Machine Learning and Global Optimization for Materials Discovery

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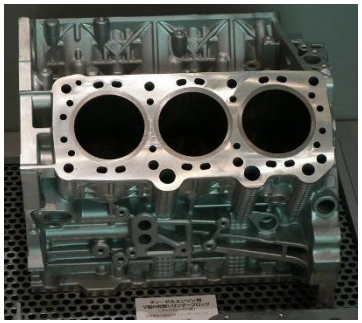
Materials Engineering: A narrow introduction

Examples of “Aluminum”

Application				
Key Properties	Conductivity	Low cost Shapeable	Castable Strong	???
Alloy	**Al + Mg**	**Al + Mg,Mn**	**Al + Si,Cu**	???

How do we tailor materials for new technologies?

How can we do this quickly?

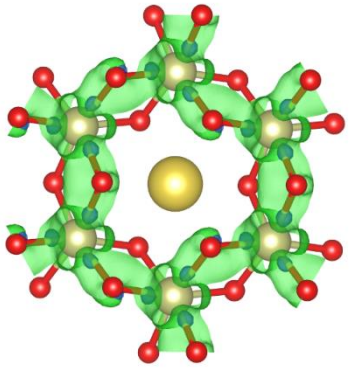
Computational Materials Engineering

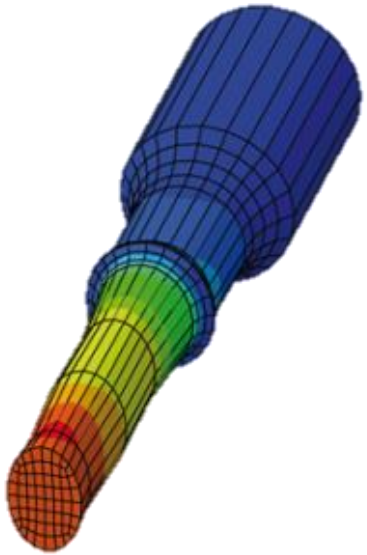
Goal: Accelerate design of materials

Method: Replace experiments with computers

Many Established Tools:

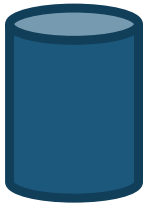
- Density Functional Theory
- Phase Field
- Finite Element Analysis
- Computational Thermodynamics





Emerging Field: **Data-driven Models**

Materials Data



Machine Learning

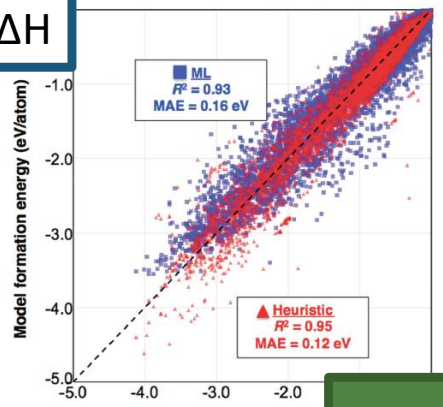
$$\sigma_Y = f(x)$$

Predictive Model

FEA Image: http://www.icams.de/content/research/index.html

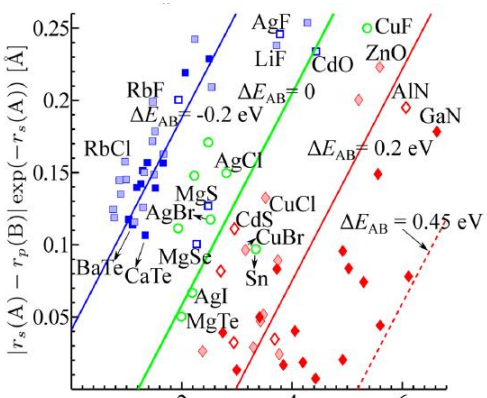
ML + Materials = “Materials Informatics”

ΔH

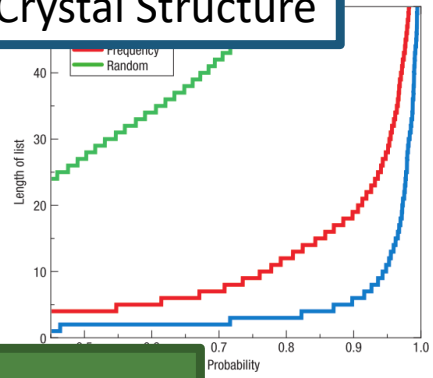


Meredig *et al*. PRB (20

ΔH



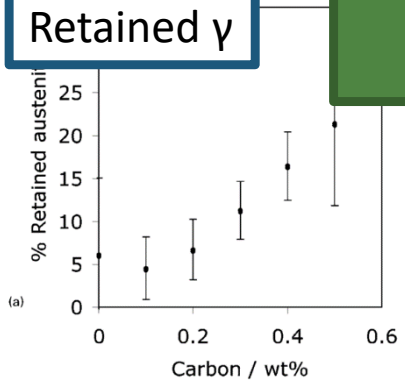
Crystal Structure



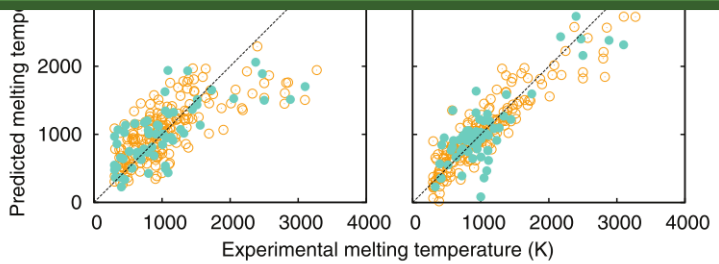
Nat. Mat. (2006), 641

Question: How can I do this, but for many more properties?

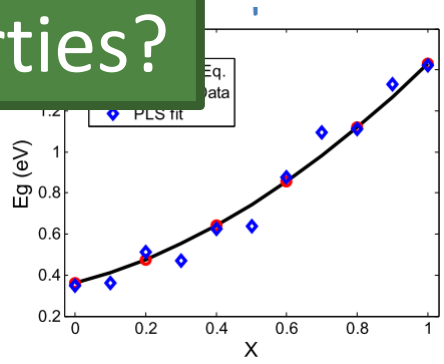
Retained γ



Chatterjee *et al*. MS&T (2007), 819



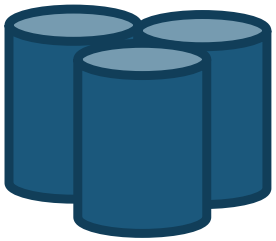
Seko *et al*. PRB (2014), 054303

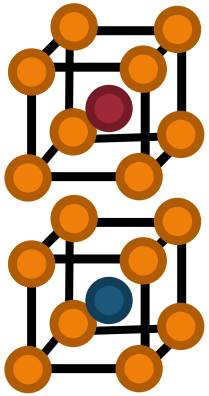


Srinivasan, Rajan. *Materials* (2013), 279

Materials Informatics Workflow

Collect → Process → Represent → Learn





$\Delta H_f = -1.0$

$\Delta H_f = -0.5$

$\vec{X}$ $\vec{y}$

Z_A	Z_B	ΔH_f
3	4	-1.0
3	5	-0.5

$$\Delta H_f = f(Z_A, Z_B)$$

Materials Informatics Challenges

Need: High-quality data

Question: How to simplify using ML in MSE?

Part 1: Automated Crystal Structure Solution

Part 2: General-Purpose Representations for Materials

Part 3: Design of New Materials

Part 1: Automated Structure Solution

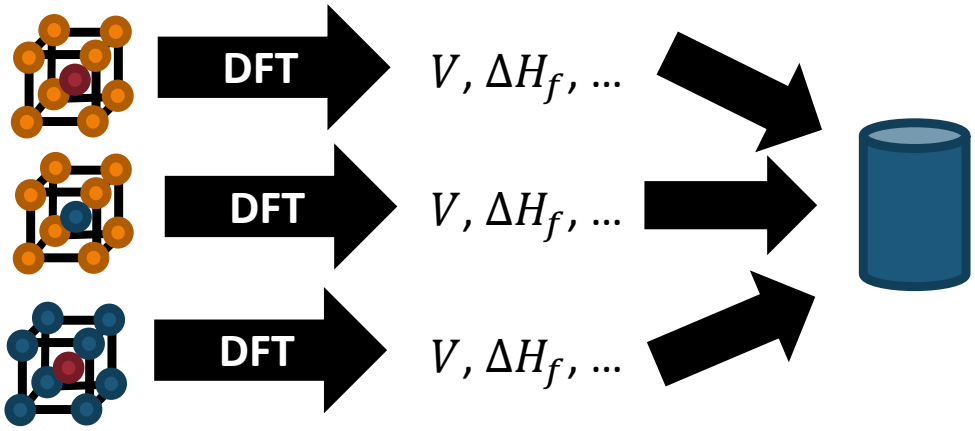
Acknowledgements: Kyle Michel

Why is this important?

One Reason: High-Throughput DFT

Automated calculation of properties

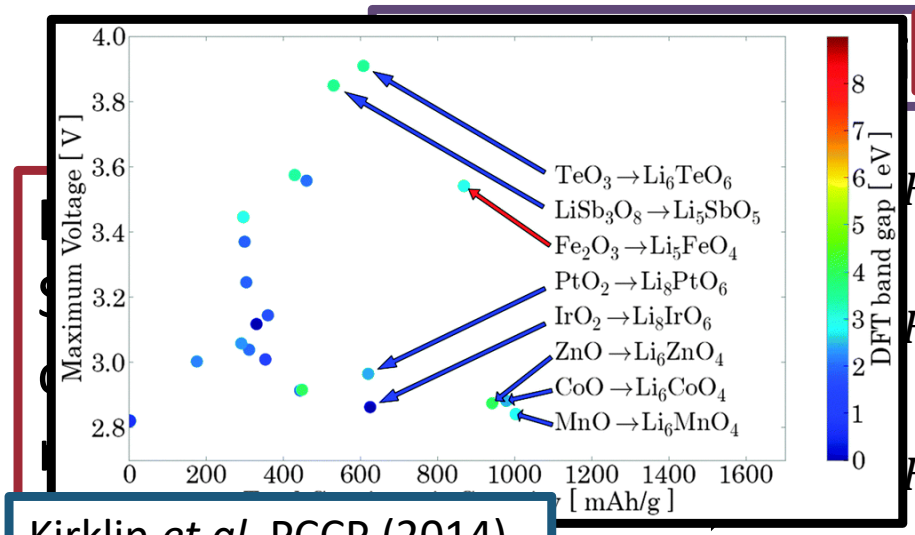
Input:
Structures of known materials



Output:
Large collection of materials data

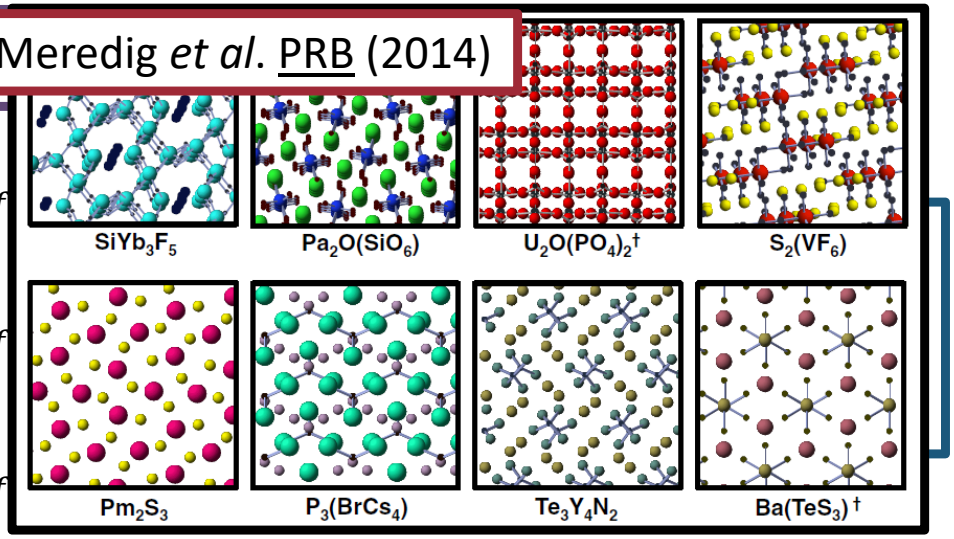
Why is this important?

One Reason: High-Throughput DFT



Kirklin *et al*. PCCP (2014)

Meredig *et al*. PRB (2014)



Any HT search is limited by the availability of high-quality crystallographic data!

Problem: Structures unknown for many materials

Crystal Structure Databases

FIZ Karlsruhe

ICSD

Inorganic Crystal Structure Database

Collection of the crystal structures of compounds

Total entries: 148818

Missing atoms: 13705 (~9%)!

