

innovating nanoscience



High-throughput electronic structure theory: are all calculations useful ?

Stefano Sanvito (sanvitos@tcd.ie)

School of Physics and CRANN, Trinity College Dublin, IRELAND

My objectives for this talk

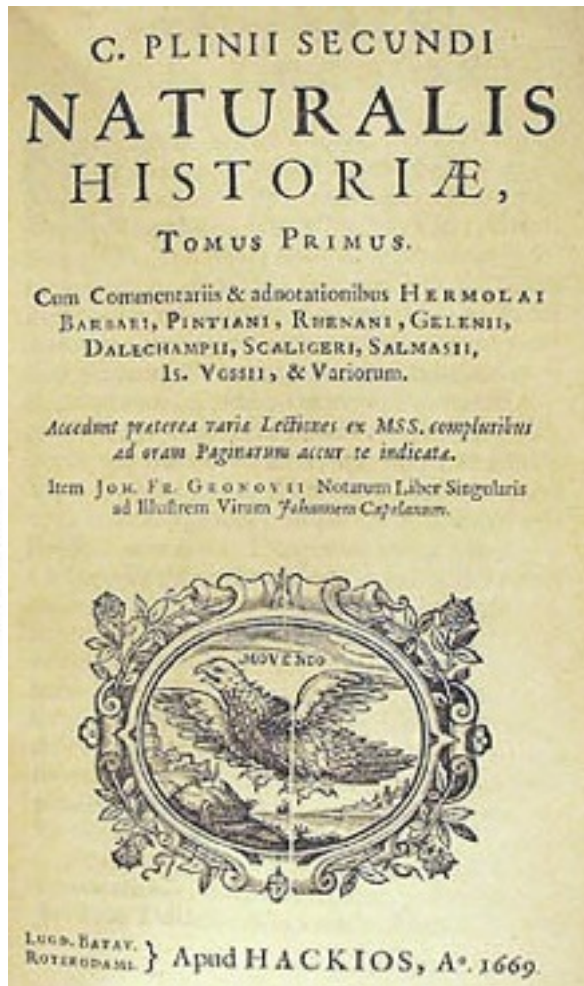
*Demonstrate that HTEST works and that **new magnets** can be discovered*

Show that, as databases grow, we will become more clever in creating and using them

Magnetism is rare



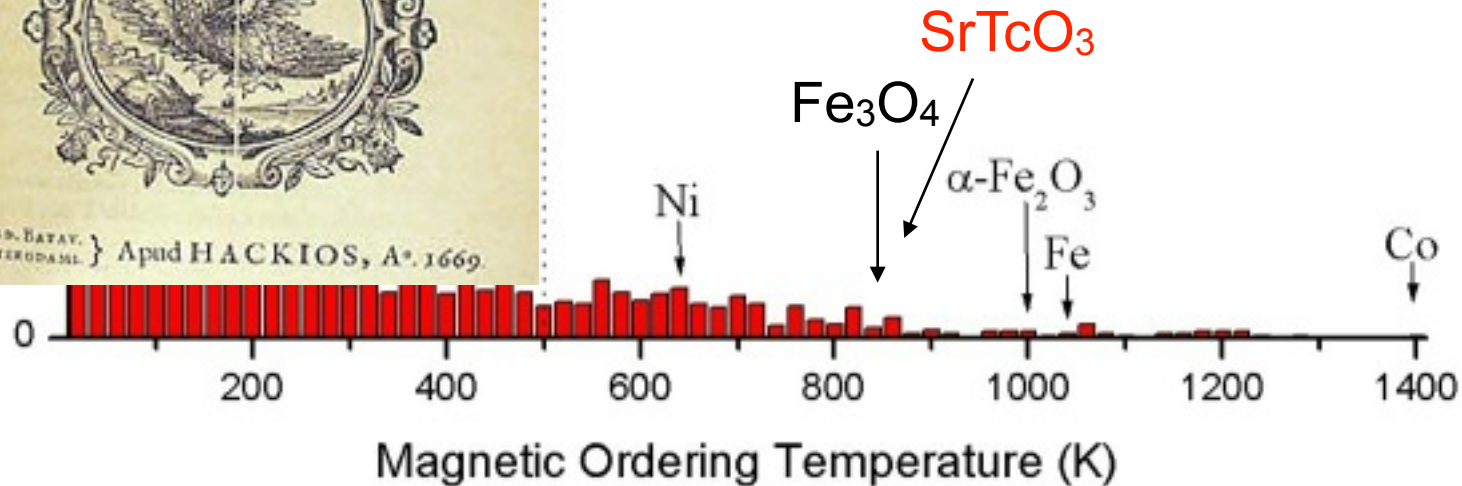
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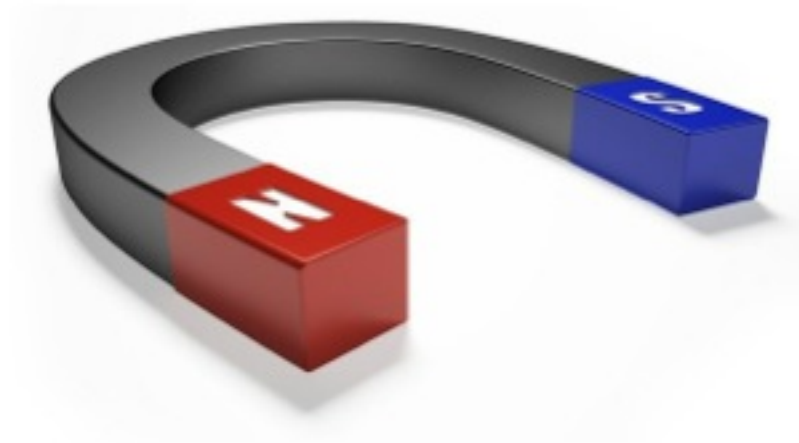
The discover a new useful magnet is a rare event

Number

Potentially useful magnets.



The magnetic genome project



The magnetic genome project



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nature
materials

REVIEW ARTICLE

PUBLISHED ONLINE: 20 FEBRUARY 2013 | DOI: 10.1038/NMAT3568

The high-throughput highway to computational materials design

Stefano Curtarolo^{1,2*}, Gus L. W. Hart^{2,3}, Marco Buongiorno Nardelli^{2,4,5}, Natalio Mingo^{2,6}, Stefano Sanvito^{2,7} and Ohad Levy^{1,2,8}

Finding *descriptors*



Materials selection

Search the database for 1) new materials, 2) physical insights

Database Creation (AFLOW)



Rational materials storage

Creating searchable database where to store information

Virtual Materials Growth

- 1) Simulating existing materials
- 2) Simulating new materials

Robust electronic structure method:
density functional theory (VASP)



The magnetic genome project



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The magnetic genome project



Virtual Materials Growth (existing materials)

Only ~150,000 are known to us

ICSD: Inorganic Crystal Structure Database

- 1,616 crystal structures of the elements
- 28,354 records for binary compounds
- 55,436 records for ternary compounds
- 54,144 records for quarternary and quinary
- About 113,000 entries (75.6%) have been assigned a structure type.
- There are currently 6,336 structure prototypes.
- **Lots of redundancy**

The magnetic genome project



Virtual Materials Growth (existing materials)

Duke calculated single elements, binary, ternary and some quaternary (about 100,000)

Calculations:

- AFLOW manages the run (large code)
- DFT done with VASP (pseudo-potential, plane-wave)
- Calculations at the DFT GGA-PBE level

- Relaxation performed → new space group worked out
- Basic electronic structures collected (including: spin-polarization, effective mass, magnetic moment, etc.)

The AFLOW consortium



CRANN

www.aflowlib.org



S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R.H. Taylor, L.J. Nelson, G.L.W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, *Comp. Mat. Sci.* **58**, 227 (2012)

Heusler alloys



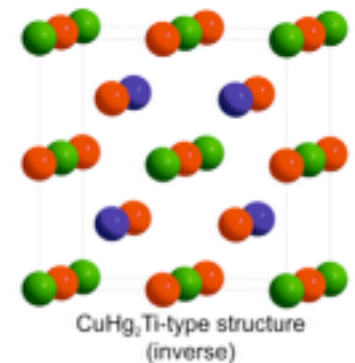
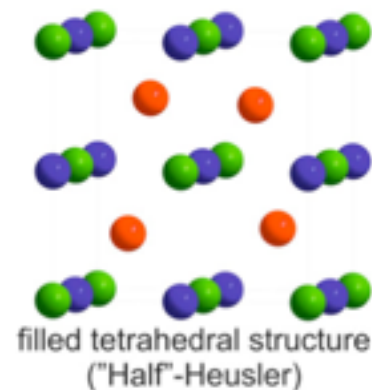
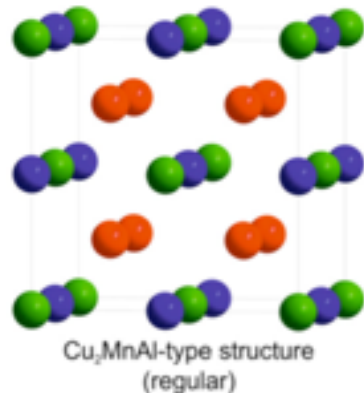
~250
known ...

