IFT UNESP, Sao Paolo August 2018

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End-to-end materials discovery with electronic structure theory and machine learning

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Trinity College
The University of Dublin

sfi
science foundation ireland
fondúireacht eolaíochta éireann

The question

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How many inorganic materials can you name?

Fe, $Fe_{1-x}C_x$, Si, U, $Nd_2Fe_{14}B$, $YBa_2Cu_3O_7$ ...

Only about 150,000

The question

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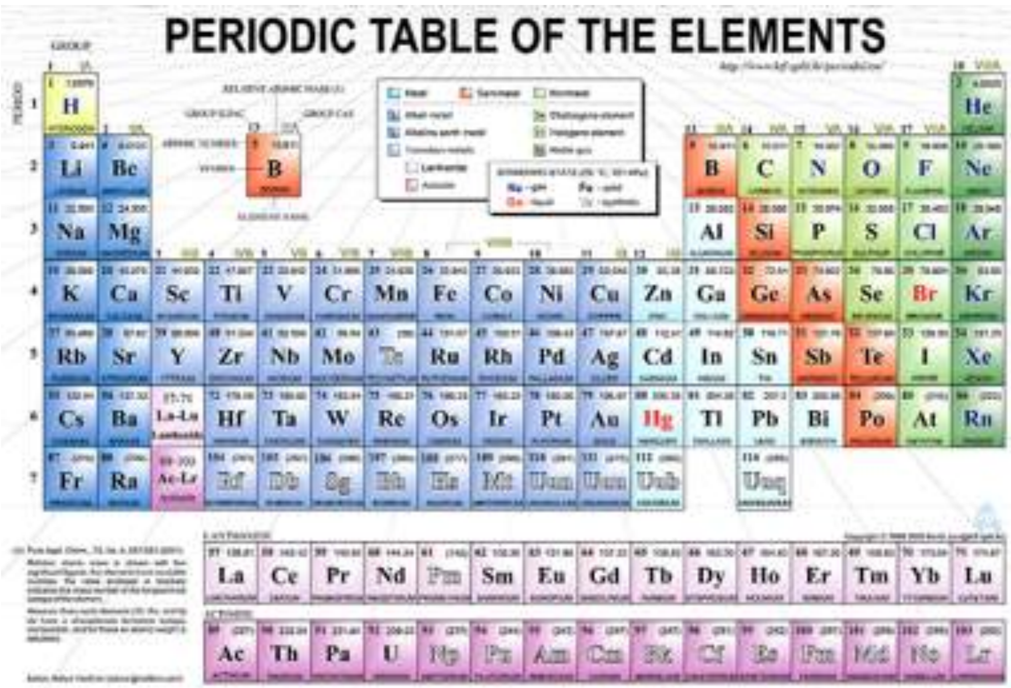
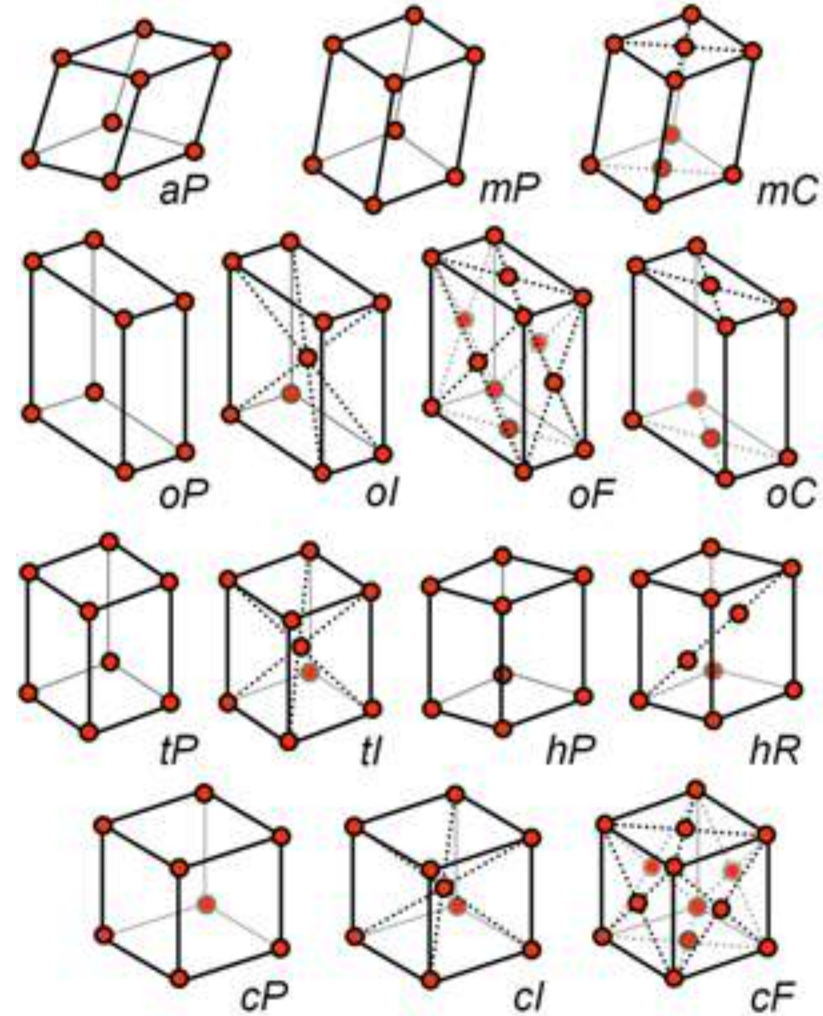
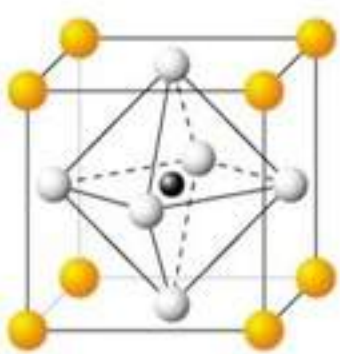
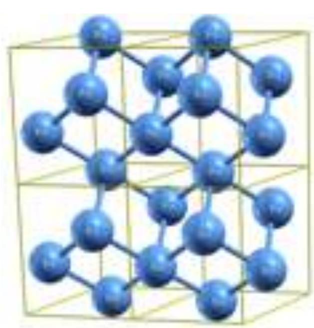


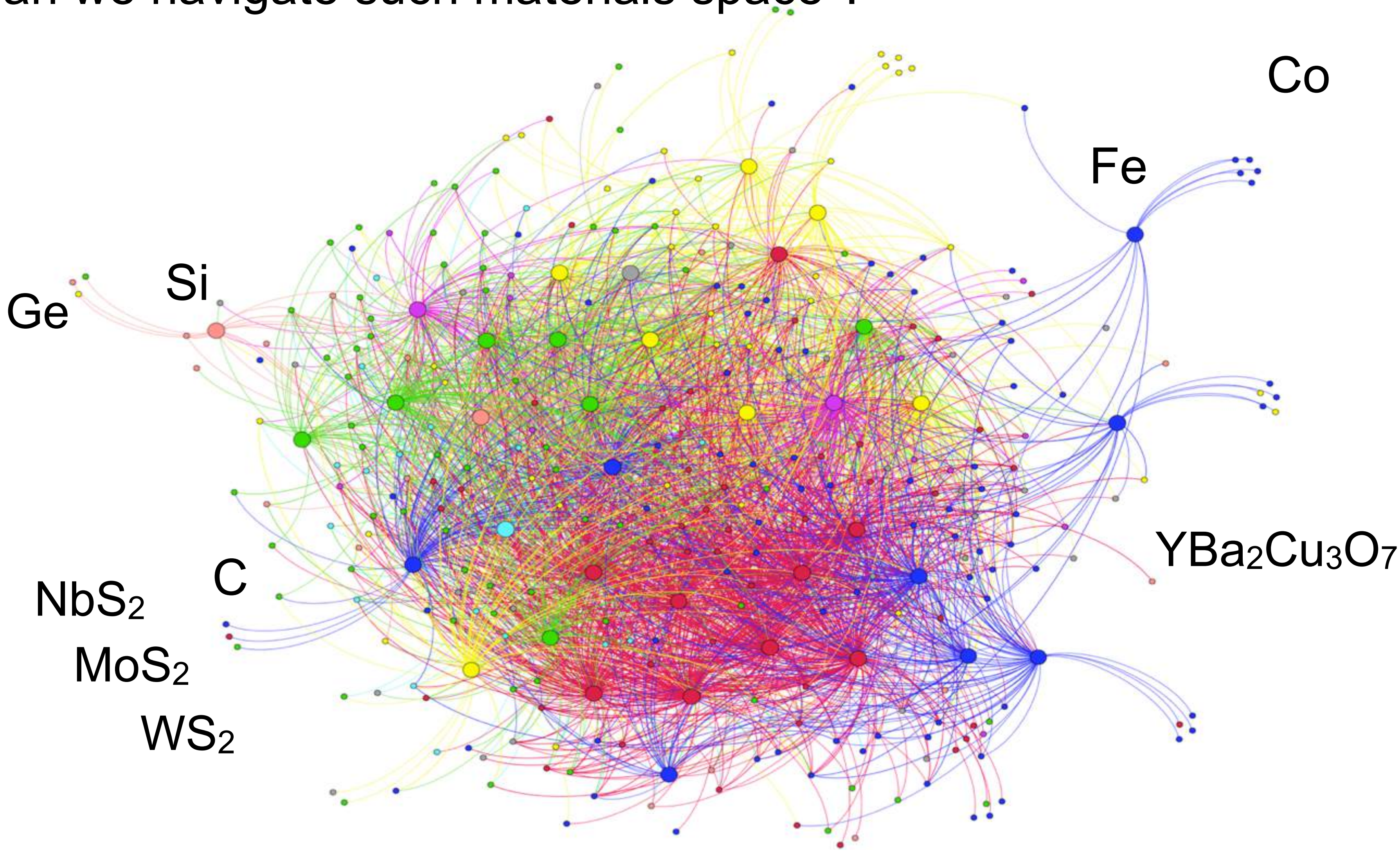






Internet of materials

Can we find out what we are looking for ?
Can we navigate such materials space ?



The question

Suppose you have a new magnetic application …. what is its ideal material(s) ?

Fe, Co, Ni, $Nd_2Fe_{14}B$, $LaMnO_3$, Fe_3O_4 ….

~4,000

Finding new magnets: why ?

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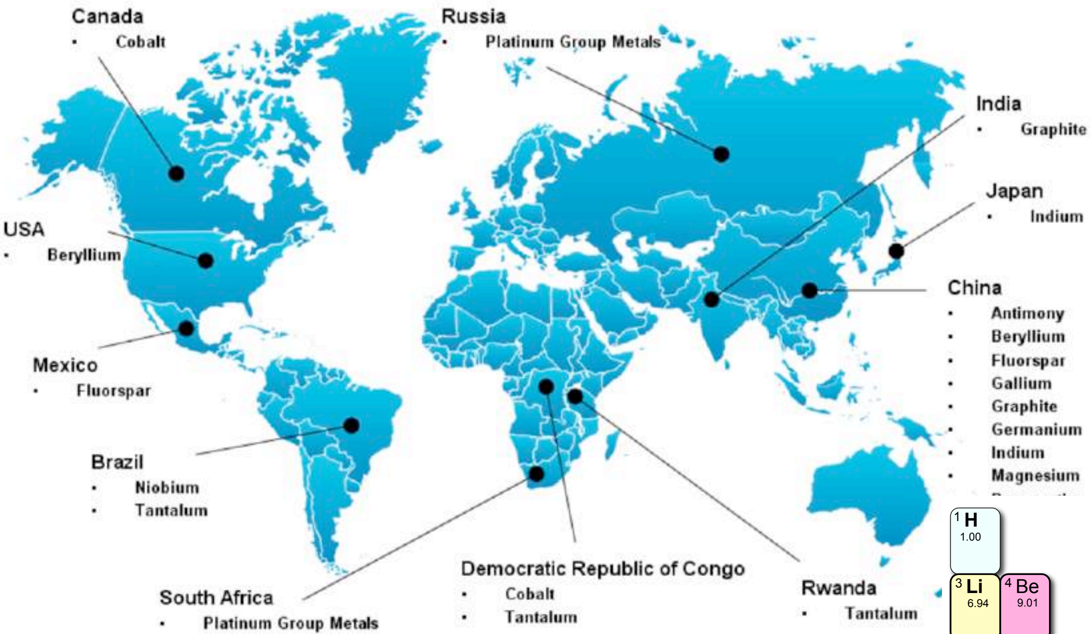
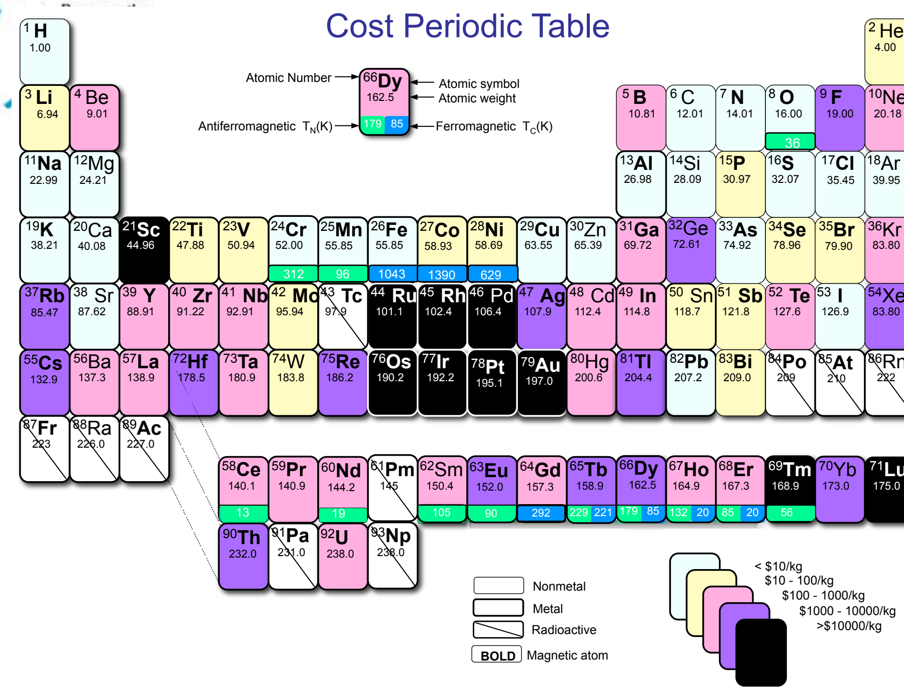


US permanent magnets market ~15B$ (2016)

~22.6B$ (2021)

Novel rare-earth-free permanent magnets

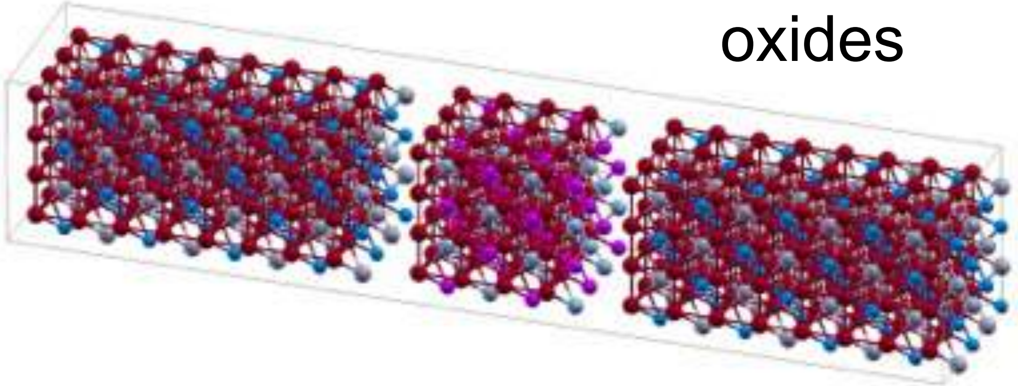


Finding new magnets: why ?