Powder Diffraction File

Database of Powder X-Ray Diffraction patterns

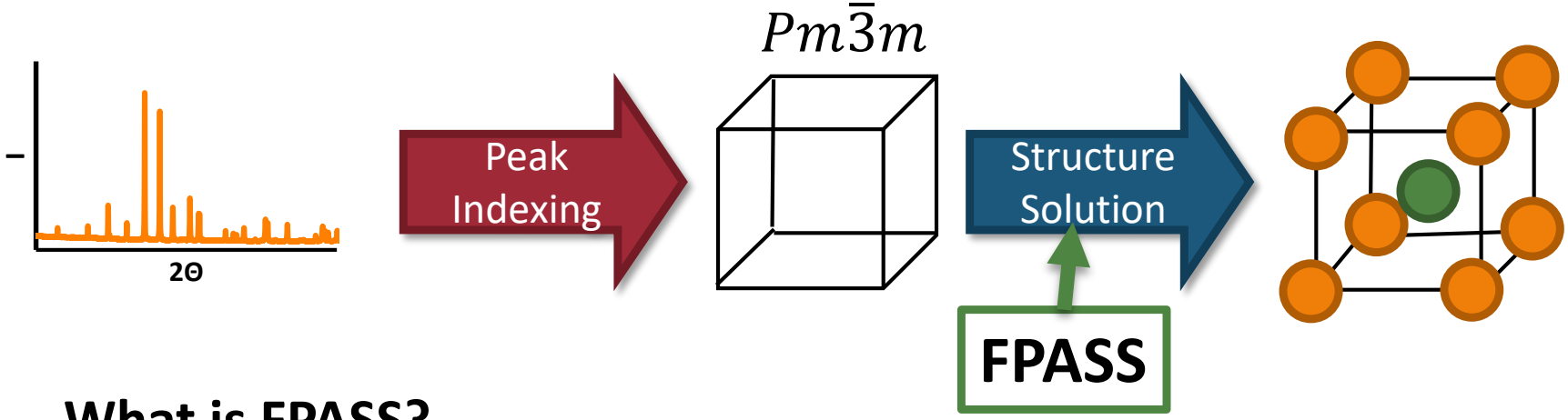
Total entries: 384613

Without structure: 113164

10^3 – 10^5 of materials not included in HT DFT Databases!

Proposed Solution: Solve them with FPASS

First-Principles-Assisted Structure Solution



What is FPASS?

Simple: Single-step method to solve structure

Accurate: Uses both energy and diffraction pattern match

Fast: Constrains search with known symmetry

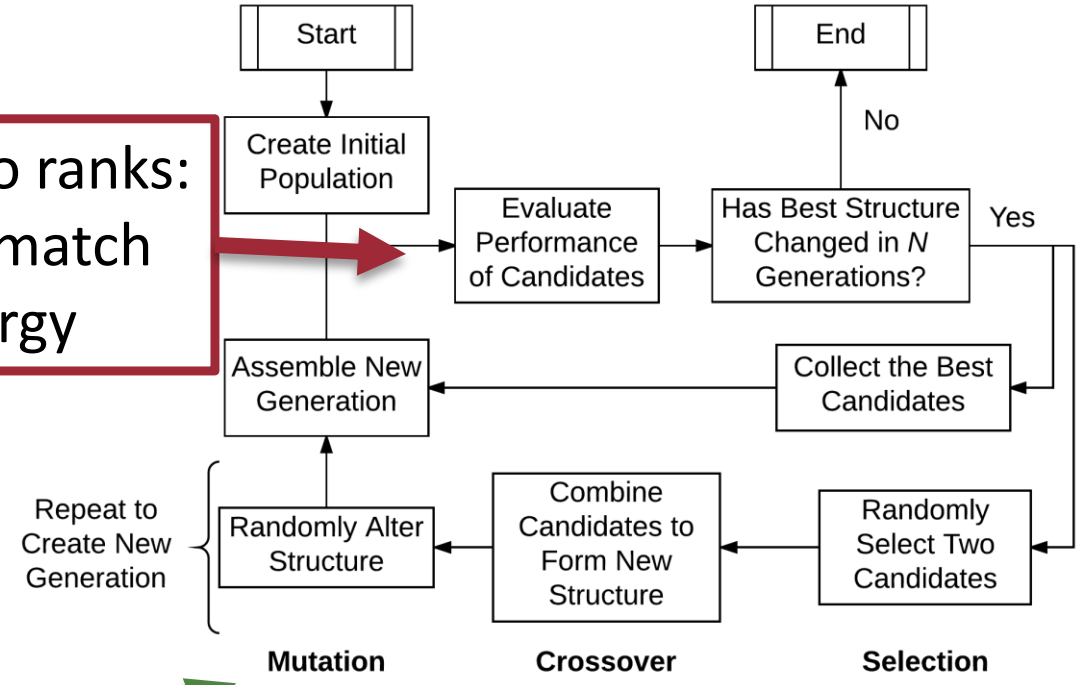
Problem: We had not used FPASS for *new* solutions

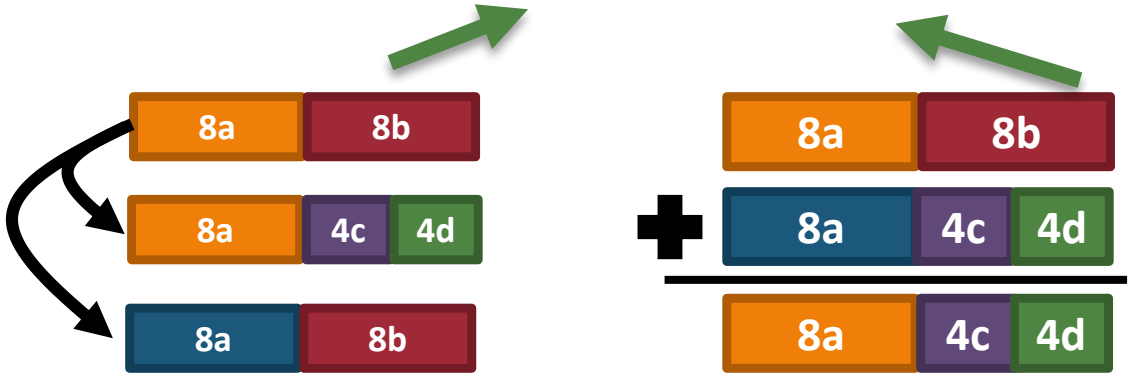
- Can we trust it in an automated scheme?
- Can we automate it?

Meredig and Wolverton. Nature Materials. (2013)

FPASS Uses a Genetic Algorithm

Sum of two ranks:
- Pattern match
- DFT energy





By maintaining Wyckoff sites, we preserve symmetry

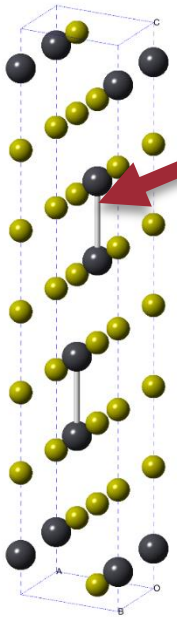
Solving Na_xPb_y Structures

Our starting point: Abstract from 1957

Phase	Ideal formula	Space group	a_0 (A)	c_0 (A)	Pb atom positions
I	Na_5Pb_2(?)	$R\bar{3}m$	5·54	23·15	6 (c): 0, 0, z; ...; z = 0·07
II	Na_9Pb_4	$P6_3/mmc$	5·47*	30·41	4 (e): 0, 0, z; ...; z = 0·05 4 (f): ⅓, ⅔, z; ...; z = 0·30
III	$Na_{13}Pb_5$	$P6_3/mmc$	5·51	40·39	2 (b): 0, 0, ¼; 0, 0, ¾ 4 (f): ⅓, ⅔, z; ...; z = 0·05 4 (f): ⅓, ⅔, z; ...; z = 0·13

indicating a true cell with $a_0' = \sqrt{3}.a_0$, $c_0' = c_0$.

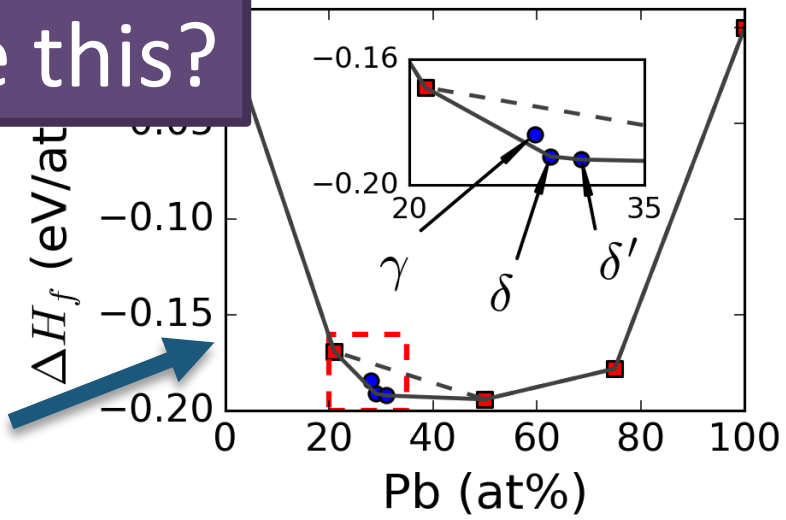
Only missing feature: Na positions



Can we automate this?

Na positions with FPASS

New structure improve computed Na-Pb diagram



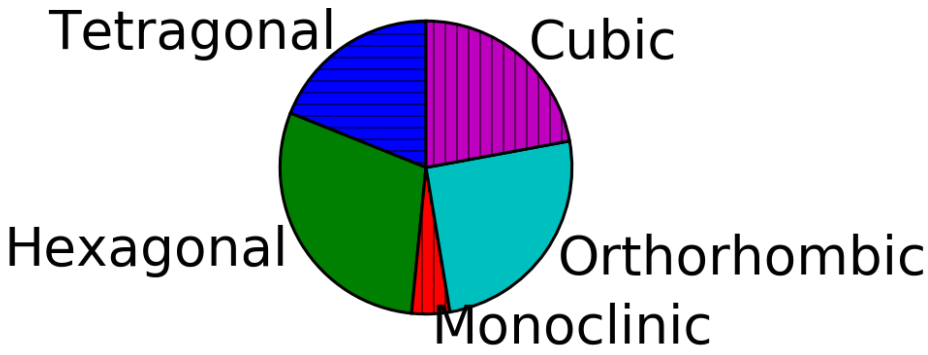
Computed T=0K Phase Diagram

Ward, Michel, Wolverton. Acta Crystal. A17 (2015), 542

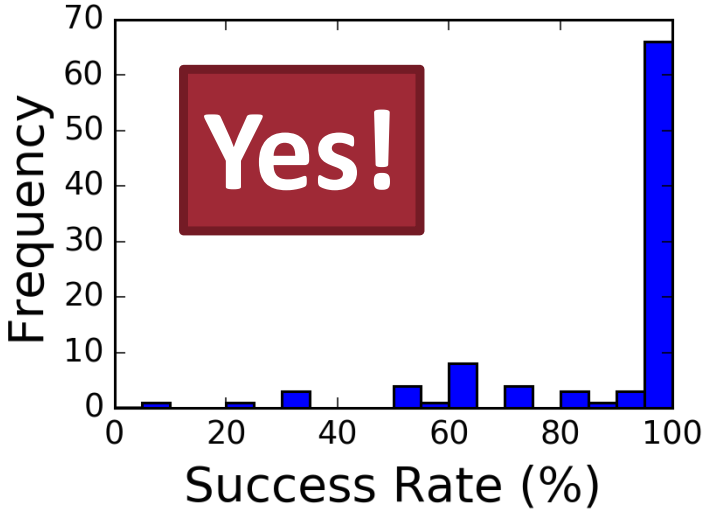
Can we trust automated FPASS?

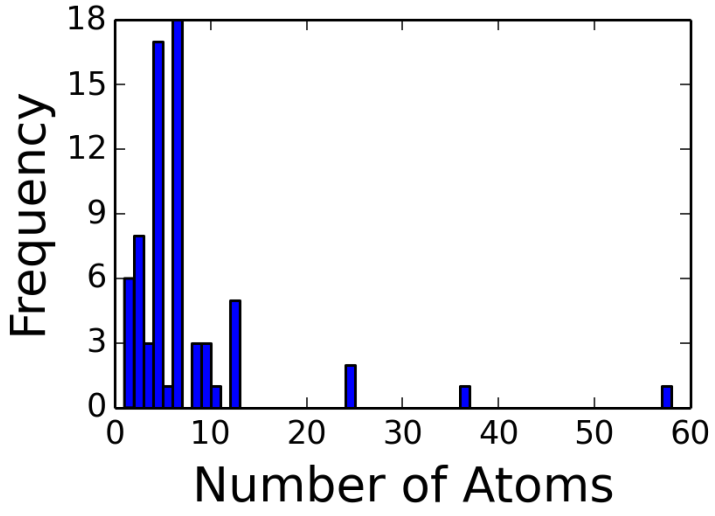
Test Cases: Strukturbericht

- Common structures
- ~100 compounds
- Variety symmetries, compositions, sizes

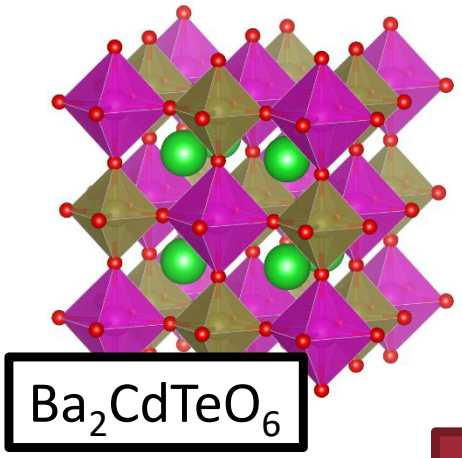


Can we solve them?

