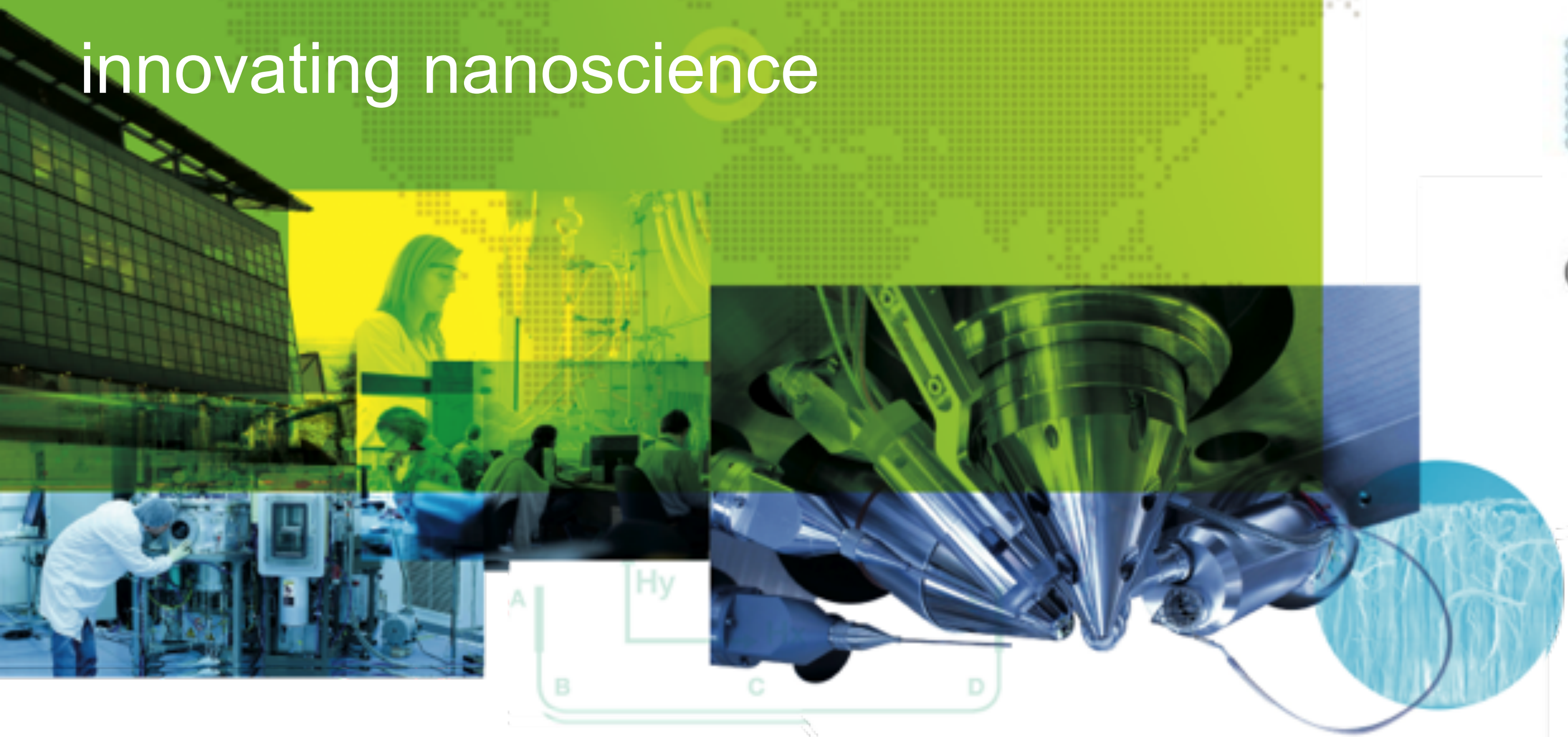
MaX Conference, Trieste January 2018



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

Stefano Sanvito (sanvitos@tcd.ie)