Data storage industry has multiple requirements

MRAM / STT Oscillators

- ❑ High T_C (>600K)
- ❑ High spin polarization (P~100%)
- ❑ Low Gilbert damping
- ❑ Low saturation magnetization
- ❑ Compatible with epitaxial growth of oxides



Magnetism is rare

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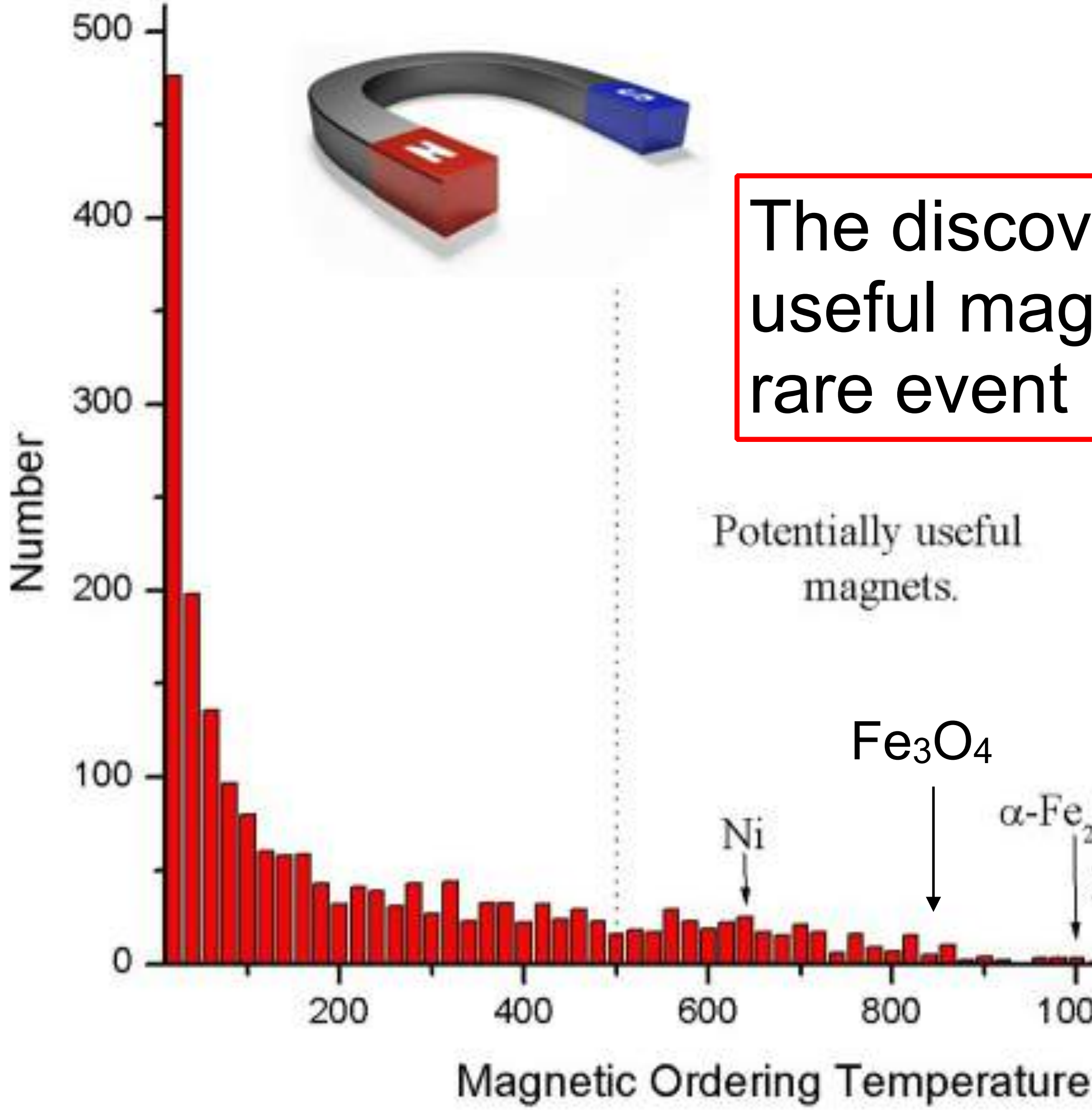
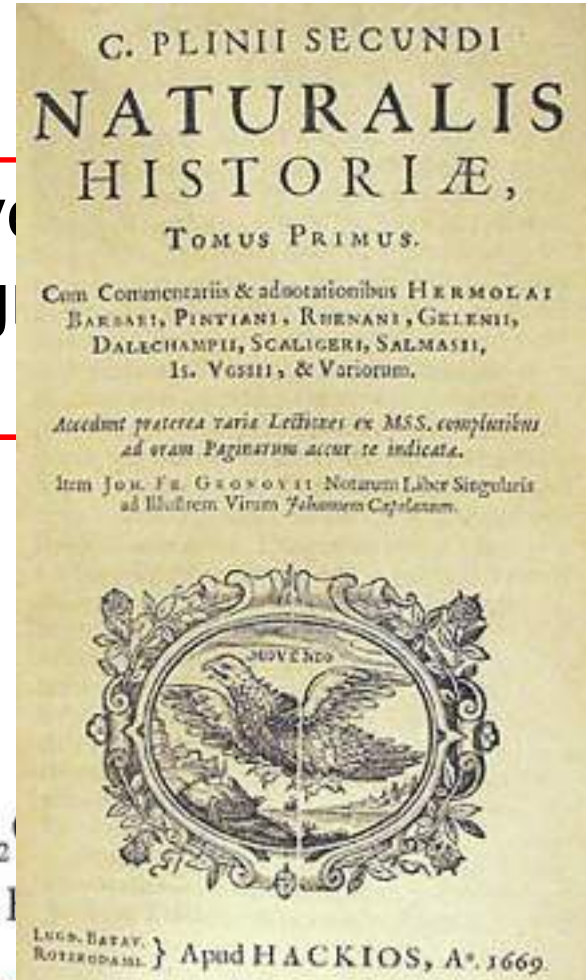


The discove
useful mag
rare event



C. PLINII SECUNDI
NATURALIS
HISTORIÆ,
TOMUS PRIMUS.

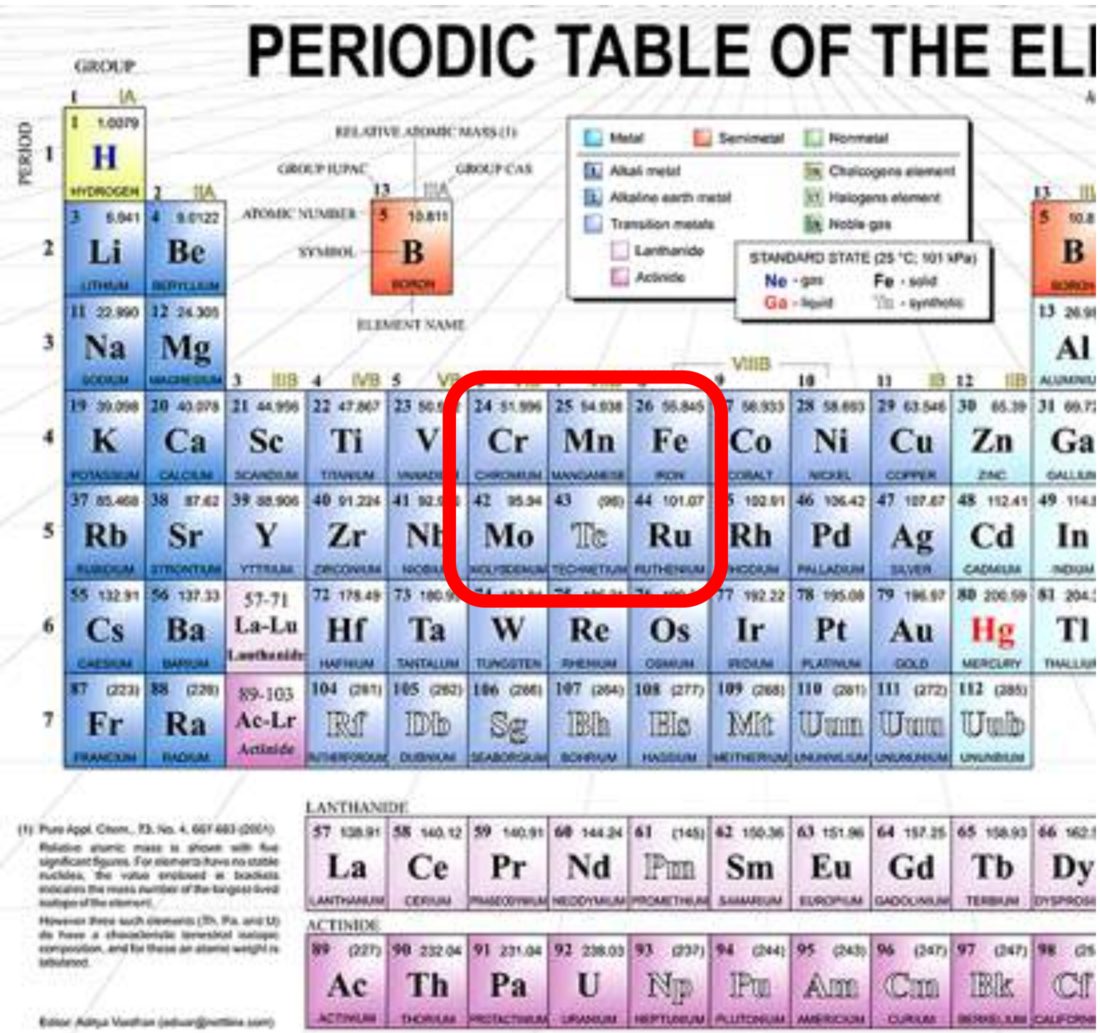
Cum Commentariis & adnotationibus HERMOLAI BARBARI, PINTIANI, RHENANI, GELENII, DALECHAMPII, SCALIGERI, SALMASII, Is. VOSSII, & Variorum.

Accedunt praeterea variæ Lectiones ex MSS. compluribus ad oram Paginarum accurate indicatæ.

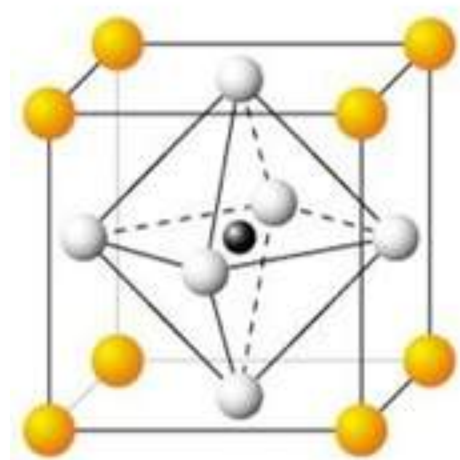
Item JOH. FR. GRONOVII Notarum Liber Singularis ad Illustrem Virum *Johannem Capellanum*.

LUGD. BATAV. ROTERODAMI. } Apud HACKIOS, A°. 1669.

Magnetism is complicated



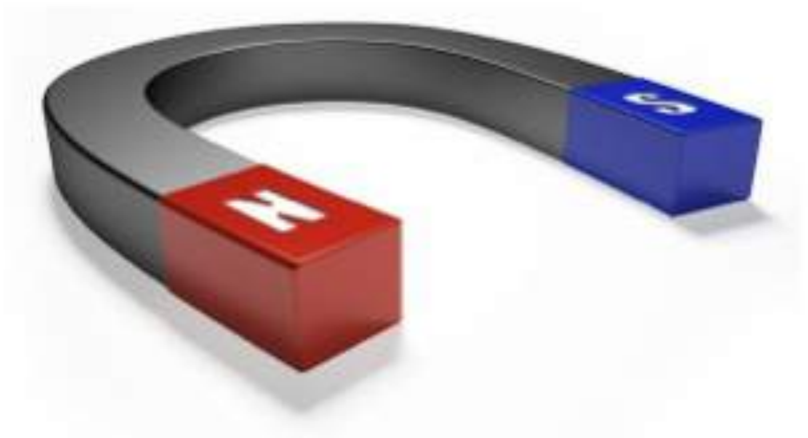
$SrCrO_3$ T_N=-230C	$SrMnO_3$ T_N=-10C	$SrFeO_3$ T_N=-140C
$SrMoO_3$	**$SrTcO_3$ T_N=500C**	$SrRuO_3$ T_C=-100C



$SrMO_3$

C. Franchini, T. Archer, J. He, X.-Q. Chen, A. Filippetti and S. Sanvito, Phys. Rev. B **83**, 220402(R) (2011)

The magnetic genome project



with Stefano Curtarolo, Duke

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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tronic structure method:
tional theory (VASP)

The AFLOW consortium

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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)

The magnetic genome project

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<u>Virtual Materials Growth (existing materials)</u>

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 quintenary
- 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

CRANN

Virtual Materials Growth (existing materials)

Duke calculated single elements, binary, ternary and some quaternary (about 60,000)

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.)

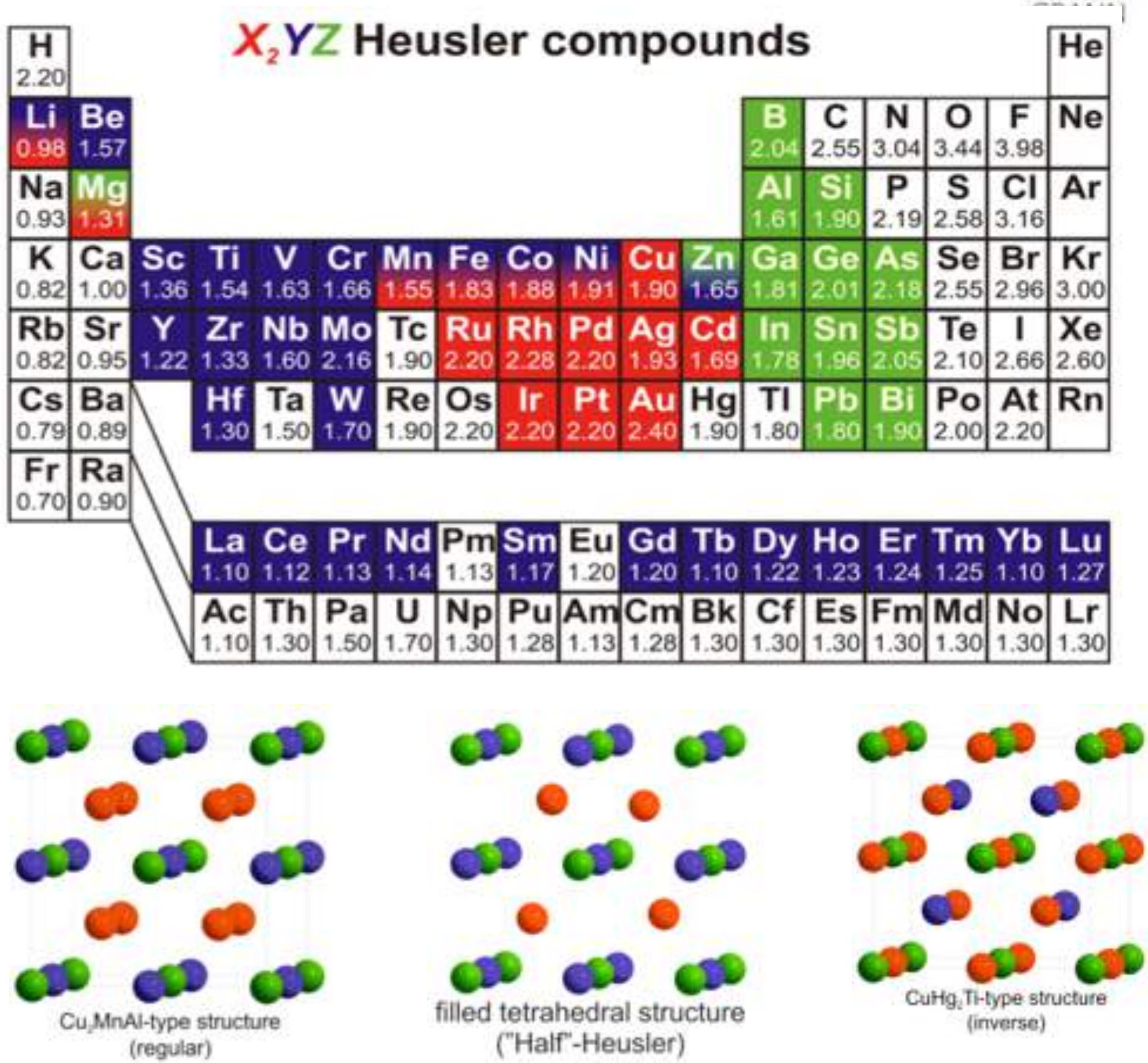
S. Curtarolo, W. Setyawan, G. L. W. Hart, M. Jahnatek, R. V. Chepulskii, R. H. Taylor, S. Wang, J. Xue, K. Yang, O. Levy, M. Mehl, H. T. Stokes, D. O. Demchenko, and D. Morgan, Comp. Mat. Sci. **58**, 218 (2012)

Heusler alloys

~250 known …

~1000 claimed …

~90 magnetic …



Heusler alloys

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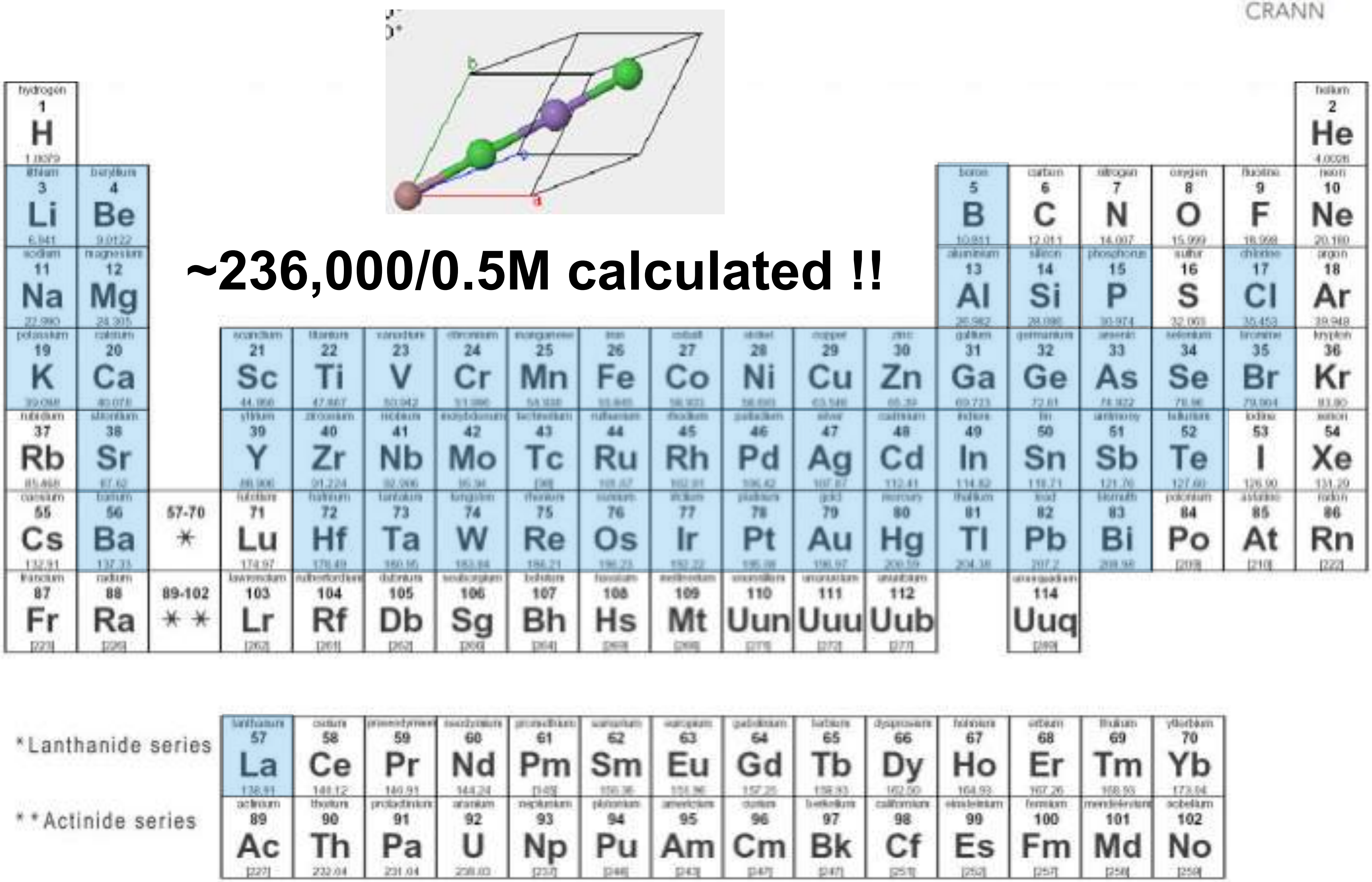


~236,000/0.5M calculated !!

