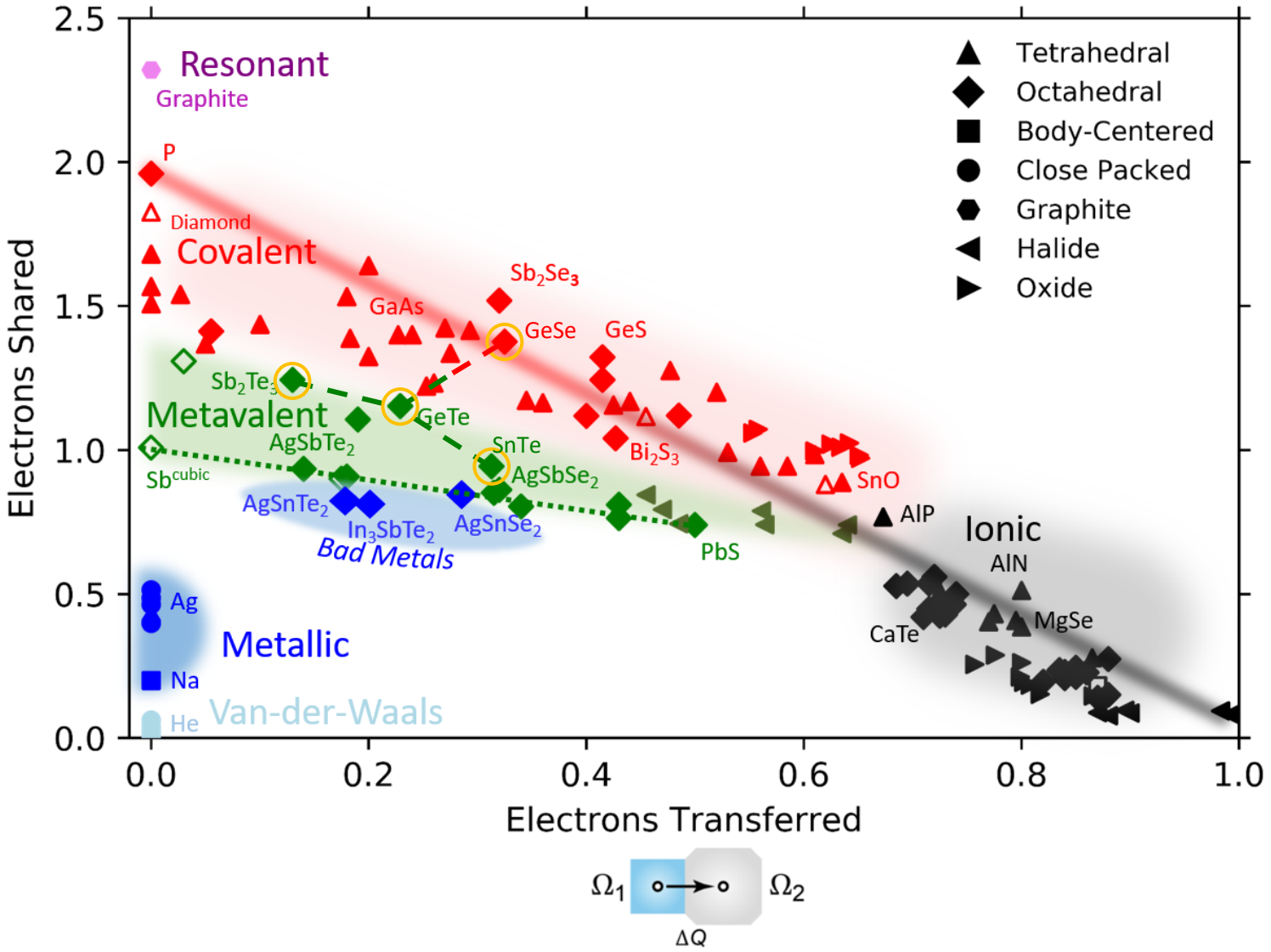
GeTe – SnTe

GeTe – Sb_2Te_3

Measurement with set-up

employing 2 different lasers

(excite - probe-setup)



