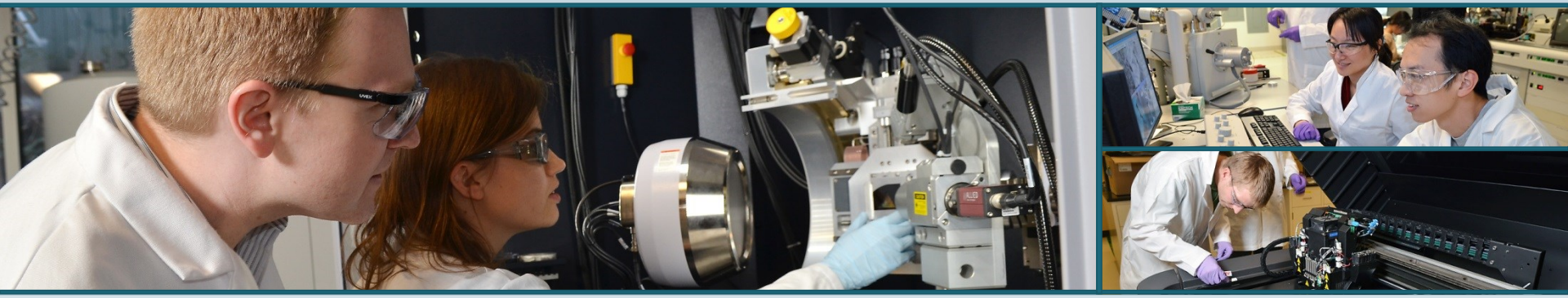


Computational Challenges In Materials Discovery

Santosh Suram, Staff Scientist, High-Throughput Experimentation Group

Joint Center for Artificial Photosynthesis

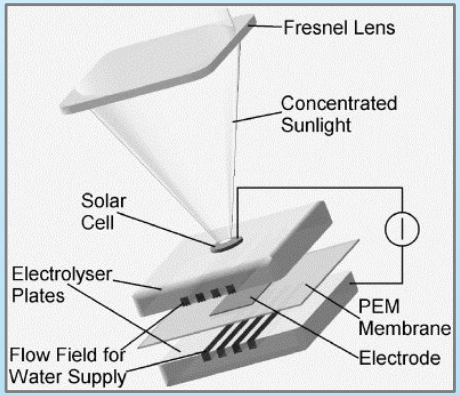
California Institute of Technology

OSG-All Hands

9 April 2014

General Approaches to Artificial Photosynthesis

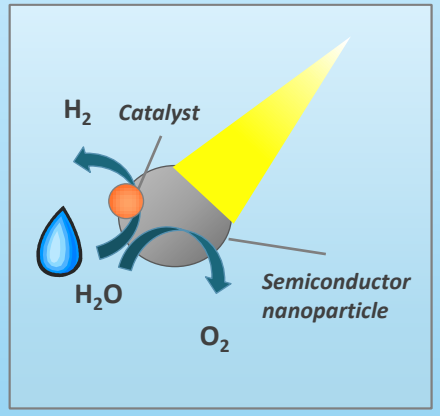
Discrete Photovoltaic Wired to Electrolyzer



Advantages: Operational system has already been demonstrated with 18% efficiency.[1]

Challenges: Demonstrated system demands expensive components; lack of integration further reduces cost efficiency.

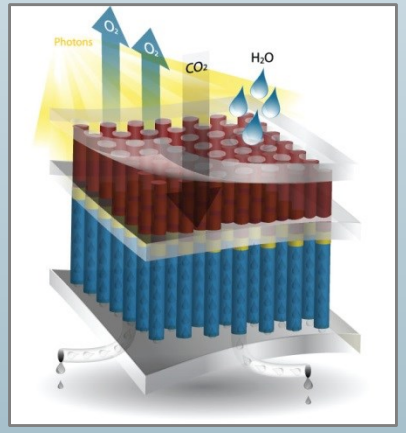
Solar-Fuel Generating Particle Dispersions



Advantages: Offers a simple architecture with the potential for low materials cost.

Challenges: Co-generation of fuel and oxidizer pose operational safety issues.