Database

<u>Rational materials storage</u> www.aflowlib.org

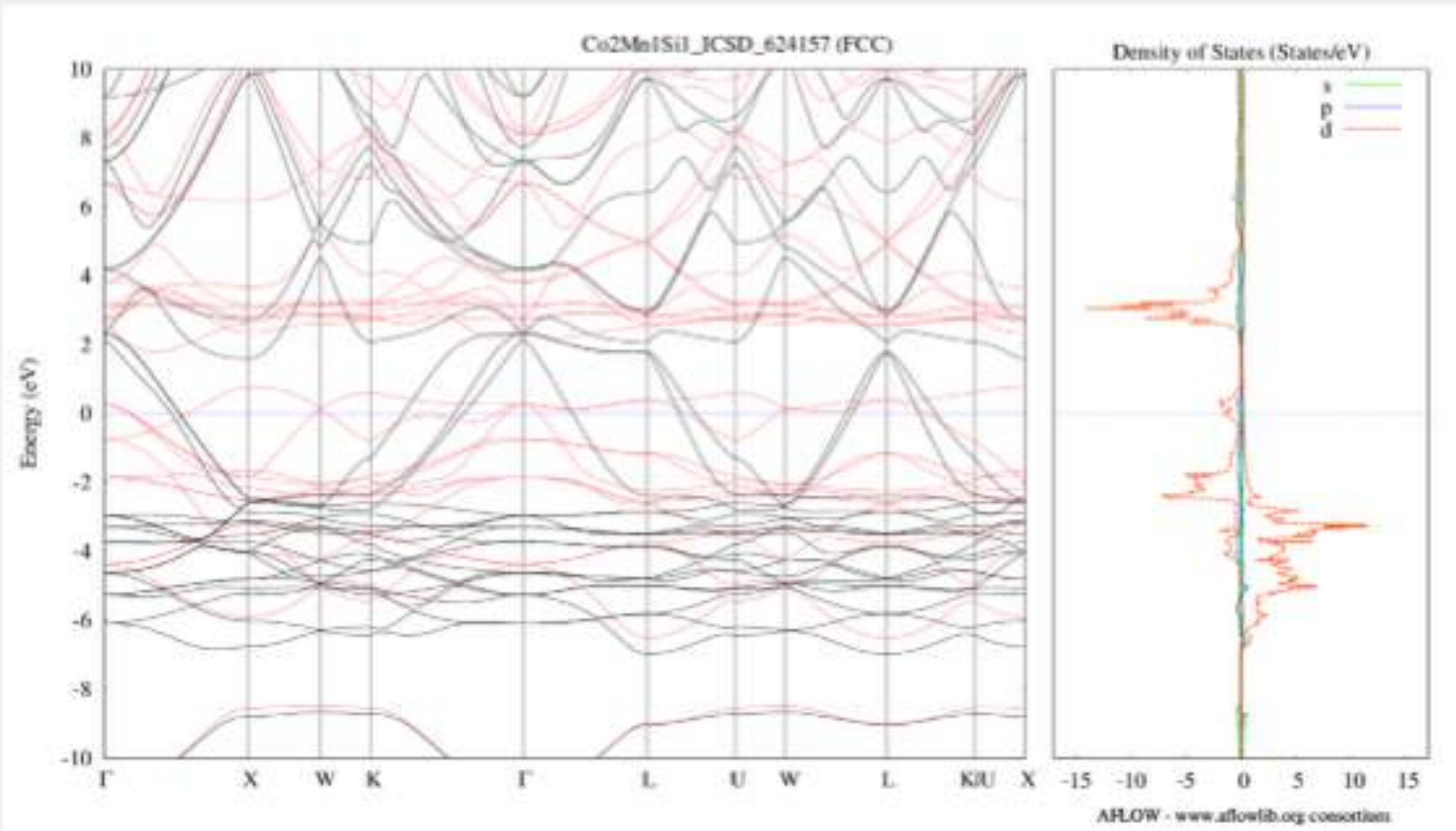
S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R.H. Taylor, L.J. Nelson, G.L.W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, Comp. Mat. Sci. **58**, 227 (2012)

Database

ELECTRONIC PROPERTIES

Band Gap:	0.000 eV (metal)	Fit Band Gap:	0.000 eV
Magnetic Moment:	7.382 μ_B	Magnetic Moment/atom:	1.845 μ_B/atom
Electron Mass(FIX):	XXX (m_0)	Hole Mass(FIX):	XXX (m_0)
Spin Polarization (E_F):	0.666	Spin Decomposition per atoms:	{1.758,1.758,4.019,-0.054} μ_B

Band Structure:



Comp. Mat. Sci. **49**, 299-312 (2010)

The magnetic genome project

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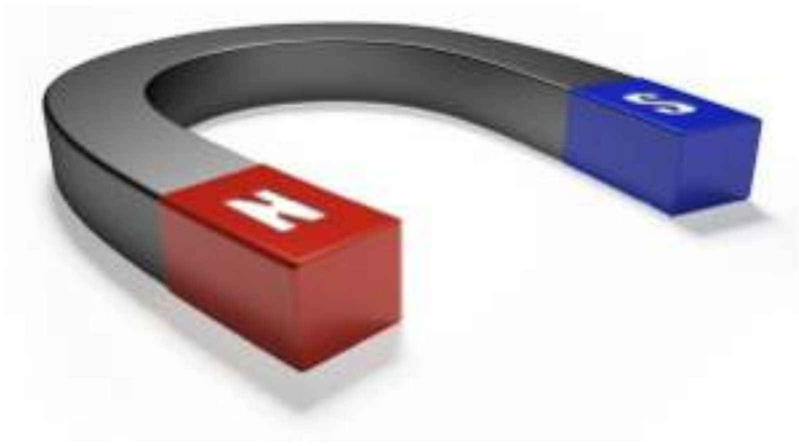
Back to the magnets …..



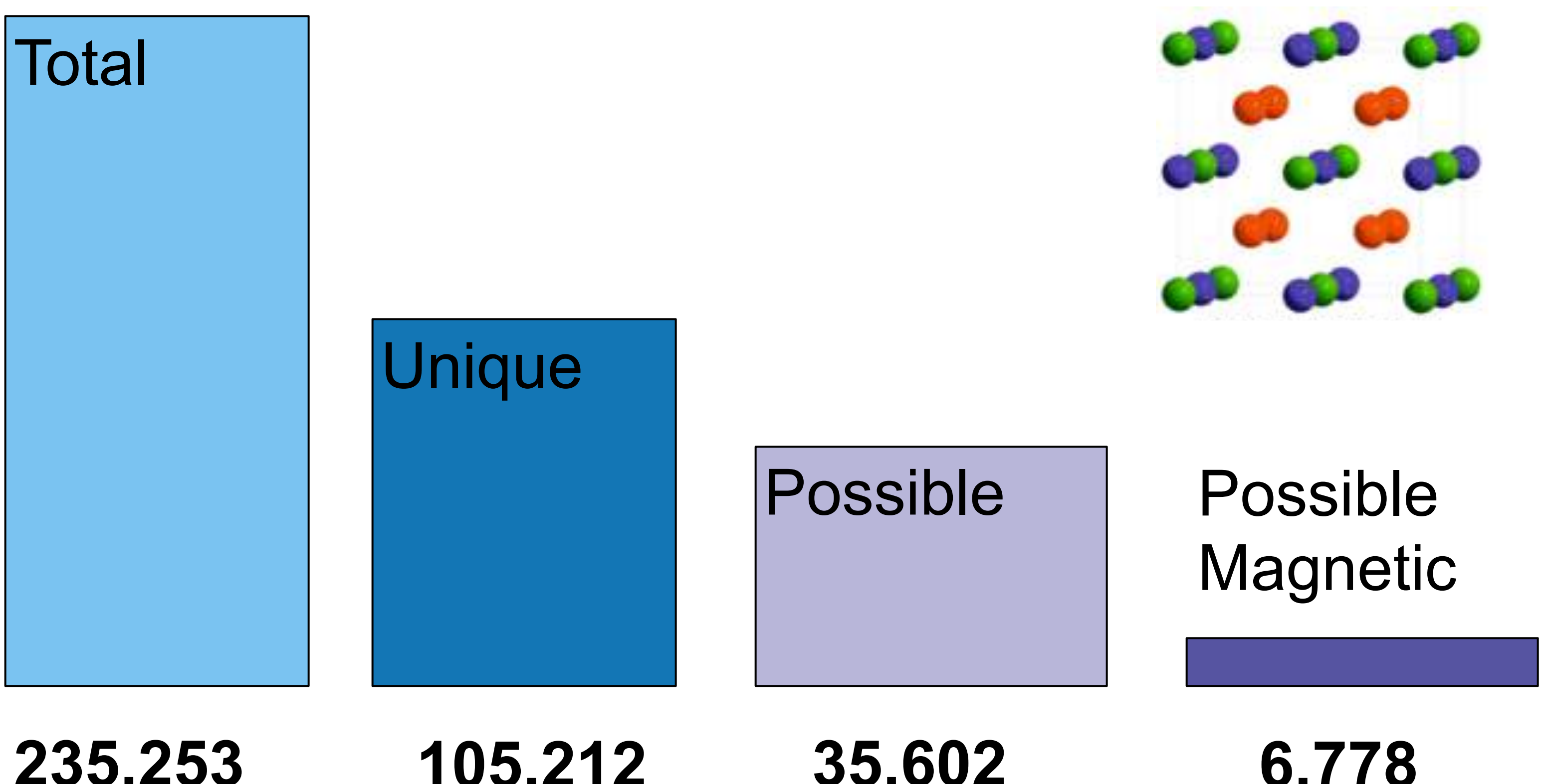
S. Sanvito et al., *Accelerated discovery of new magnets in the Heusler alloy family*, Science Advances **3**, e1602241 (2017)

A look at the full database

Property: Can be made ?

Descriptor 0:
Enthalpy of formation

Energy (Ni_2MnAl) < Energy (2Ni + Mn +Al)



Stability analysis

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Descriptor 1: Enthalpy of formation

2 Ni + Mn + Al

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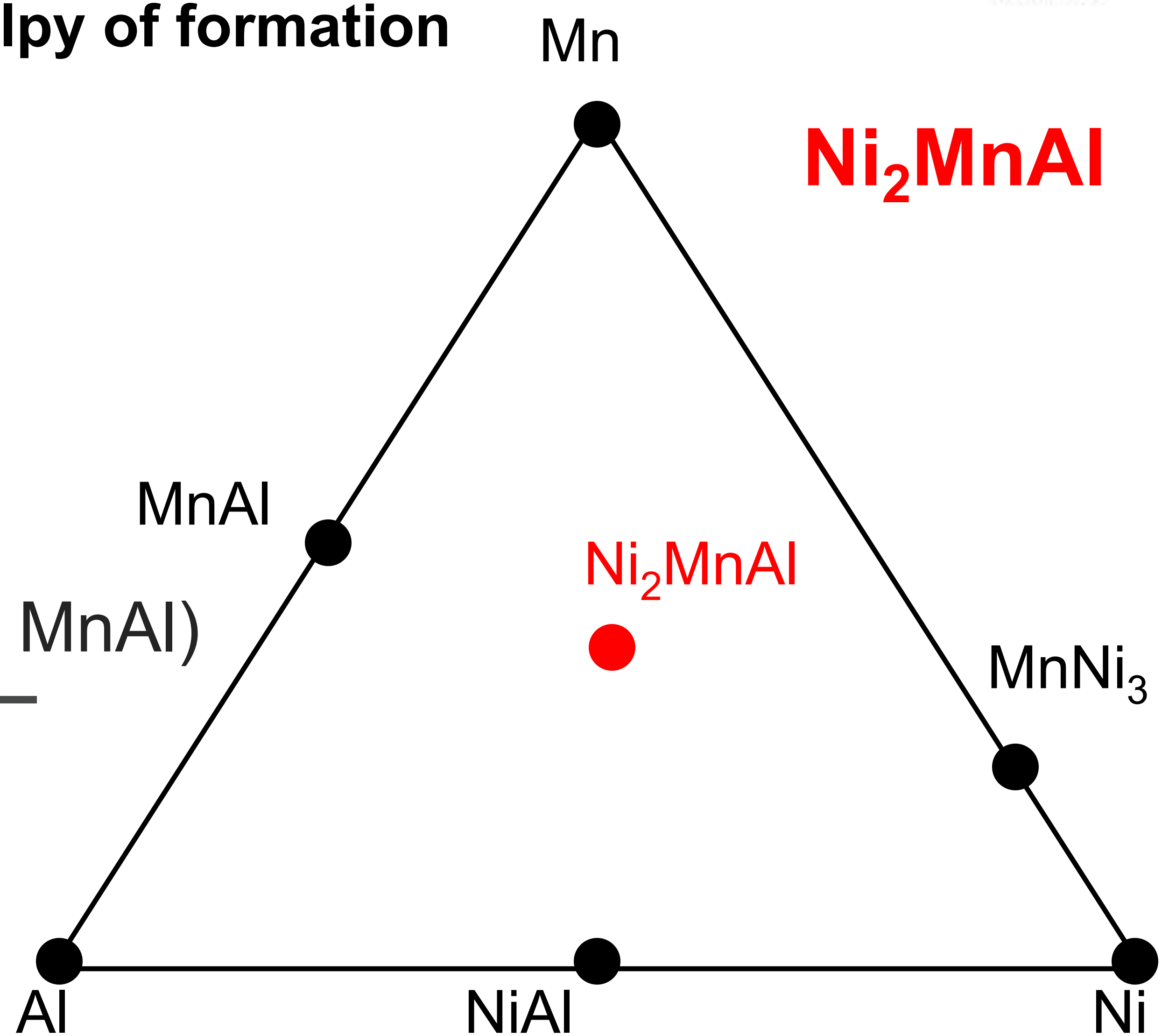
2 Ni + MnAl

1/2 ($MnNi_3$ + NiAl + MnAl)