School of Physics and CRANN, Trinity College Dublin, IRELAND

Trinity College
The University of Dublin

sfi
science foundation ireland

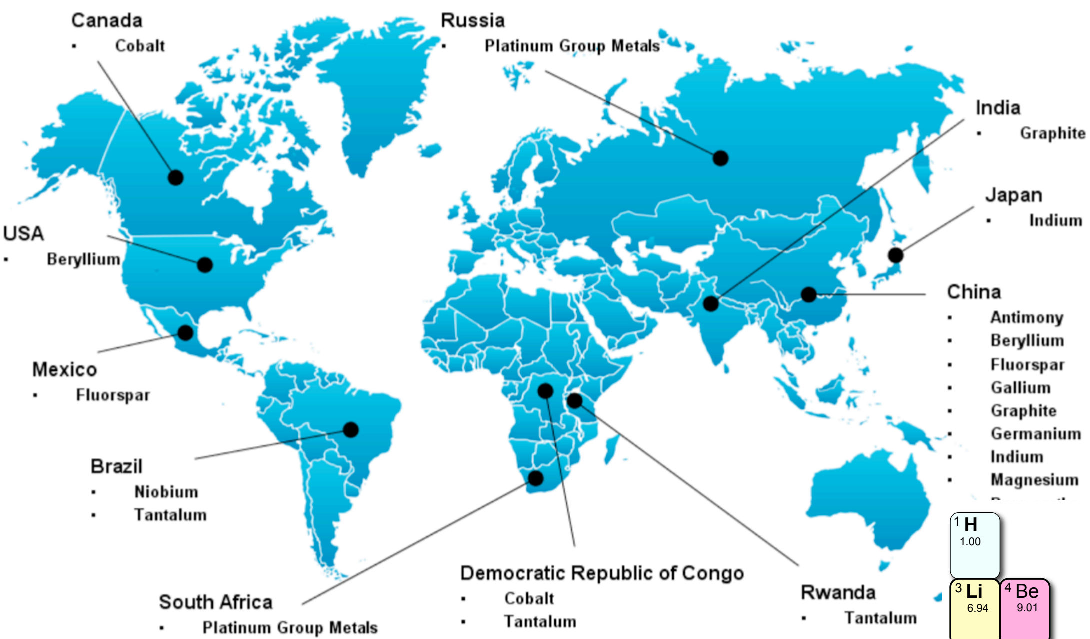
My objectives for this talk

CRANN

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

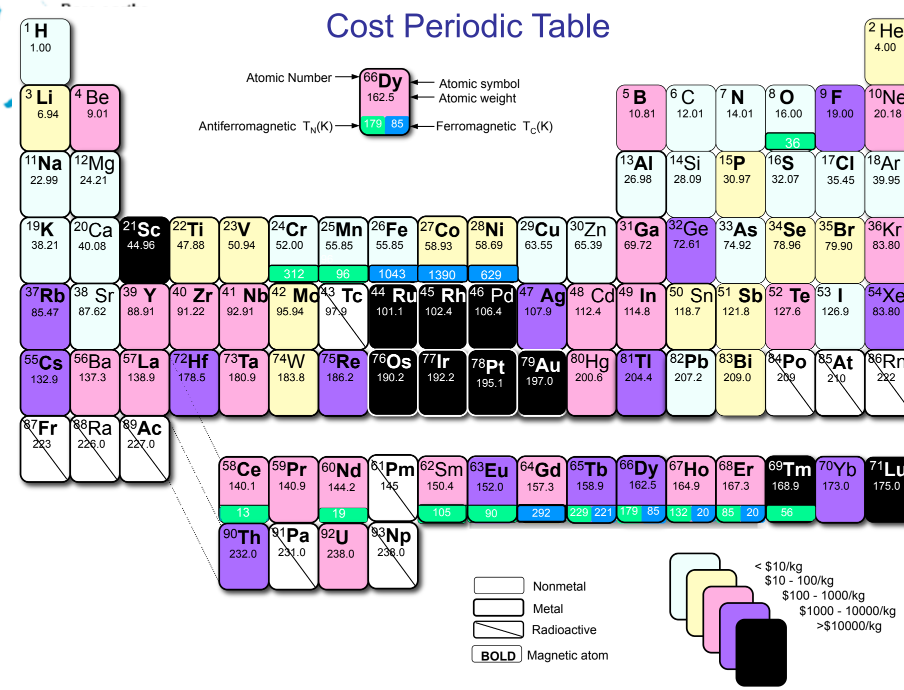
Show that, as databases grow, we will become more clever in creating and using them

Finding new magnets: why ?

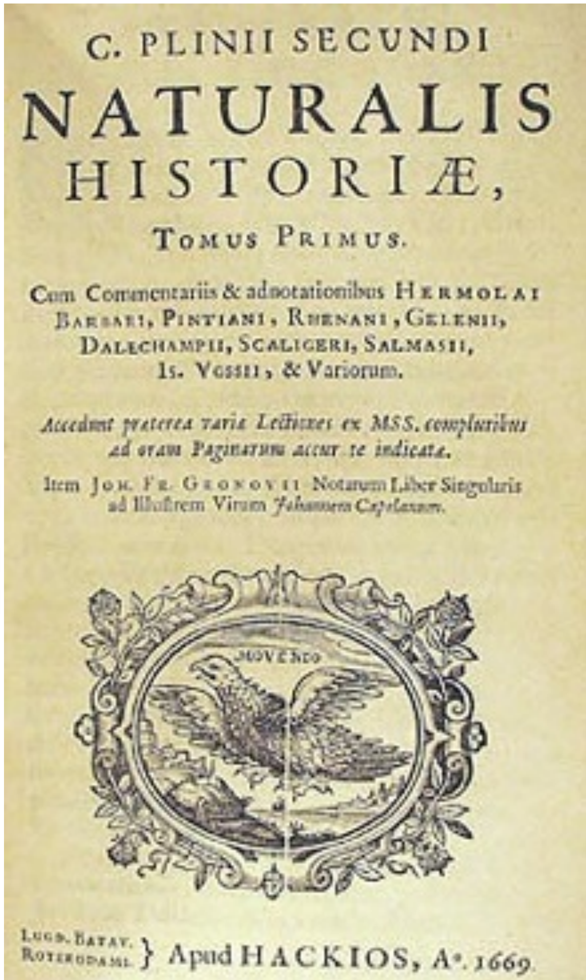


US permanent magnets market ~22.6B$ (2021)