Integrated Photoelectrochemical Solar-Fuel Generator

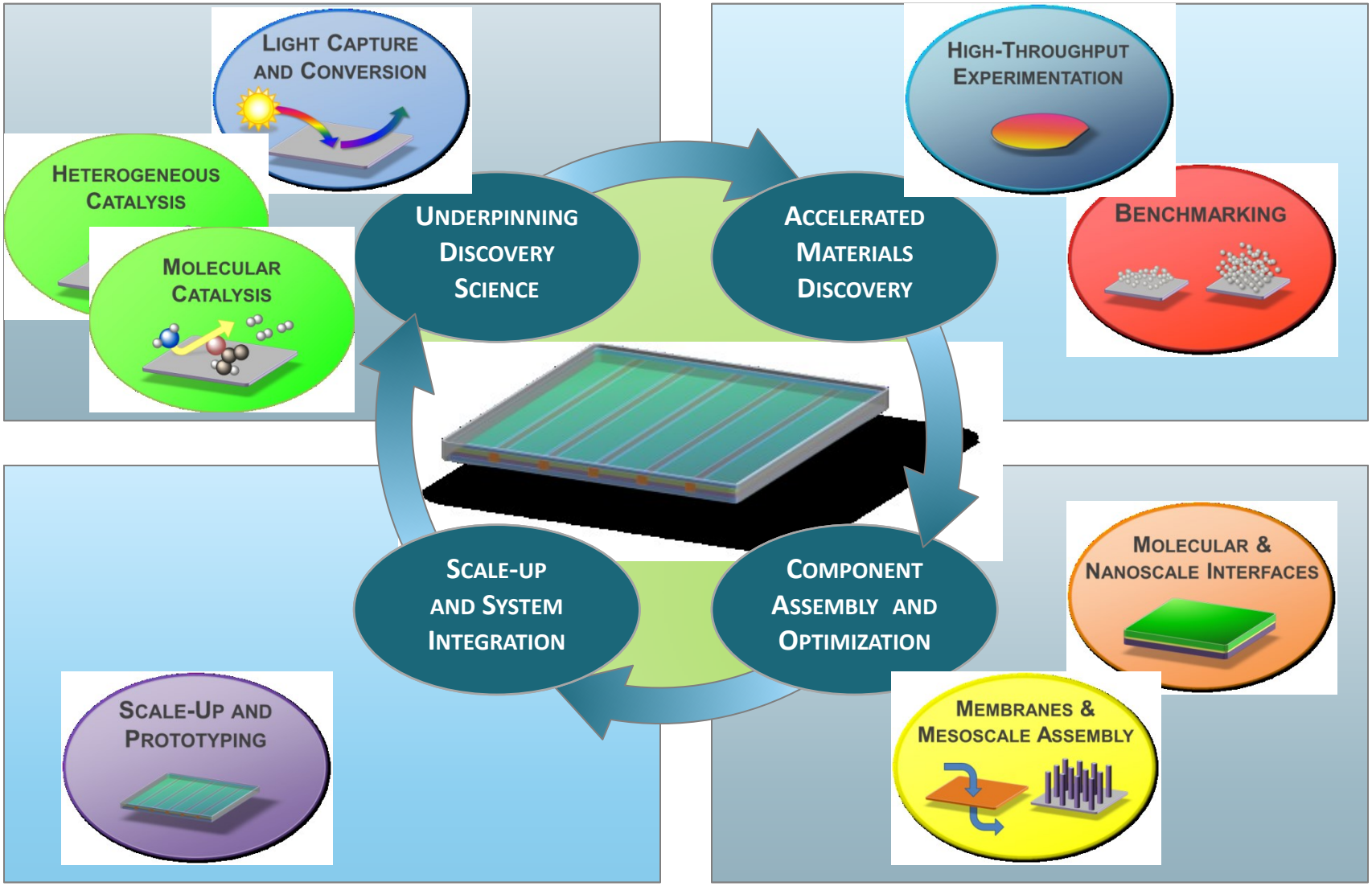


Advantages: Potentially lower component costs than a discrete system with reduced complexity.

Challenges: Requires that semiconductor, catalysts, and membranes operate efficiently under identical conditions.

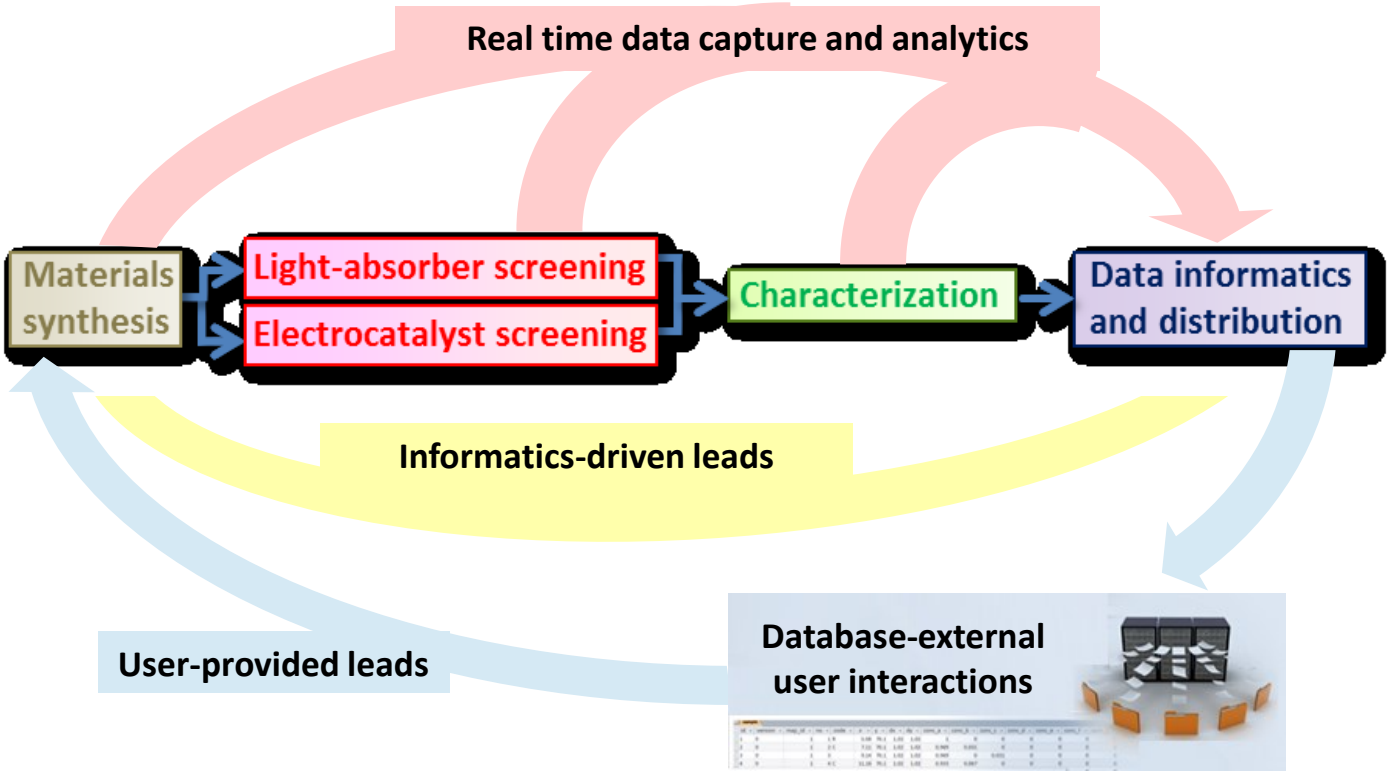
[1] G. Peharz, F. Dirmouth, and U. Wittstadt *Int. J. of Hydrogen Energy* **2007**, *32*, 3248-3252 (DOI: 10.1016/j.ijhydene.2007.04.036)

Discovery and Integrated System Development In JCAP



JCAP Joint Center for Artificial Photosynthesis

HTE Pipeline



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Combinatorial and High Throughput Material Science

- **Measurement of material properties as a function of composition and/or processing**
  - **JCAP-HTE employs 2 complementary deposition methods and has developed several processing methods**
    - PVD synthesis of continuous composition libraries
    - Inkjet printing of elemental precursors and post-calcinations

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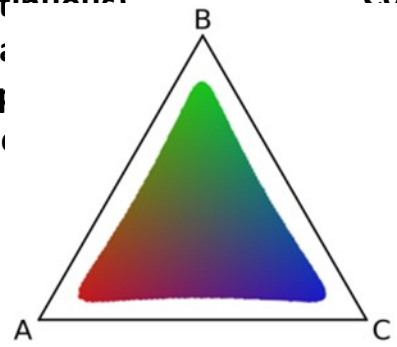