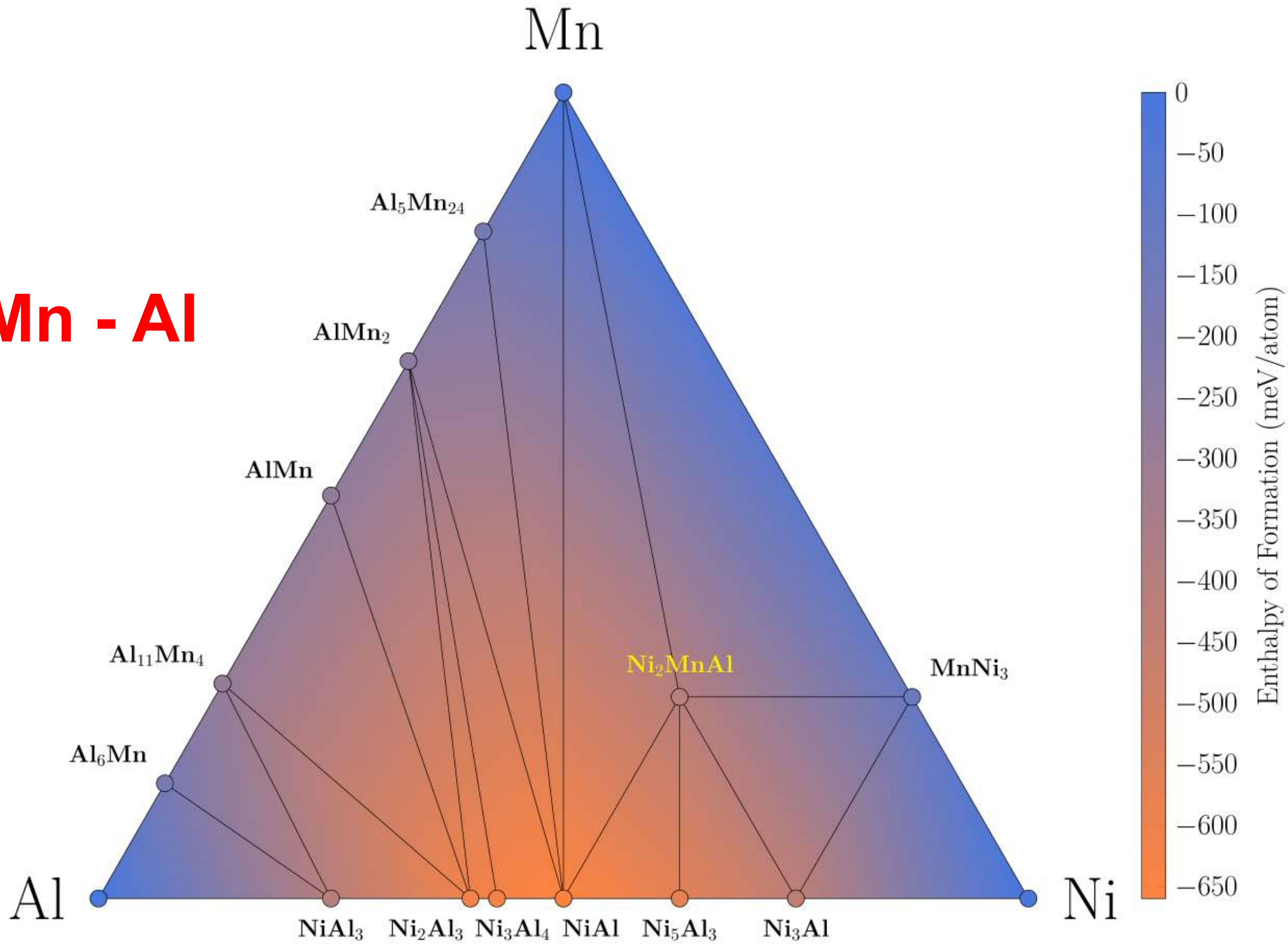
Ni_2MnAl

Ni_2MnAl



Stability analysis

Ni - Mn - Al



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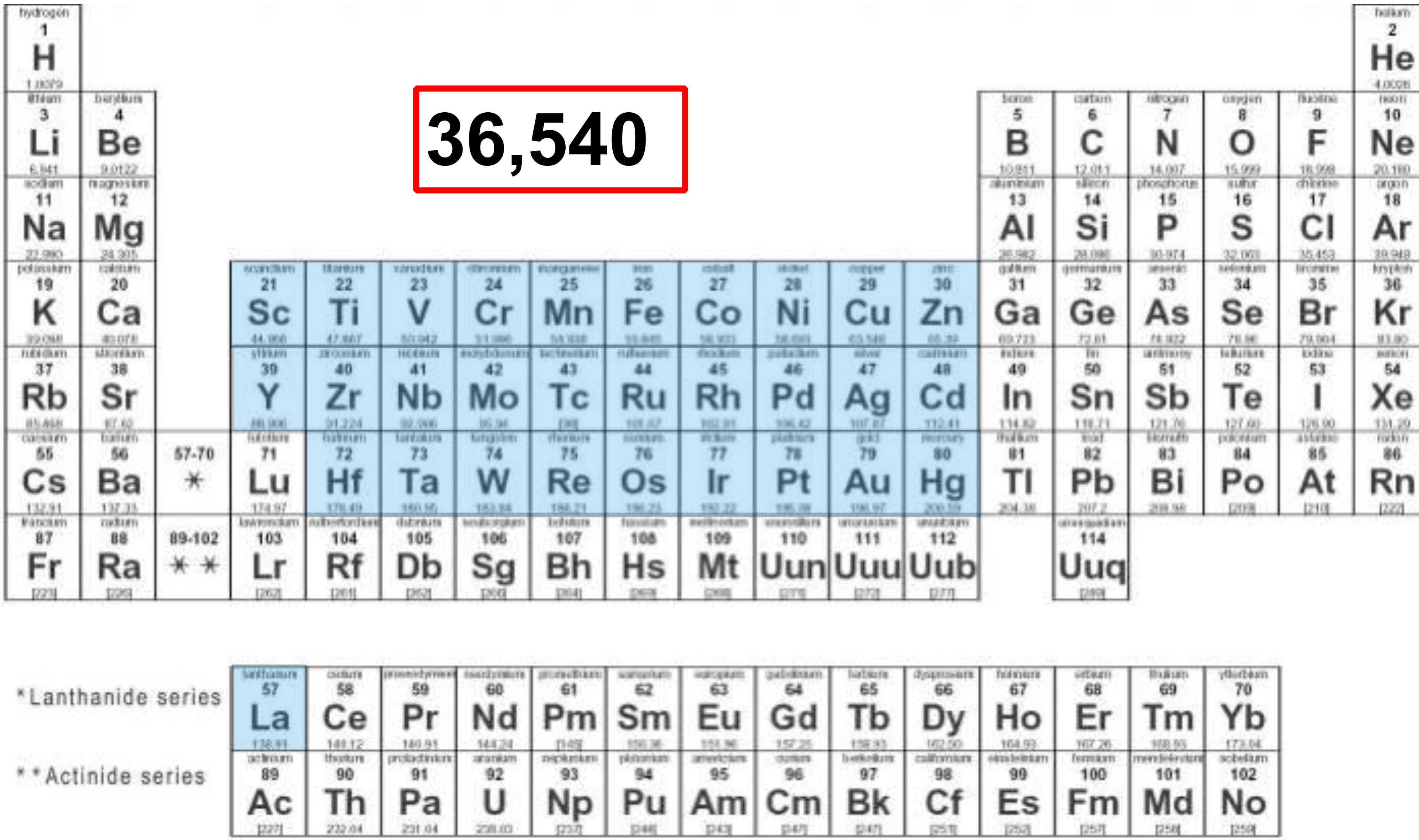
Look at the transition metal intermetallics

36,540



In summary …

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36,540 possible → 248 stable

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

↓ Extrapolating

236,000 possible → 1550 stable

138 magnetic → 50 Robust

↓ For real ….

52 magnetic

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

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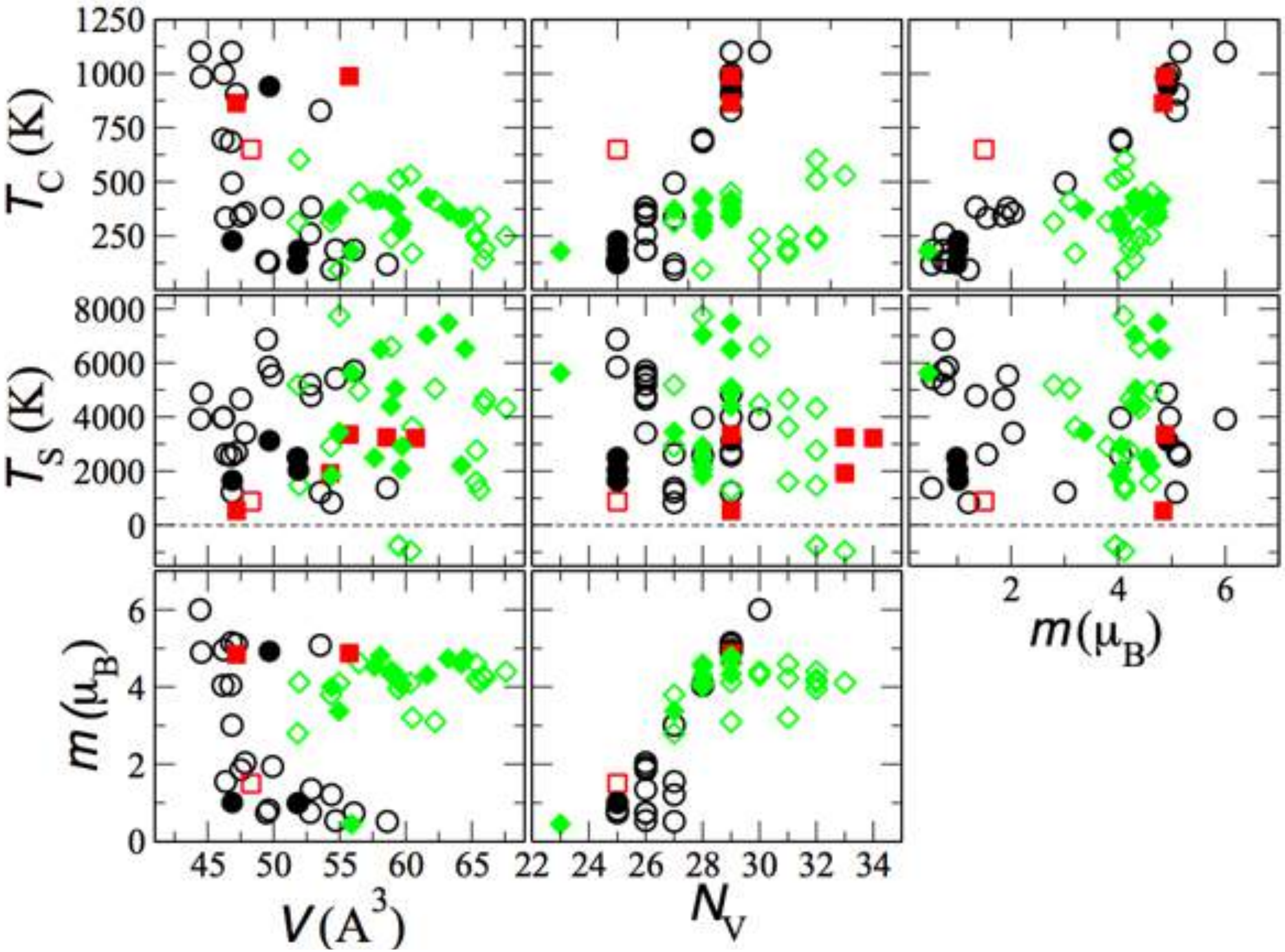
Co_2XY

Mn_2XY

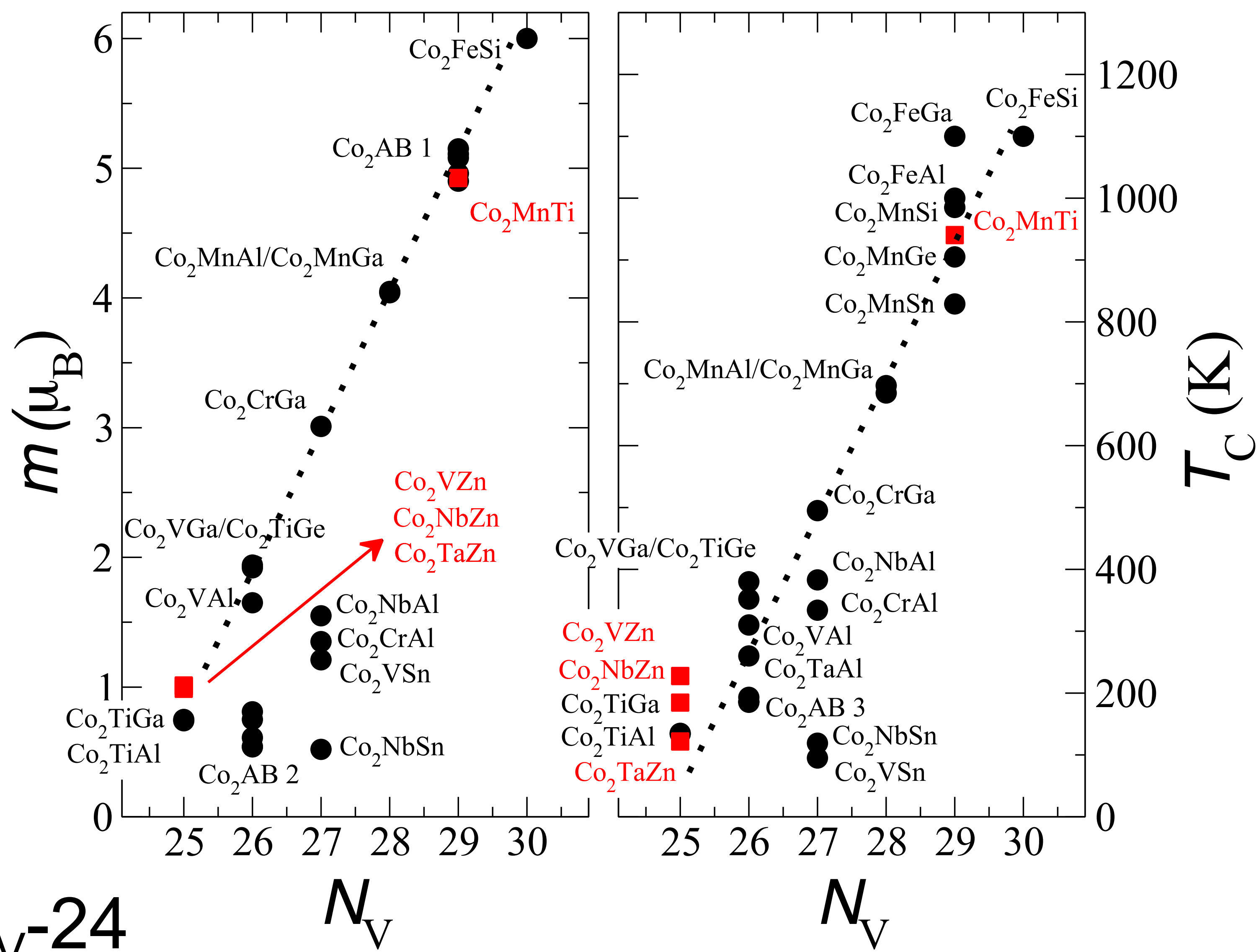
X_2MnY



Co_2YZ

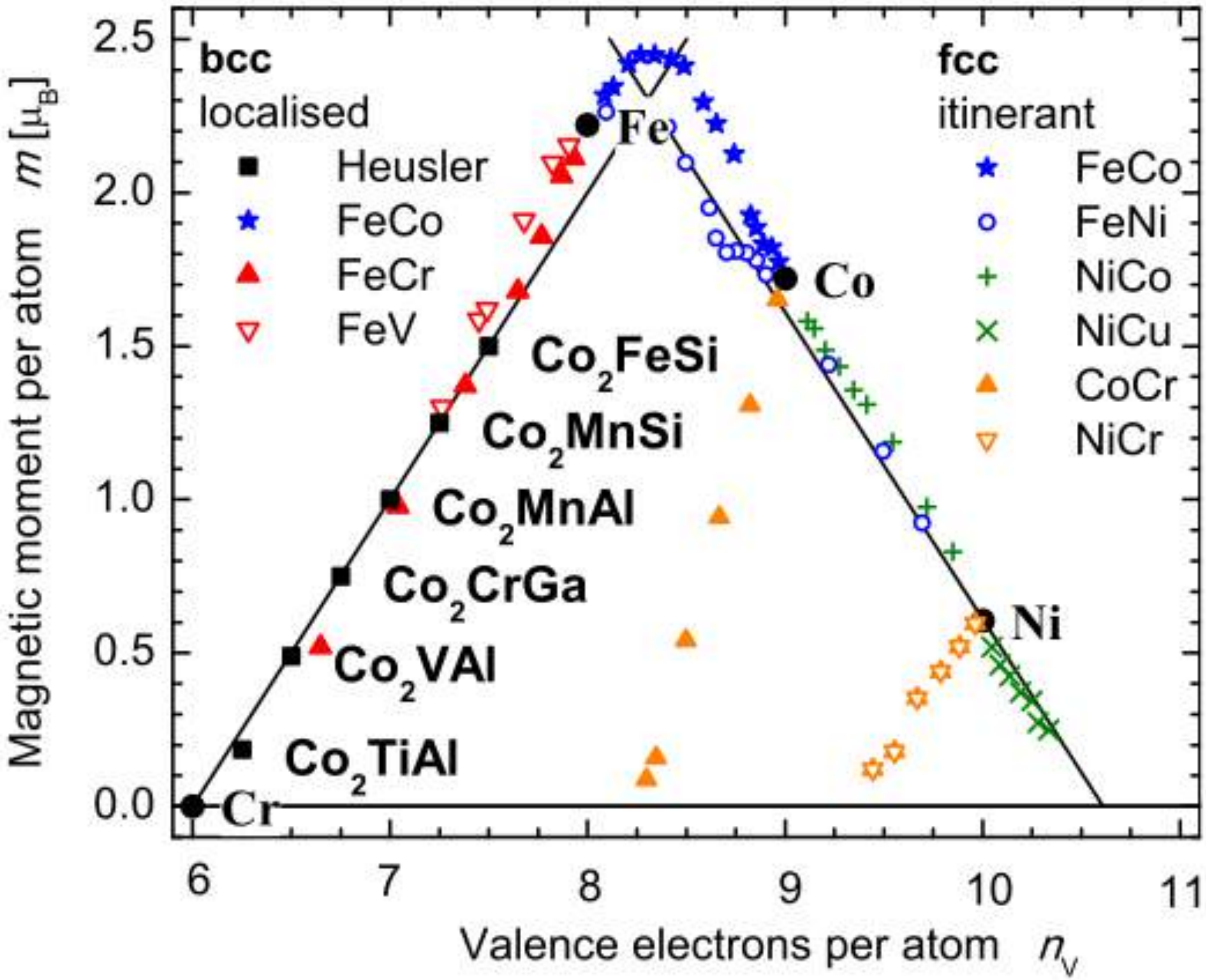
Co_2YZ

Slater-Pauling



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

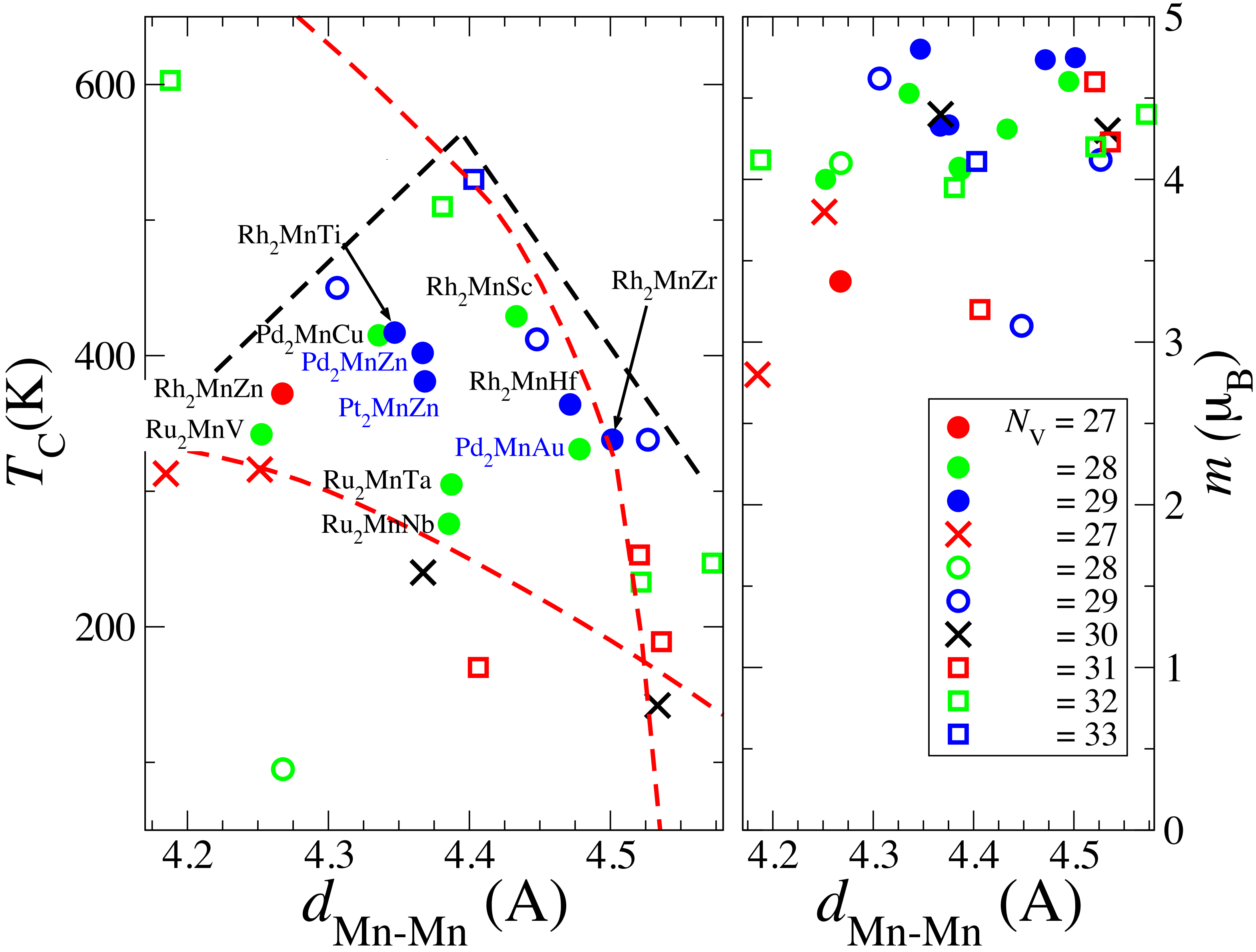
Co_2YZ



bcc
localised
Heusler
FeCo
FeCr
FeV
fcc
itinerant
FeCo
FeNi
NiCo
NiCu
CoCr
NiCr
Fe
Co
Ni
Cr
Co2FeSi
Co2MnSi
Co2MnAl
Co2CrGa
Co2VAl
Co2TiAl
Magnetic moment per atom m [μB]
Valence electrons per atom nV
2.5
2.0
1.5
1.0
0.5
0.0
6
7
8
9
10
11