~1000
claimed ...

~90
magnetic ...

X_2YZ Heusler compounds

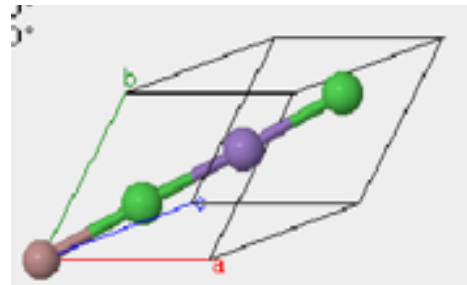
H 2.20																	He	
Li 0.98	Be 1.57											B 2.04	C 2.55	N 3.04	O 3.44	F 3.98	Ne	
Na 0.93	Mg 1.31											Al 1.61	Si 1.90	P 2.19	S 2.58	Cl 3.16	Ar	
K 0.82	Ca 1.00	Sc 1.36	Ti 1.54	V 1.63	Cr 1.66	Mn 1.55	Fe 1.83	Co 1.88	Ni 1.91	Cu 1.90	Zn 1.65	Ga 1.81	Ge 2.01	As 2.18	Se 2.55	Br 2.96	Kr 3.00	
Rb 0.82	Sr 0.95	Y 1.22	Zr 1.33	Nb 1.60	Mo 2.16	Tc 1.90	Ru 2.20	Rh 2.28	Pd 2.20	Ag 1.93	Cd 1.69	In 1.78	Sn 1.96	Sb 2.05	Te 2.10	I 2.66	Xe 2.60	
Cs 0.79	Ba 0.89			Hf 1.30	Ta 1.50	W 1.70	Re 1.90	Os 2.20	Ir 2.20	Pt 2.20	Au 2.40	Hg 1.90	Tl 1.80	Pb 1.80	Bi 1.90	Po 2.00	At 2.20	Rn
Fr 0.70	Ra 0.90																	
		La 1.10	Ce 1.12	Pr 1.13	Nd 1.14	Pm 1.13	Sm 1.17	Eu 1.20	Gd 1.20	Tb 1.10	Dy 1.22	Ho 1.23	Er 1.24	Tm 1.25	Yb 1.10	Lu 1.27		
		Ac 1.10	Th 1.30	Pa 1.50	U 1.70	Np 1.30	Pu 1.28	Am 1.13	Cm 1.28	Bk 1.30	Cf 1.30	Es 1.30	Fm 1.30	Md 1.30	No 1.30	Lr 1.30		



Heusler alloys



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~236,000/0.5M calculated !!

hydrogen 1 H 1.0079																	helium 2 He 4.0026	
lithium 3 Li 6.941	beryllium 4 Be 9.0122																	neon 10 Ne 20.180
sodium 11 Na 22.990	magnesium 12 Mg 24.305																	argon 18 Ar 39.948
potassium 19 K 39.098	calcium 20 Ca 40.078	scandium 21 Sc 44.956	titanium 22 Ti 47.867	vanadium 23 V 50.942	chromium 24 Cr 51.996	manganese 25 Mn 54.938	iron 26 Fe 55.845	cobalt 27 Co 58.933	nickel 28 Ni 58.693	copper 29 Cu 63.546	zinc 30 Zn 65.39	gallium 31 Ga 69.723	germanium 32 Ge 72.61	arsenic 33 As 74.922	selenium 34 Se 78.96	bromine 35 Br 79.904	krypton 36 Kr 83.80	
rubidium 37 Rb 85.468	strontium 38 Sr 87.62	yttrium 39 Y 88.906	zirconium 40 Zr 91.224	niobium 41 Nb 92.906	molybdenum 42 Mo 95.94	technetium 43 Tc [98]	ruthenium 44 Ru 101.07	rhodium 45 Rh 102.91	palladium 46 Pd 106.42	silver 47 Ag 107.87	cadmium 48 Cd 112.41	indium 49 In 114.82	tin 50 Sn 118.71	antimony 51 Sb 121.76	tellurium 52 Te 127.60	iodine 53 I 126.90	xenon 54 Xe 131.29	
caesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	thallium 81 Tl 204.38	lead 82 Pb 207.2	bismuth 83 Bi 208.98	polonium 84 Po [209]	astatine 85 At [210]	radon 86 Rn [222]
francium 87 Fr [223]	radium 88 Ra [226]	89-102 **	103 Lr [262]	rutherfordium 104 Rf [261]	dubnium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	unnilennium 110 Uun [279]	ununennium 111 Uuu [277]	unbinilium 112 Uub [277]	unquadium 114 Uuq [289]					

* Lanthanide series

** Actinide series

lanthanum 57 La 138.91	cerium 58 Ce 140.12	praseodymium 59 Pr 140.91	neodymium 60 Nd 144.24	promethium 61 Pm [145]	samarium 62 Sm 150.36	europium 63 Eu 151.96	gadolinium 64 Gd 157.25	terbium 65 Tb 158.93	dysprosium 66 Dy 162.50	holmium 67 Ho 164.93	erbium 68 Er 167.26	thulium 69 Tm 168.93	ytterbium 70 Yb 173.04
actinium 89 Ac [227]	thorium 90 Th 232.04	protactinium 91 Pa 231.04	uranium 92 U 238.03	neptunium 93 Np [237]	plutonium 94 Pu [244]	americium 95 Am [243]	curium 96 Cm [247]	berkelium 97 Bk [247]	californium 98 Cf [251]	einsteinium 99 Es [252]	fermium 100 Fm [257]	mendelevium 101 Md [258]	nobelium 102 No [259]

Database



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Rational materials storage

www.aflowlib.org

Search Aflowlib (50522 Compounds)

icsd elements binaries Heuslers

Periodic table interface with search filters and logical operators.

Atomic # element [electrons] [density] [T_m] [lattice] [crystal] [Debye] mass

and not or xor () Right Click for Wikipedia Link

Periodic table elements: H, He, Li, Be, Na, Mg, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Kr, Rb, Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te, I, Xe, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu.

Show 40 results per table. Limit to 1000 total results.

of Species:

All Metals Alkali Metals Alkaline Earths Transition Metals Lanthanides Other Metals

Nonmetals Group 3A Group 4A Group 5A Chalcogens Halogens

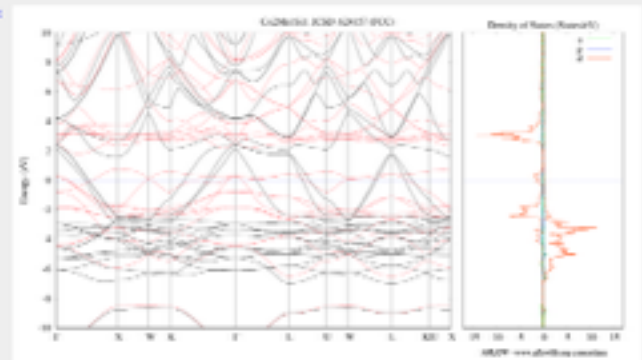
Chemistry Crystal Electronics Thermodynamics Magnetics Scintillation

Mechanical Calculation

ELECTRONIC PROPERTIES

Band Gap: 0.000 eV (metal) **Dir Band Gap:** 0.000 eV
Magnetic Moment: 7.382 μ_B **Magnetic Moment/atom:** 1.845 μ_B/atom
Electron Mass(TEX): XXX (m_0) **Hole Mass(TEX):** XXX (m_0)
Spin Polarization (S_z): 0.666 **Spin Decomposition per atom:** (1.758,1.758,4.018,-0.054) μ_B

Band Structure:



AFLIB - www.aflowlib.com

S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R.H. Taylor, L.J. Nelson, G.L.W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, *Comp. Mat. Sci.* **58**, 227 (2012)

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Finding *descriptors*

The high-throughput highway to computational materials design

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Materials selection

Search the database for 1) new materials, 2) physical insights

Database Creation (AFLOW)

Rational materials storage

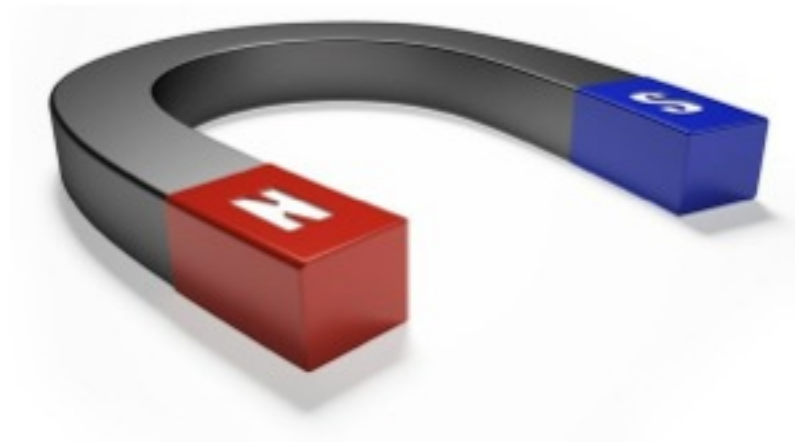
Creating searchable database where to store information