New tech. to deploy

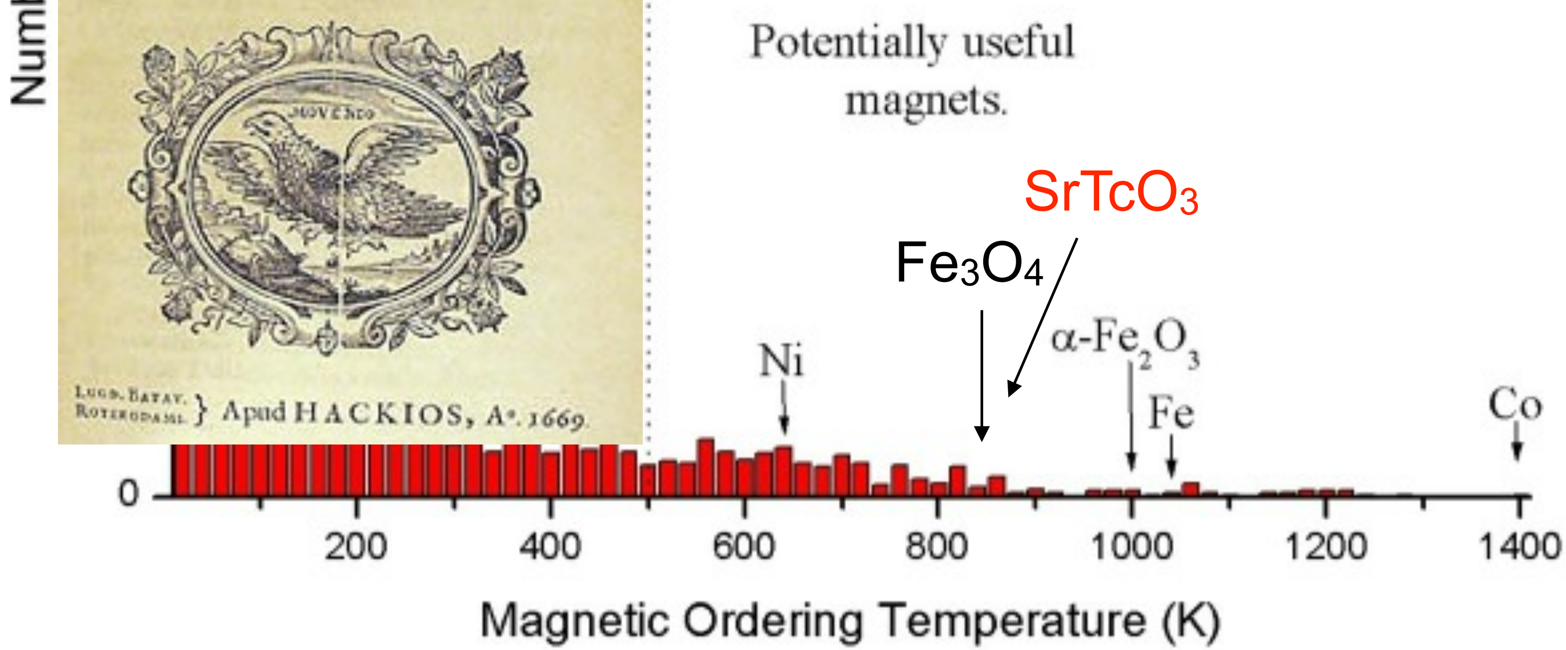


Magnetism is rare

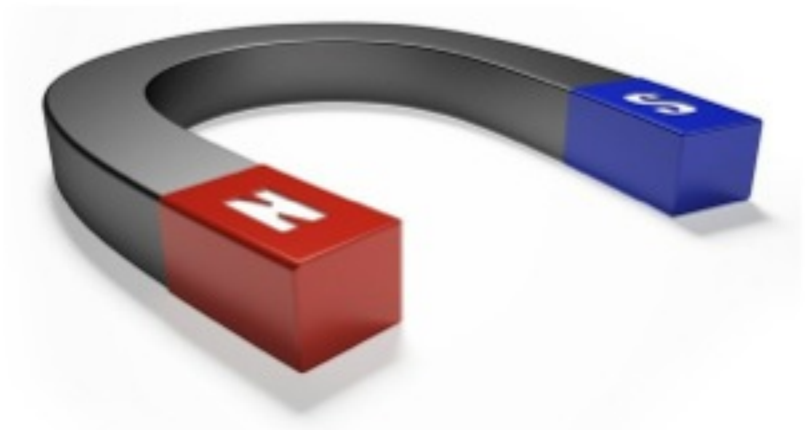

C. PLINII SECUNDI
NATURALIS HISTORIÆ,
TOMUS PRIMUS.
Cum Commentariis & adnotationibus HERMOLAI BARBARI, PINTIANI, RHENANI, GELENII, DALECHAMPII, SCALIGERI, SALMASII, IS. VOSSII, & Variorum.
Accedunt præterea variæ Lectiones ex MSS. compluribus ad oram Paginarum accur te indicatæ.
Item JOH. FR. GRONOVII Notarum Liber Singularis ad Illustrem Virum *Johannem Capellanum.*

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

The discover a new useful magnet is a rare event



The magnetic genome project



with Stefano Curtarolo, Duke

The magnetic genome project

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

CRANN

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...tronic structure method:
...ctional theory (VASP)

The magnetic genome project

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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 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

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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.)

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)

The AFLOW consortium

CRANN

www.aflowlib.org

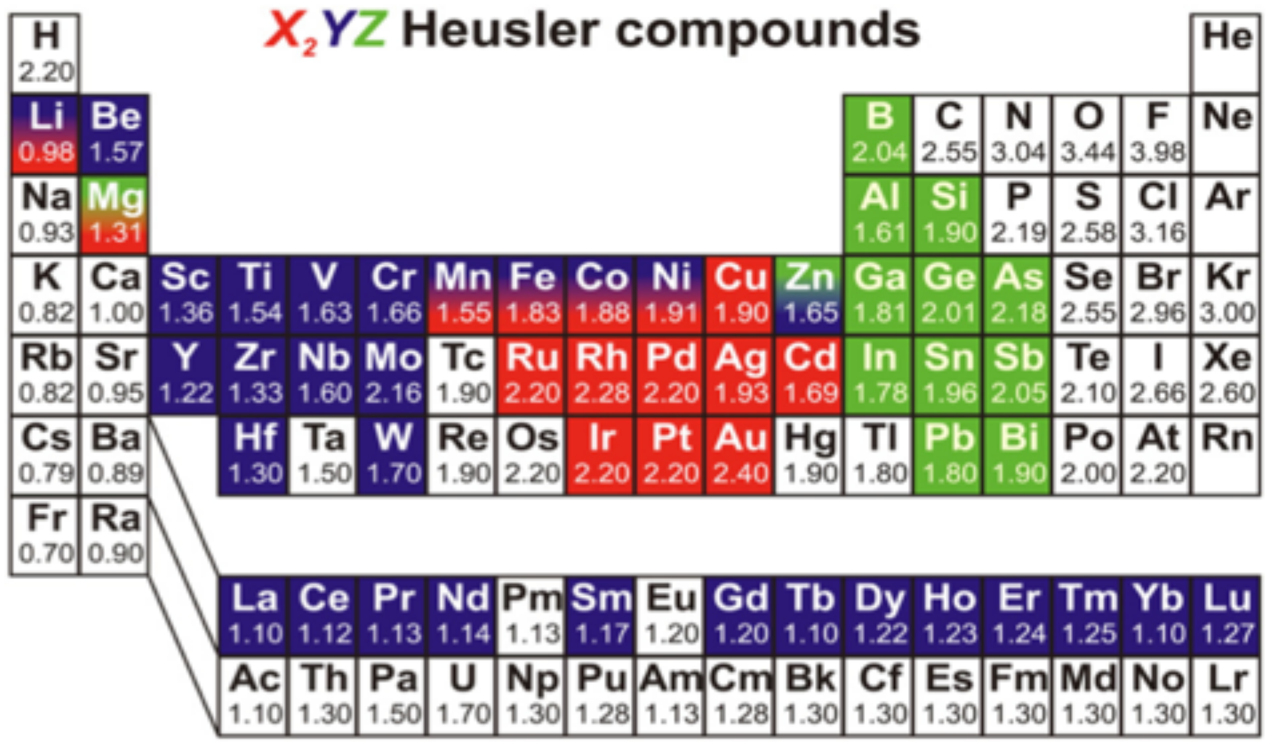
S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R.H. Taylor, L.J. Nelson, G.L.W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, Comp. Mat. Sci. **58**, 227 (2012)