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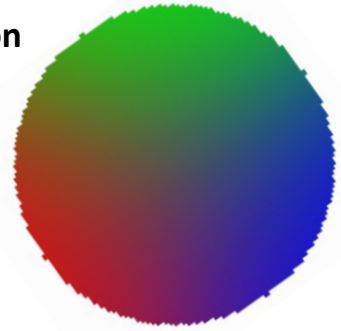
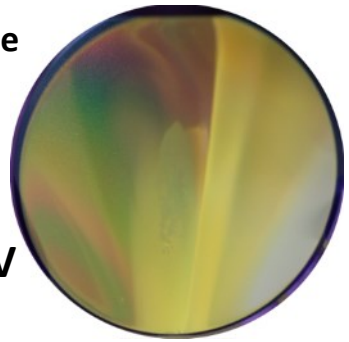
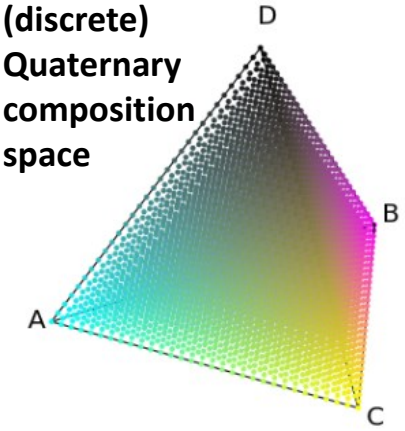


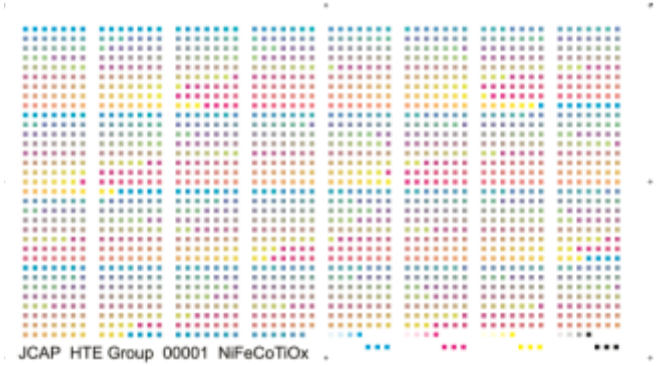
Example PVD library



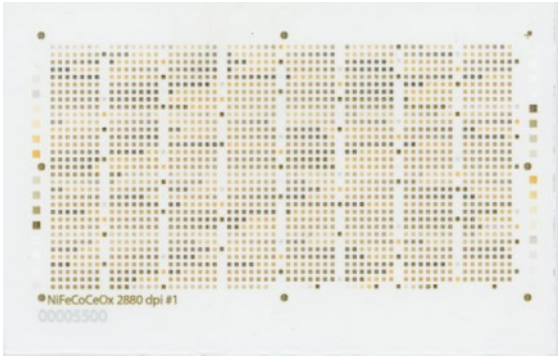
(discrete) Quaternary composition space



Composition map onto substrate



Example inkjet printed library



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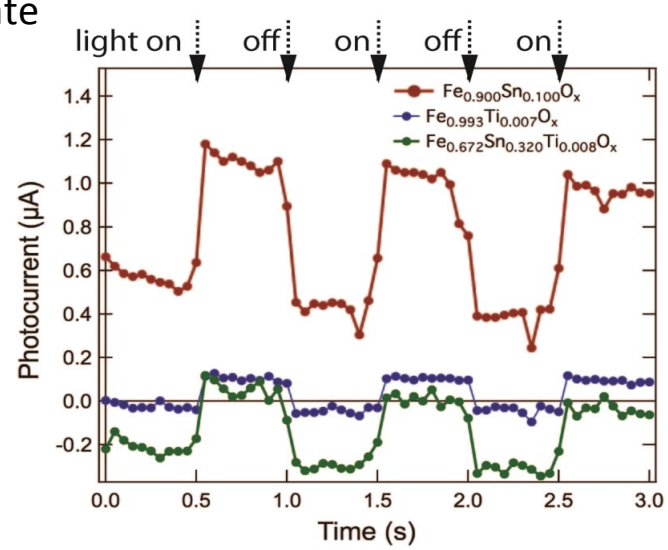
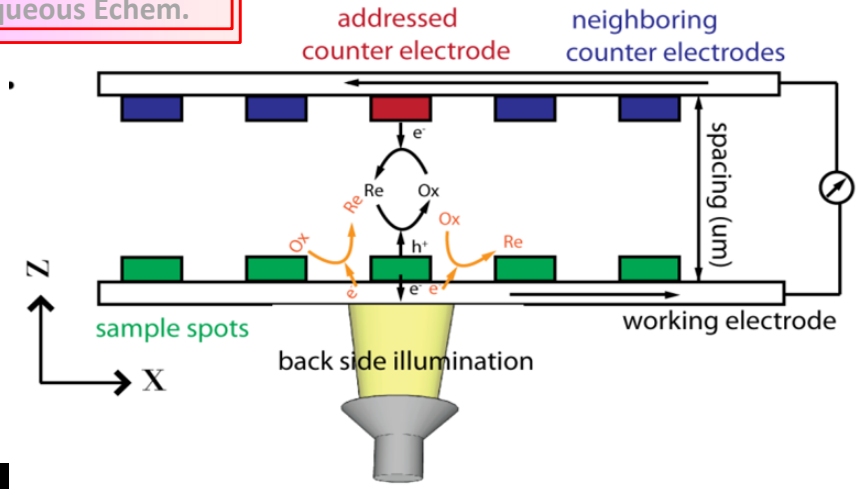
Primary PEC Screen for Photoabsorbers: Quantum Efficiency

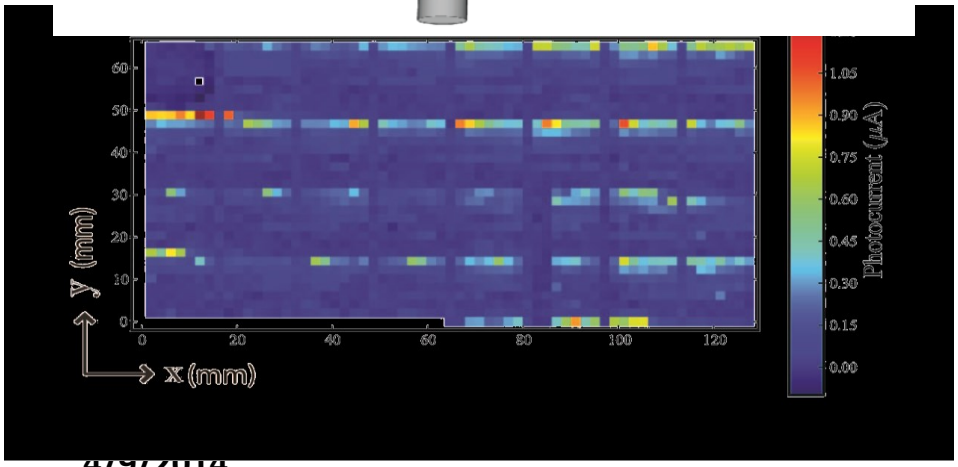
Materials synthesis → Light-absorber screening / Electrocatalyst screening