Virtual Materials Growth

- 1) Simulating existing materials
- 2) Simulating new materials

Robust electronic structure method: density functional theory (VASP)

Back to the magnets



S. Sanvito et al., *Accelerated discovery of new magnets in the Heusler alloy family*, *Science Advances* **3**, e1602241 (2017)

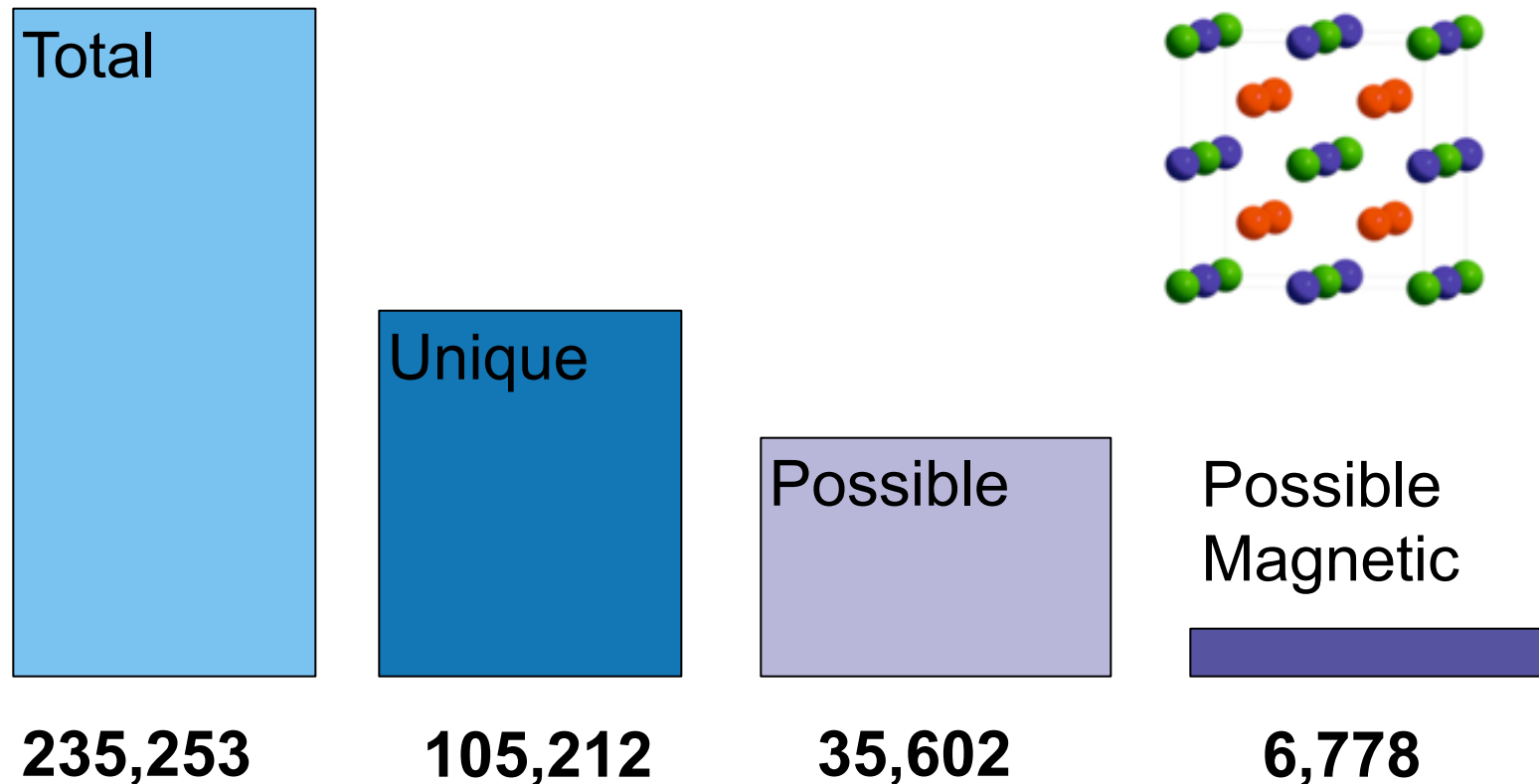
A look at the full database



Property: Can be made ?

Descriptor 0:
Enthalpy of formation

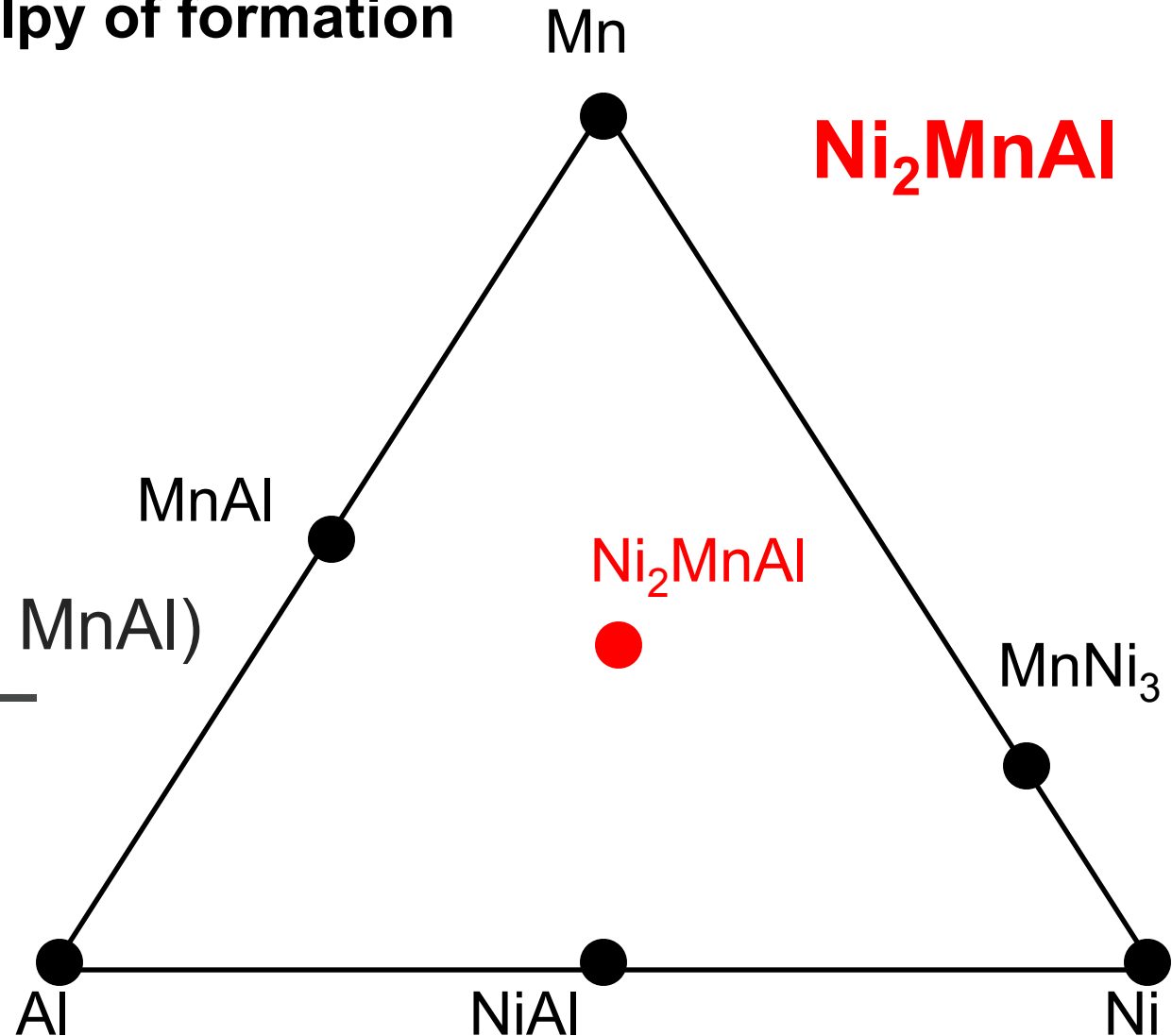
$$\text{Energy (Ni}_2\text{MnAl)} < \text{Energy (2Ni + Mn + Al)}$$