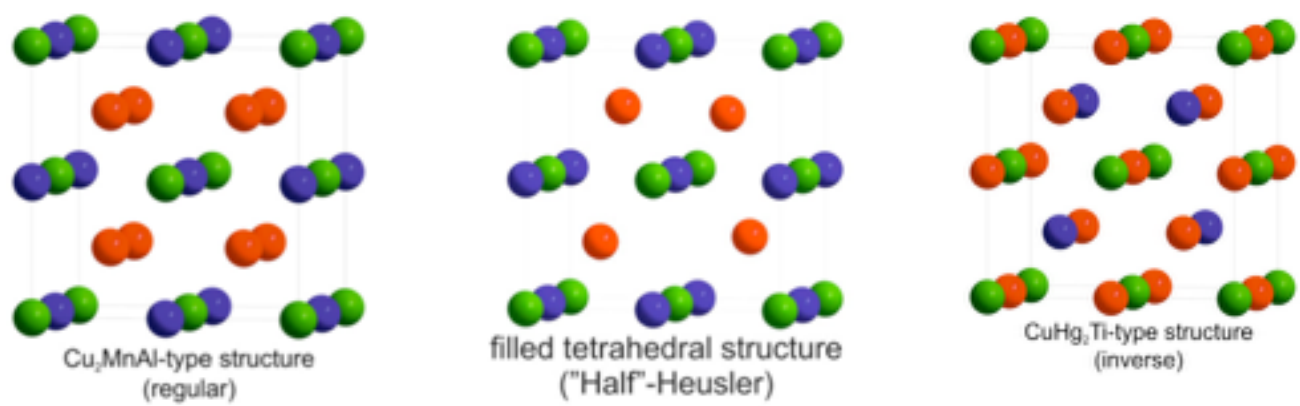
Heusler alloys

~250 known …

~1000 claimed …

~90 magnetic …





Heusler alloys

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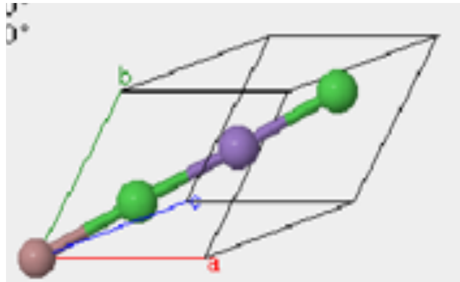
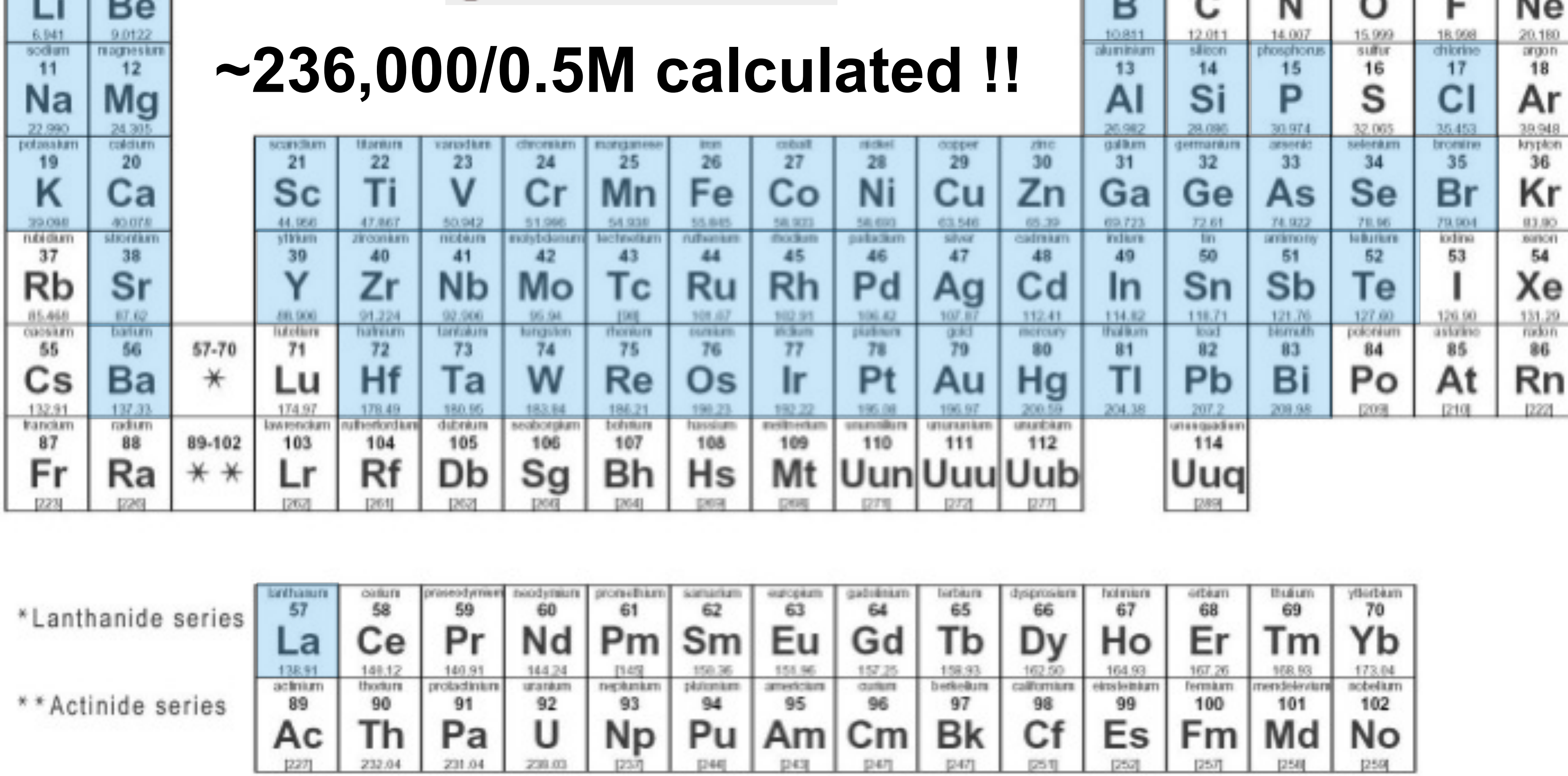


~236,000/0.5M calculated !!



Database

Rational materials storage

www.aflowlib.org

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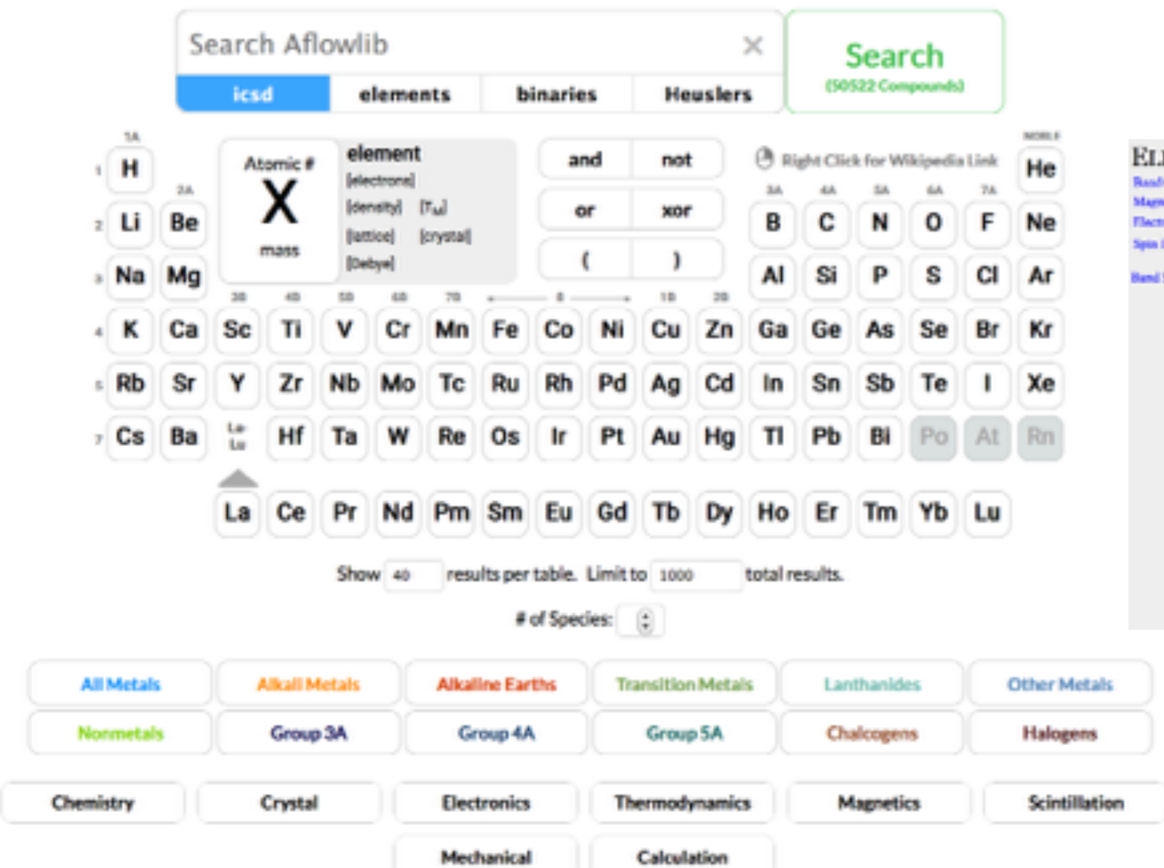