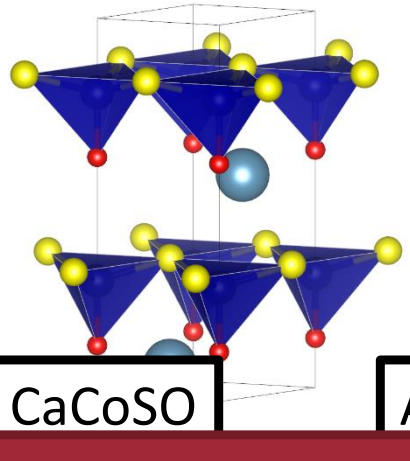




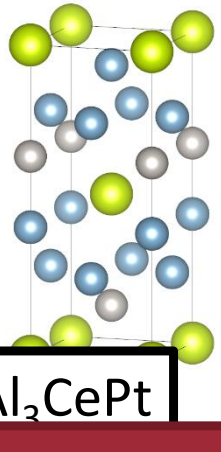
New Solutions



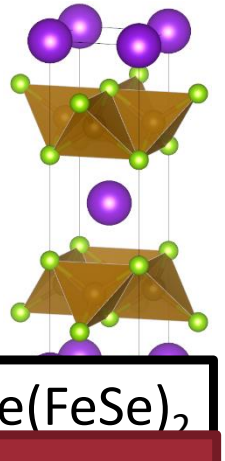
Ba_2CdTeO_6



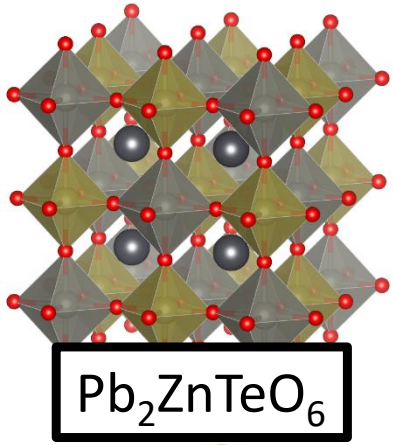
CaCoSO



Al_3CePt

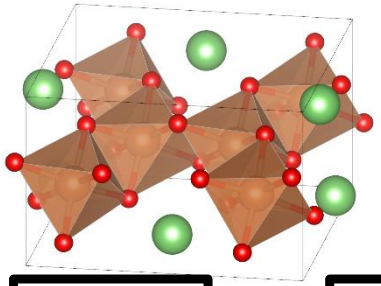


$Ke(FeSe)_2$

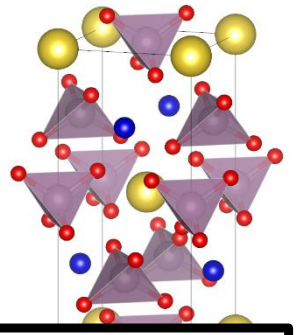


Pb_2ZnTeO_6

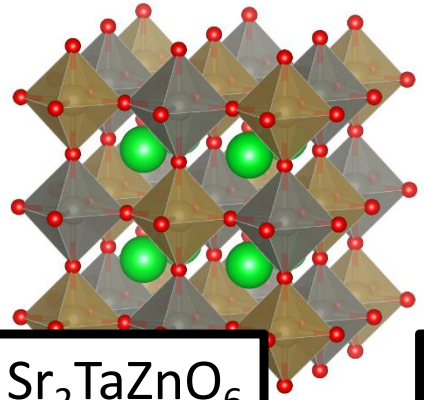
And there are more on the way!



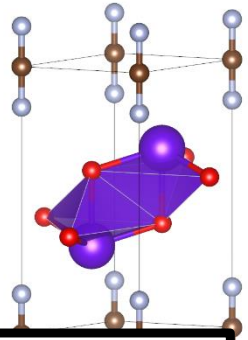
$LiSbO_3$



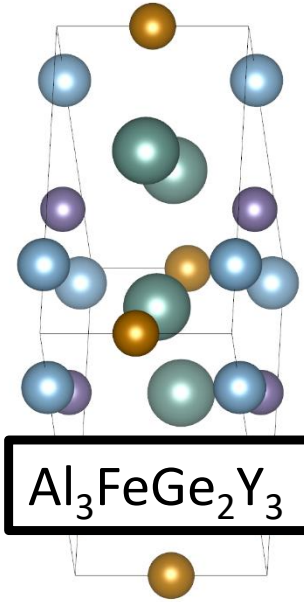
$NaTm(MoO_4)_2$



Sr_2TaZnO_6



$Tb_2O_2CN_2$



$Al_3FeGe_2Y_3$

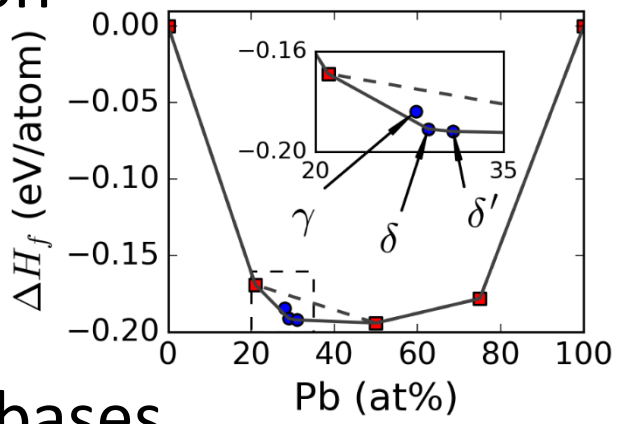
Ward, Michel, Wolverton, *in review*

Summary

Goal: Reduce the number of unsolved compounds

Method: First-Principles-Assisted Structure Solution

1. Created automated implementation
2. Validated against ~100 structures
3. Solved 13 crystal structures



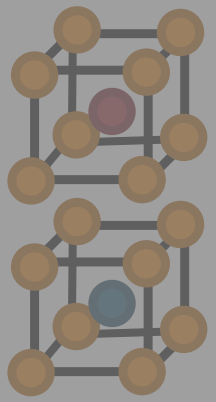
Result: Improved accuracy of HT-databases

Ward, Michel, Wolverton. Physical Review Materials, *in review*

Part 2: General-Purpose ML Methods for Materials

Materials Informatics Workflow

Collect → Process → Represent → Learn



$\Delta H_f = -1.0$

$\Delta H_f = -0.5$

$\vec{X}$ $\vec{y}$

Z_A	Z_B	ΔH_f
3	4	-1.0
3	5	-0.5

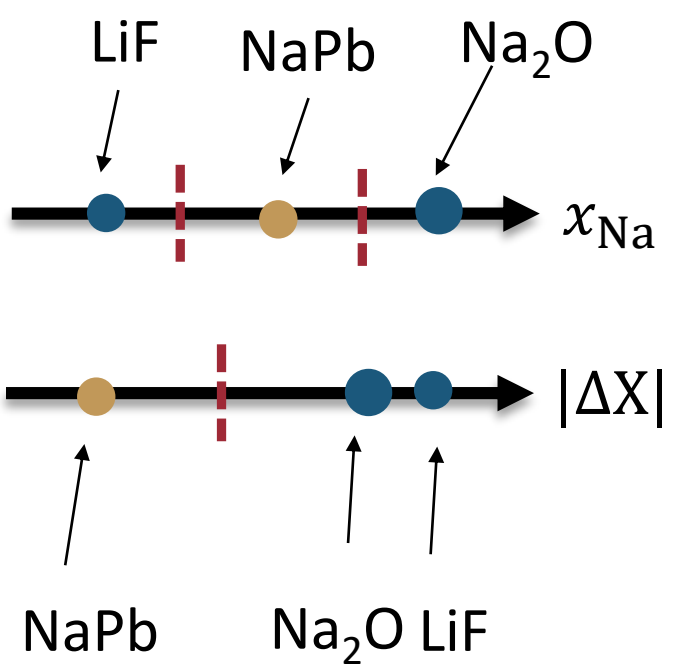
$\Delta H_f = f(Z_A, Z_B)$

How can one create problem-independent representations?

What is a representation?

Set of quantitative attributes that describe a material

$$Property = \boldsymbol{f}(Attributes)$$



Representation of material

Ex: *Attributes* = $\boldsymbol{g}(x_H, x_{He}, \ldots)$

What does a representation need?

Completeness: Differentiate materials

Efficiency: Quick to compute

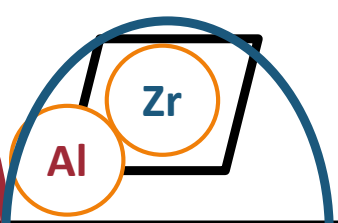
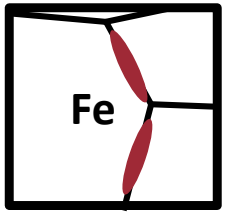
Accuracy: Capture important effects

Diversity: Many possible properties

How do we create "general-purpose" representations?

Variety of Types of Materials Data

How to differentiate materials?

	Zr	Al,Zr	Al_4Zr		
Available Information	Element	Phase Diagram	Composition	Crystal Structure	μ-Structure
Example Properties	Dilute ΔH_{mix}	# Eutectics	Glass-Forming Ability	ΔH_f	σ_Y

Need: A suite of general-purpose representations

Focus #1: Composition

$$Property = f(Composition)$$

Property	Attributes	Reference
Crystal Structure	VE, ΔX, n_{av}, $\Delta n_{ws}^{1/3}$	Kong et al., 2012
Band Gap	ΔX, Z, T_m, R, n_{av}	Srinivasan & Rajan, 2013
Formation Energy	ΔX, Z, $n_{s\|p\|d\|f}$, row, col	Meredig *et al.*, 2014
Melting Point	Z, *m*, *n*, r^{cov}, *I*, *X*, …	Seko *et al.*, 2014
ΔH_f: Rocksalt – Wurtzsite	IP, EA, r_s, r_p, …	Ghiringhelli *et al.*, 2015

Observations:

- Different properties, different attributes
- All based on elemental property statistics

Our Strategy: Create set that includes all of these and more

General-Use Attributes

Elemental Property Stats.: *Mean T_m, Range Z, …*

6 Statistics: Mean, variance, max, min, range, mode

22 Elemental Properties: Z, EN, Row, Column, Radius, …

Stoichiometric: *# Components,* $\|x_Z\|_p$

Electronic Structure Based: *Fraction p Electrons, …*

Ionicity: *Can form Ionic, % Ionic Character, …*

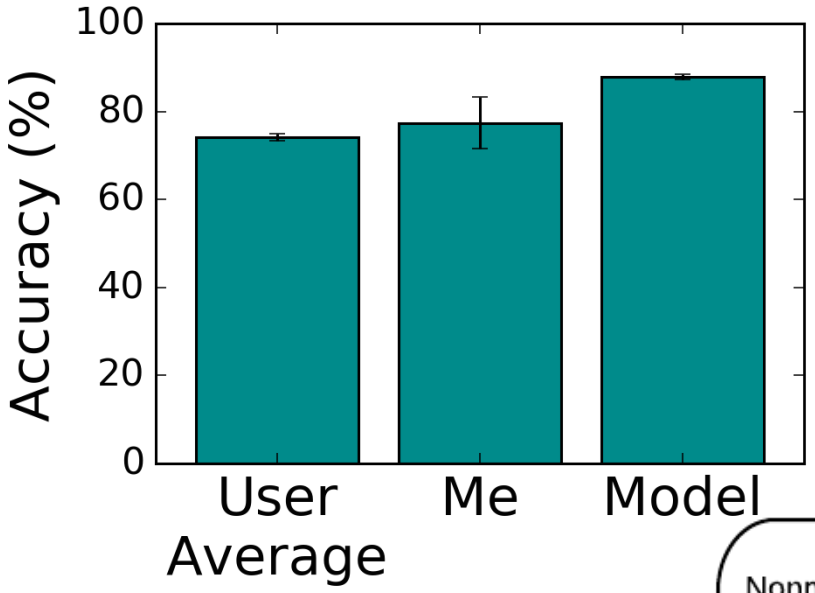
Ref: Ward *et al*. npj Comp. Mat., (2016) 28

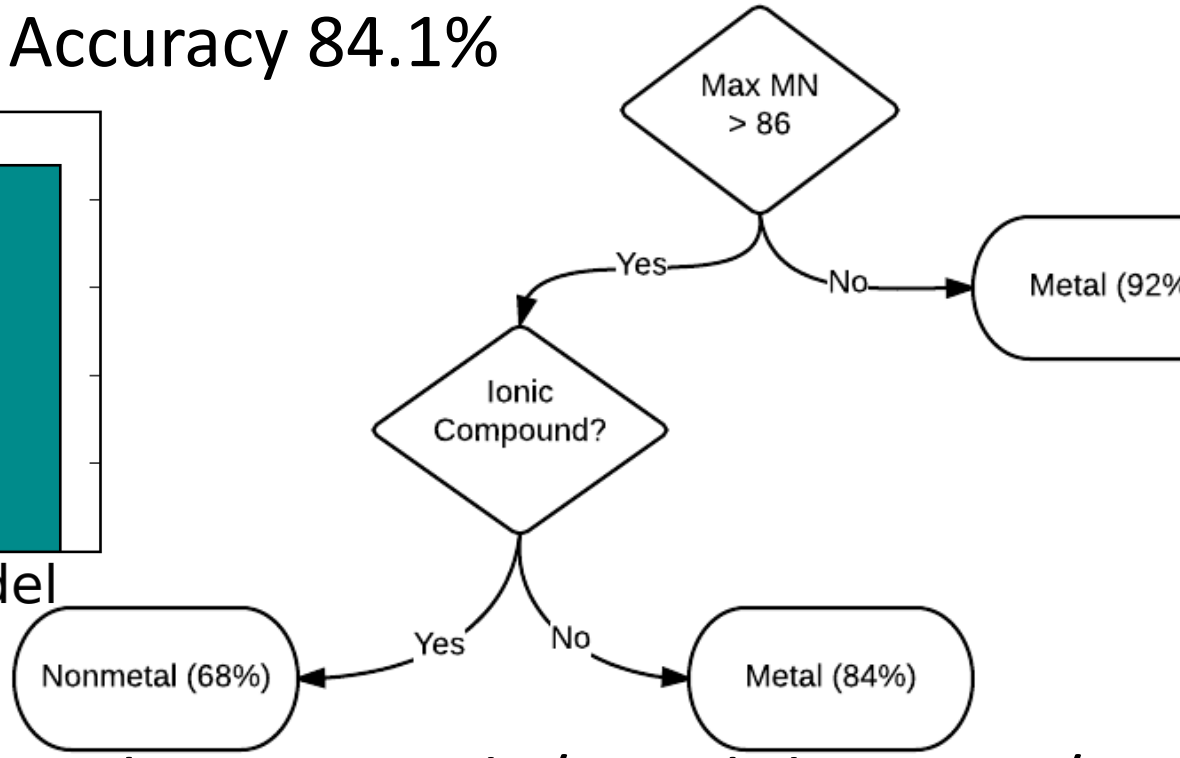
Simple Example: Is it a Metal?