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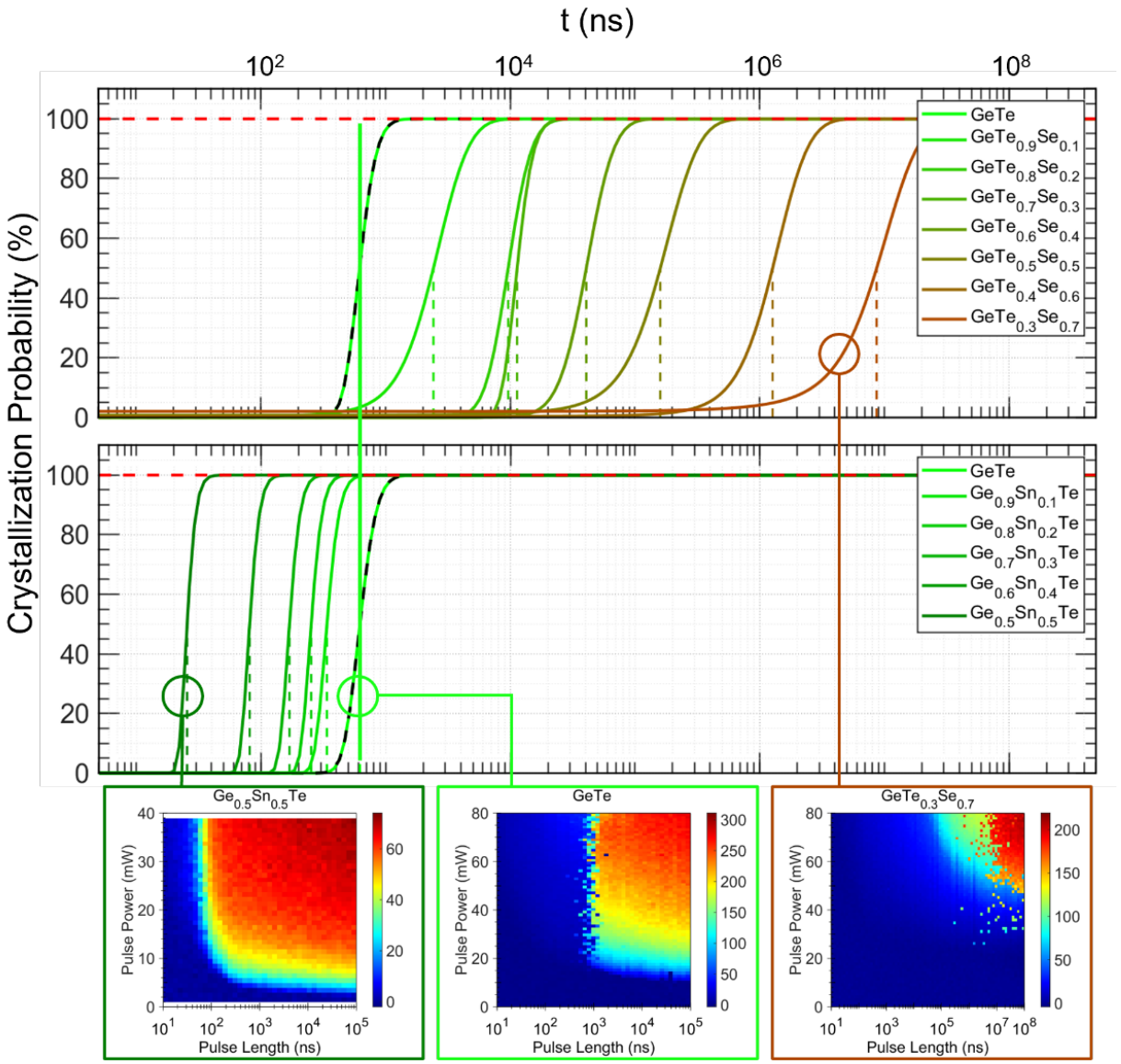
Minimum Crystallization Time τ to Quantify the Switching Speed