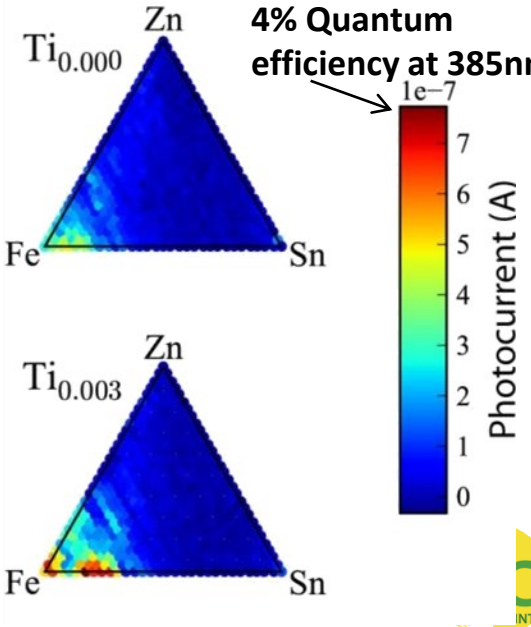
- Colorimetry
- Optical Band Gap
- **Quantum Efficiency**
- Aqueous Echem.

- Facile redox couples alleviate catalytic limitations
- Nonaqueous environment removes aqueous stability requirement



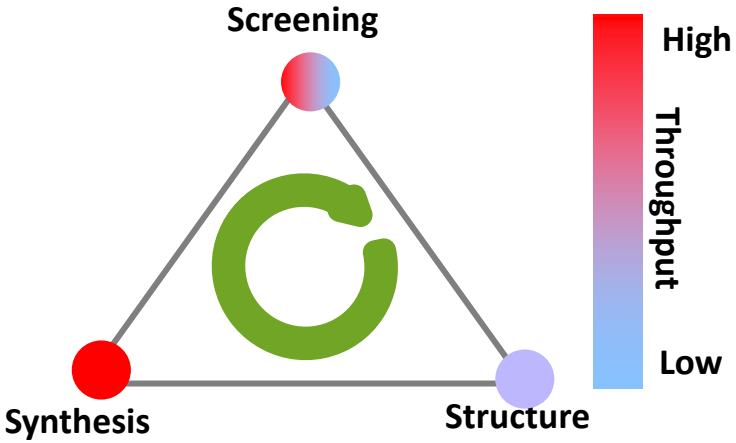




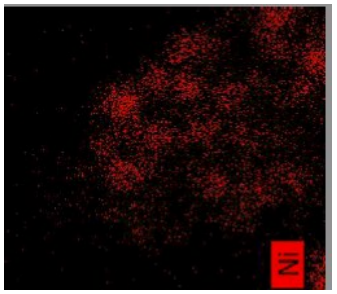


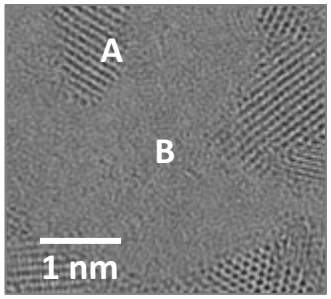
The Bottleneck

Accelerated Discovery Paradigm



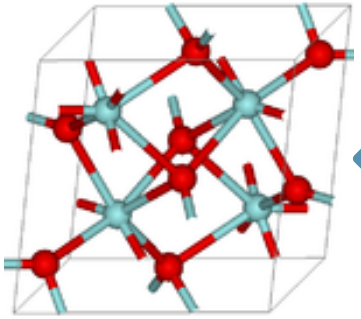
Structure

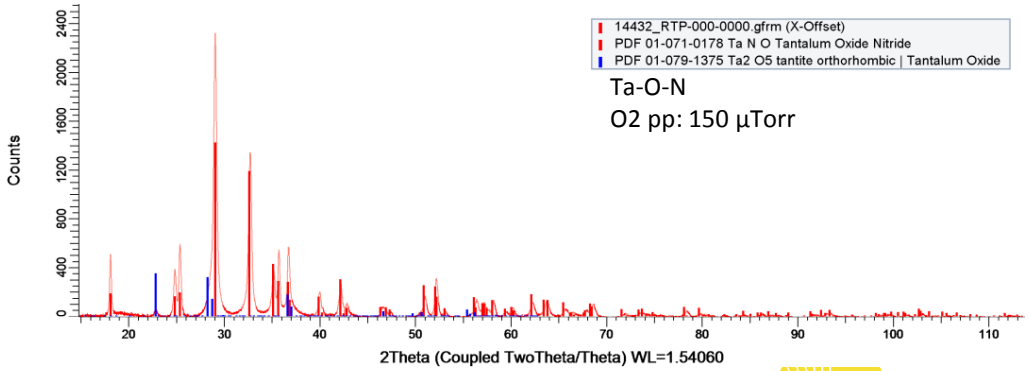




Two approaches to overcome the bottleneck:

- Develop High-Throughput Experimentation methods for determining structure
- Develop informatics tools that work on synthesis-screening space to select the most informative materials for the low-throughput structure analysis.