Task: Given composition, $E_g > 0$?

Training Set Dataset: 3000 entries from the OQMD

Simple ML Model: Accuracy 84.1%



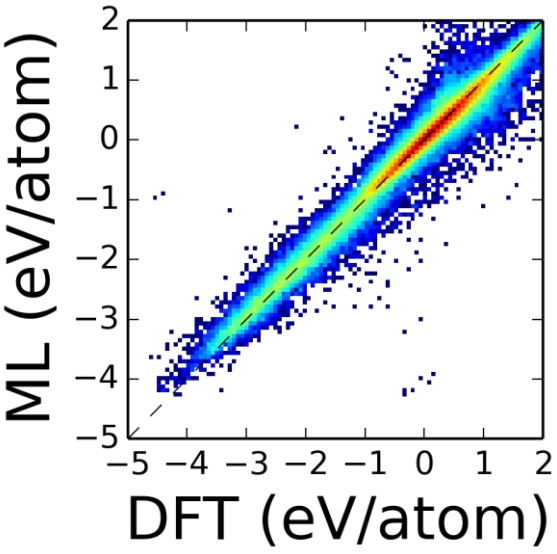


Game: palestrina.northwestern.edu/metal-detection/

Application to the OQMD

Dataset: 240000 DFT Calculations (OQMD.org)

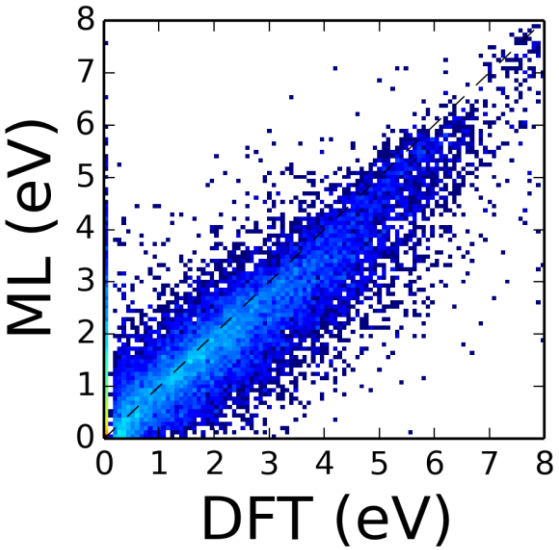
ΔH_f



R: 0.944

MAE: 80.5 meV/atom

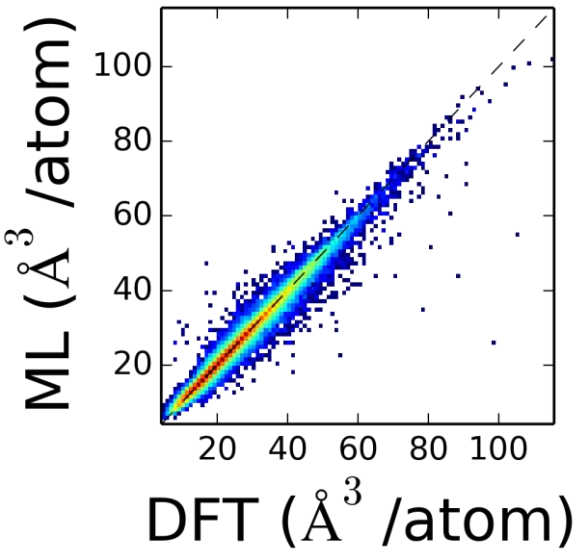
E_g



R: 0.924

MAE: 0.21 eV

V



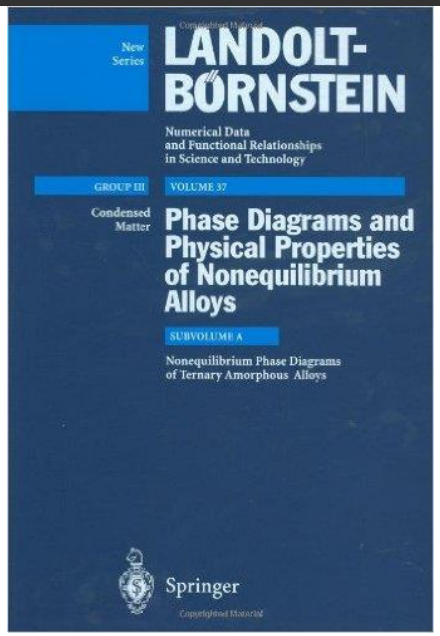
R: 0.993

MAE: 0.452 Å^3/atom

Ref: Ward *et al*. npj Comp. Mat., (2016) 28

Predicting Glass Forming Ability

Application: Metallic Glasses



Goal: Predict glass-forming ability

Dataset: Landolt-Börnstein

- 6836 experimental measurements
- 295 ternary systems
- Binary property: **[Can Form Glass]** | **[Cannot Form]**

Model: Random Forest

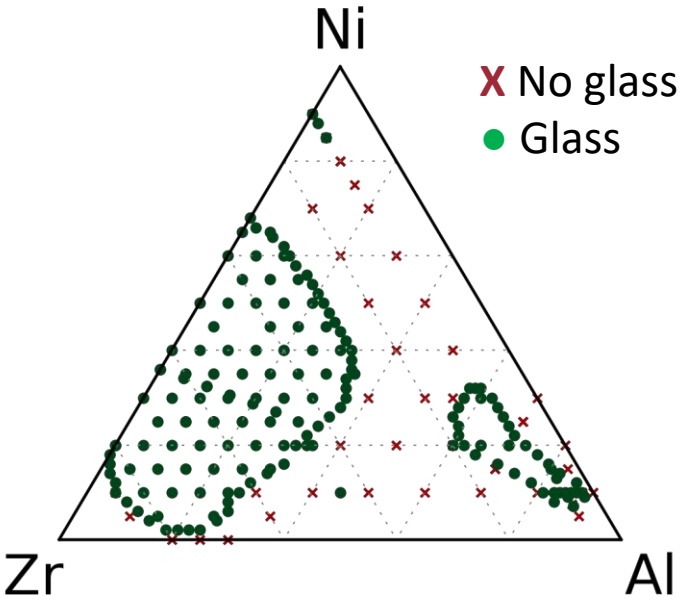
- 90% accurate in 10-fold cross-validation

Ref: Ward *et al*. npj Comp. Mat., (2016) 28

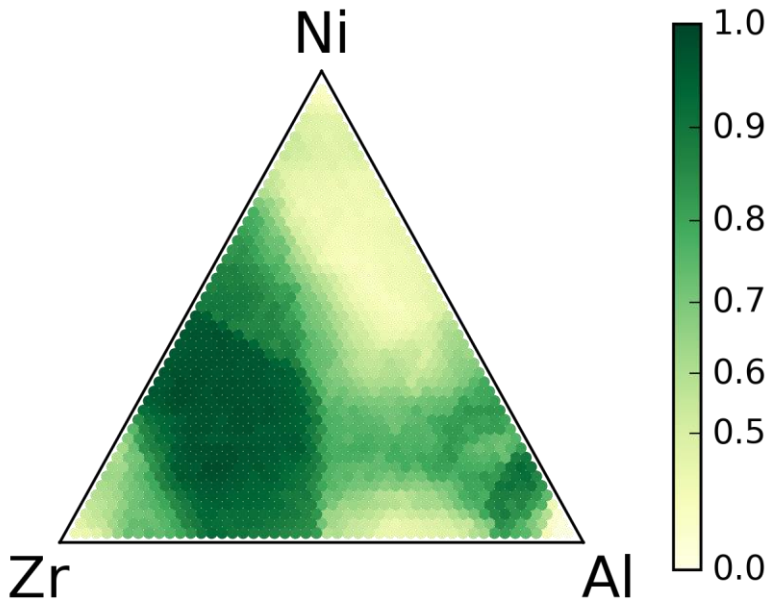
Predicting Glass-Forming Ability

Test: Remove Al-Ni-Zr data from training data, try to predict

Measured



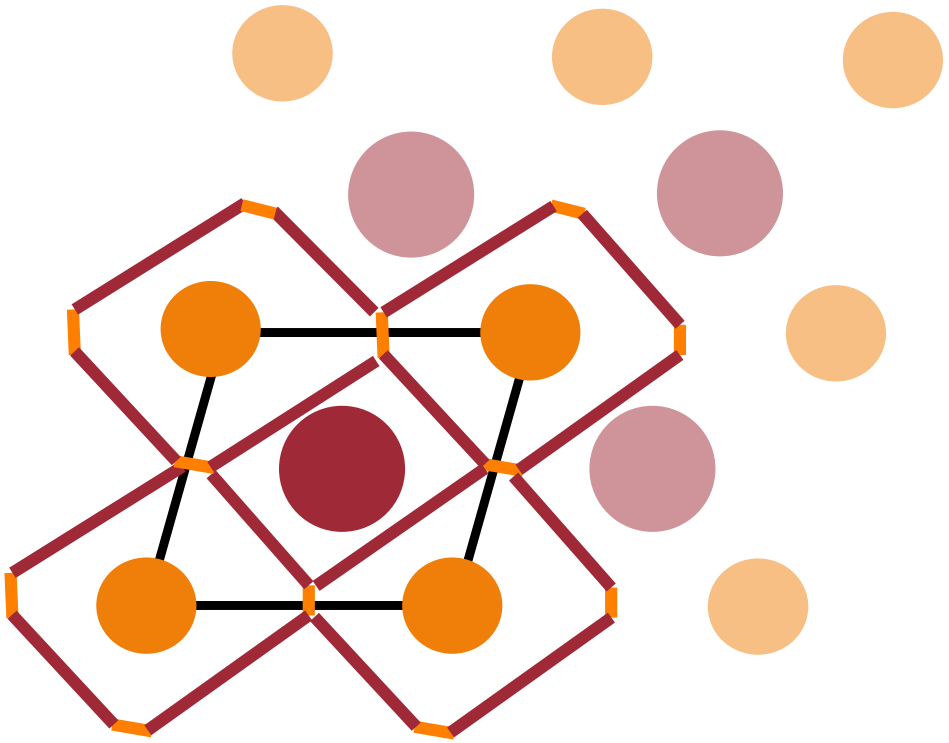
Predicted



Same representation, very different material.

Ref: Ward *et al*. npj Comp. Mat., (2016) 28

Focus #2: Crystal Structure



Our Approach:

Voronoi-tessellation-based attributes

Atomic Characteristics:

1. Element identity
2. Coordination number
3. Bond length
4. Cell size

…

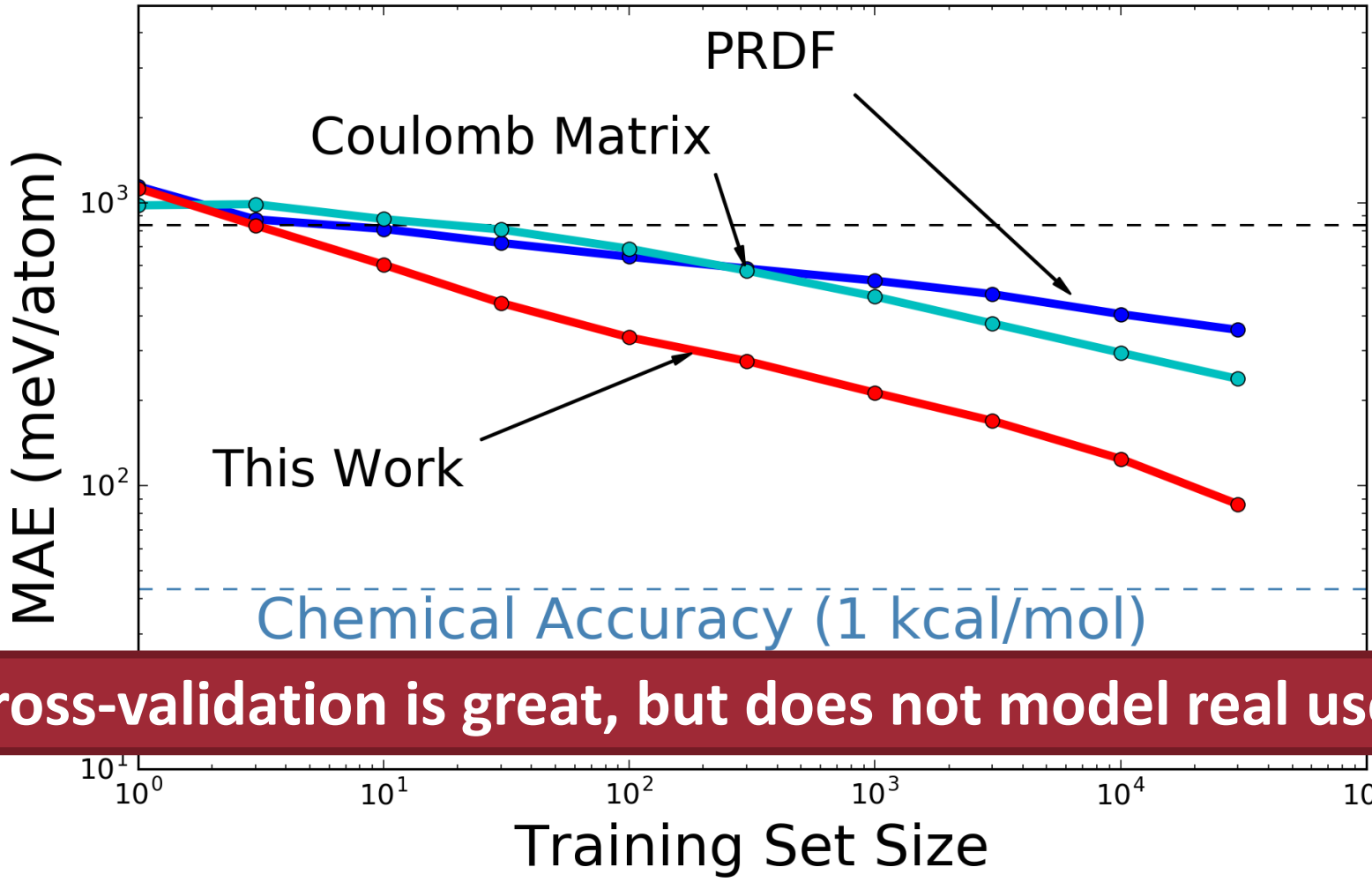
Atomic Characteristics + Descriptive Statistics = 275 Attributes