Minimum crystallization time τ

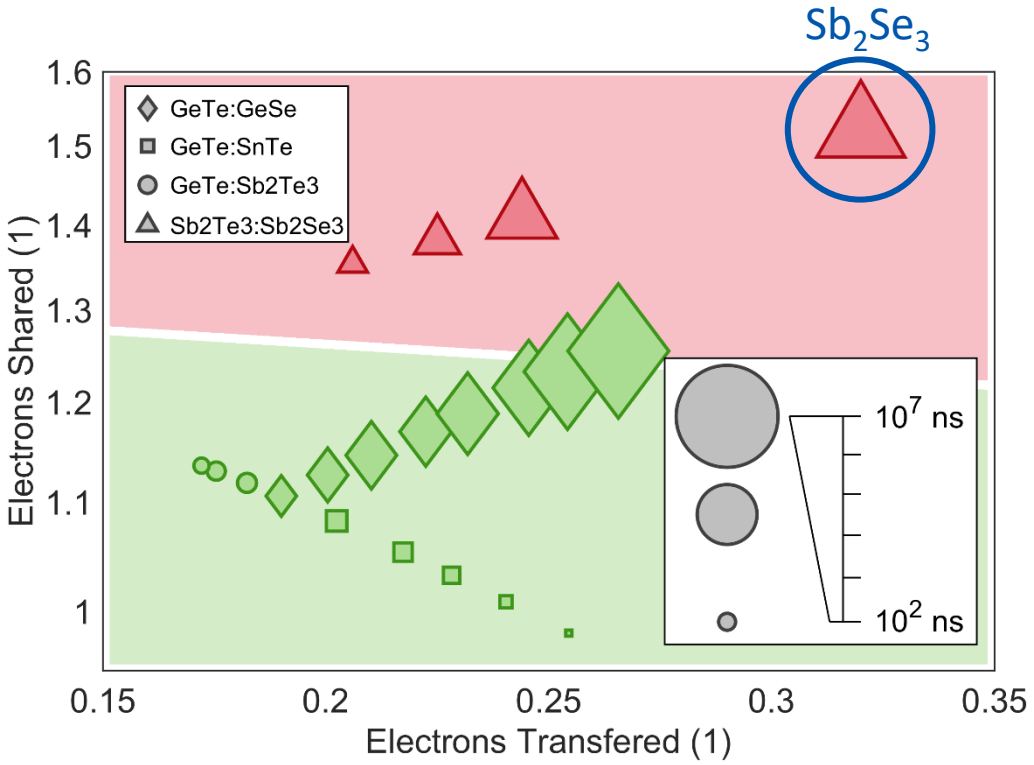
GeTe → GeSe: Increase over 4 orders of magnitude