X_2MnZ

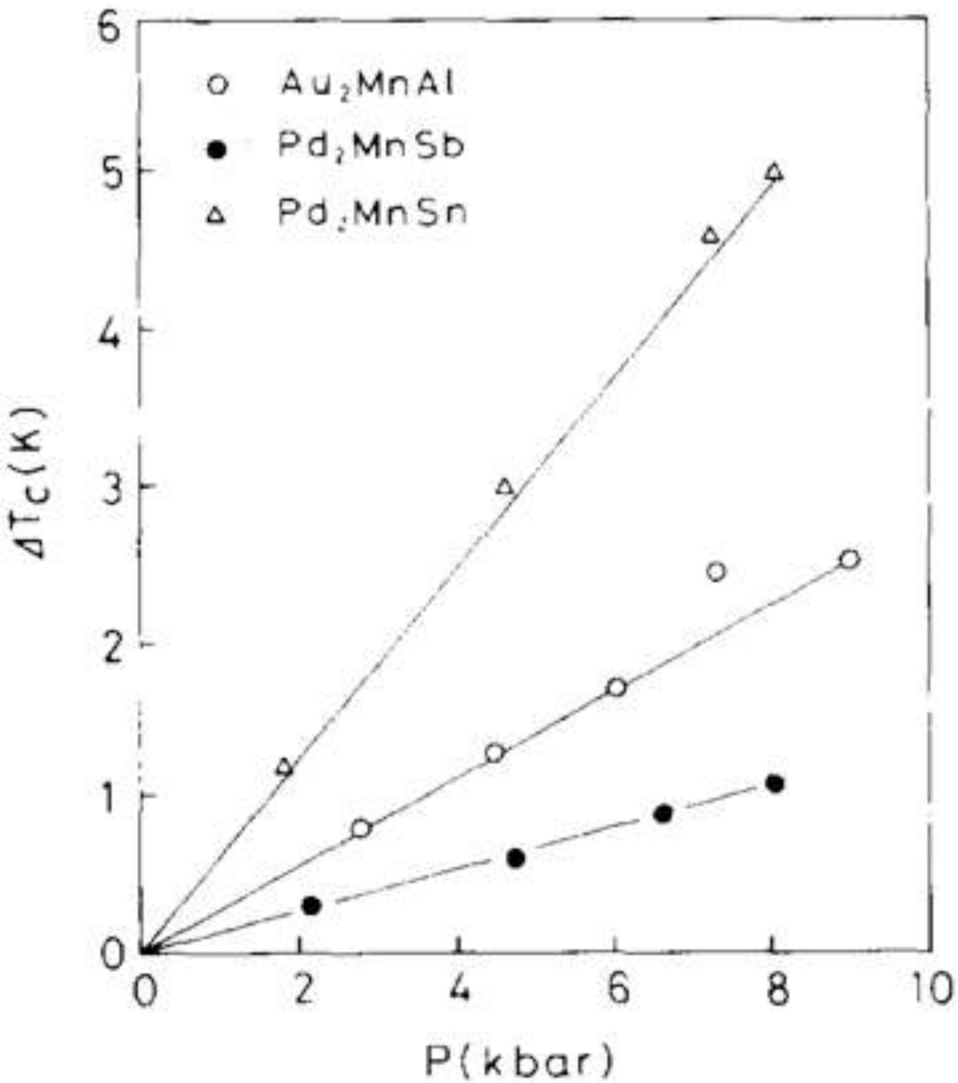
X_2MnZ



Castelliz-Kanomata curve

X_2MnZ

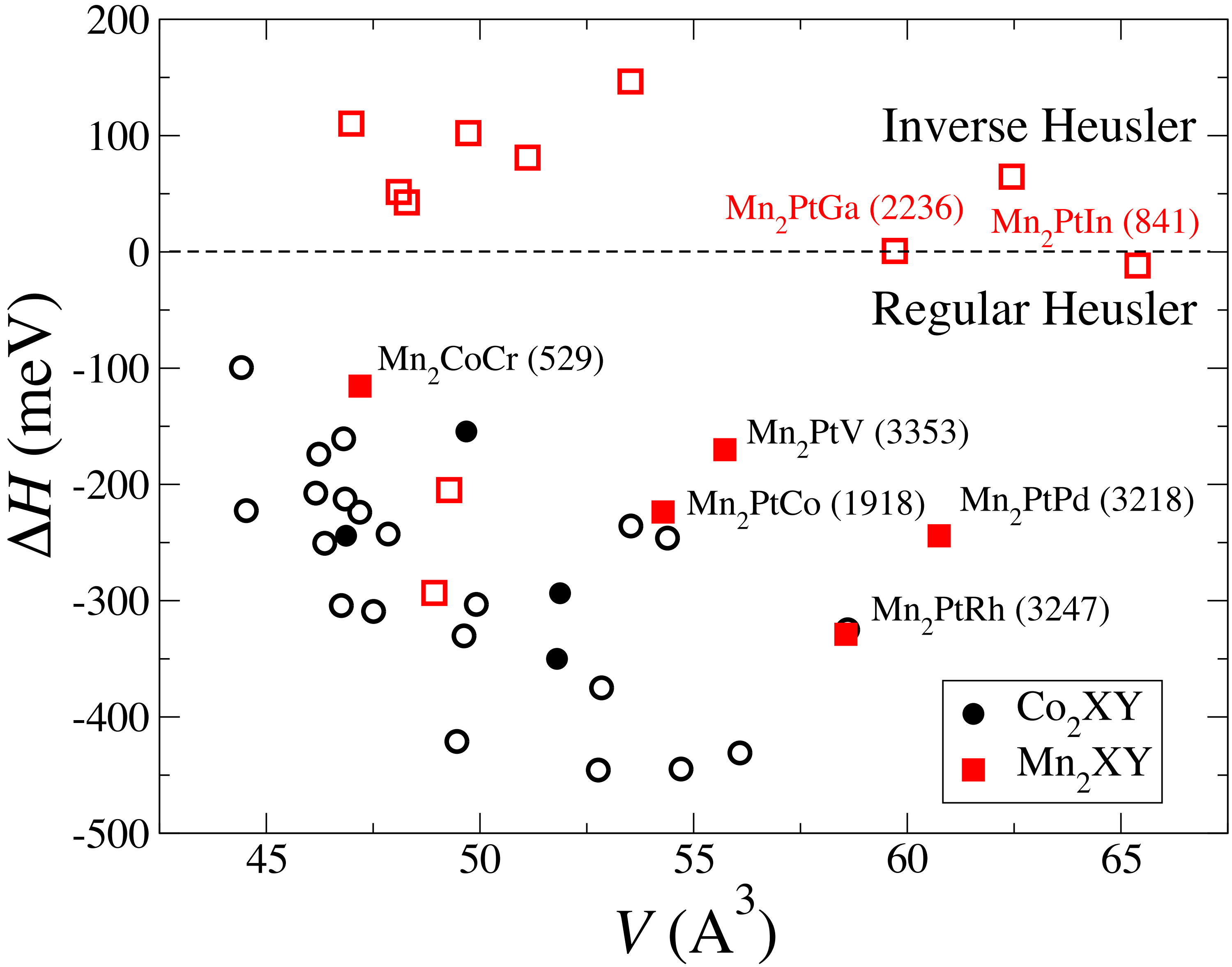
X_2MnZ



K. Shirakawa et al., J. Magn. Magn. Mater. **70**, 421 (1987)

Mn_2YZ

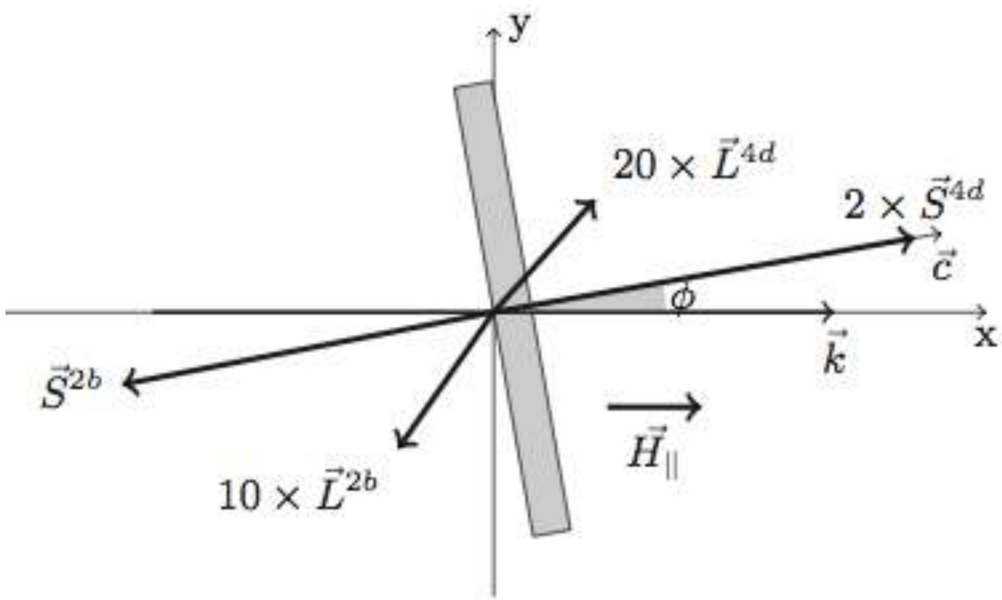
Mn_2YZ

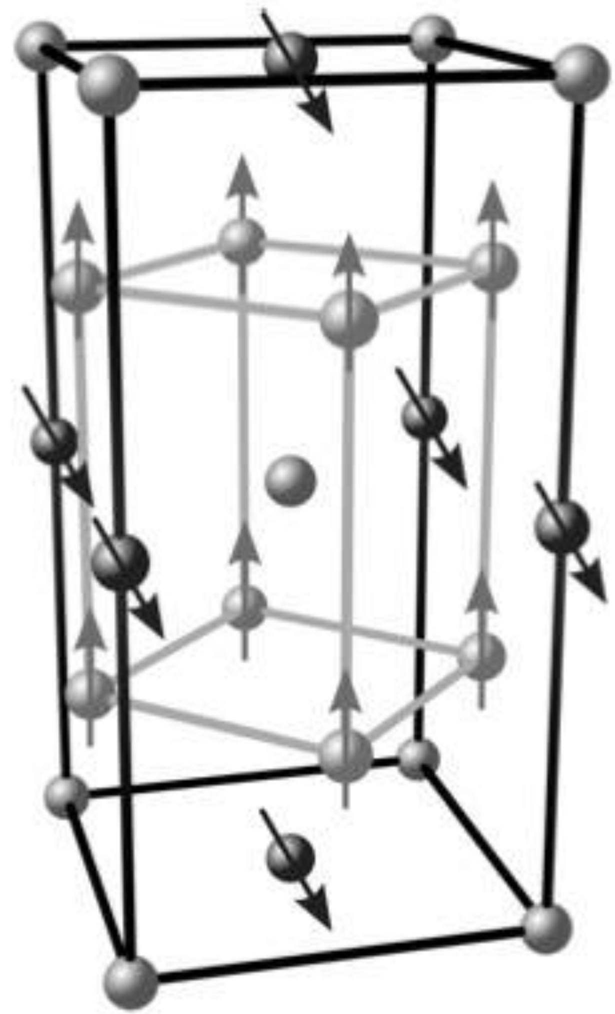


Inverse Heusler
Regular Heusler
Mn2PtGa (2236)
Mn2PtIn (841)
Mn2CoCr (529)
Mn2PtV (3353)
Mn2PtCo (1918)
Mn2PtPd (3218)
Mn2PtRh (3247)
Co2XY
Mn2XY
ΔH (meV)
V (A3)
200
100
0
-100
-200
-300
-400
-500
45
50
55
60
65

Mn_3Ga

Mn_3Ga





K. Rode et al., Phys. Rev. B **87**, 184429 (2013)

Tetragonal distortion

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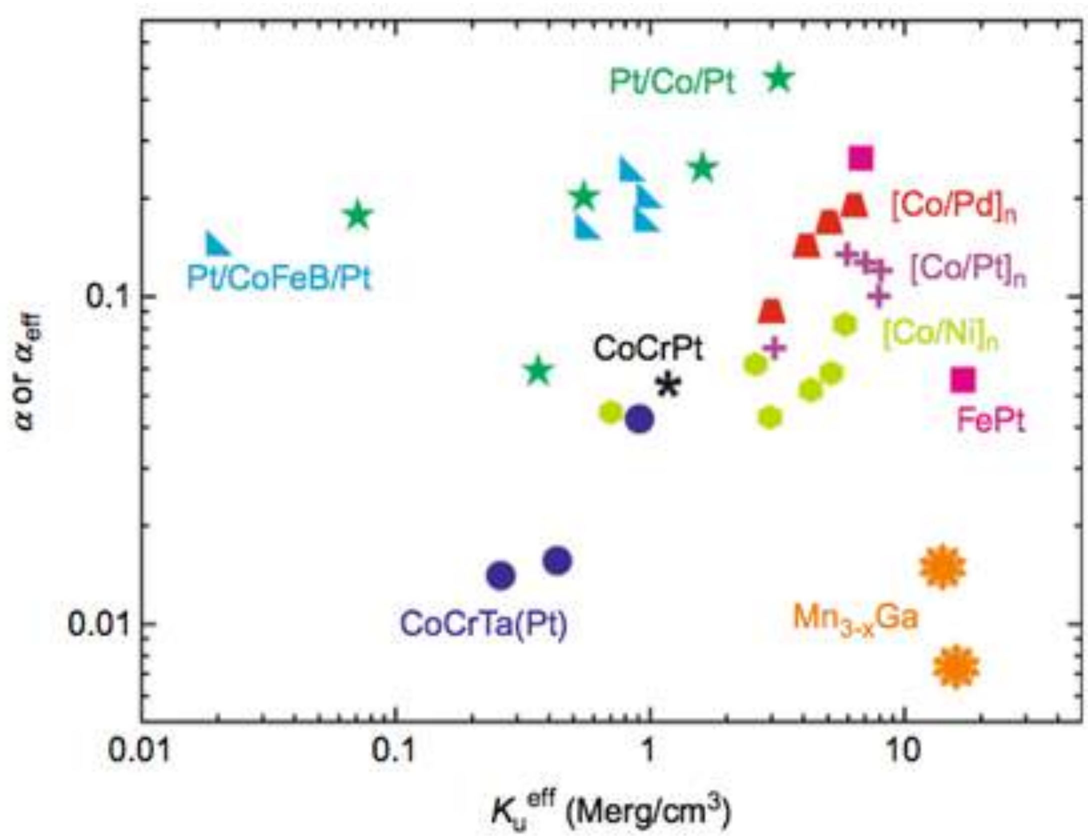
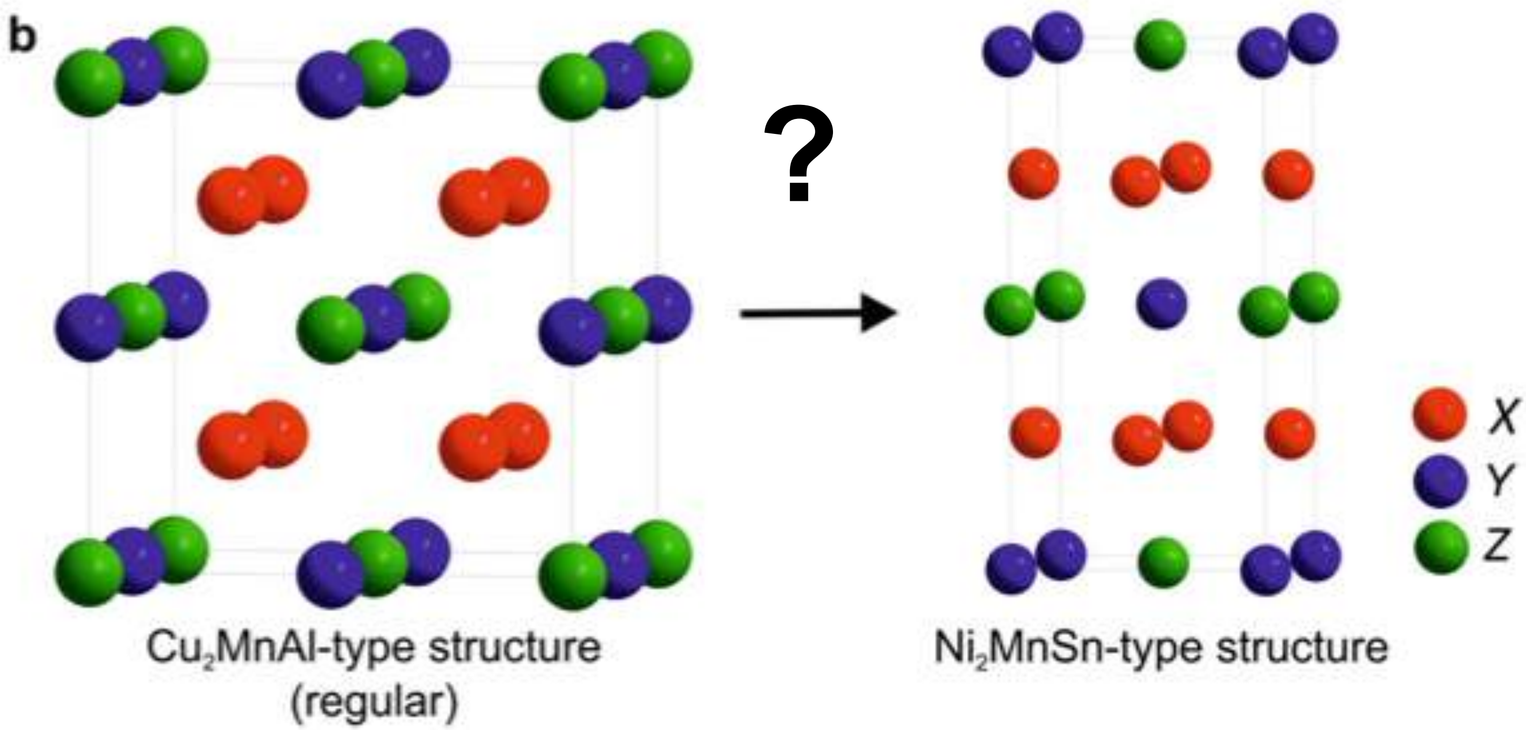
Descriptor 3: Magneto-crystalline anisotropy

Little magnetic anisotropy in cubic symmetry.
Tetragonal distortion does not much better!