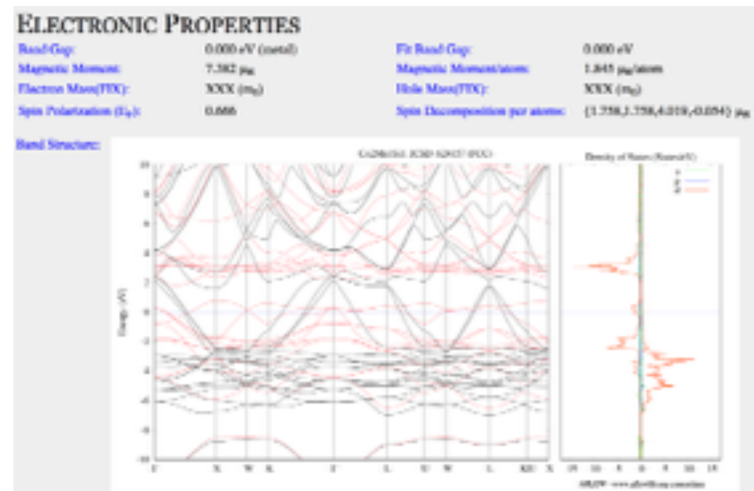

S. Curtarolo, W. Setyawan, S. Wang, J. Xue, K. Yang, R.H. Taylor, L.J. Nelson, G.L.W. Hart, S. Sanvito, M. Buongiorno-Nardelli, N. Mingo, O. Levy, Comp. Mat. Sci. **58**, 227 (2012)

The magnetic genome project

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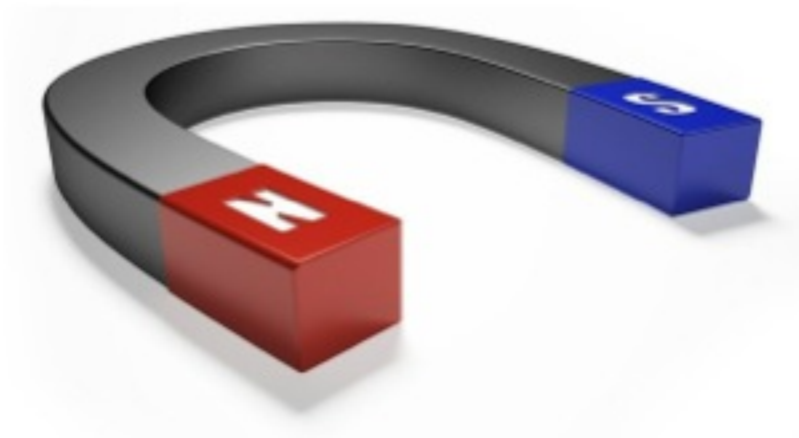
Back to the magnets …..



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

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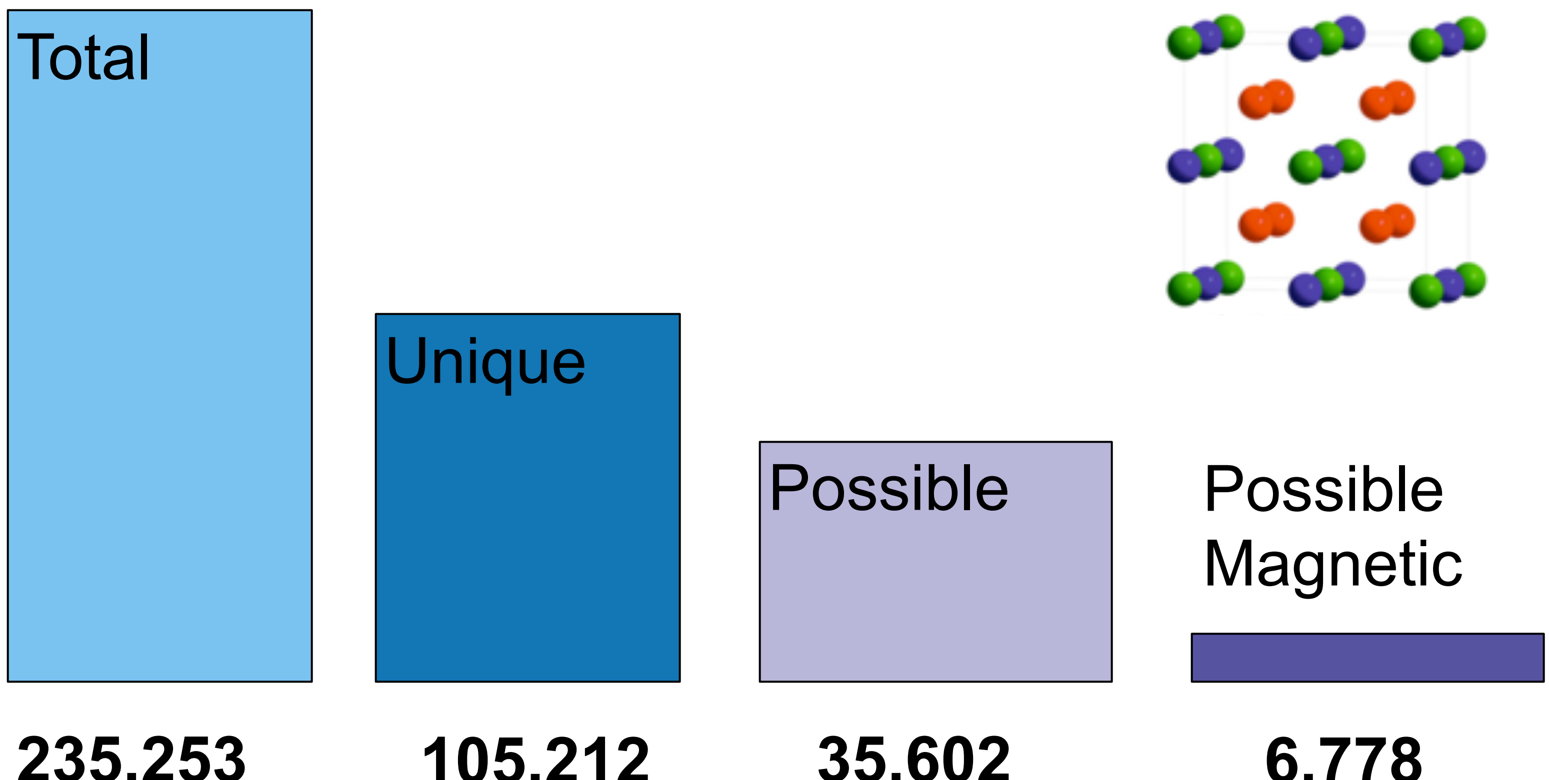
Property: Can be made ?