GeTe → SnTe: Decrease over 1.5 orders of magnitude

C. Persch et al.,
Nature Communications 12, 4978 (2021)

Treasure Map for Crystallization Kinetics / Switching Speed

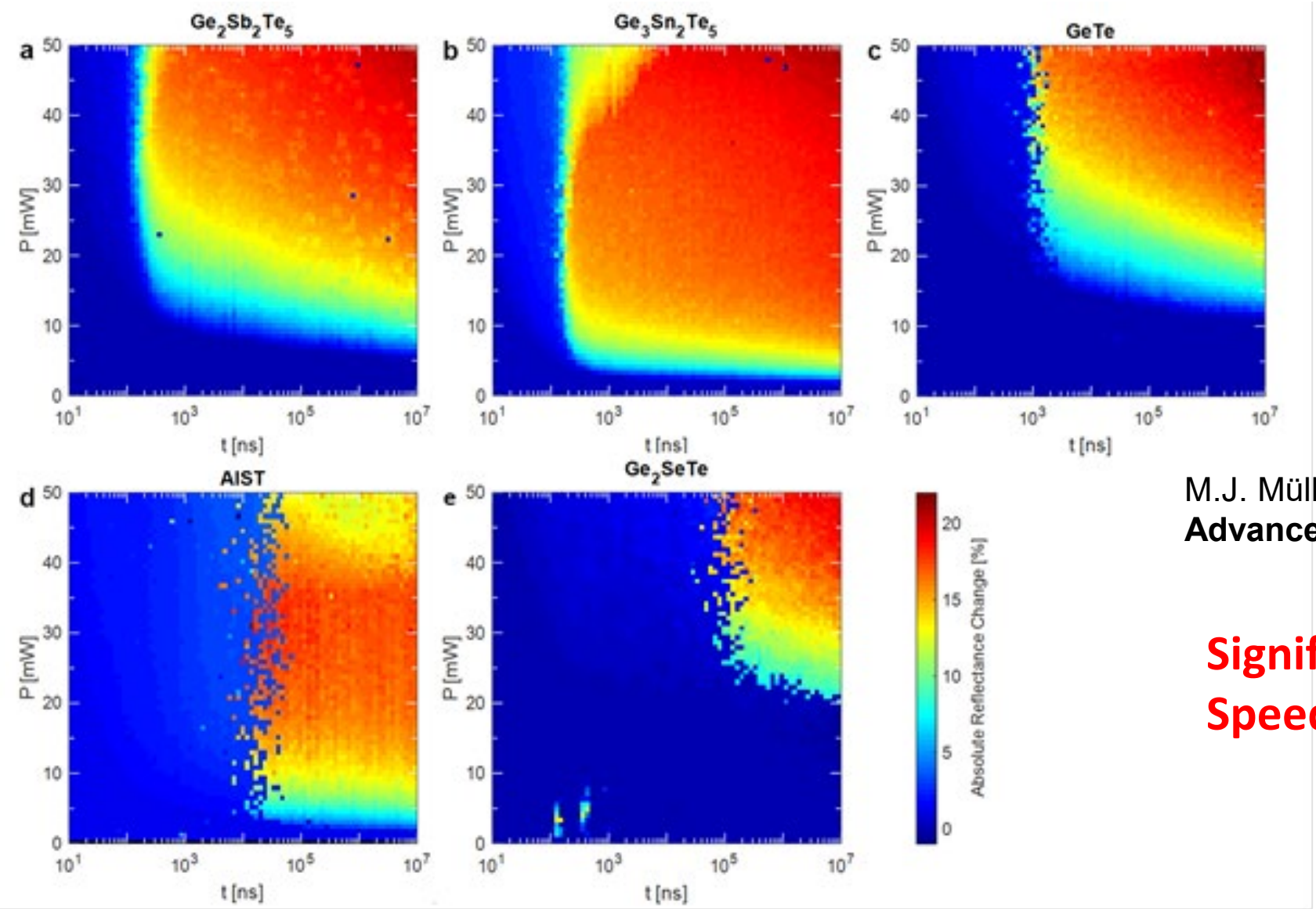
Minimum Crystallization Time τ



C. Persch et al.,
Nature Communications 12, 4978 (2021)

Sb_2Se_3 still shows reasonably fast switching below ms-regime, although being strictly covalently bonded.

Unraveling Crystallization Kinetics (Statistics) beyond Speed?

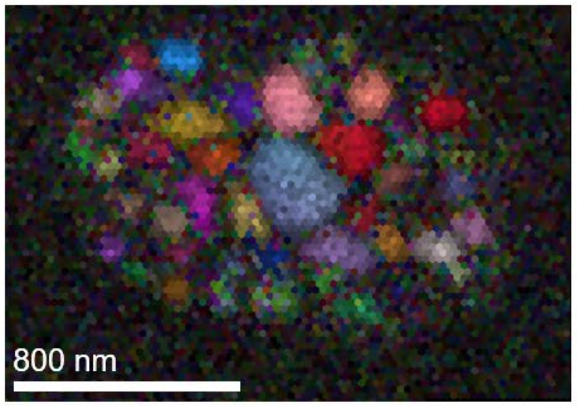


M.J. Müller et al.,
Advanced Functional Materials 2403476 (2024)

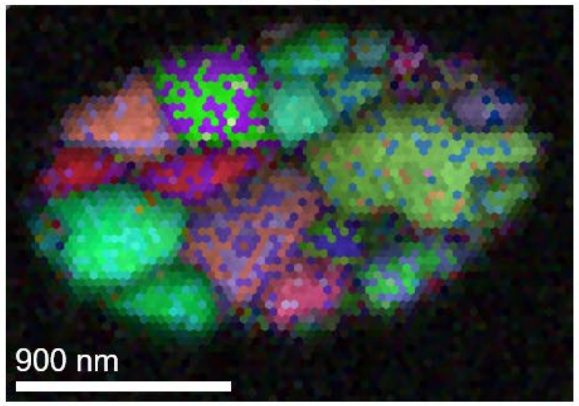
Significant Differences in Speed and Stochasticity