Ref: Ward *et al*. PRB. (2017) 024104

Learning Rate Comparison

Dataset: 32k DFT ΔH_f from the OQMD

Test: Remove 1000, train on *N* remaining



Cross-validation is great, but does not model real use

CM: Faber *et al*. Int J Quantum Chemisty. 2015; PRDF: Schutt *et al.* PRB. (2014)

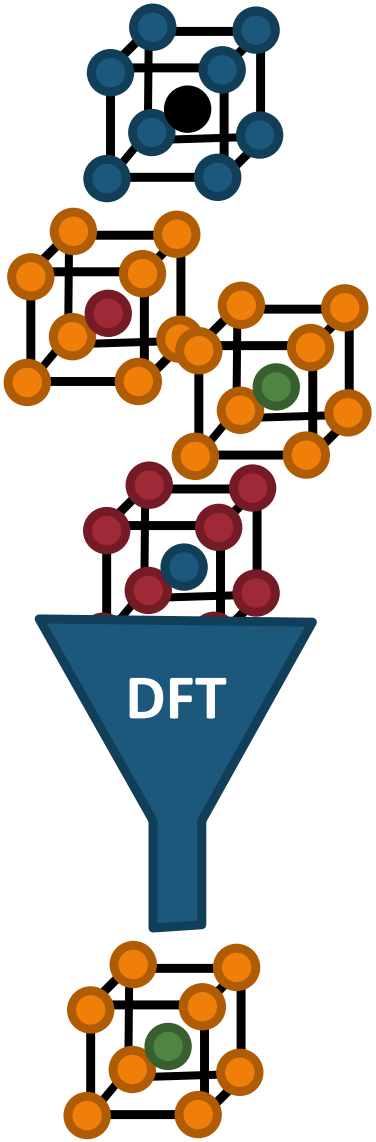
Application: The Prototype Search

Common Method: Prototype Search

1. Select a crystal structure
2. Evaluate **all** possibilities with DFT
3. Select only stable ones

Challenge: Computational cost

Possible Solution: Guide with ML

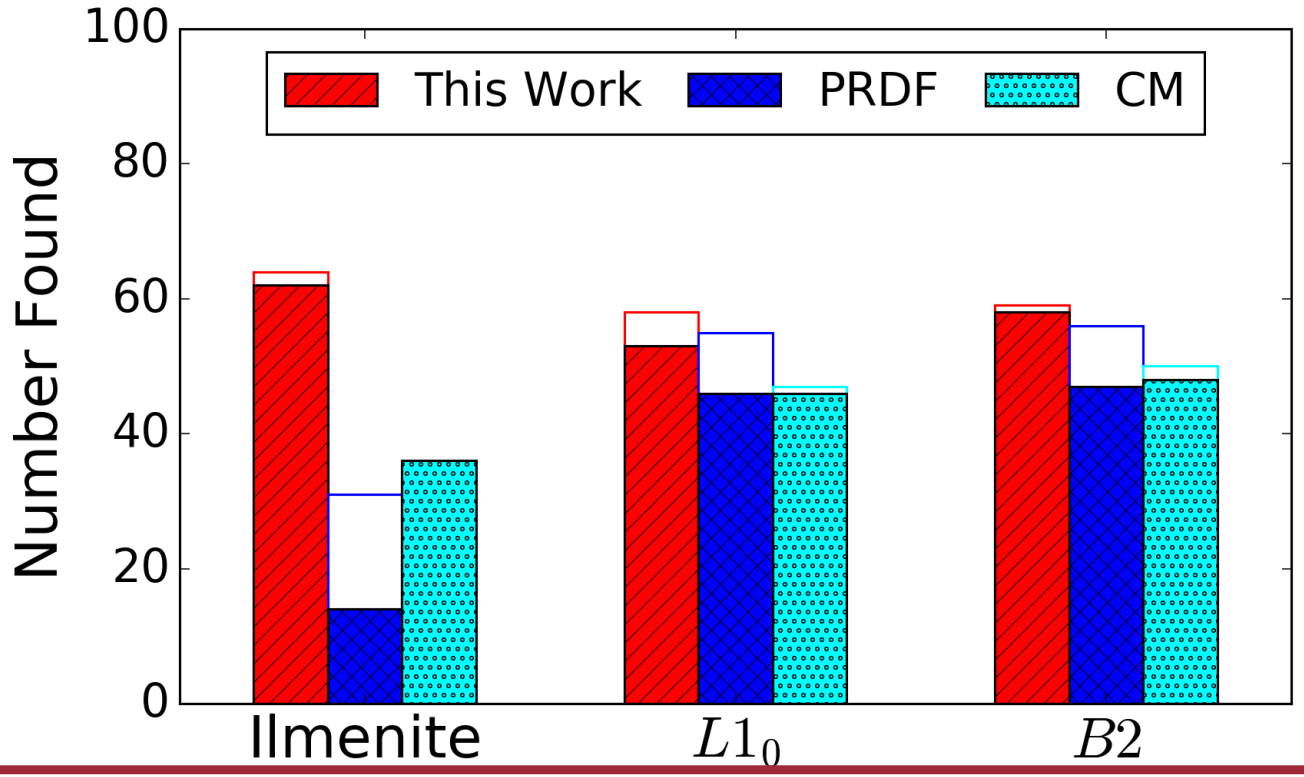


Ref: Ward *et al*. PRB. (2017) 024104

Ranking Candidate Materials

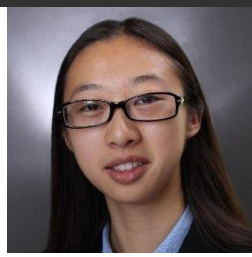
Training Set: 32k entries from OQMD

Test: Select top 100 entries from test set



Ranks entries better than existing methods, good choice for accelerating combinatorial searches

Future Steps: Open Benchmarks



Jiming Chen (UIUC)

Challenge: Which method is best for a certain dataset?

Our proposal: Develop a set of benchmark challenges

Approach: Host data, models using MDF and WholeTale

Current Progress: Compared several composition-based models

WholeTale.org

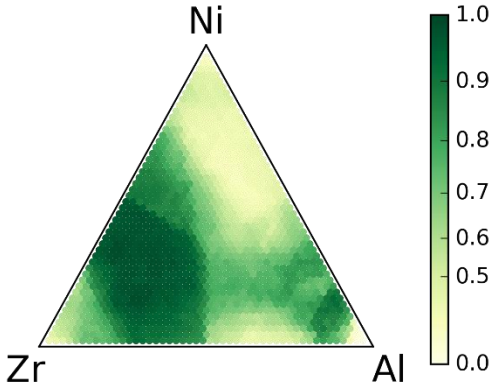
MATERIALS DATA FACILITY

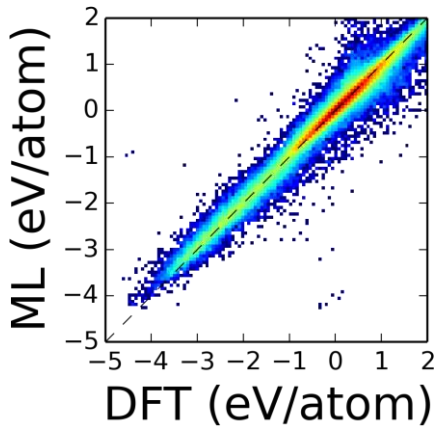
Datasets	Methods			
	Ward 2016	**Deml 2016**	**Meredig 2014**	**deJong 2016**
Full OQMD	**0.161**	0.180	0.214	0.187
Deml 2016	0.170	0.160	**0.144**	0.176
Meredig 2014	**0.072**	0.140	0.087	0.091

Summary

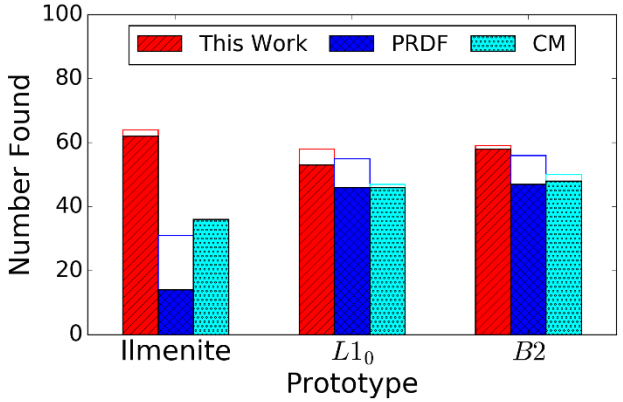
Collect → Process → Represent → Learn

Composition-Based Attributes





Crystal Structure Attributes



How do I use this to find a new material?

Part 3: Using ML To Find Materials

Acknowledgments:

Liquidmetal: S. O'Keeffe, J. Stevik, G. Jelbert

NU: M. Aykol, K. Kim, J. He

Example Design Problems

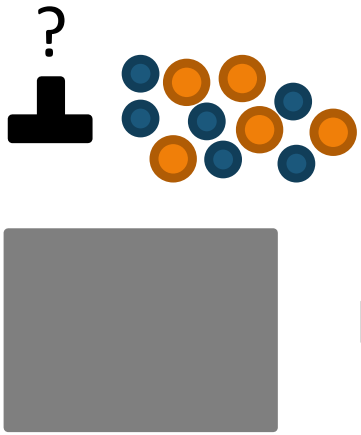
Designing Metallic Glasses Alloys

1. Optimizing commercial alloys
2. Locating new glasses with sputtering

Discovering Quaternary Heuslers

Application: Bulk Metallic Glasses

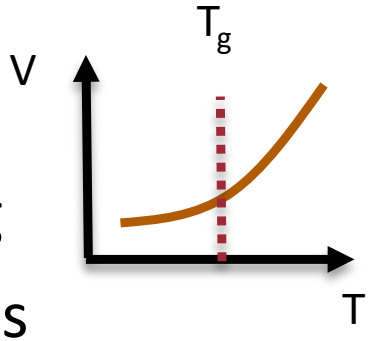
Metals with Amorphous Structures



No Dislocations -> High σ_Y

Glass Transition -> Net Shape Casting

No Grain Boundaries -> Low Hysteresis



Applications: Surgical tools, flight control surfaces, …

Source: liquidmetal.com

What are the tradeoffs?

Source: Wikipedia.org

Problems with BMGs

Main Problem: Metastability of glassy phase

GFA *Few Alloy Systems Known to Form Glasses*

<2% possible ternaries in Landolt-Börnstein handbook

D_{max} *Small Critical Casting Thicknesses*

Typical thickness < 10 mm

ΔT_x *Small Time/Temperature Processing Window*

BMGs may crystallize during processing

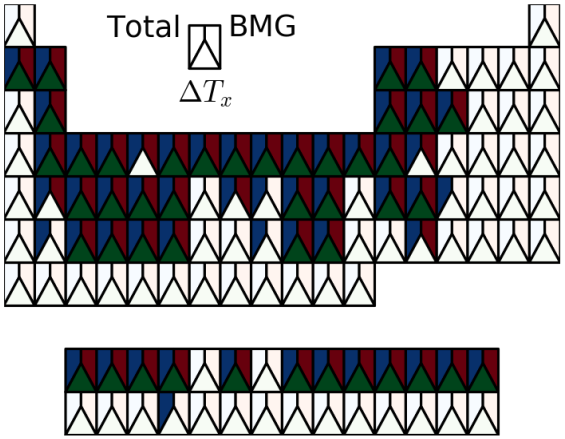
Design Issue: Lack of Composition -> Properties Links