Ta-O-N
O2 pp: 150 µTorr

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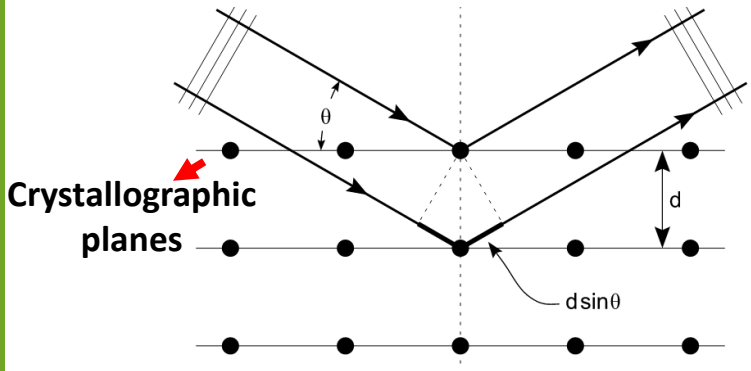
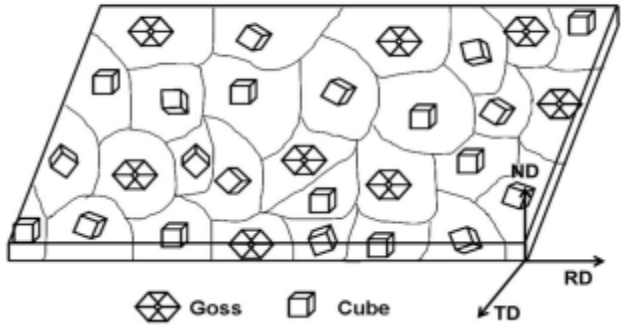
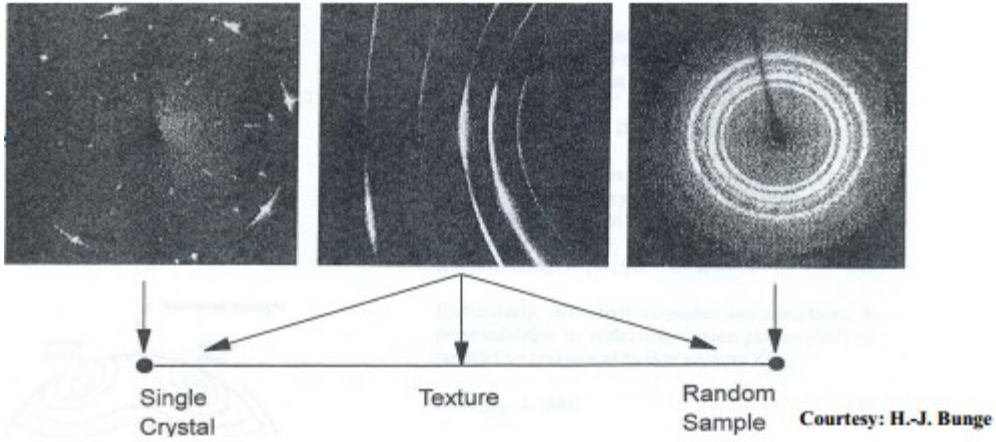
Crystal Structure: Background

X-rays are used to produce the diffraction pattern because their wavelength λ is typically the same order of magnitude (1–100 angstroms) as the spacing *d* between planes



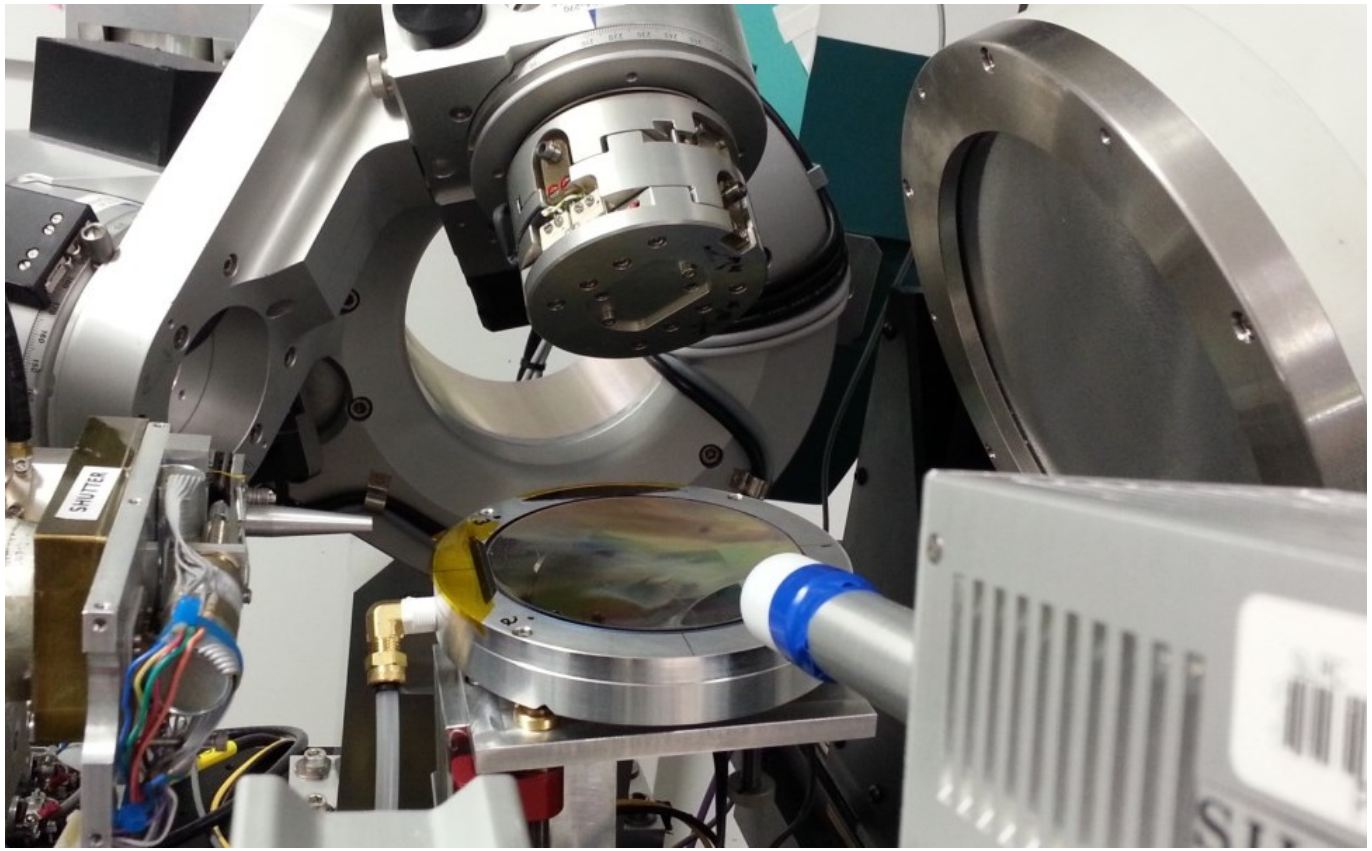
$$n\lambda = 2d\sin\theta$$





High-Throughput Experimentation methods for determining structure

High – Throughput X-Ray Diffraction Setup



Current setup: 1,000-2,000 samples/day
x10 flux -> 10,000 samples/day
Faster detector -> 100,000 samples/day
May become motor speed limited