Stability analysis



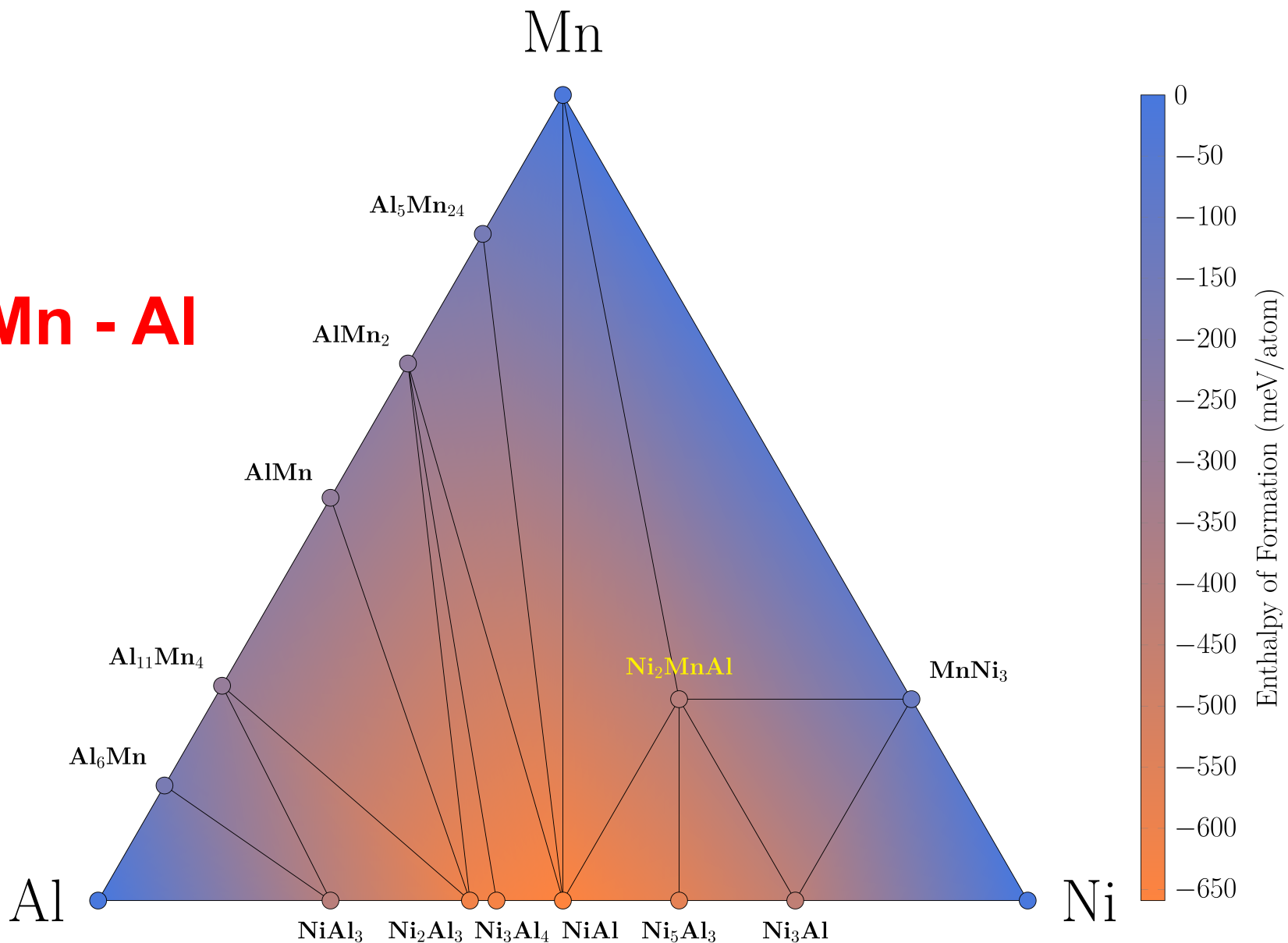
Descriptor 1: Enthalpy of formation



Stability analysis



Ni - Mn - Al





Look at the transition metal intermetallics

36,540

hydrogen 1 H 1.0079																	helium 2 He 4.0026						
lithium 3 Li 6.941	beryllium 4 Be 9.0122																	boron 5 B 10.811	carbon 6 C 12.011	nitrogen 7 N 14.007	oxygen 8 O 15.999	fluorine 9 F 18.998	neon 10 Ne 20.180
sodium 11 Na 22.990	magnesium 12 Mg 24.305																	aluminum 13 Al 26.982	silicon 14 Si 28.086	phosphorus 15 P 30.974	sulfur 16 S 32.065	chlorine 17 Cl 35.453	argon 18 Ar 39.948
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cesium 55 Cs 132.91	barium 56 Ba 137.33	57-70 *	71 Lu 174.97	hafnium 72 Hf 178.49	tantalum 73 Ta 180.95	tungsten 74 W 183.84	rhenium 75 Re 186.21	osmium 76 Os 190.23	iridium 77 Ir 192.22	platinum 78 Pt 195.08	gold 79 Au 196.97	mercury 80 Hg 200.59	81 Tl 204.38	82 Pb 207.2	83 Bi 208.98	84 Po [209]	85 At [210]	86 Rn [222]					
francium 87 Fr [223]	radium 88 Ra [226]	89-102 **	103 Lr [262]	rutherfordium 104 Rf [261]	dundubium 105 Db [262]	seaborgium 106 Sg [266]	bohrium 107 Bh [264]	hassium 108 Hs [269]	meitnerium 109 Mt [268]	unnilium 110 Uun [270]	ununium 111 Uuu [272]	unbibium 112 Uub [277]	114 Uuq [289]										

* Lanthanide series

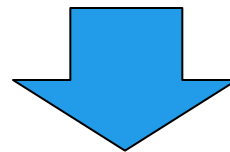
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In summary ...

36,540 possible \rightarrow 248 stable

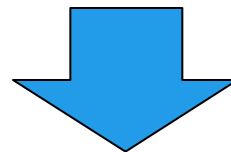
22 magnetic \rightarrow 8 Robust (Δ^{30} criterion)