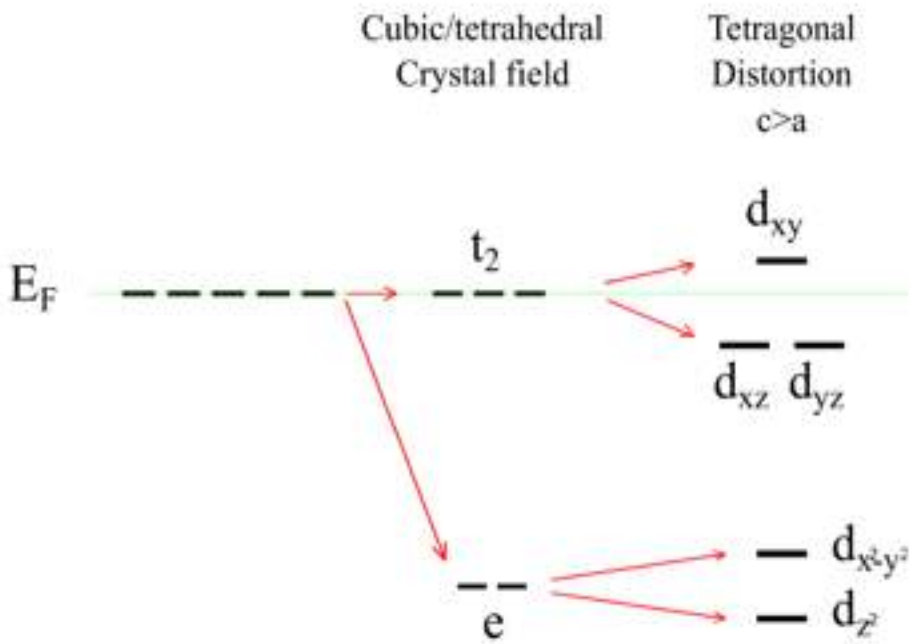


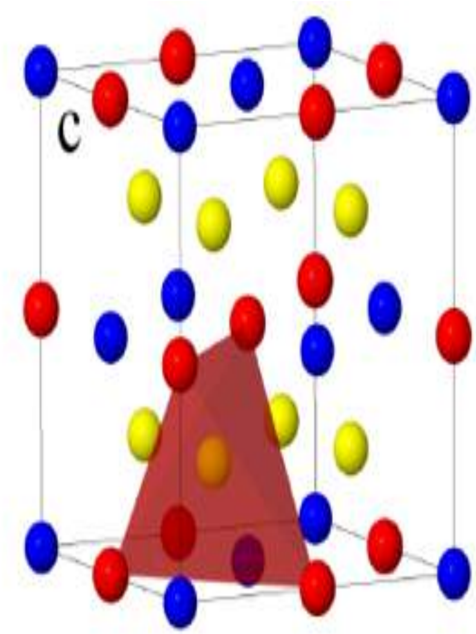
T. Graf et al., in *Handbook of Magnetic Materials*, Elsevier (2013)

Tetragonal distortion



Tetragonal distortion





Mn_3Ga Ni_2MnGa Mn_2NiGa Rh_2VSn Pd_2NbSn

Mn_3Ge Ni_2MnSn Rh_2CrSn Pd_2TbSn

Ni_2MnIn Co_2NbSn Rh_2FeSn Pd_2DySn

Rh_2CoSn

Mechanism for tetragonal distortion

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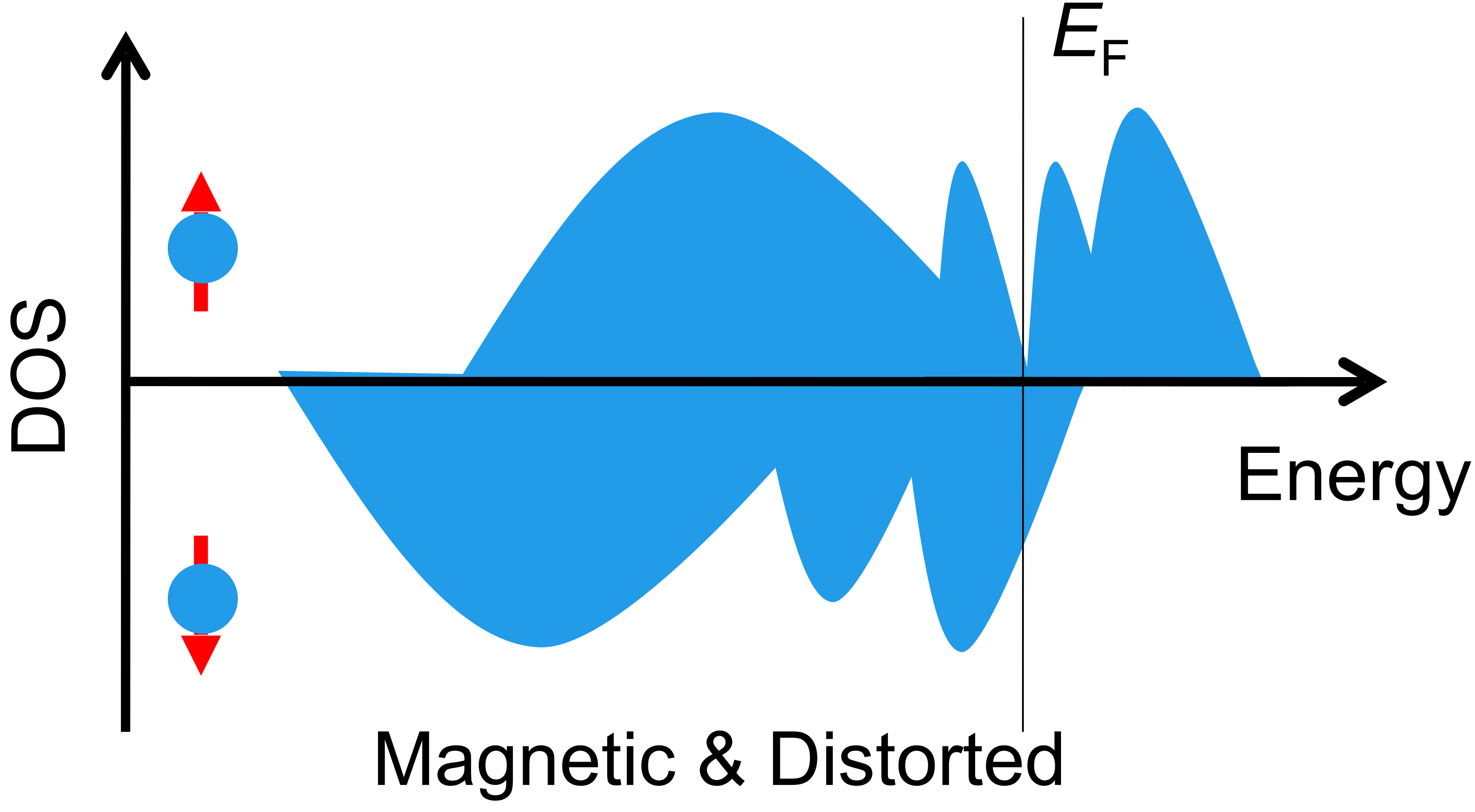
Stoner Criterion vs band Jahn-Teller distortion



Tetragonal distortion

Among our 22:

<u>2 turn diamagnetic</u>

Co_2NbZn

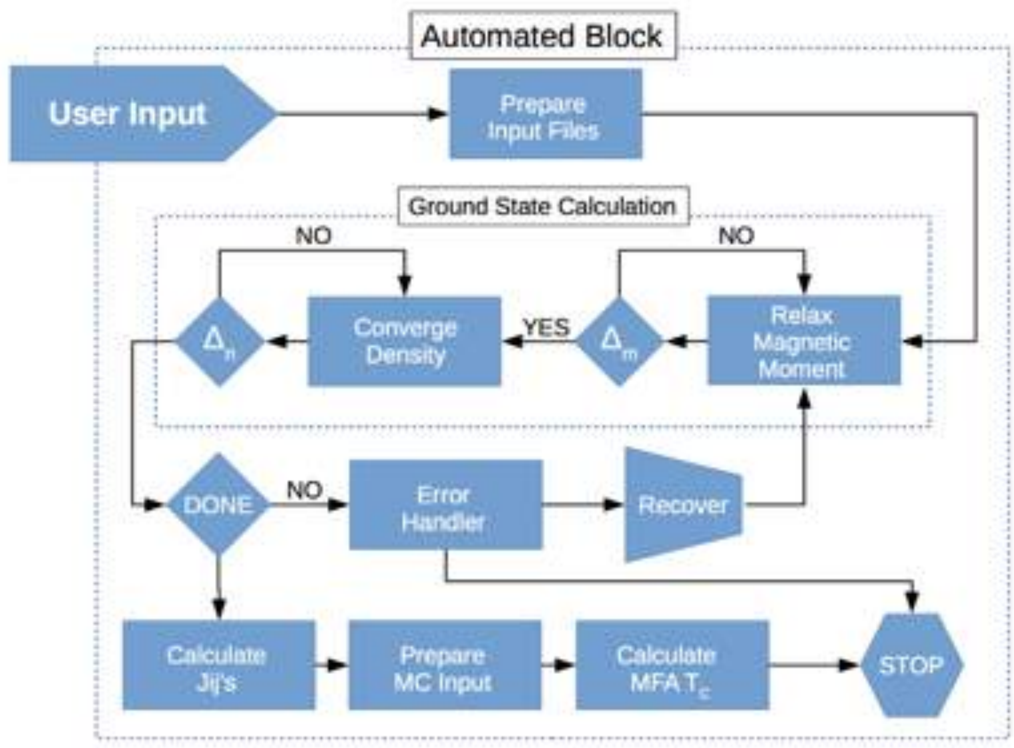
Co_2TaZn

<u>3 remain magnetic</u>

X_2MnZ

P_F~0, T_C~300-400, T_N~2000-5000

A different workflow



Machine Learning

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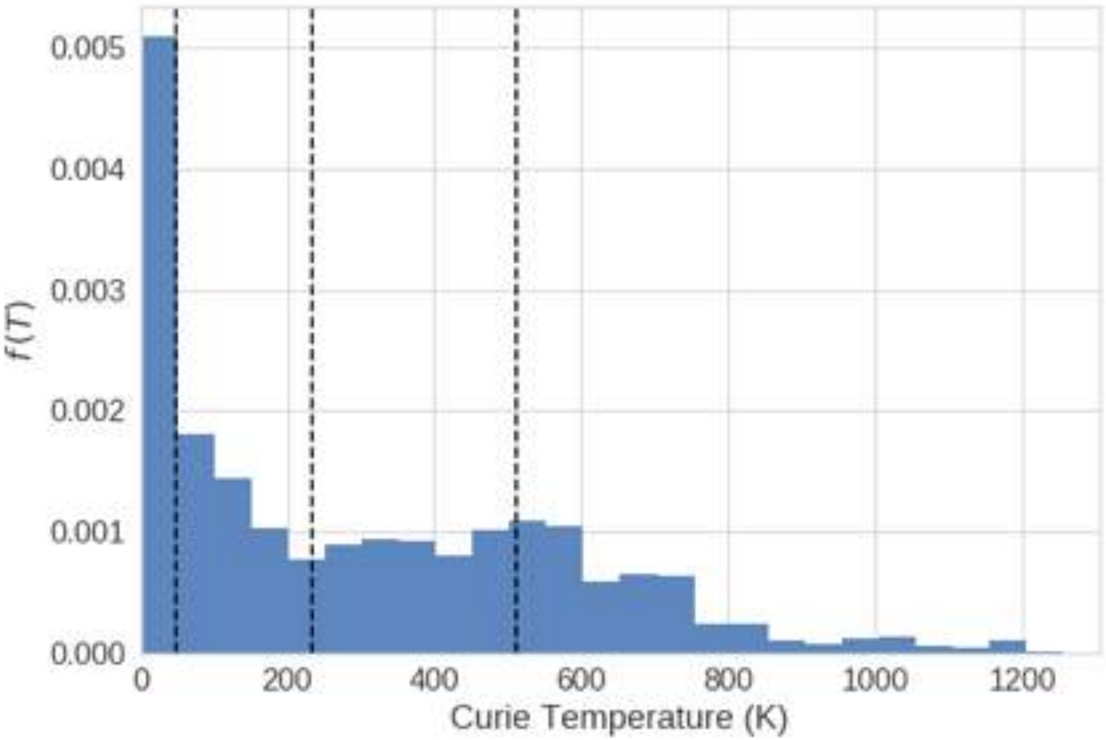
This is all about processing data

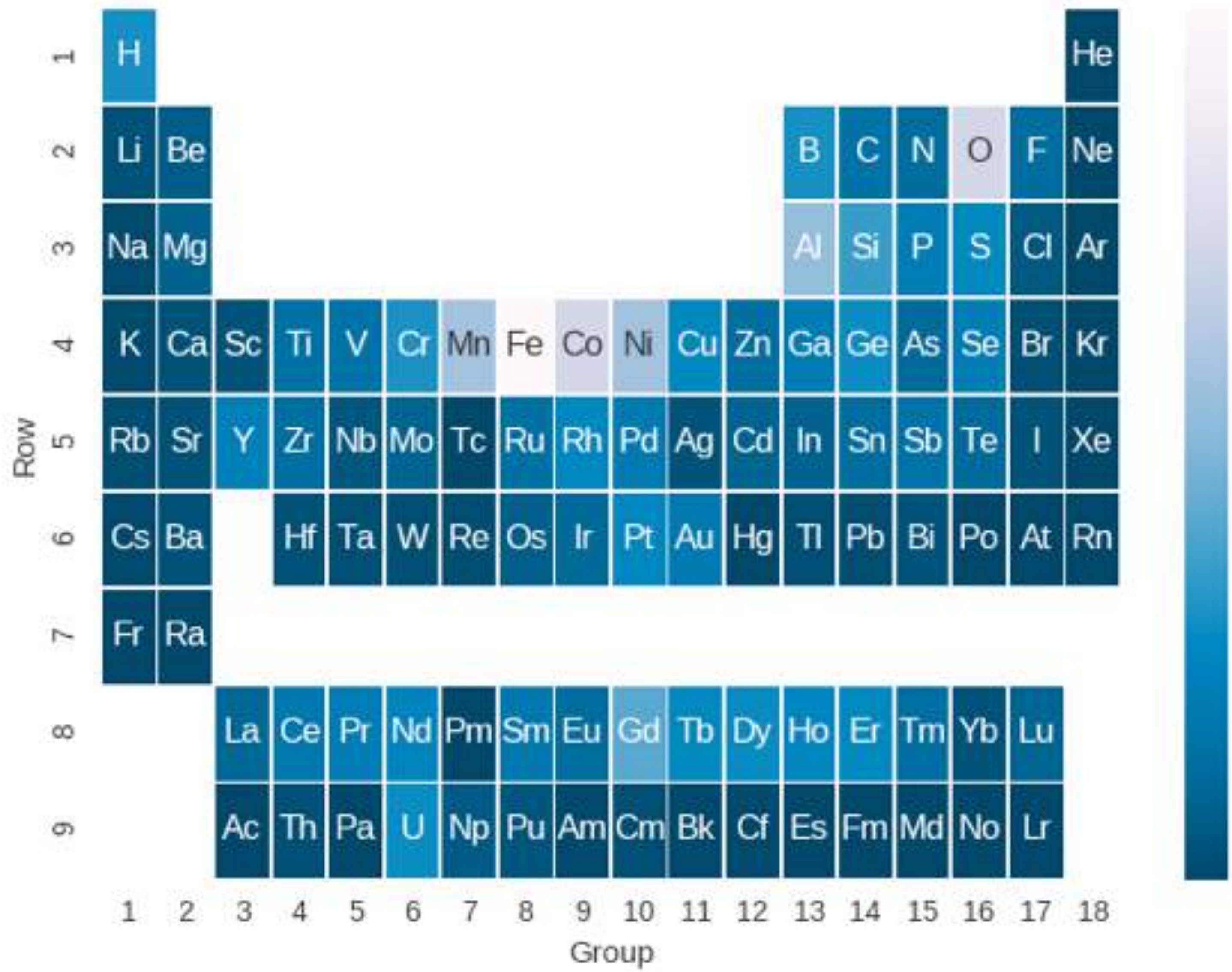
$$(\{Z_i\}, \{N_i\}, \{\zeta_i\}, \{M_i\}, V) \xrightarrow{\text{ML}} y$$