Descriptor 0:
Enthalpy of formation

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



Stability analysis

Descriptor 1: Enthalpy of formation

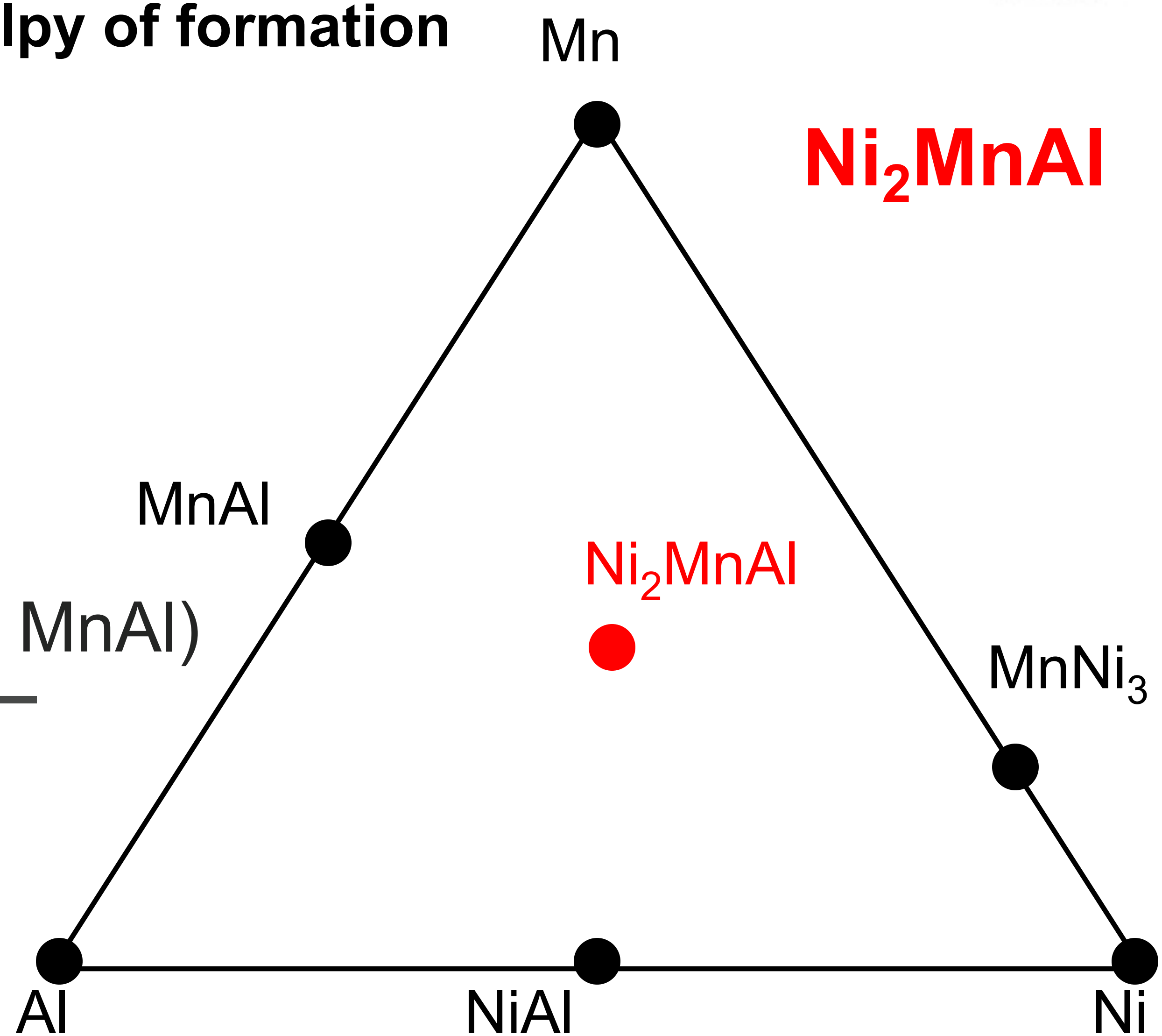
2 Ni + Mn + Al

2 Ni + MnAl

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

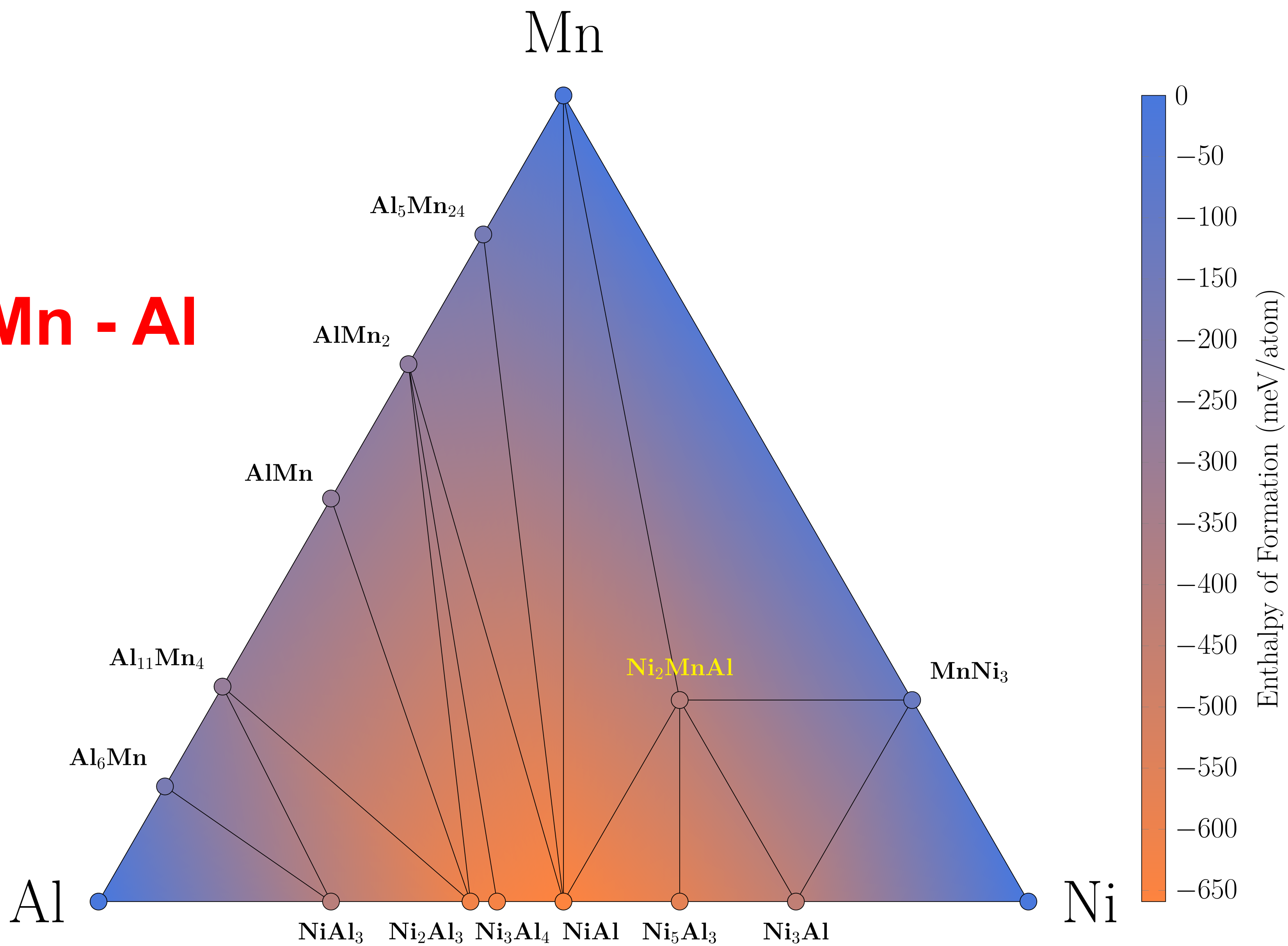