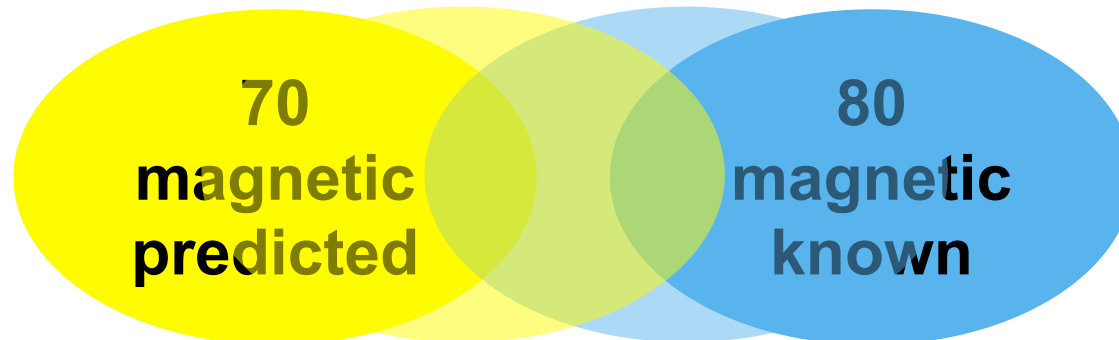
Extrapolating

236,000 possible \rightarrow 1550 stable

138 magnetic \rightarrow 50 Robust



For real



Critical temperature magnetism



Descriptor 2: Critical temperature

Known Heusler
ferromagnets

Co_2XY

Fe_2MnY

Ni_2MnY

Mn_2XY

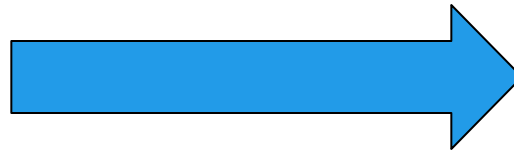
Rh_2MnY

Cu_2MnY

Pd_2MnY

Au_2MnY

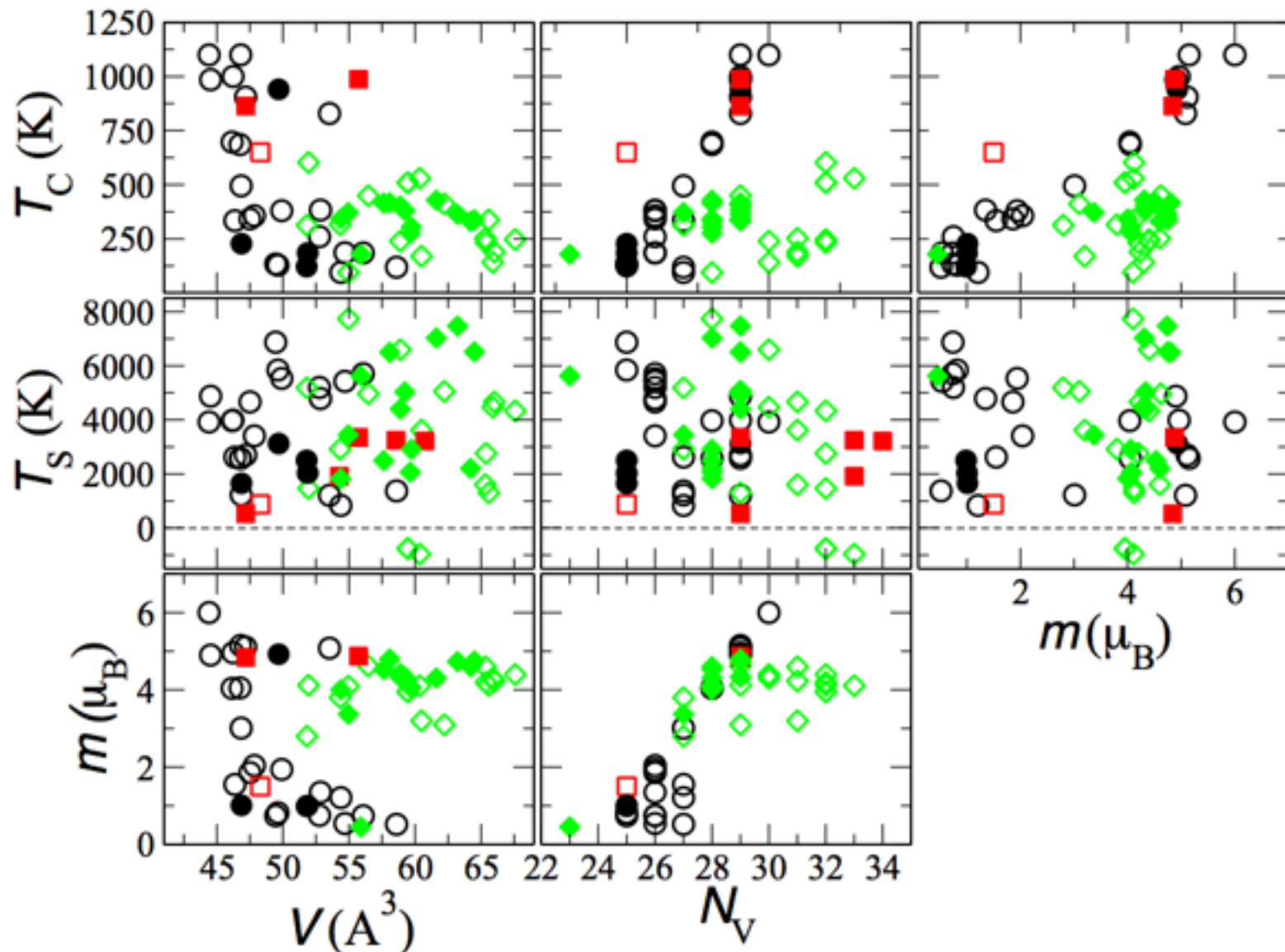
Generalized regression model based on
valence, volume, spin decomposition



Prediction of T_C

Material	V (Å)	μ	ΔE (eV)	T	T
Co	47.85	2.0	-0.30	3007		352
Mn	48.93	2.0	-0.32	3524		760
...
Mn	54.28	9.03	-0.17	1918		?

Analysis



Co_2XY

Mn_2XY

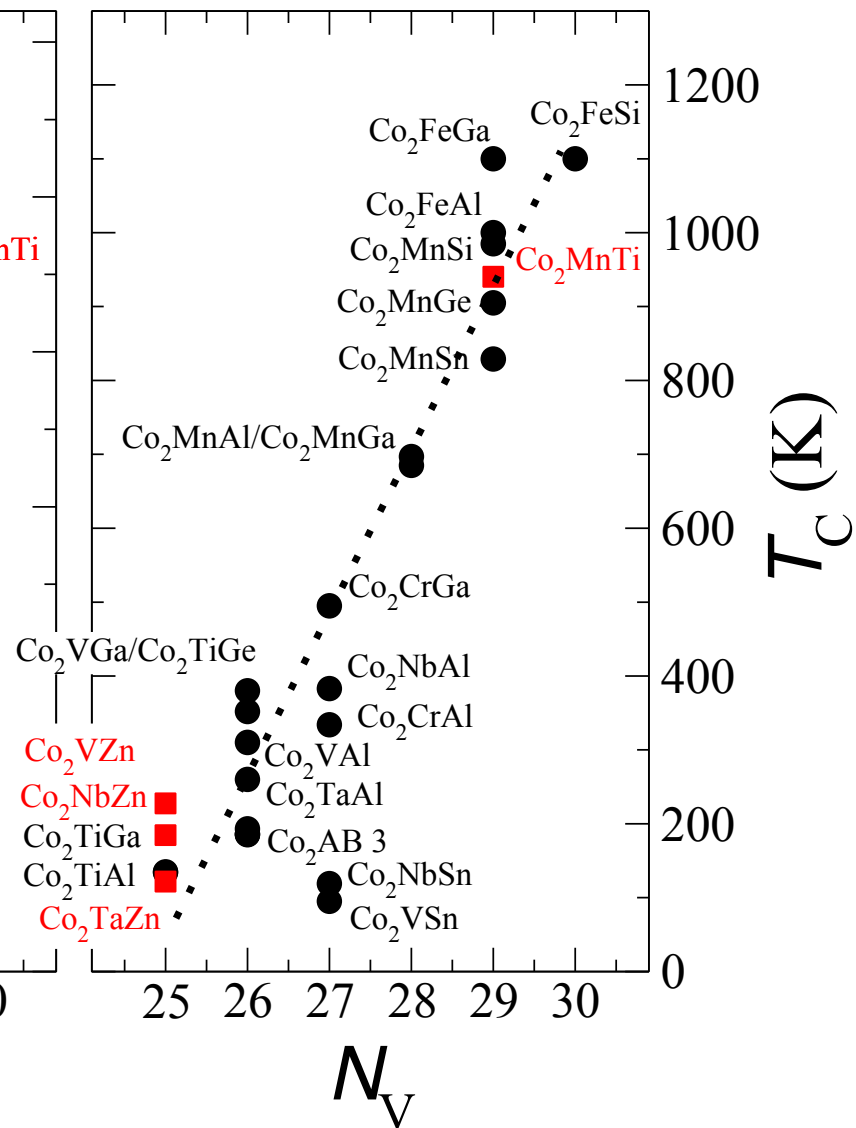
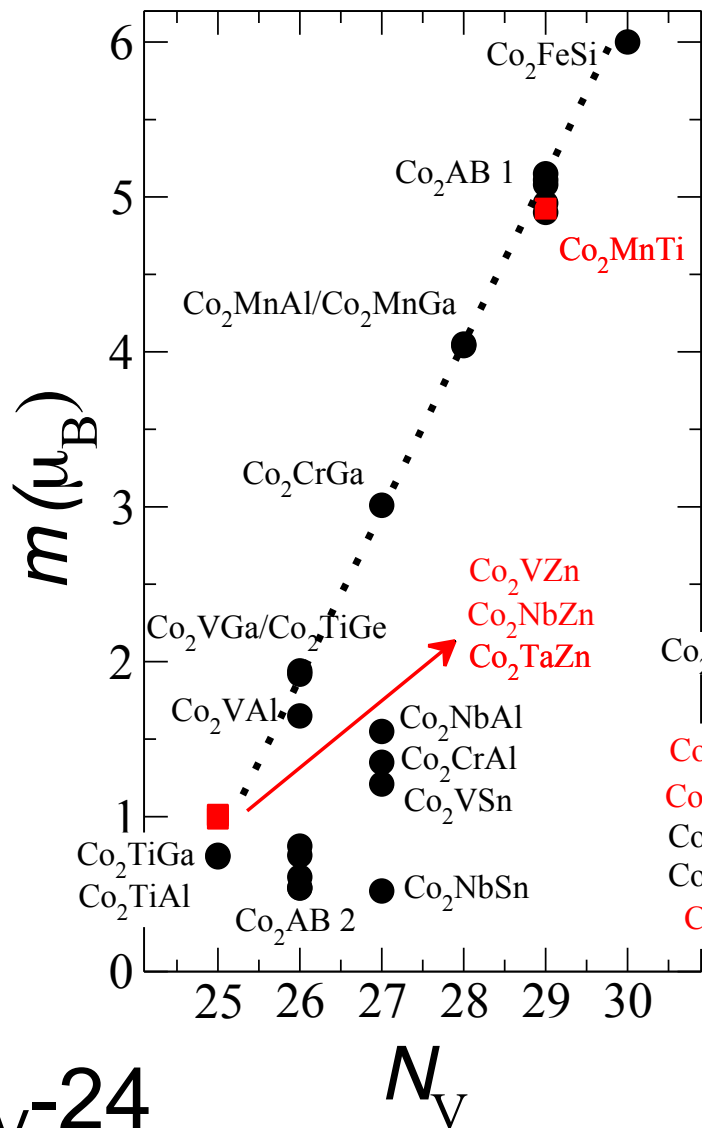
$X_2\text{Mn}Y$