Our Proposed Solution: Data analytics

Goal: Optimize D_{max} and ΔT_x

Materials Informatics Workflow

Collect → Process → Represent → Learn

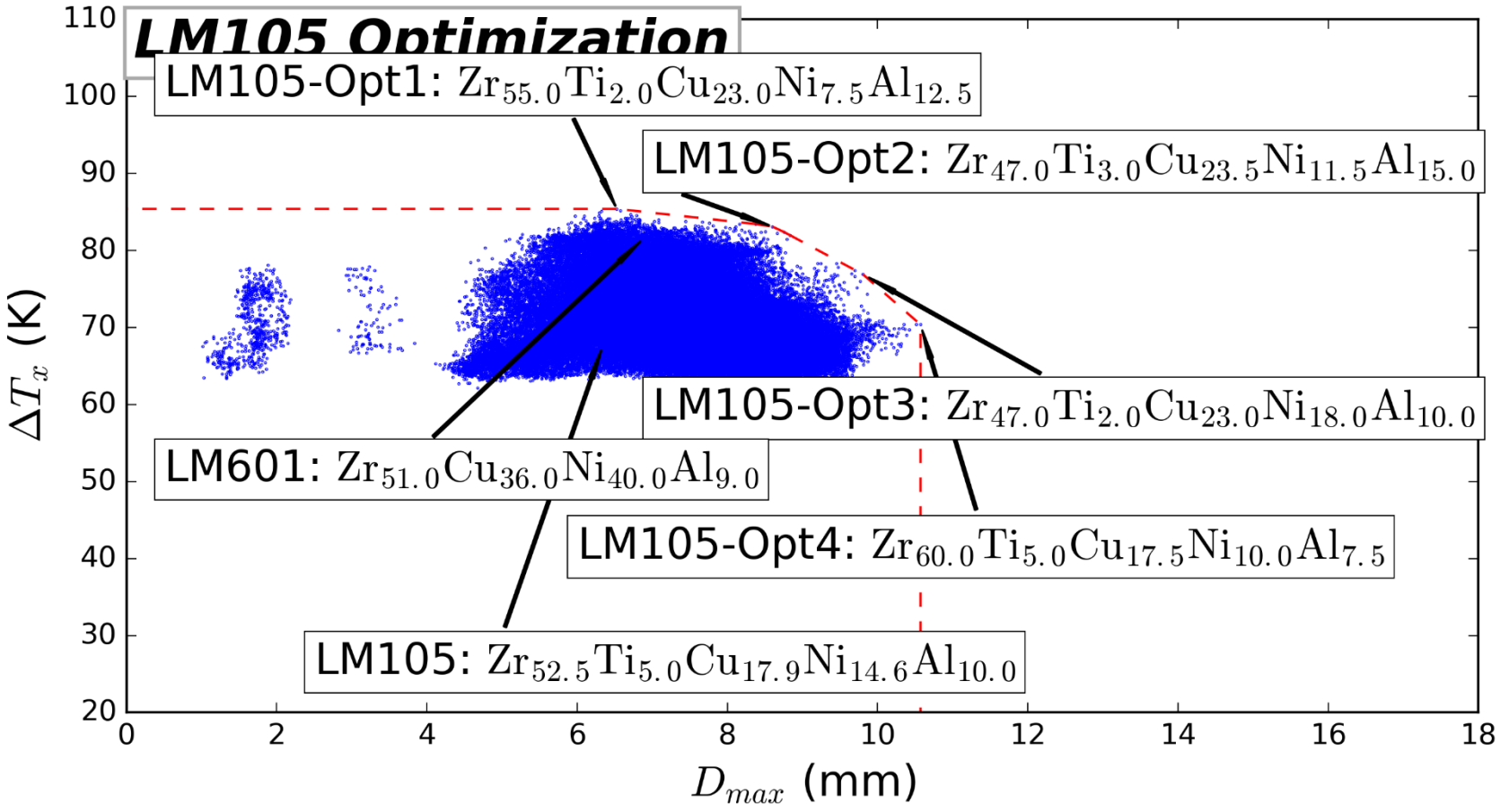


$$\vec{X} = f(Composition)$$

"General Purpose" Attributes
+ Cluster Packing Models
+ DFT ΔH_f from OQMD

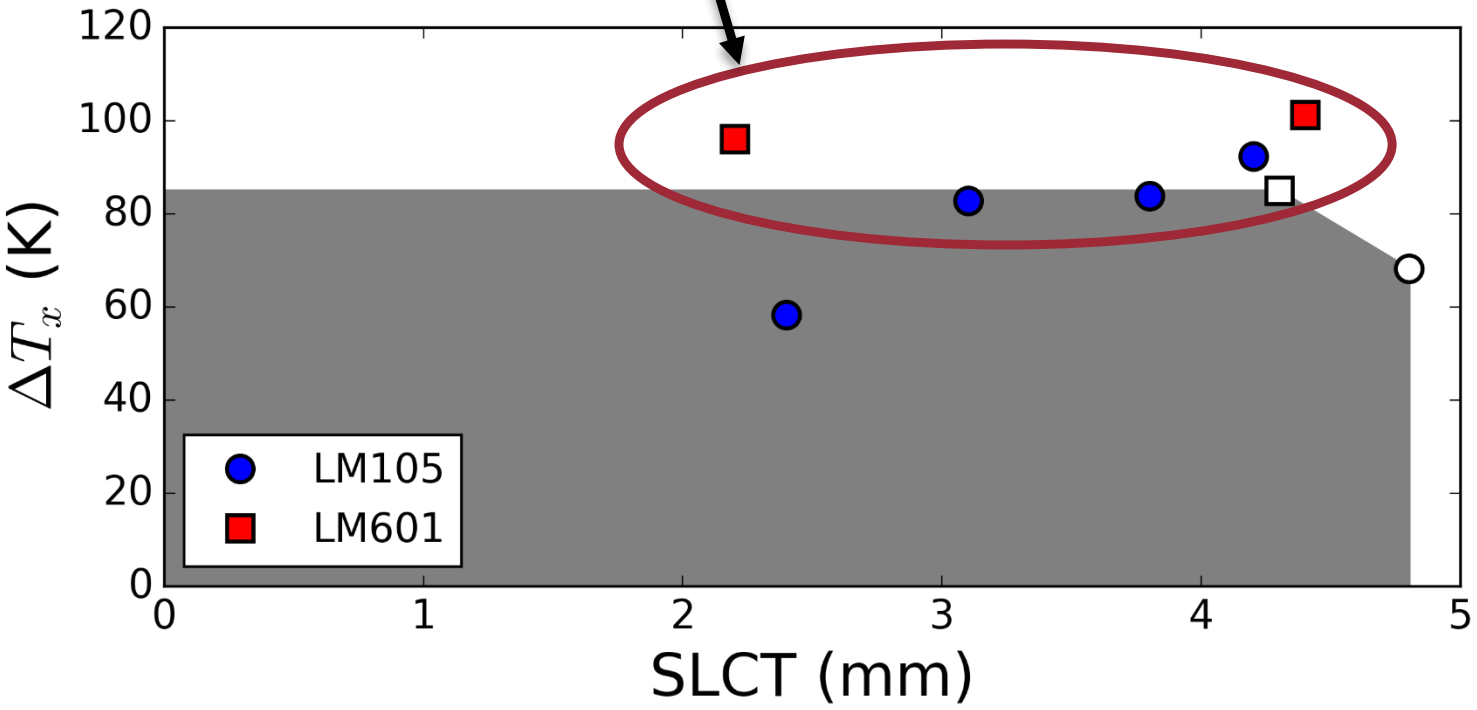
GFA	6315 entries	GFA	Random Forest
D_{max}	5916 entries	D_{max}	Random Forest + Additive Regression
ΔT_x	621 entries	ΔT_x	

Task #1: Tuning Known Alloys



Tuned Alloys: Pareto Analysis

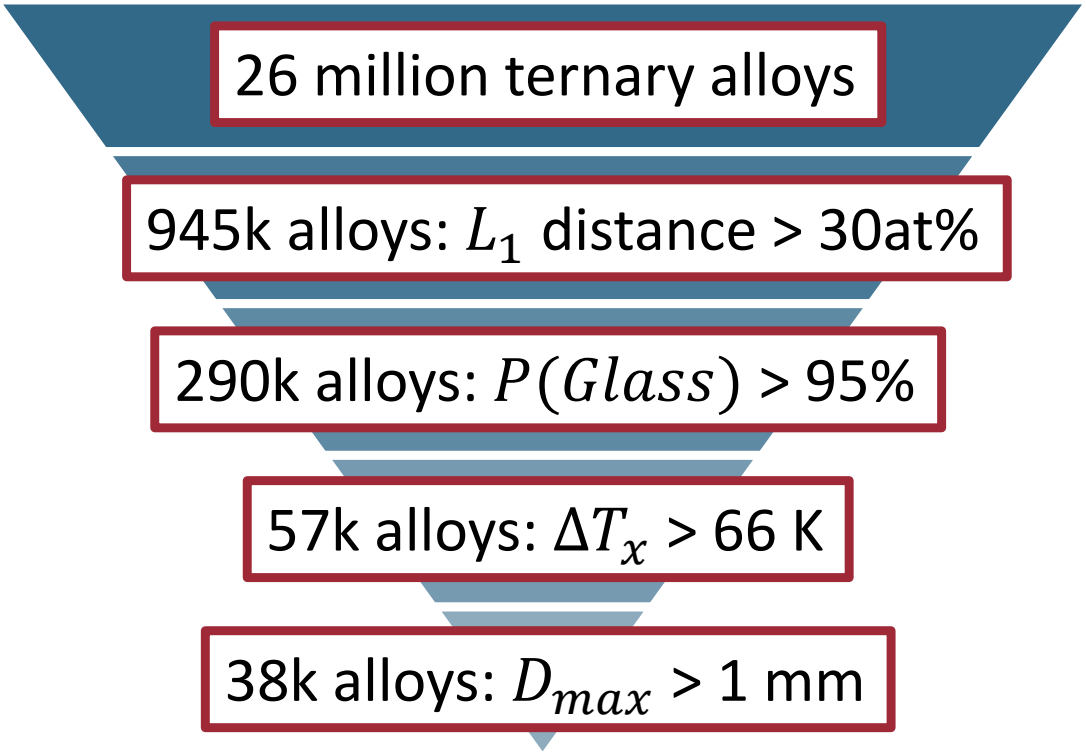
Predicted alloys have superior properties



Results: Improved design space

Task #2: Discovering New Alloys

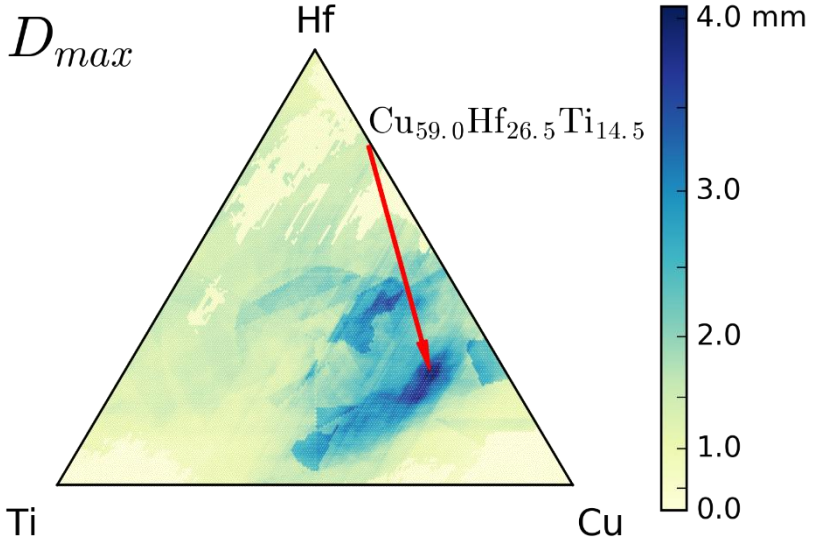
Search Space: Ternary alloys of 53 elements

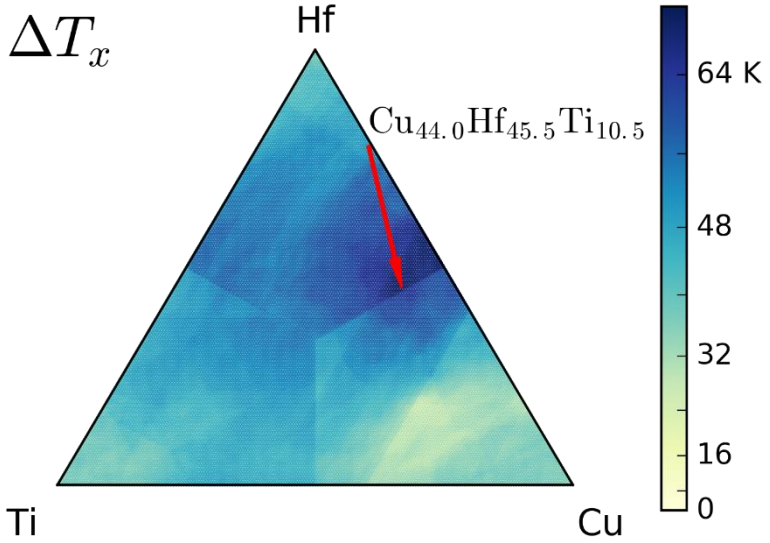


Run Time: ~ 2 days

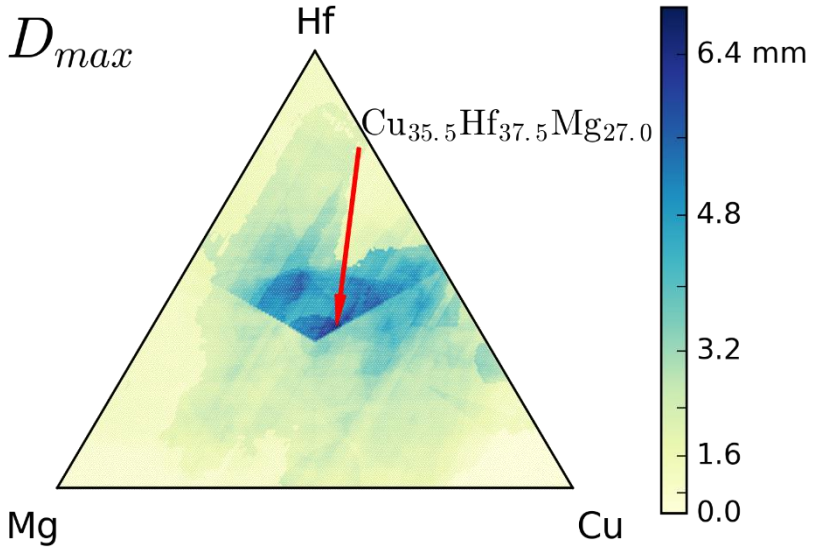
Chosen System: Cu-Hf-[Mg,Ti]