Ni_2MnAl

Ni_2MnAl



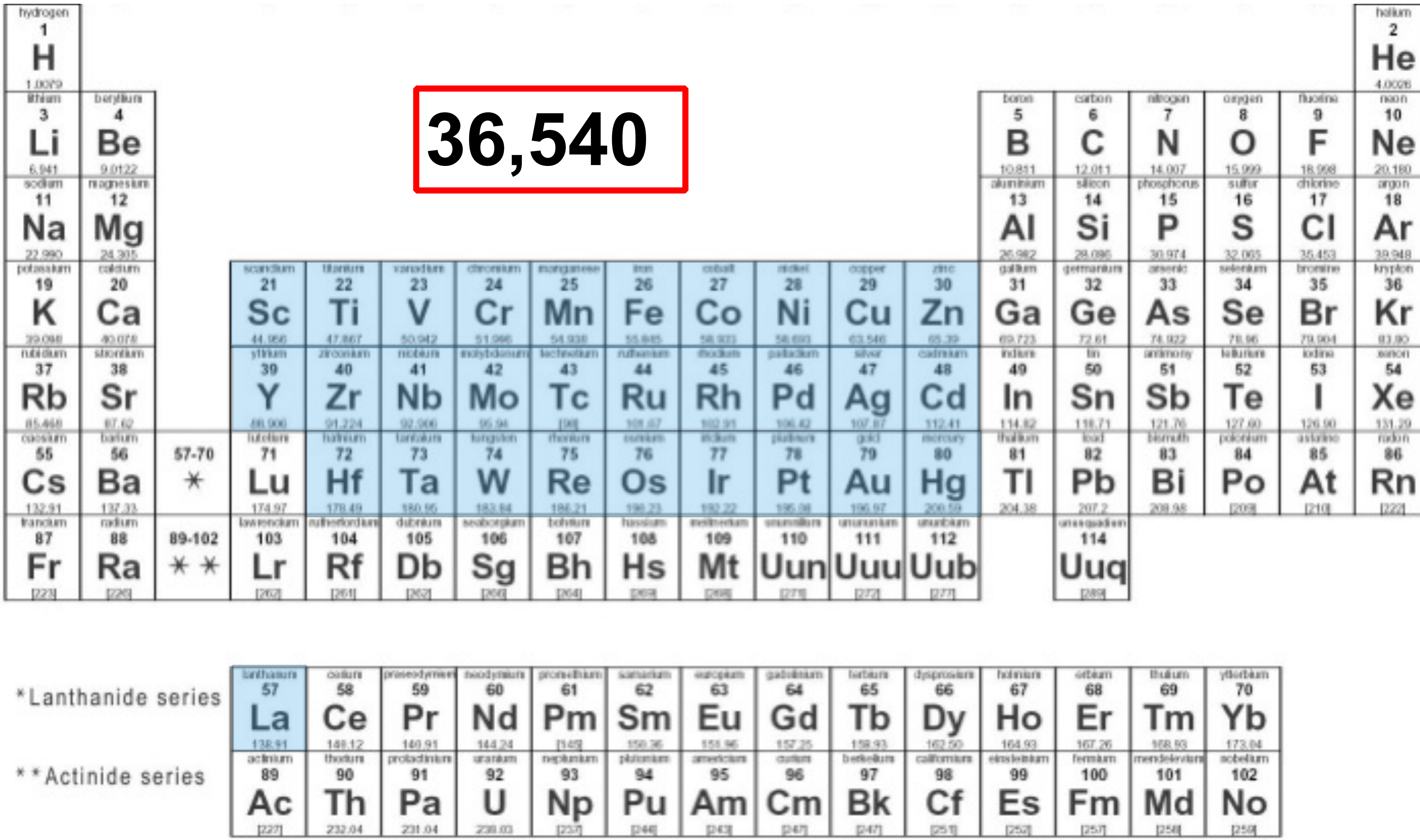
Stability analysis

Ni - Mn - Al



TM[3]

Look at the transition metal intermetallics



In summary …

36,540 possible → 248 stable

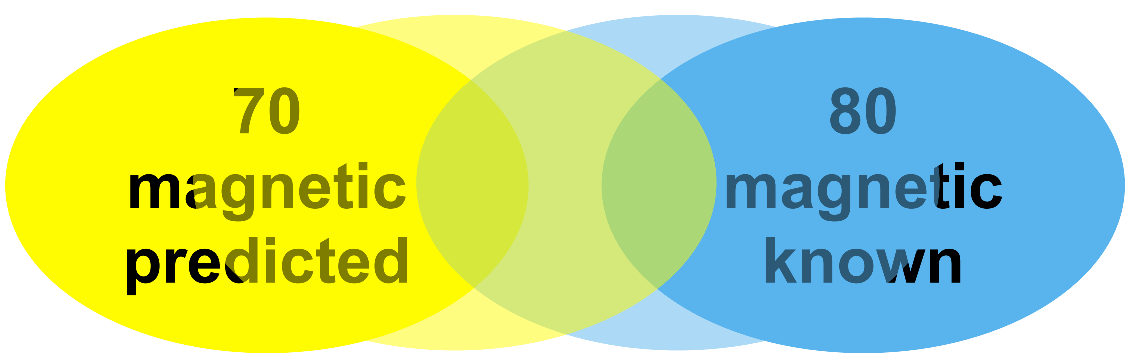
22 magnetic → 8 Robust (Δ^{30} criterion)