Co₂YZ



Co₂YZ

Slater-Pauling

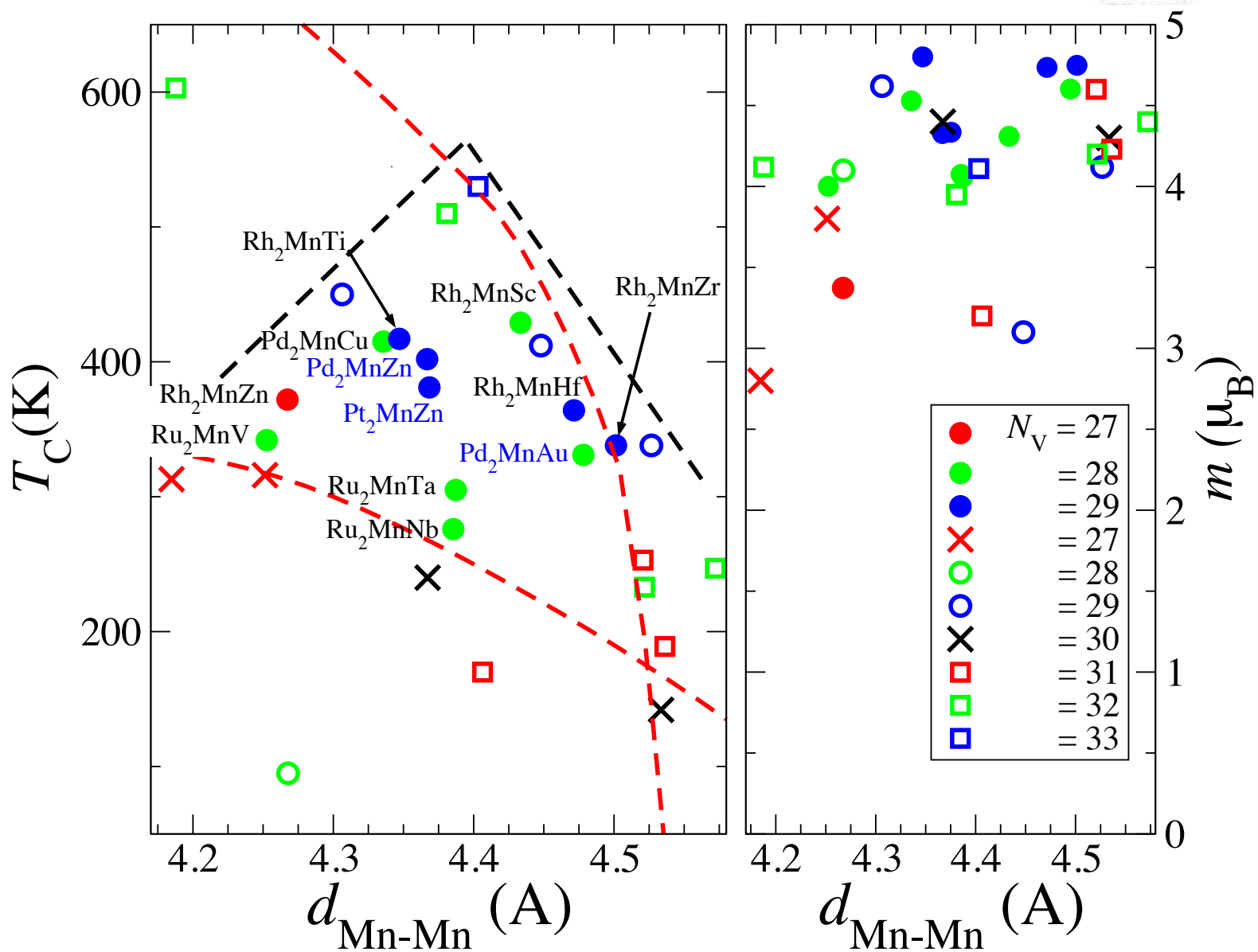


$$m_{X_2YZ} = N_V - 24$$

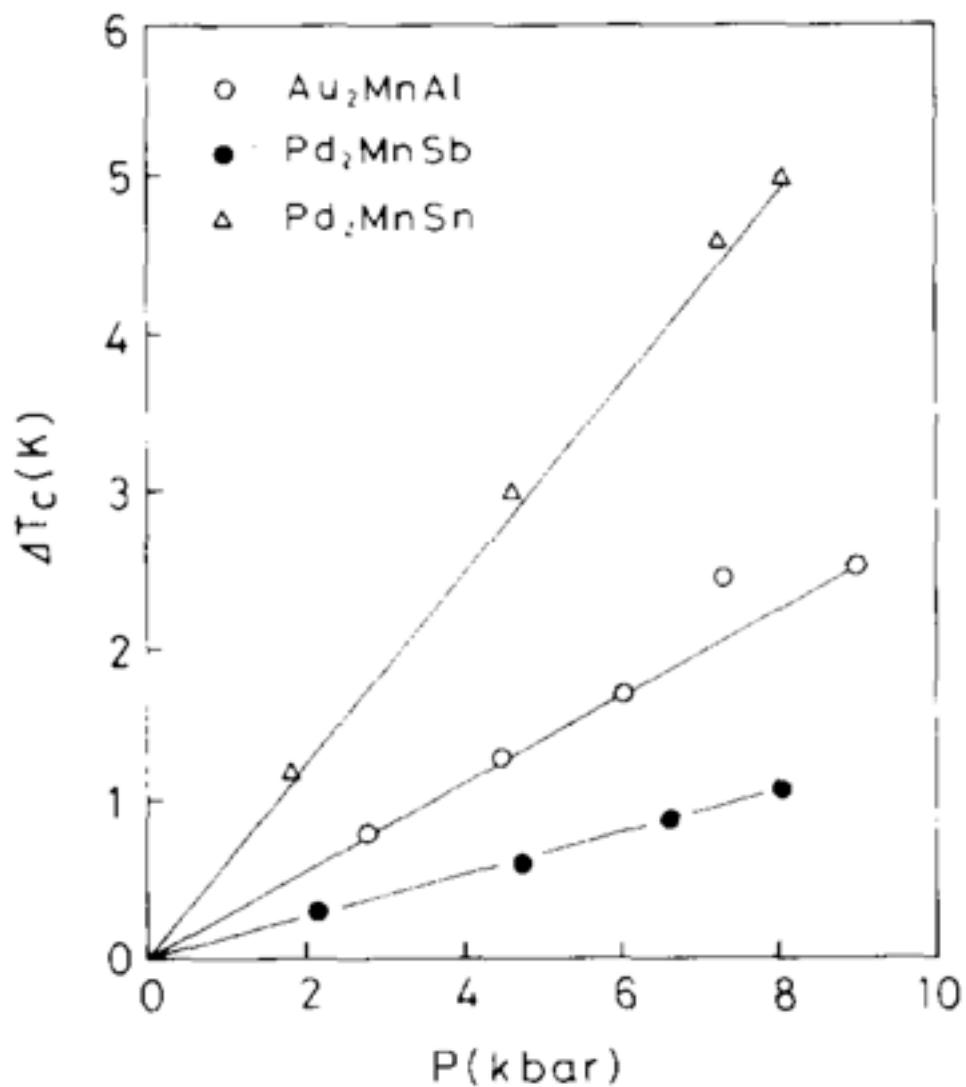
X_2MnZ



X_2MnZ



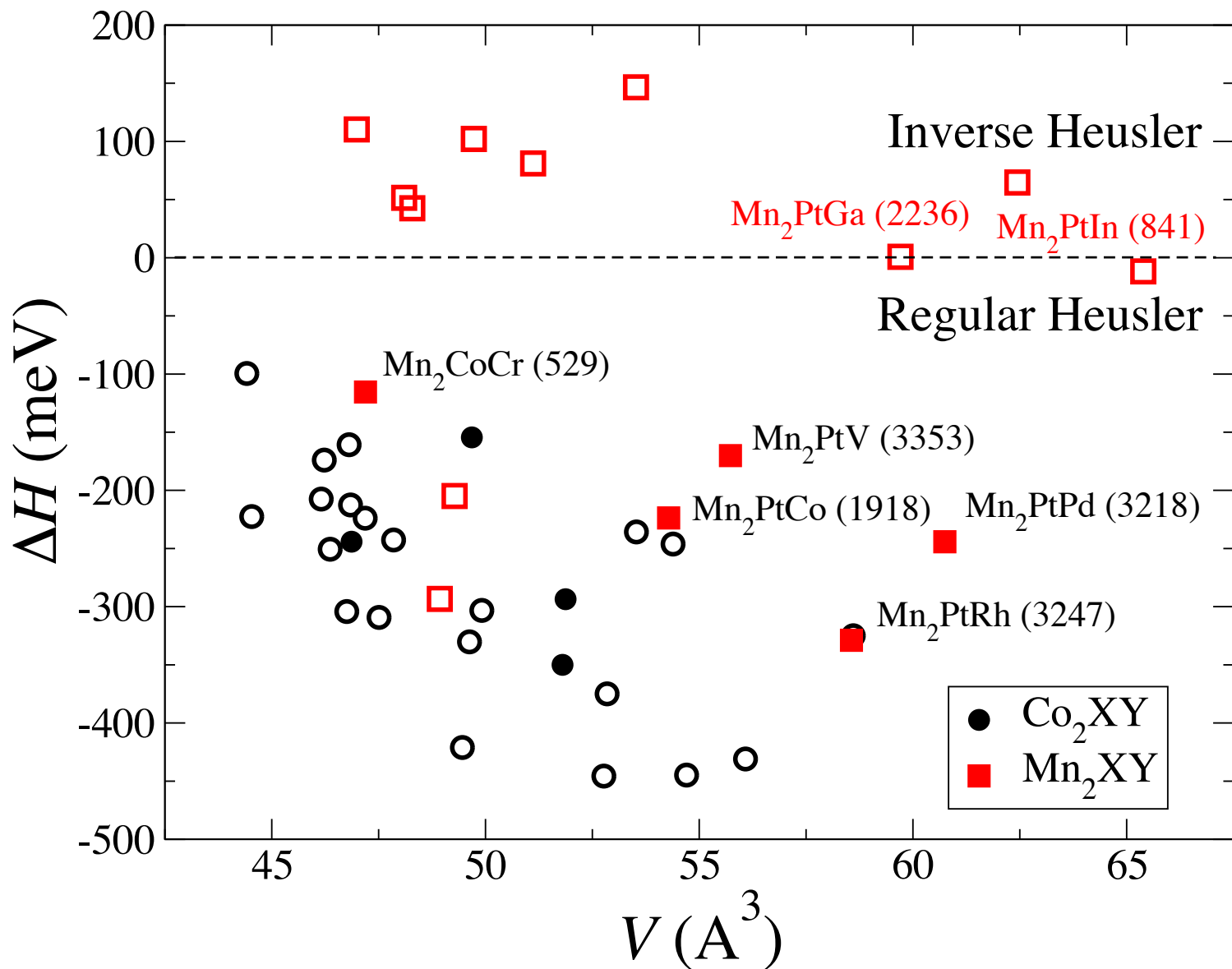
X_2MnZ



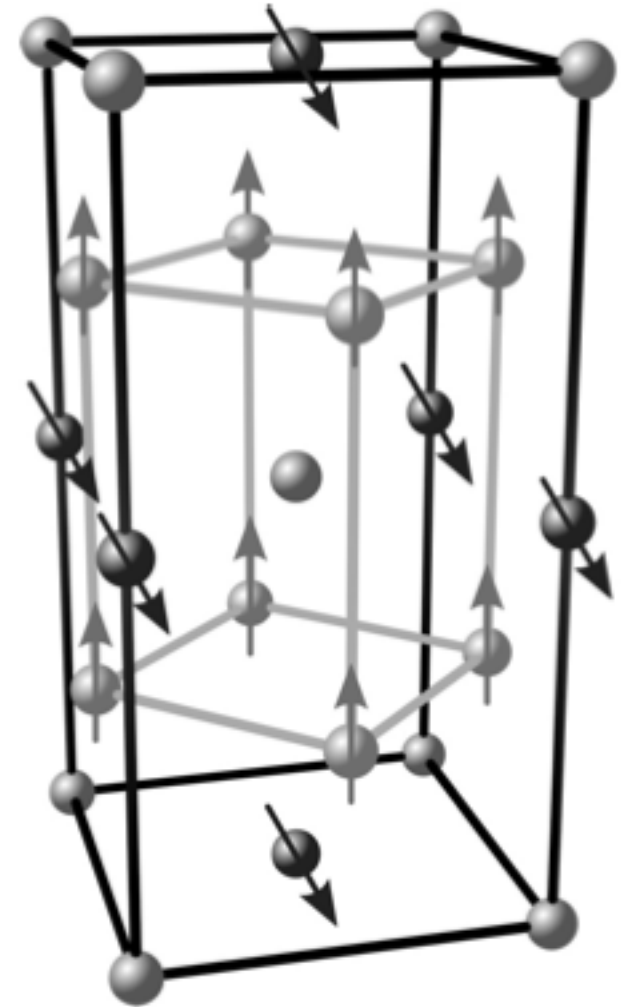
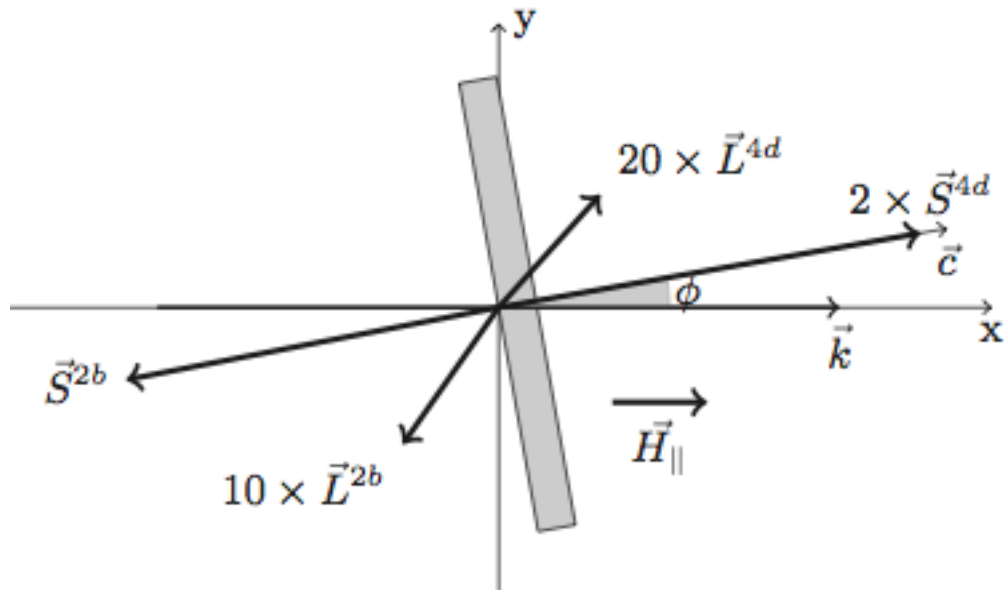
Mn₂YZ



Mn₂YZ



Mn₃Ga



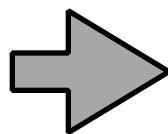
Machine learning *workflow*