Example: T_C of magnets



James Nelson

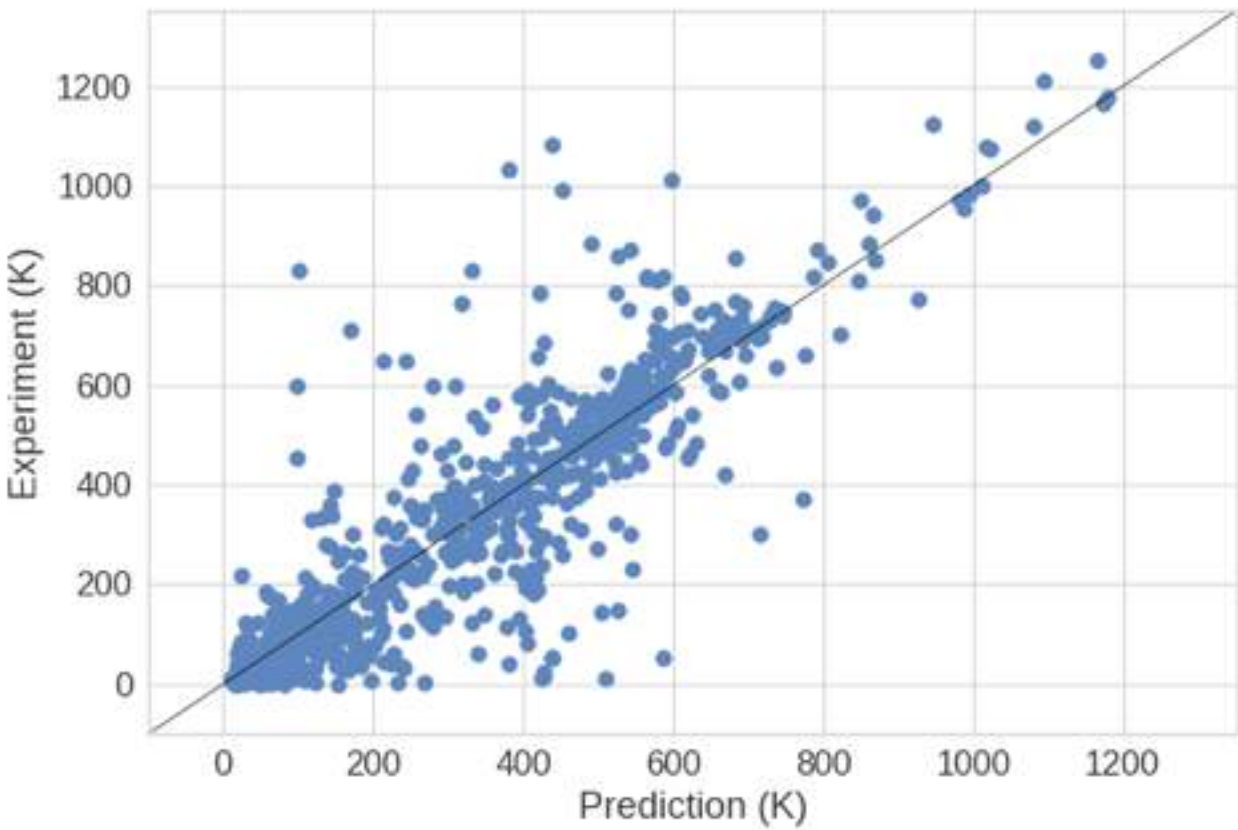
Example: T_C of magnets



Example: T_C of magnets

ML algorithm

$$(\{Z_i\}, \{N_i\}, \{\zeta_i\}, \{M_i\}, V) \Rightarrow T_C$$



Example: T_C of magnets

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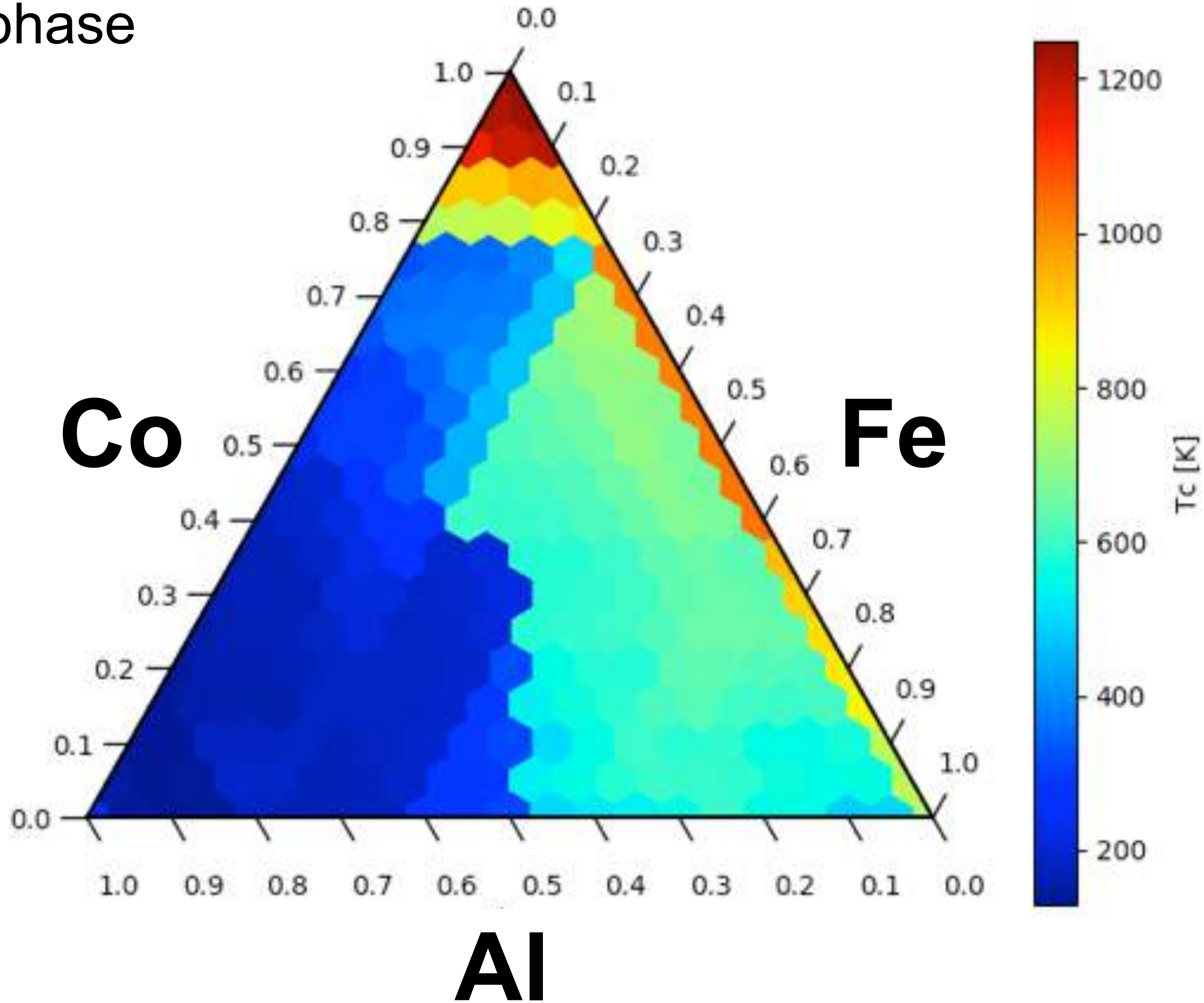
Al-Fe-Co phase diagram



Targeted screening

Looking for new permanent magnets:

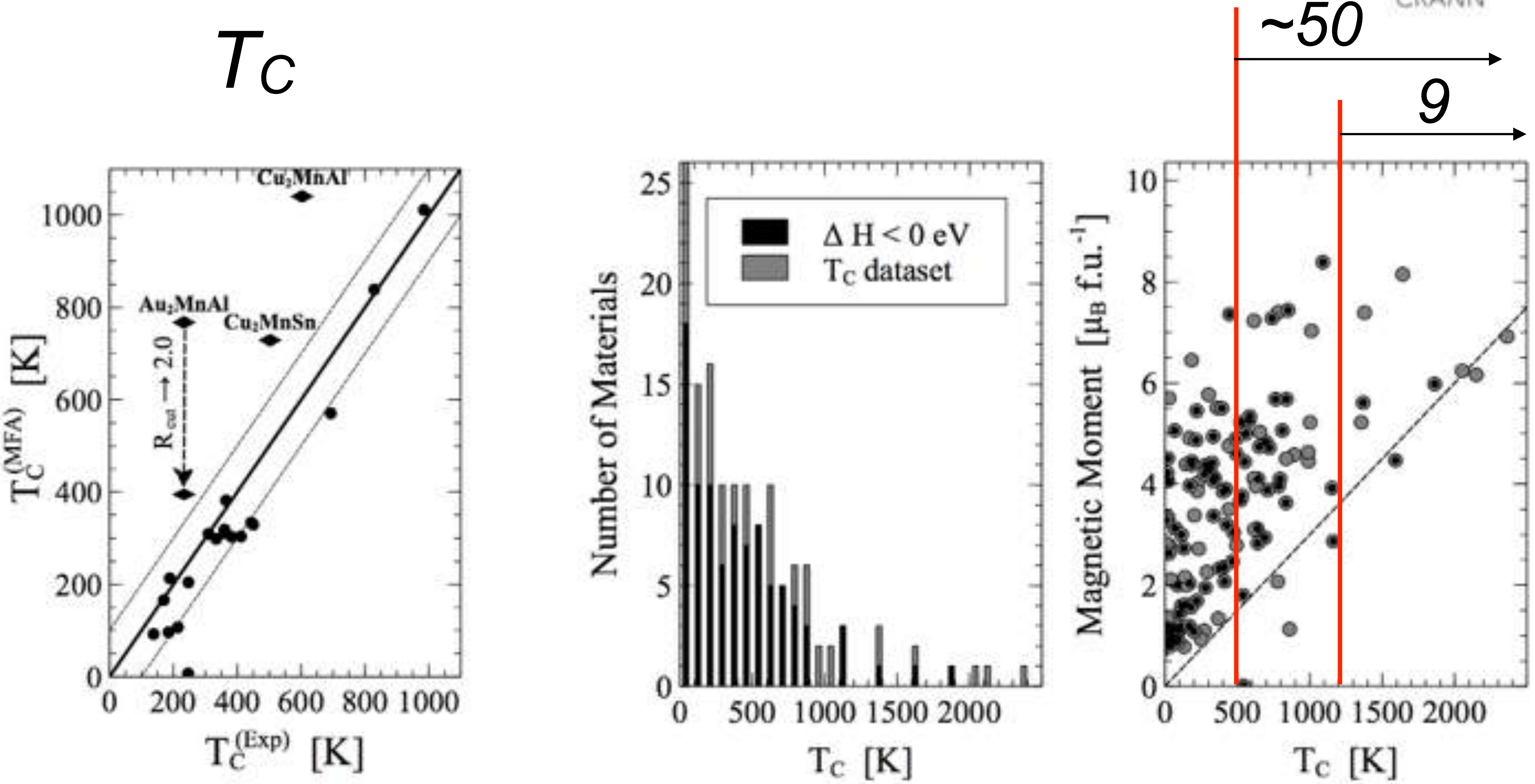
$$M_S > 0.5\,\mu_\mathrm{B}\,f.u.^{-1}$$
$$c/a \neq 1.0$$
$$\Delta H < 500\,\mathrm{meV}$$

229 candidates → T_C, *MCA* ?

$$\mathrm{MCA} > 1\,\mathrm{MJ\,m^{-3}}$$
$$M_S > 6\,\mu_\mathrm{B}\,f.u.^{-1}$$
$$T_\mathrm{C} > 500\,\mathrm{K}$$

Targeted screening

T

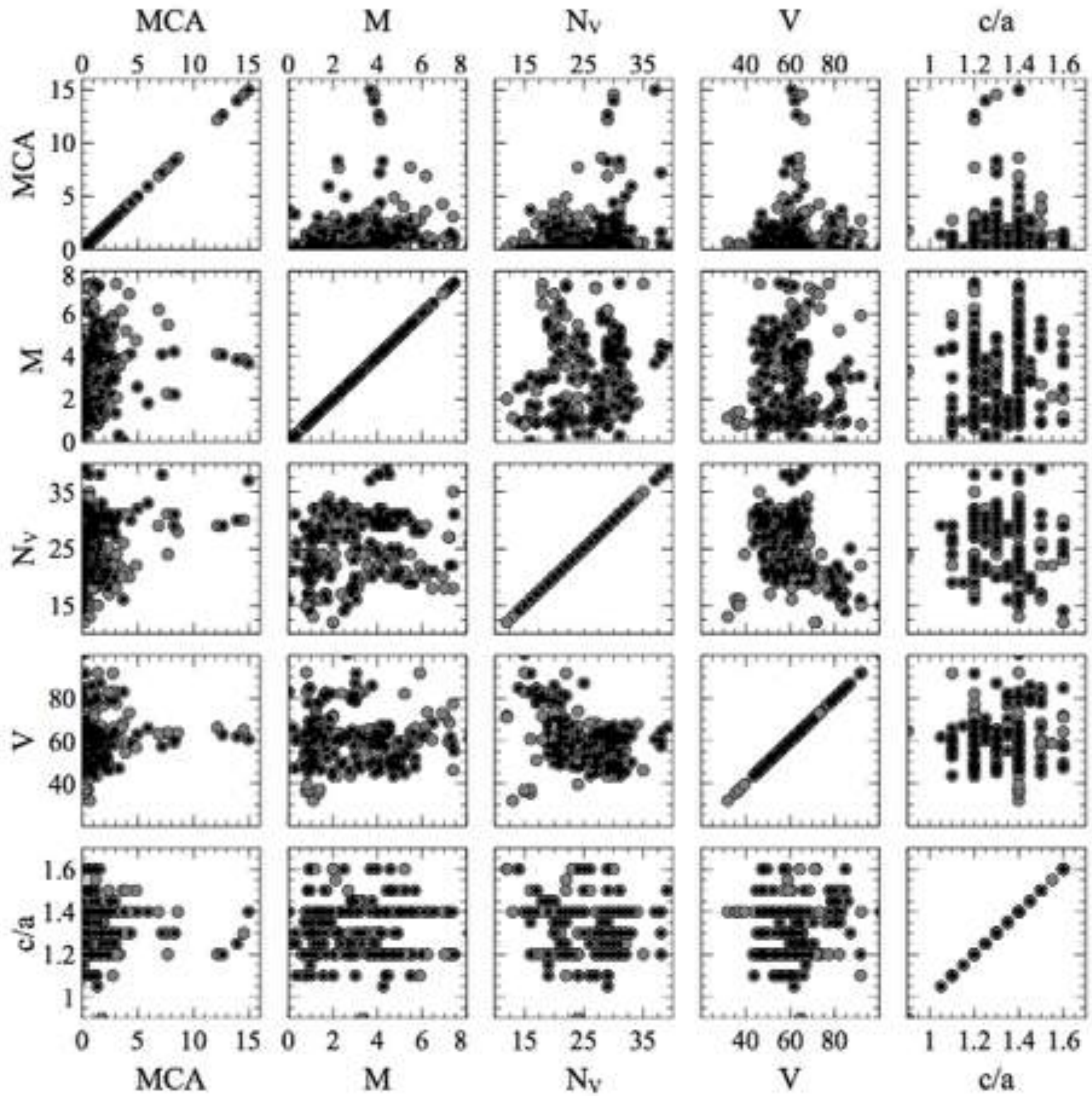


From DFT + MFA

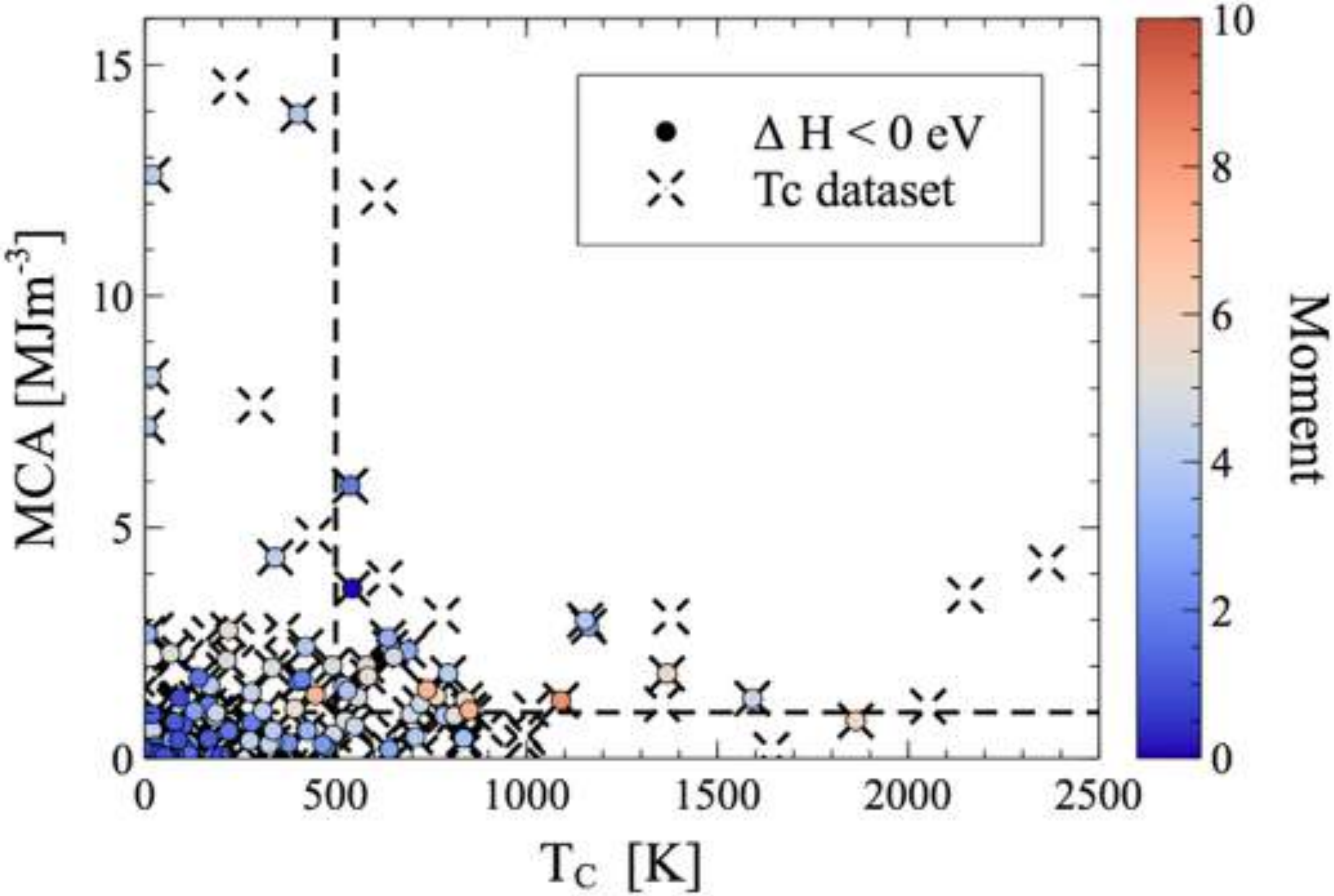
Targeted screening

MCA

From DFT-SO + magnetic force theorem



Targeted screening



Fe_2MnS
Mn_2CuS
Mn_2MgS
Fe_2GeCo
Mn_2CuAl
Cr_2ZnSb
Mn_2ZnAl
Fe_2NiSb

Lots of data: machine learning

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ML algorithm

$$(\{Z_i\}, \{N_i\}, \{\zeta_i\}, \{M_i\}, V) \Rightarrow$$ *Property (MCA)*

Accurate = Selective ⇒ Useful materials will be left out

The lottery example

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Who won the last National lottery ?