Unraveling Crystallization Kinetics: EBSD

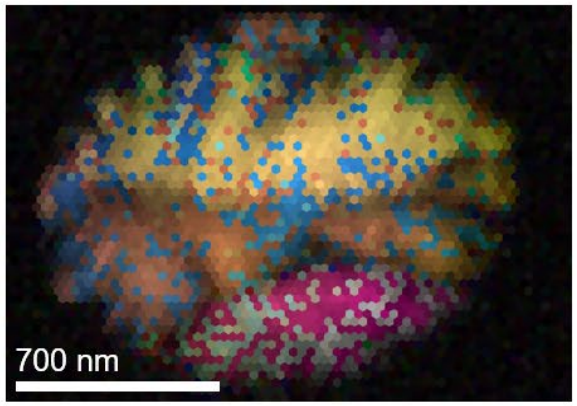
a $Ge_2Sb_2Te_5$



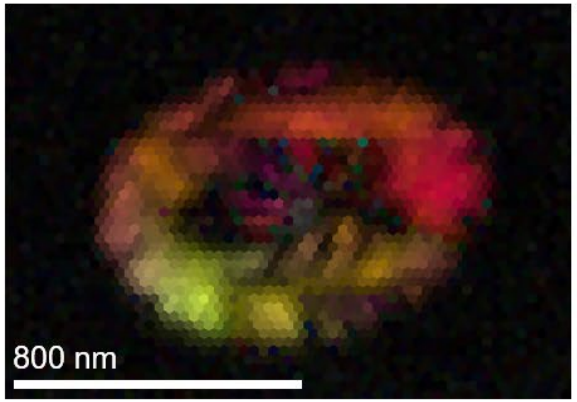
b $Ge_3Sn_2Te_5$



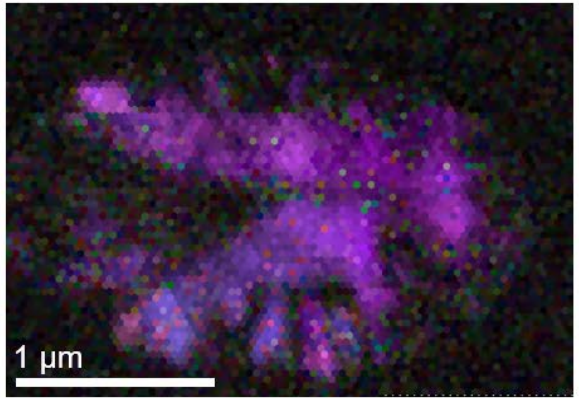
c GeTe



d AIST



e Ge_2SeTe



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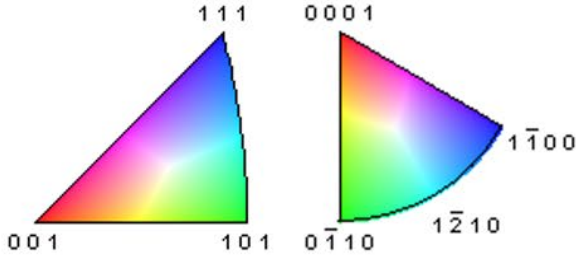

Significant Differences in Nucleation Density

Mission and Vision of Team: Focus on Applications

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Phase Change Materials, Thermoelectrics, Topological Insulators, Photovoltaics,….

Our Vision: Devising and employing (treasure) maps,

i.e. relationship between chemical bonding and material properties

Google Map for Materials

We have learned to the use this map to design PCMs and thermoelectrics

Summary / Designing Chalcogenides for PCM Applications

- a novel 2D map (3rd Generation Map) has been presented, which differs substantially from previous generations.
- based on QM calculations,
  which show that ES and ET are good chemical bonding descriptors
- separates different bonding mechanisms (ionic, metallic, covalent, vdW,..) and supports the idea of a new bonding mechanism, i.e. metavalent bonding (MVB)
- reveals pronounced correlations and explains link between bonding and properties,
  which shows that ES and ET are good property predictors (3D map)
- PCMs are Quantum Materials, where glass is covalent and crystal metavalent
  We can use these insights to design PCMs!