250,000 candidates

2000 candidates

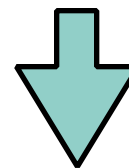
229 candidates



1000 used for DFT + ML

80 used for DFT + ML

80 used for DFT + ML



249,000 remaining

1920 remaining

149 remaining



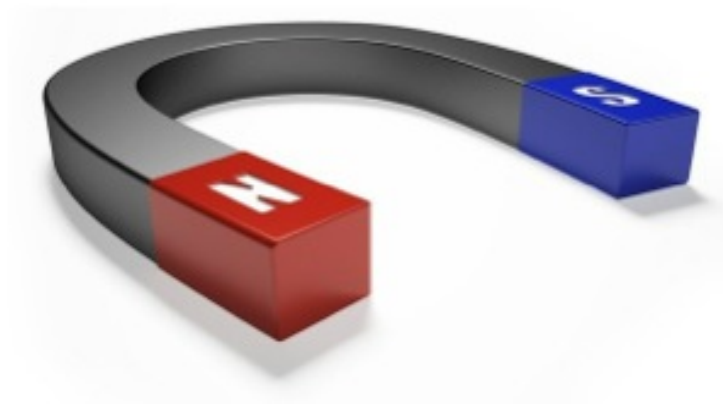
ML TPR 60% (50:50 population)

Don't calculate 30% = 50

Don't calculate 30% = ~650

Don't calculate 30% = ~80,000

OK, but does all that work?