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Data Storage

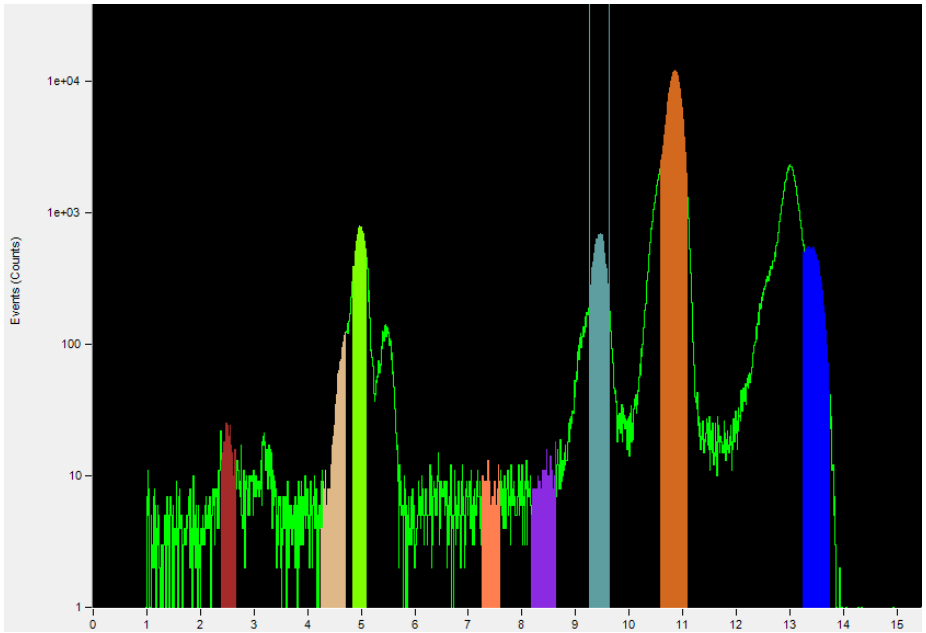
XRD

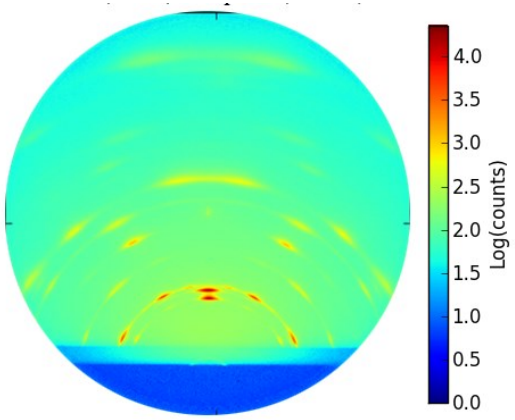
- XRD image (~4MB/sample)
- XRD unwarped image (~4MB/sample)
- XRD 1D pattern (~15kB)

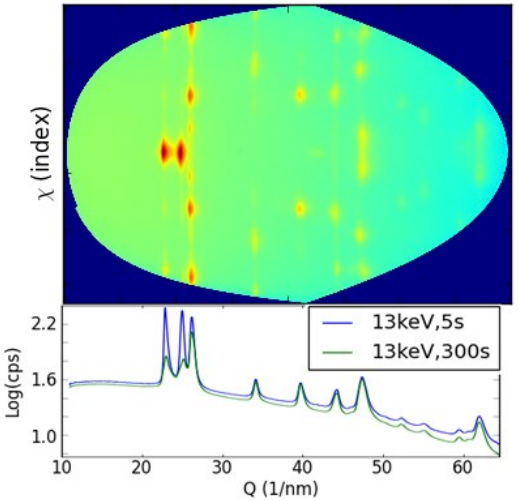
XRF

- SCA, set of ~10 intensities (10)
- MCA, full fluorescence spectrum (~16KB)

Daily data throughput ~10GB
Eventually:10TB







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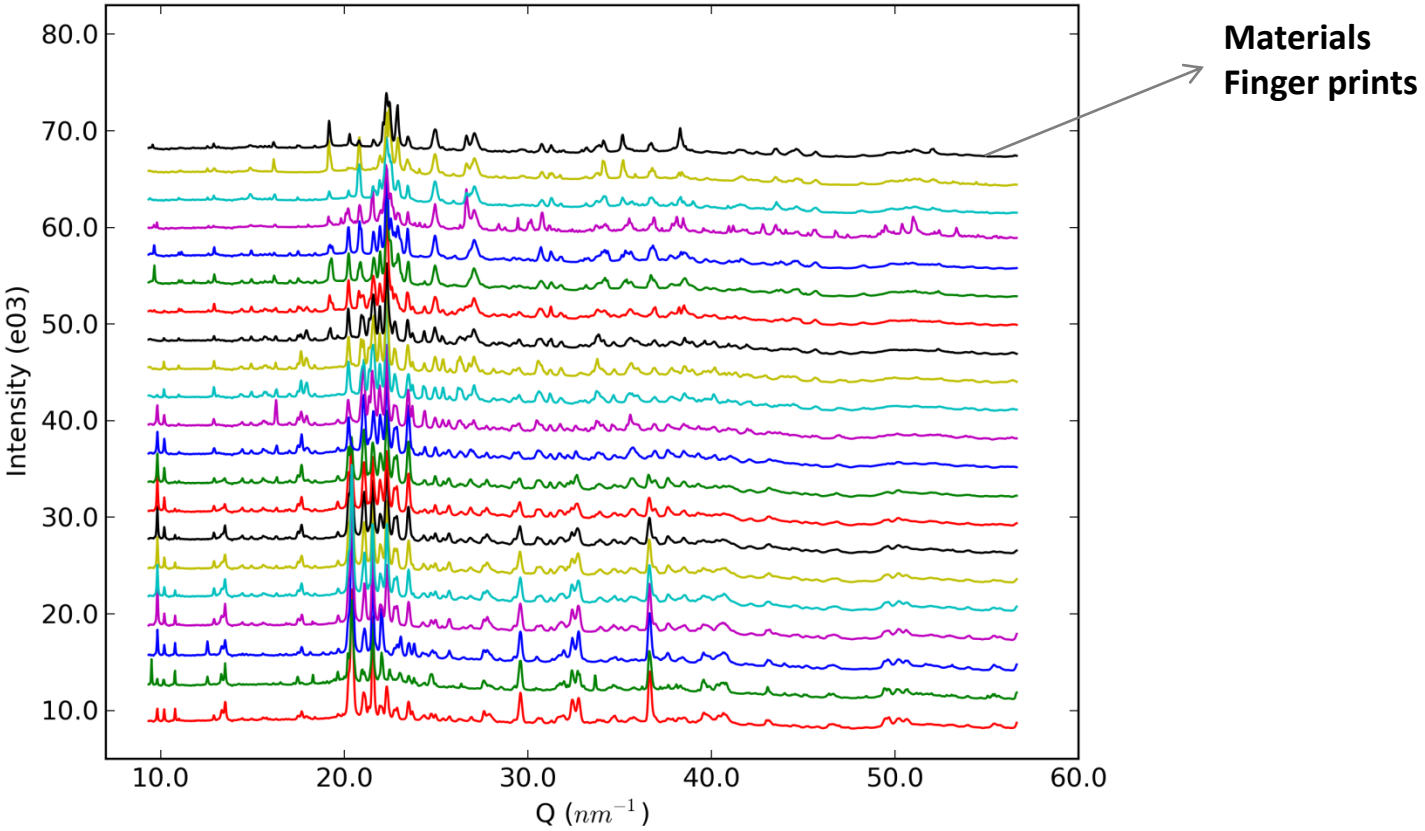
X-RAY DIFFRACTION DATA ANALYSIS

- Grouping compositions with similar XRD patterns into individual clusters.
- Identify the patterns that form the basis for the observed XRD patterns.