NO

Lots of data: machine learning

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Strategy: use machine learning to screen the ***NOT*** hard magnets

ML algorithm

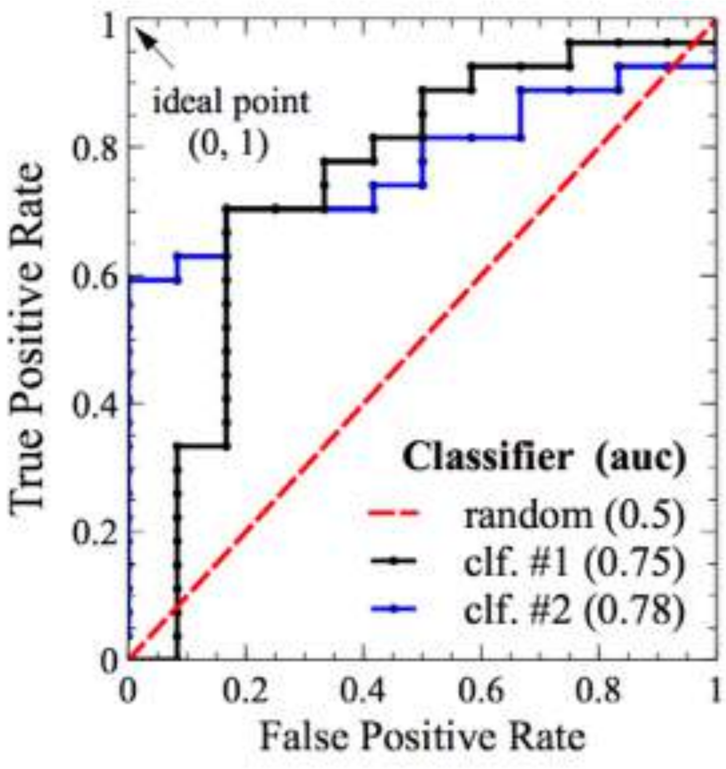
$(\{Z_i\}, \{N_i\}, \{\zeta_i\}, \{M_i\}, V)$ → *Property (MCA)*

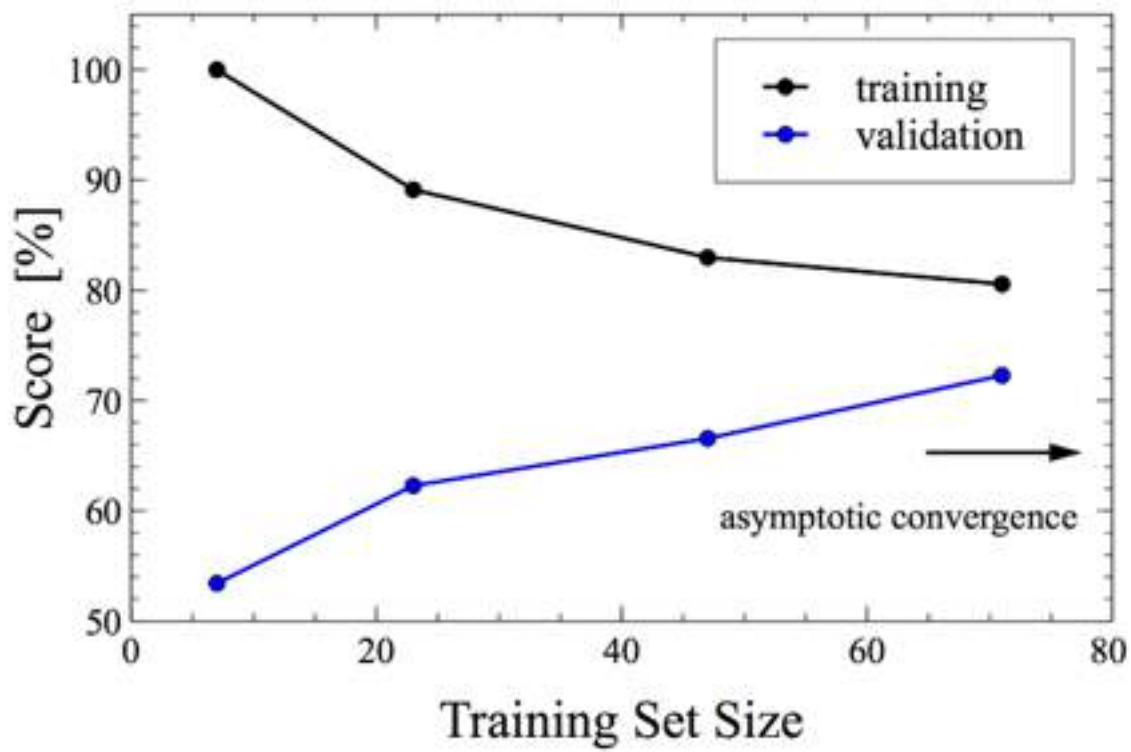
Accurate = Selective → Useful materials will be left out

Selecting hard magnets		Selecting soft magnets	
Hard	Soft + Hard	Soft	Hard + Soft

Lots of data: machine learning

Receiver operating curve (ROC)





60% TPR with 0 FPR

Need ~80 data for learning

Machine learning *workflow*

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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

Limitations

Two issues in materials science

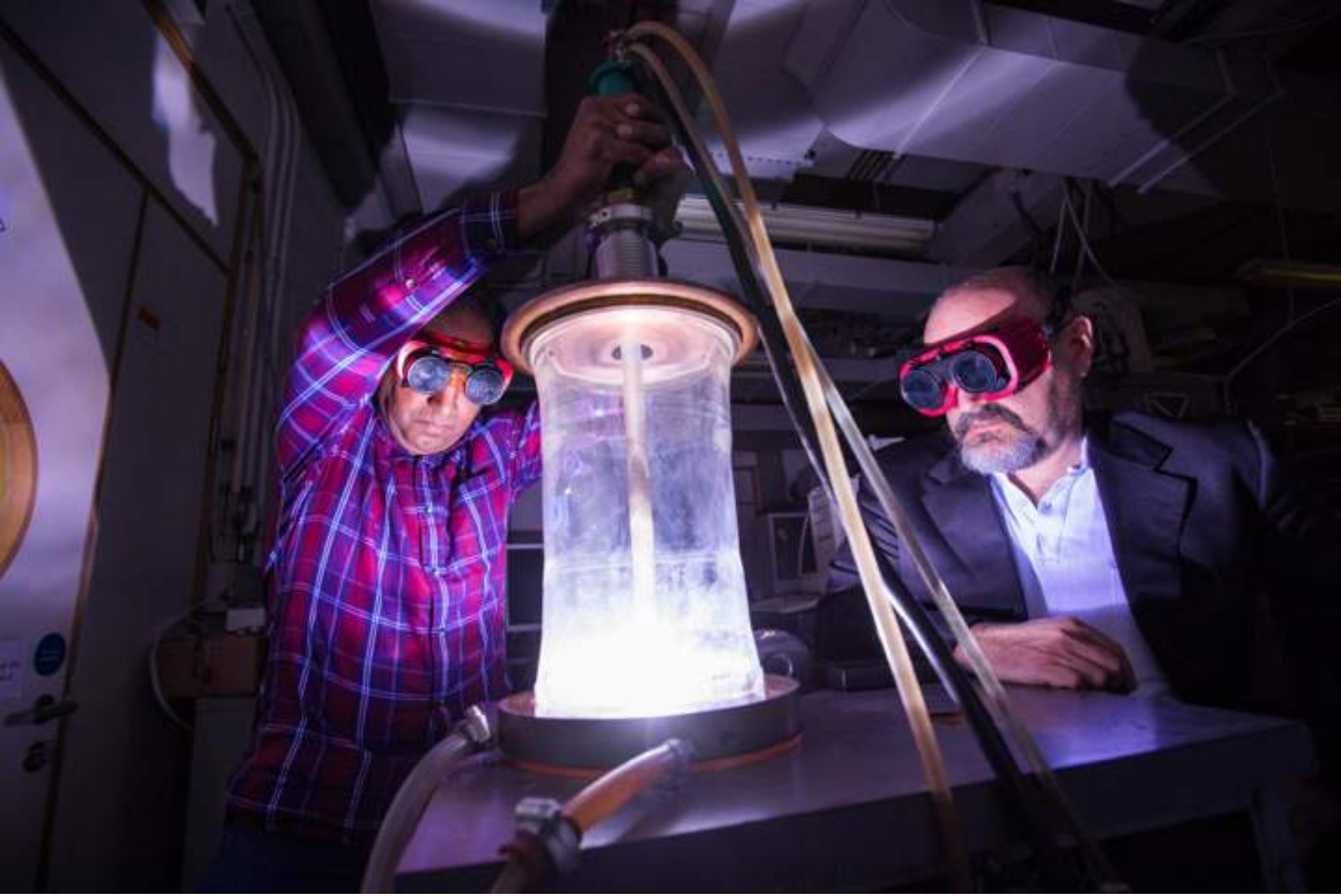
$$(\{Z_i\}, \{N_i\}, \{\zeta_i\}, \{M_i\}, V)$$

N

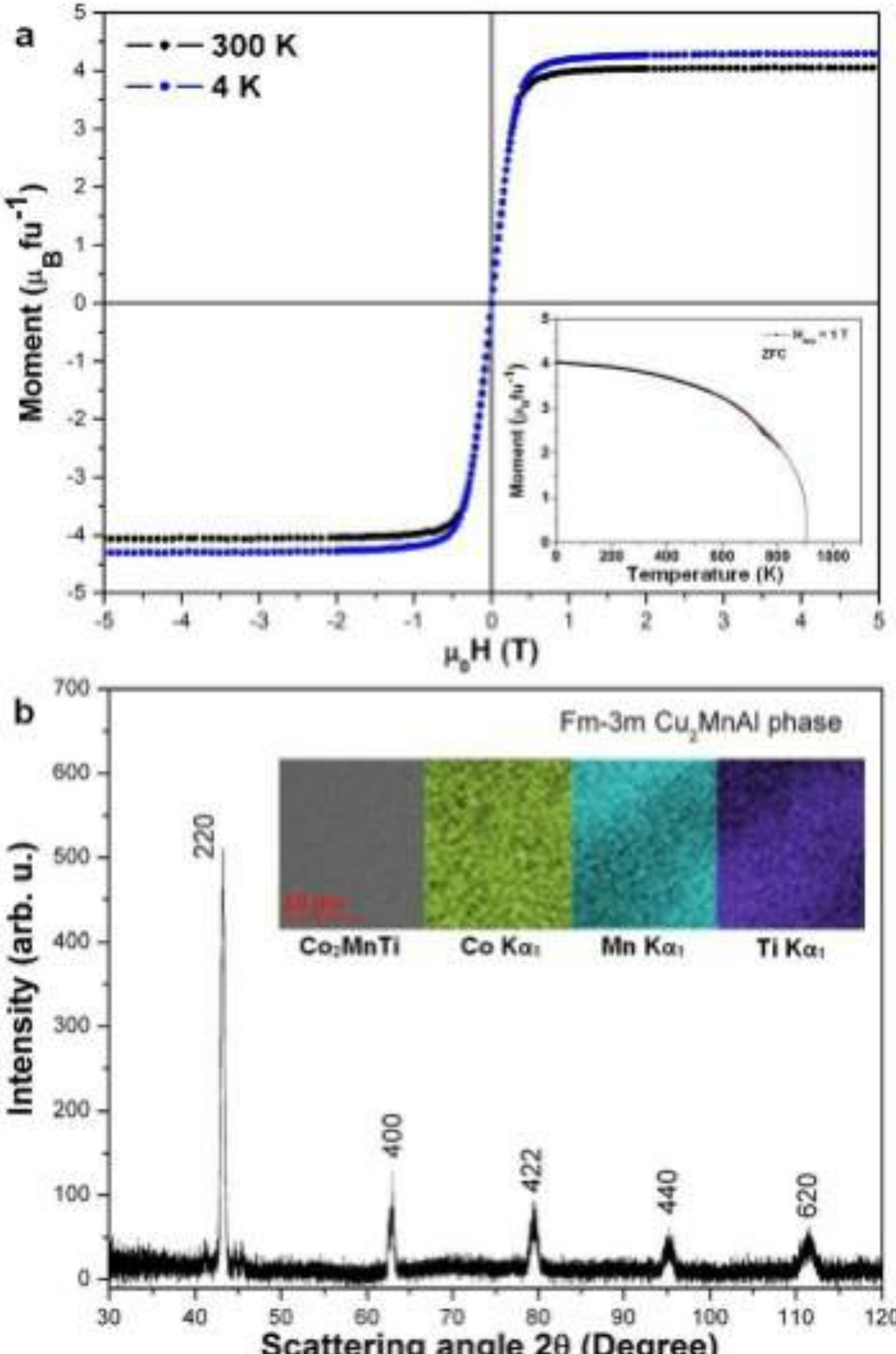
curse of dimensionality

$$\rho \longrightarrow m^N$$

Our “big data” are not big …. yet



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



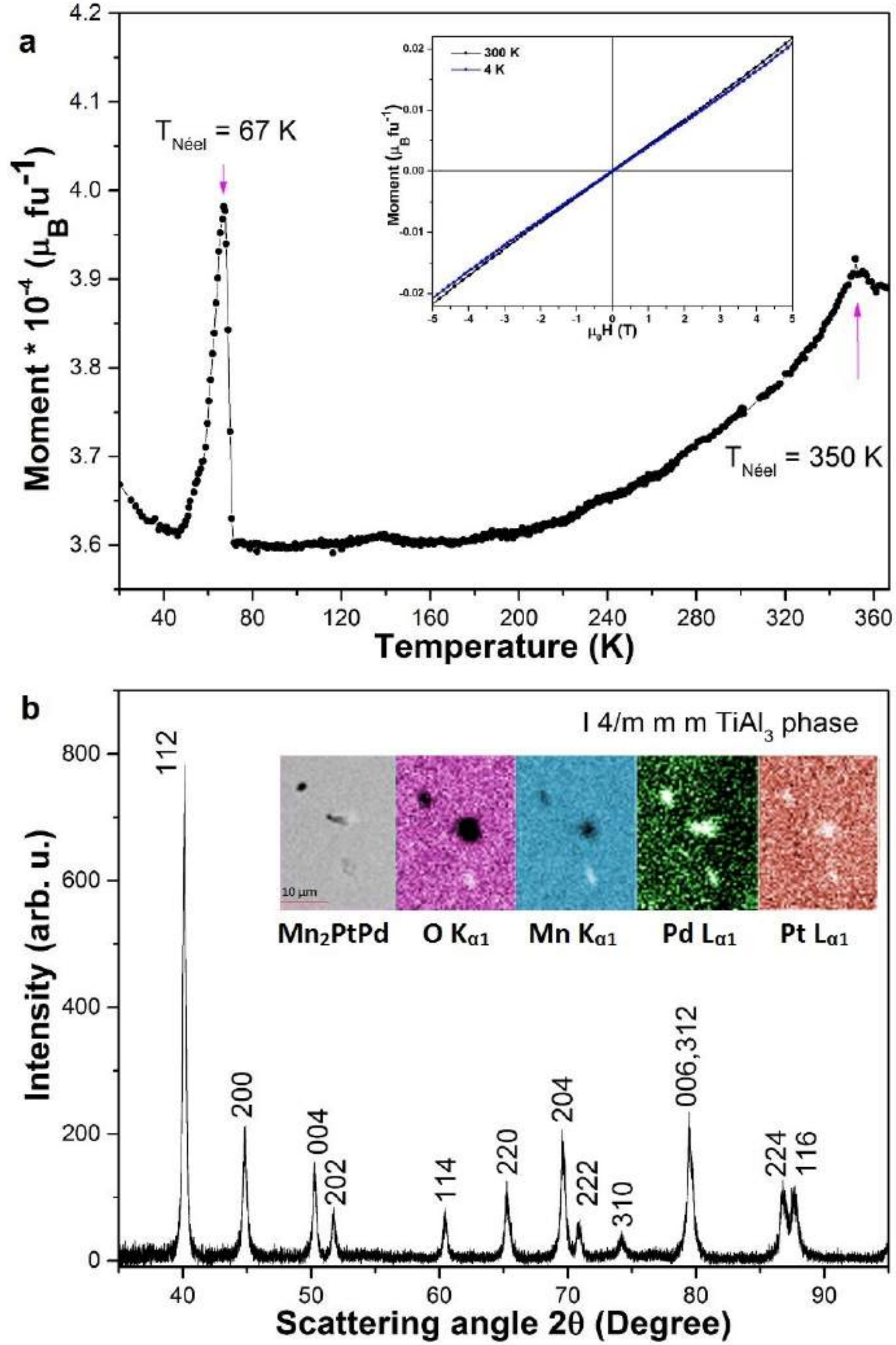
Co_2MnTi

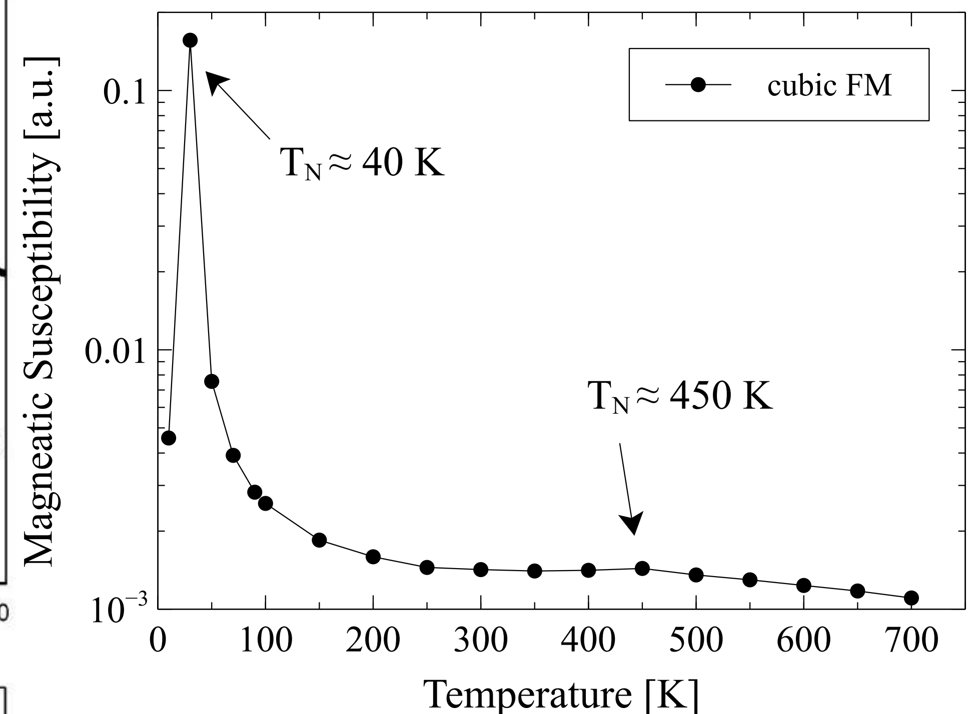
$T_C^{measured}$ = 940K

$T_C^{predicted}$ = 938K

Prepared by arc melting in an Ar atmosphere

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





Complex antiferromagnetic order

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

Bottom line ….

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Did we find one ?

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