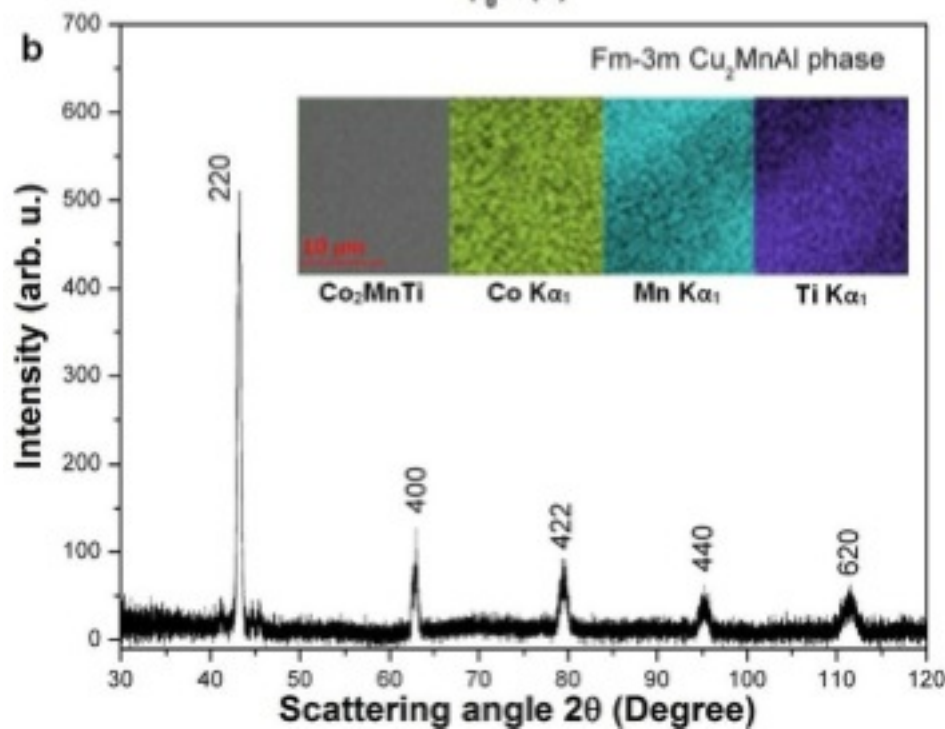
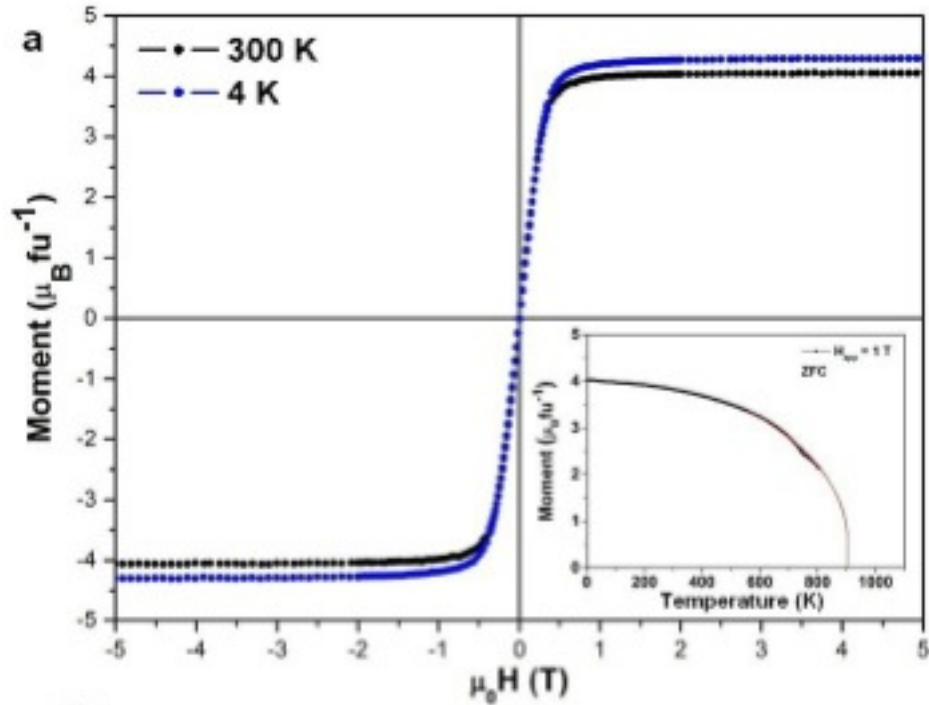
Co₂MnTi

$T_C^{\text{measured}} = 940\text{K}$

$T_C^{\text{predicted}} = 938\text{K}$

Prepared by arc melting in
an Ar atmosphere

Courtesy J.M.D. Coey's Lab
(P. Tozman, M. Venkatesan)



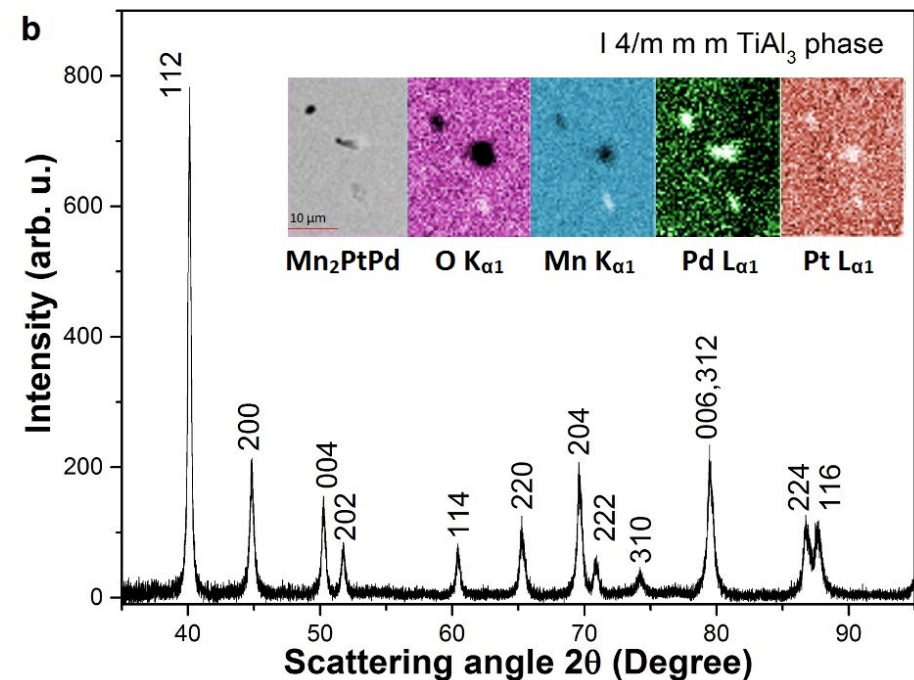
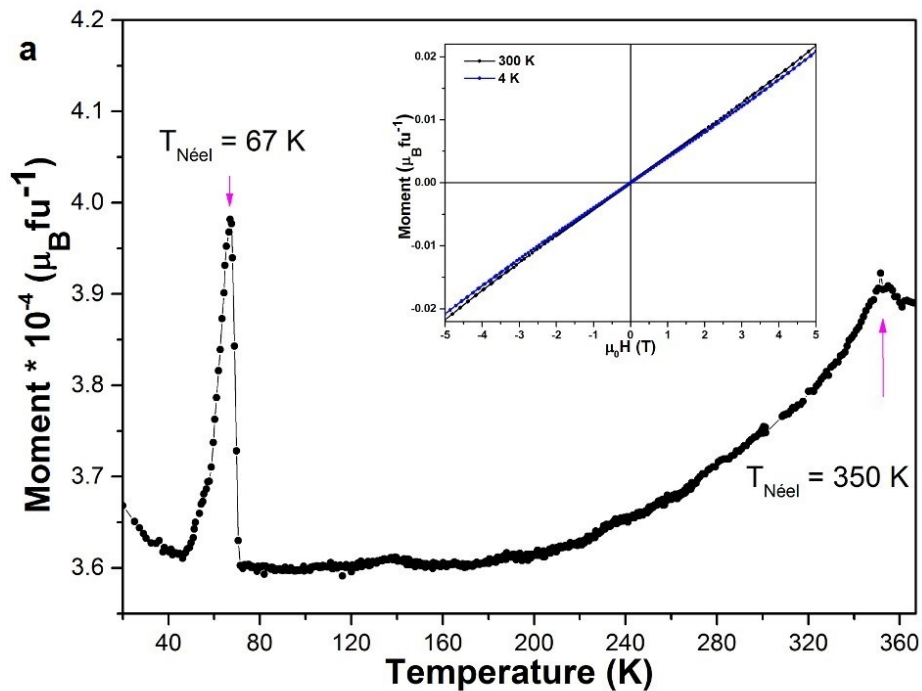
Mn₂PtPd

$T_{N1}^{\text{measured}} = 67\text{K}$

$T_{N1}^{\text{measured}} = 350\text{K}$

Complex antiferromagnetic order

Courtesy J.M.D. Coey's Lab
(P. Tozman, M. Venkatesan)



Demonstrate that HTEST works and that new magnets can be discovered

Yes, it works!! But a massive effort is needed!

Show that, as databases grow, we will become more clever in creating and using them

Maybe ... the algorithm will be clever if the researcher is



COMPUTATIONAL SPINTRONICS

SANVITO RESEARCH GROUP
TRINITY COLLEGE, DUBLIN



CRANN

TCD Team:

Tom Archer, Anurag Tiwari, Mario Zic, James Nelson

Duke Team:

Stefano Curtarolo, Junkai Xue, Kevin Rasch, Corey Oses