MATERIALS GENOME



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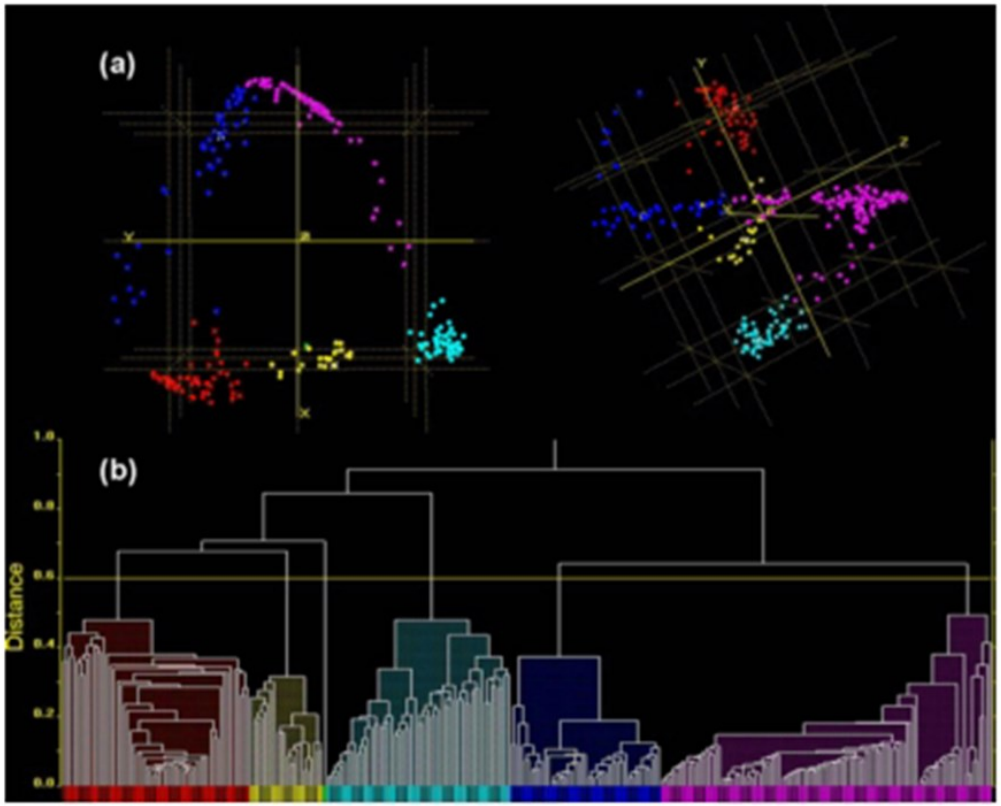
Correlation Clustering

❖Correlation coefficients are dominated by the correlation amongst intense peaks whereas information regarding growth of minor peaks is lost.

❖Peak shifts due to continuous variation of lattice parameter with composition within a phase region is not considered.

❖Amorphous materials with small crystallinity is not properly analyzed.

❖Identification of basis patterns is not considered.



Barr, G.; Dong, W.; Gilmore, C. J. *Journal of Applied Crystallography* 2009, *42*, 965–974.

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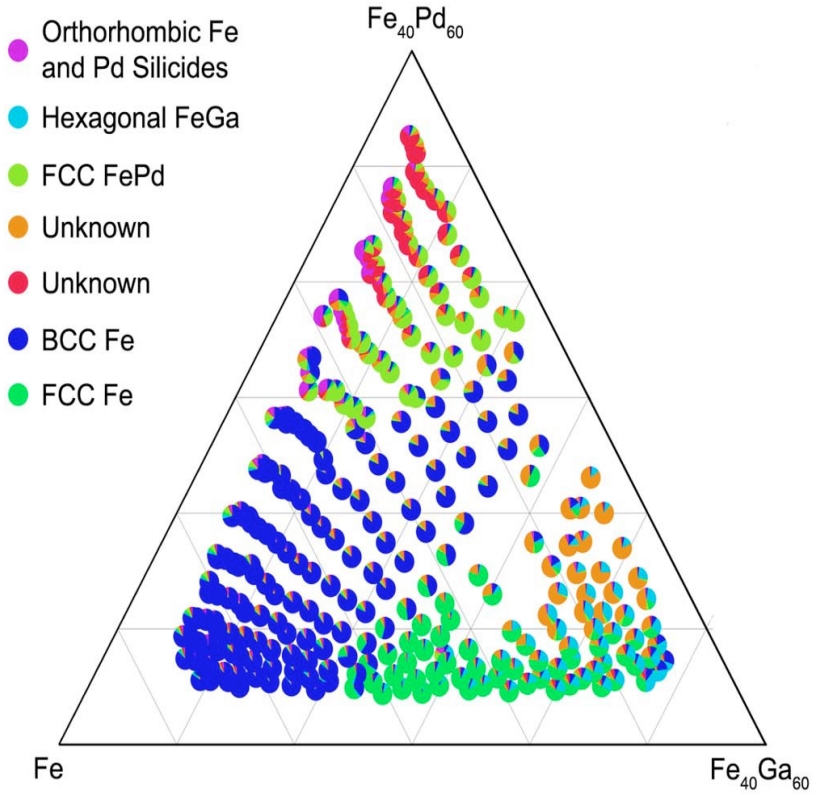
Non-Negative Matrix Factorization

- ❖Information regarding growth of small peaks is misinterpreted/lost.
- ❖Amorphous materials with small crystallinity is not properly analyzed.
- ❖Linear factorization does not take peak-shifts into account.
- ❖Spurious basis patterns due to misinterpretation of peak shifting.



Long, C. J.; Bunker, D.; Li, X.; Karen, V. L.; Takeuchi, I. *Rev. Sci. Instrum.* 2009, *80*, 103902.