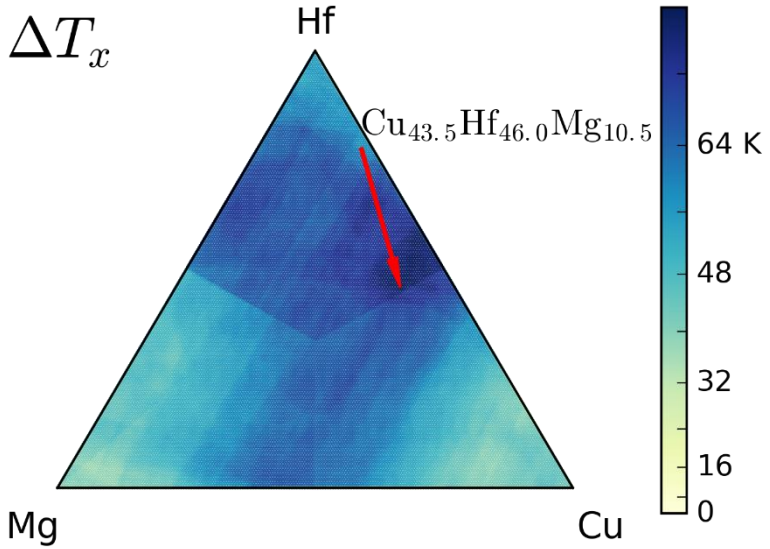
Cu-Hf-Ti





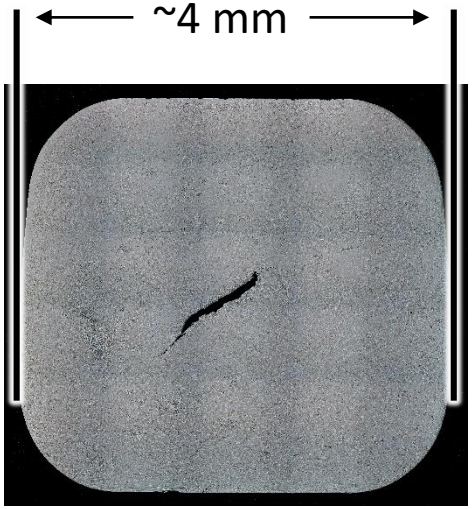
Cu-Hf-Mg





Cu-Hf-[Ti/Mg]: Current results

Cu-Hf-Ti



Sample crystallized and/or shattered during injection molding

- $Cu_{60}Hf_{25}Ti_{15}$ could be formed with copper mold casting*

Cu-Hf-Mg

Mg ignited during arc melting

Lessons Learned:

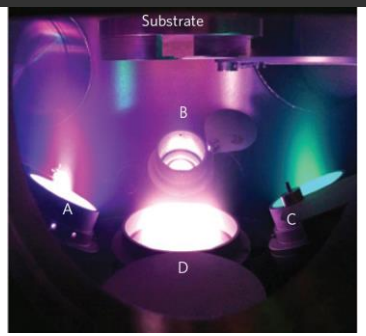
- Integrate data from target process
- Screen based on processing technique

*Inoue *et al*. Acta Mat. (2001), 2645

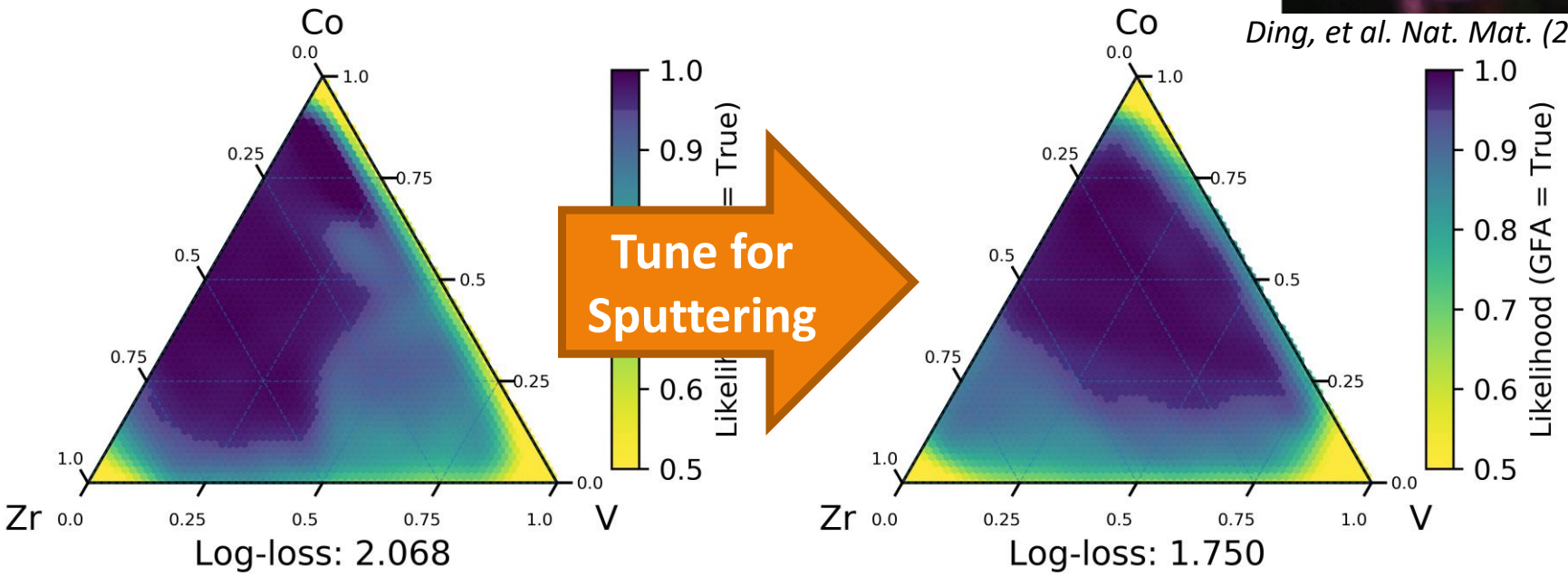
Example #2: Finding New Glasses

Goal: Find new sputtering glasses

Step 1: Include processing information

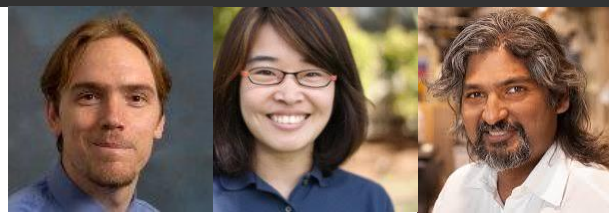


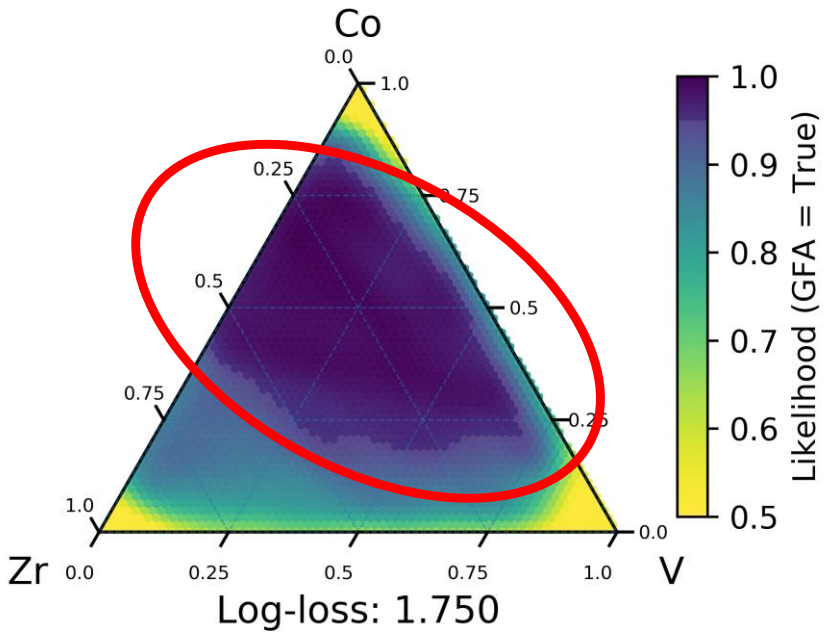
Ding, et al. Nat. Mat. (2014)

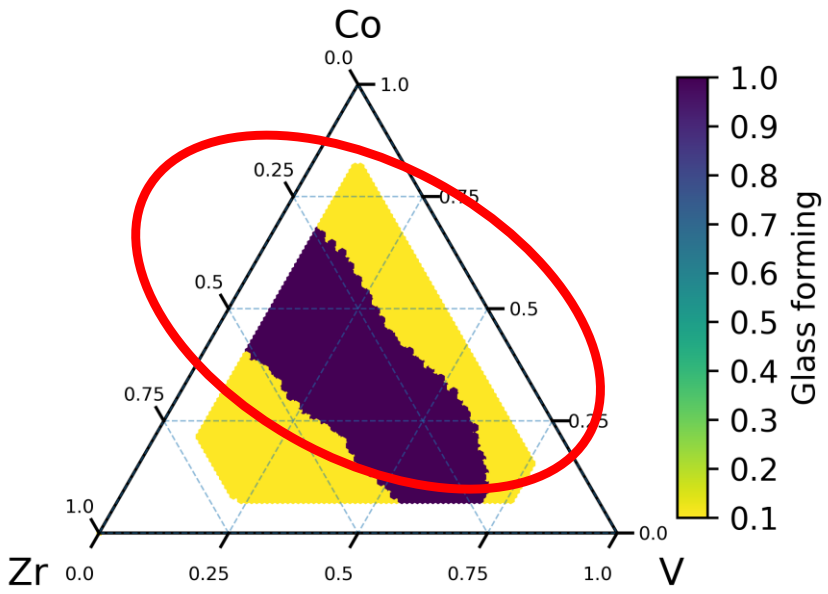


Step 2: Scan 2.4M alloys (~5 min) -> Selected Co-V-Zr

Comparison to Experiment

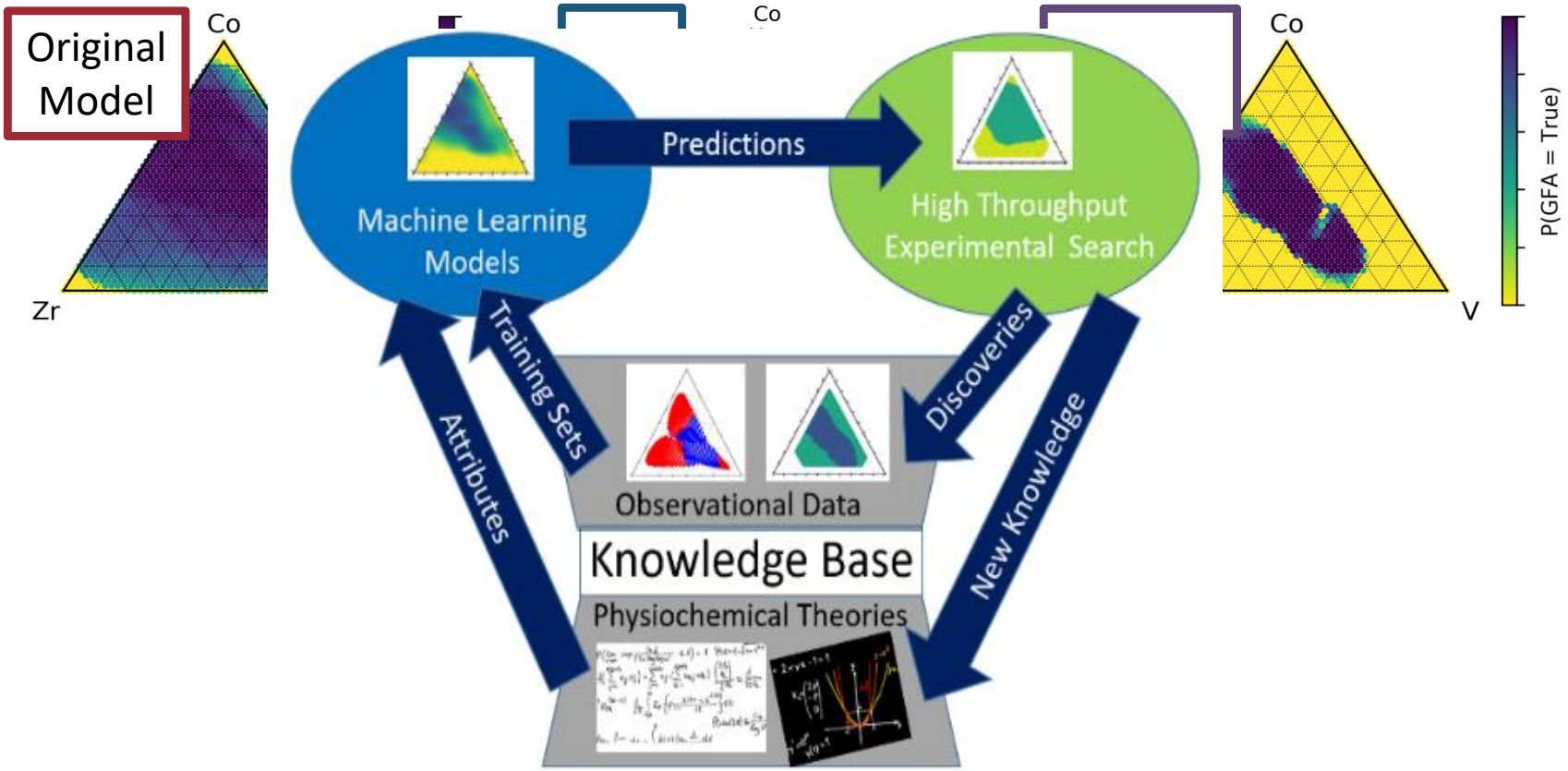






Decent agreement. ML model “Zr-lean”, but close enough for success

Repeat, with Improved Model



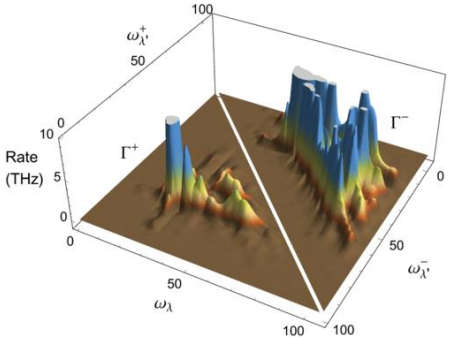
Initial Model → Add Co-V-Zr → Improved Accuracy

Example #2: Finding Compounds

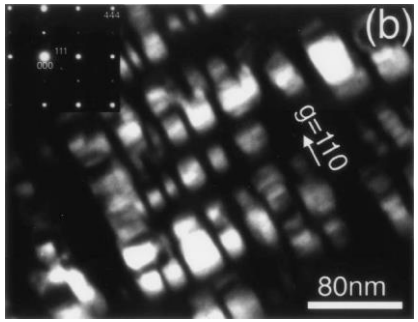
Goal: Find more Quaternary Heuslers (QHs)

Why quaternary Heuslers?