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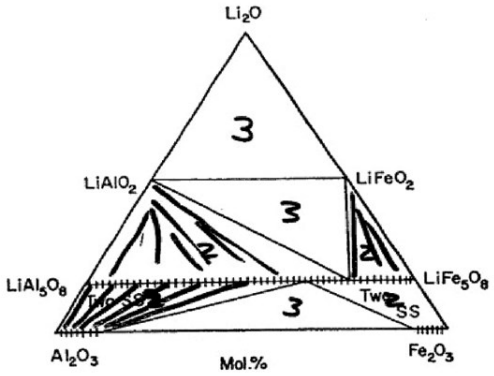
CONSTRAINT PROGRAMMING

Objective: Identification of K basis patterns given M observed patterns.

- Number of basis patterns is known apriori.
- The presence of one-peak within a shift width in several observed patterns is used as the basis for identifying basis patterns.
- Penalties are posed for peaks present in basis patterns but missing in observed patterns.
- Every peak has to be explained by some basis pattern.



Constraints

Maximum Missing Bounds, maximum number of phases, Shift Continuity, Shift Monotonicity, Zero-degrees of freedom regions, phase connectivity,

Lebras, R.; Damoulas, T.; Gregoire, J. M.; Sabharwal, A.; Gomes, C. P.; Dover, R. B. Van. *Proceedings of the 17th international conference on Principles and practice of constraint programming* 2011, 508–522.

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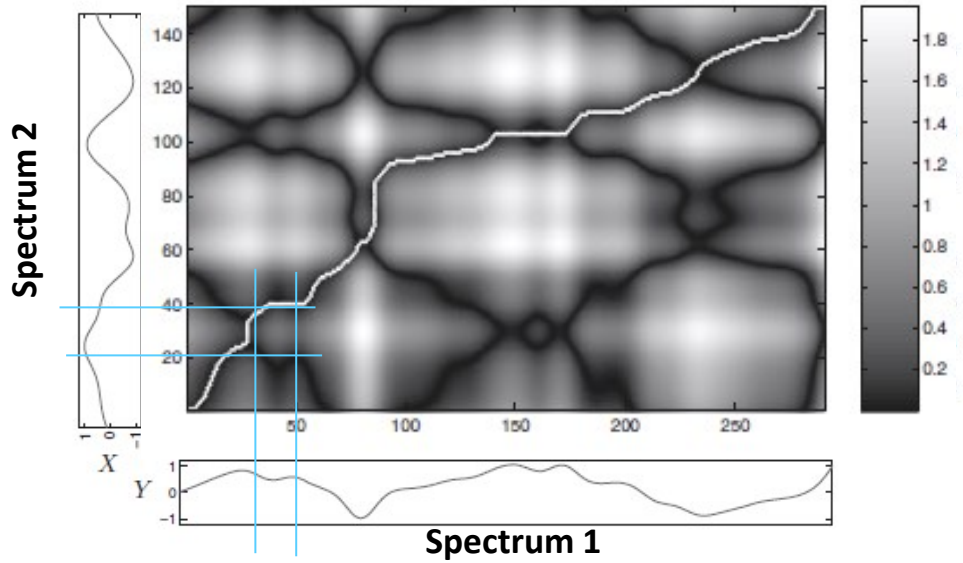
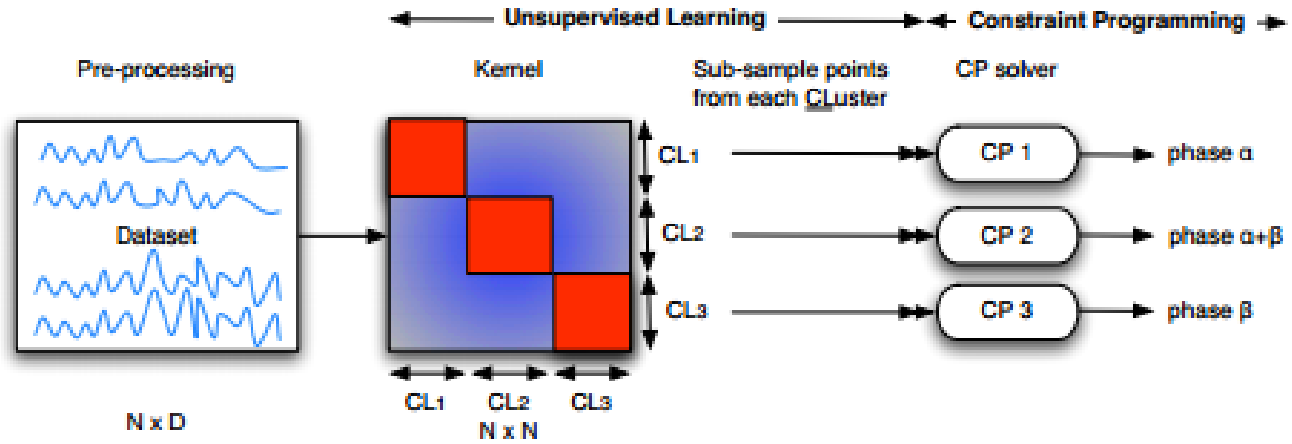
CONSTRAINT PROGRAMMING





Schematic representation of the integration of CP and ML.

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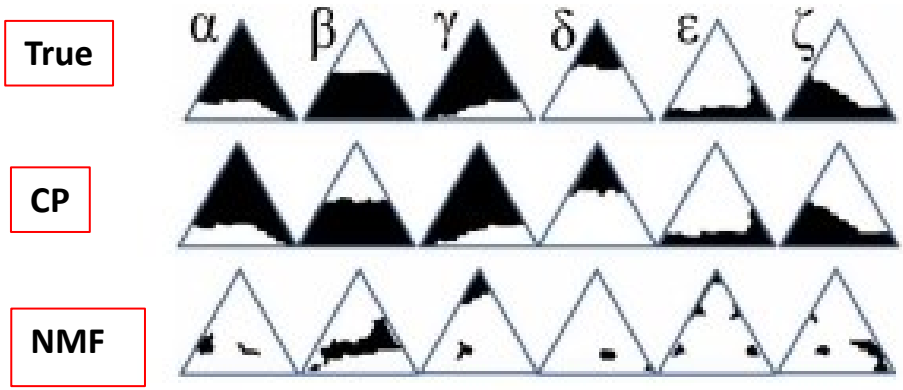

Precision: Fraction of retrieved results that are relevant.
Recall performance: Fraction of relevant results retrieved
Hybrid-CP: 77.4% / 84.2%.
NMF: 39.5% / 77.9%.

Results: appearance (white) or not (black) of the 6 phases underlying the Al-Li-Fe system. Top: the true values. Middle: phases found by our hybrid method. Bottom: phases found by the competing NMF approach.