Ternary Heuslers have great properties,



He *et al*. PRL. (2016), 046602

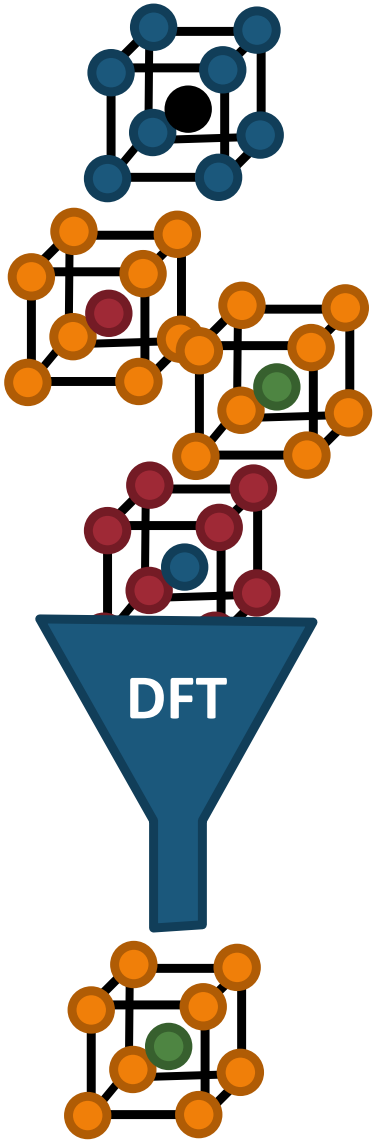


Jung *et al*. MMTA. (2003), 1221

…so a 4th degree of freedom could be better?

Problem: ~3M possible combinations

Solution: Guide search with ML



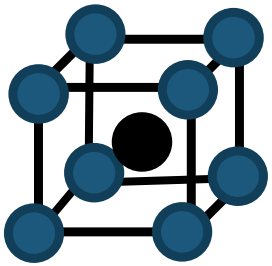
Ref: Kim *et al*., *in preparation*

Materials Informatics Workflow

Collect → Process → Represent → Learn

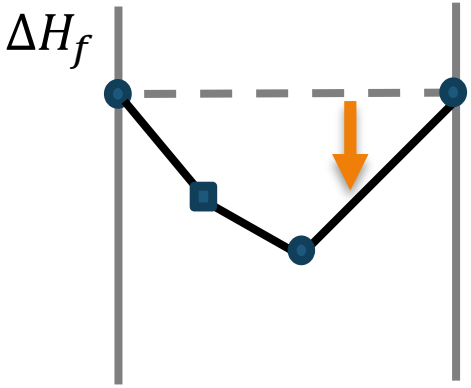
OQMD

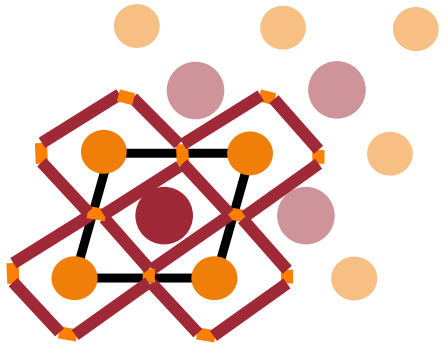
Input: Crystal structure
Output: Stability



Training Data: OQMD

Entries: 410981
96160 QHs





Voronoi-Tessellation Method

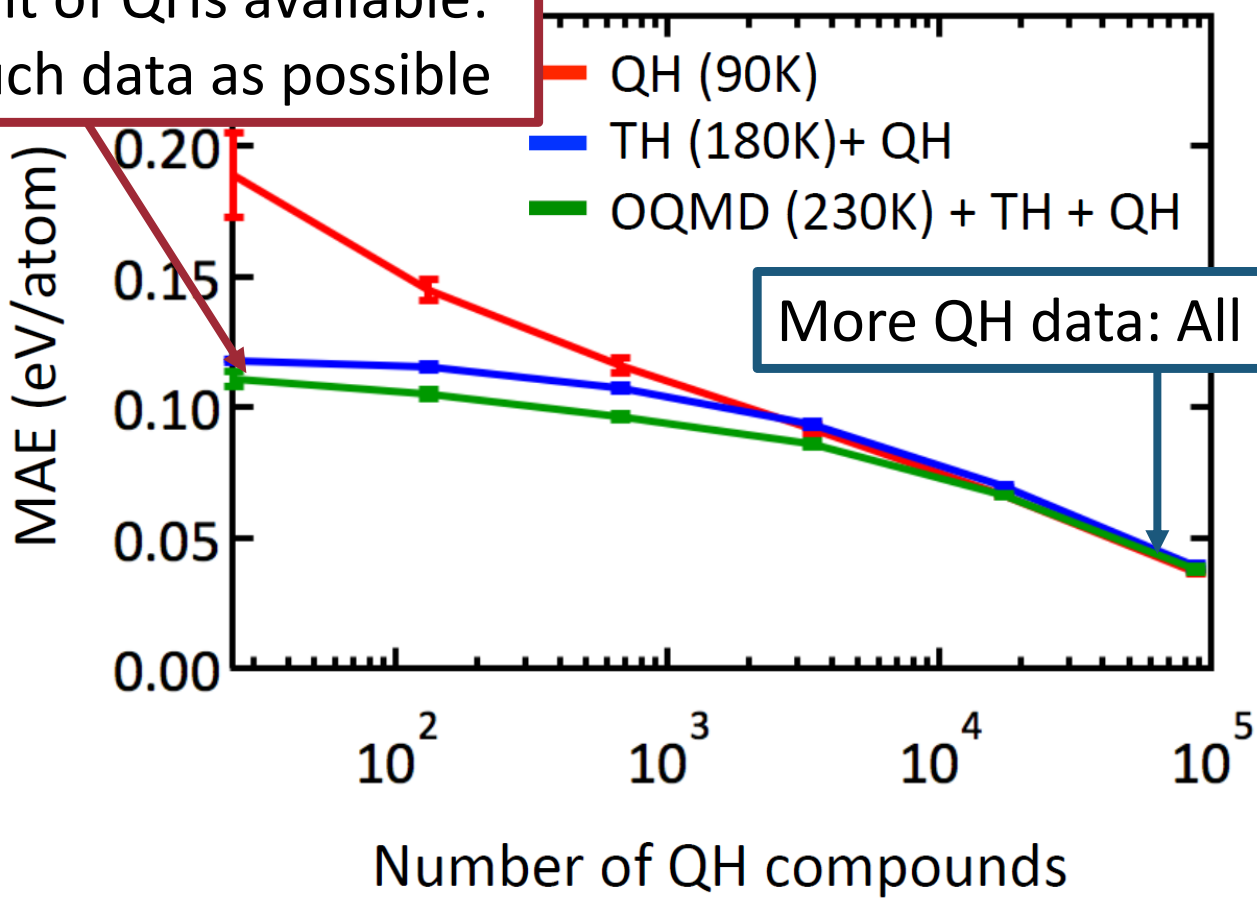
275 attributes

Random Forest

Step 1: Select Training Set, Validate

Training data: All data available, or only similar materials?

Small amount of QHs available:
Select as much data as possible



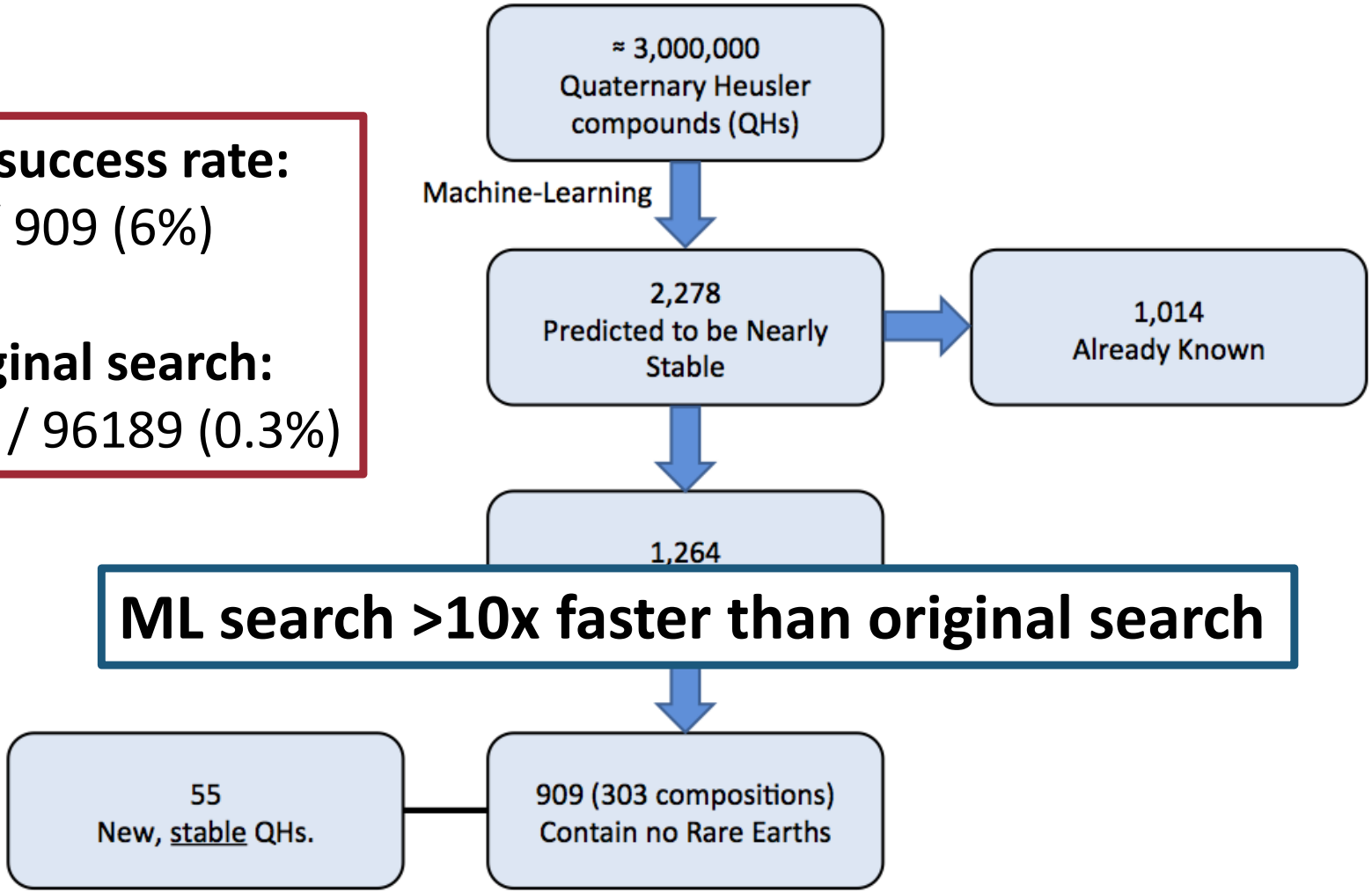
More QH data: All equivalent

Bottom Line: Training on *all available data* is advantageous

Step 2: Find New Heuslers

ML success rate:
55 / 909 (6%)

Original search:
353 / 96189 (0.3%)



Summary

Automated Crystal Structure Solution

- ✓ Validated FPASS method, created automatic framework
- ✓ Solved 13 crystal structures

General-Purpose Representations

Using Composition: DFT ΔH_f, E_g, V; Glass-forming ability

Using Crystal Structure: 2x better than existing approaches

Example applications:

- ✓ Improved ΔT_x of 2 commercial BMGs alloys
- ✓ Discovered many new metallic glasses
- ✓ Found dozens of new quaternary Heuslers

Open Source Software: Magpie, Matminer

http://bitbucket.org/wolverton/magpie

http://github.com/hackingmaterials/matminer

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