Extrapolating

236,000 possible → 1550 stable

138 magnetic → 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

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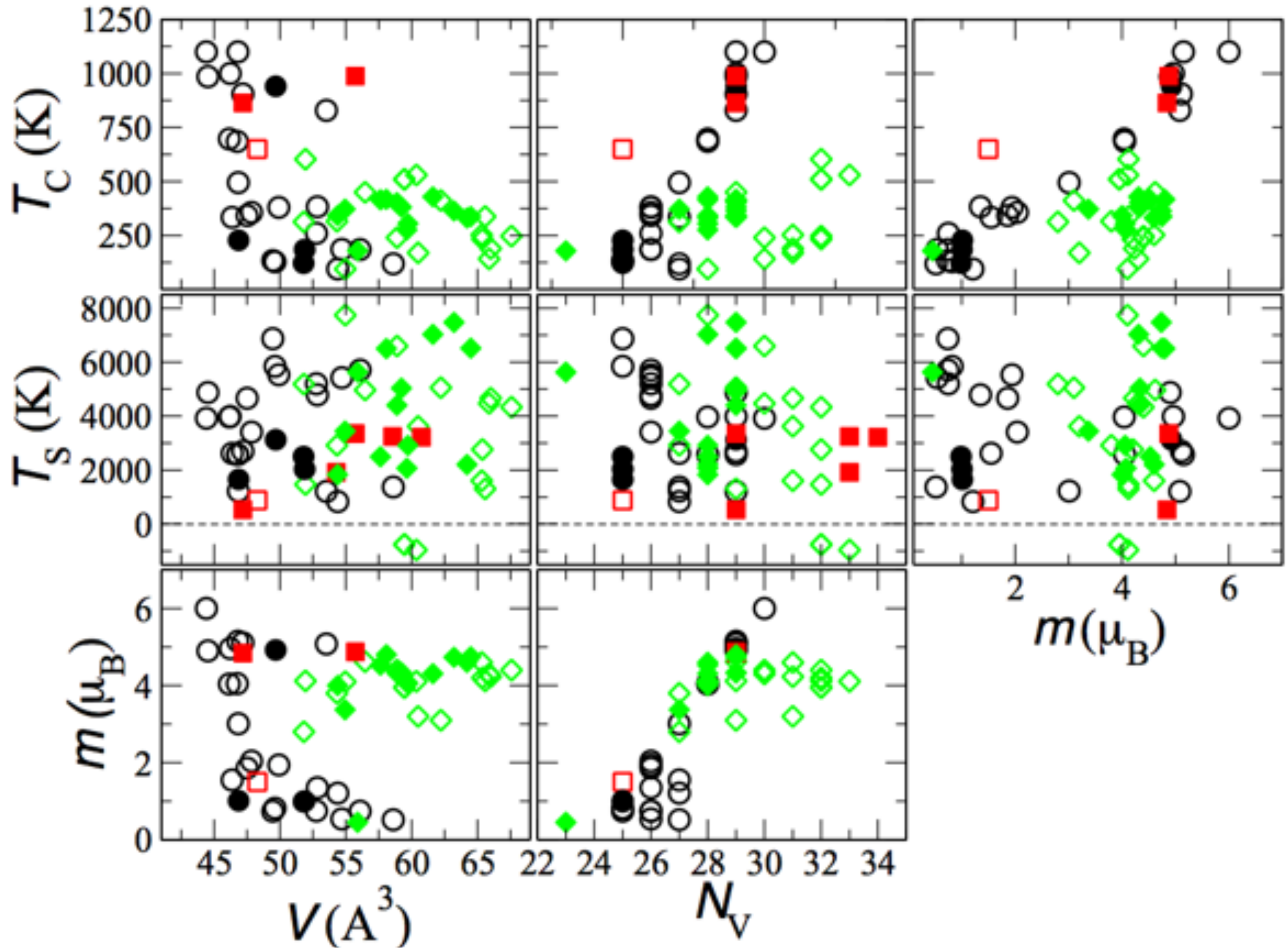
Co_2XY

Mn_2XY

X_2MnY

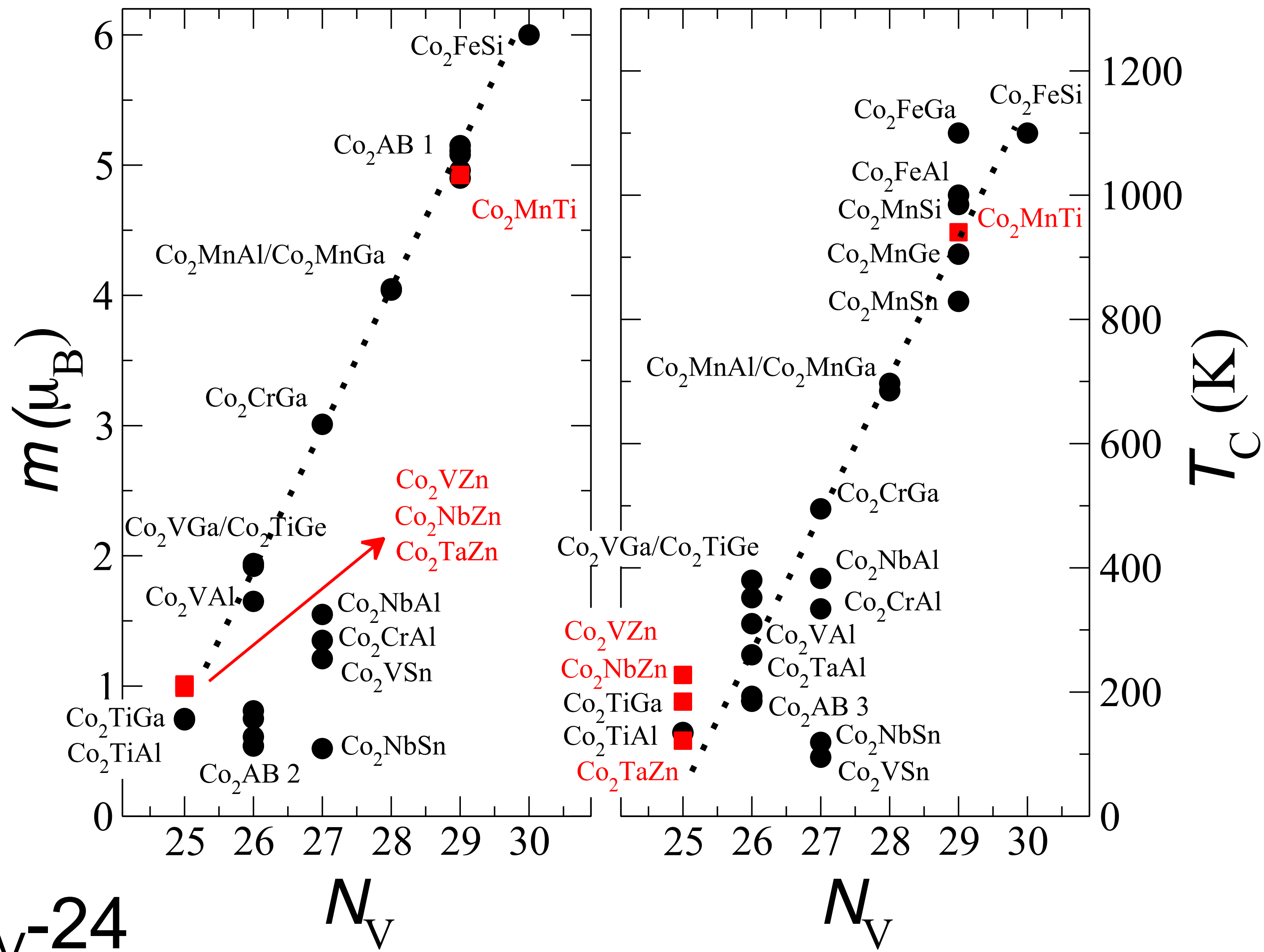


Co_2YZ

Co_2YZ

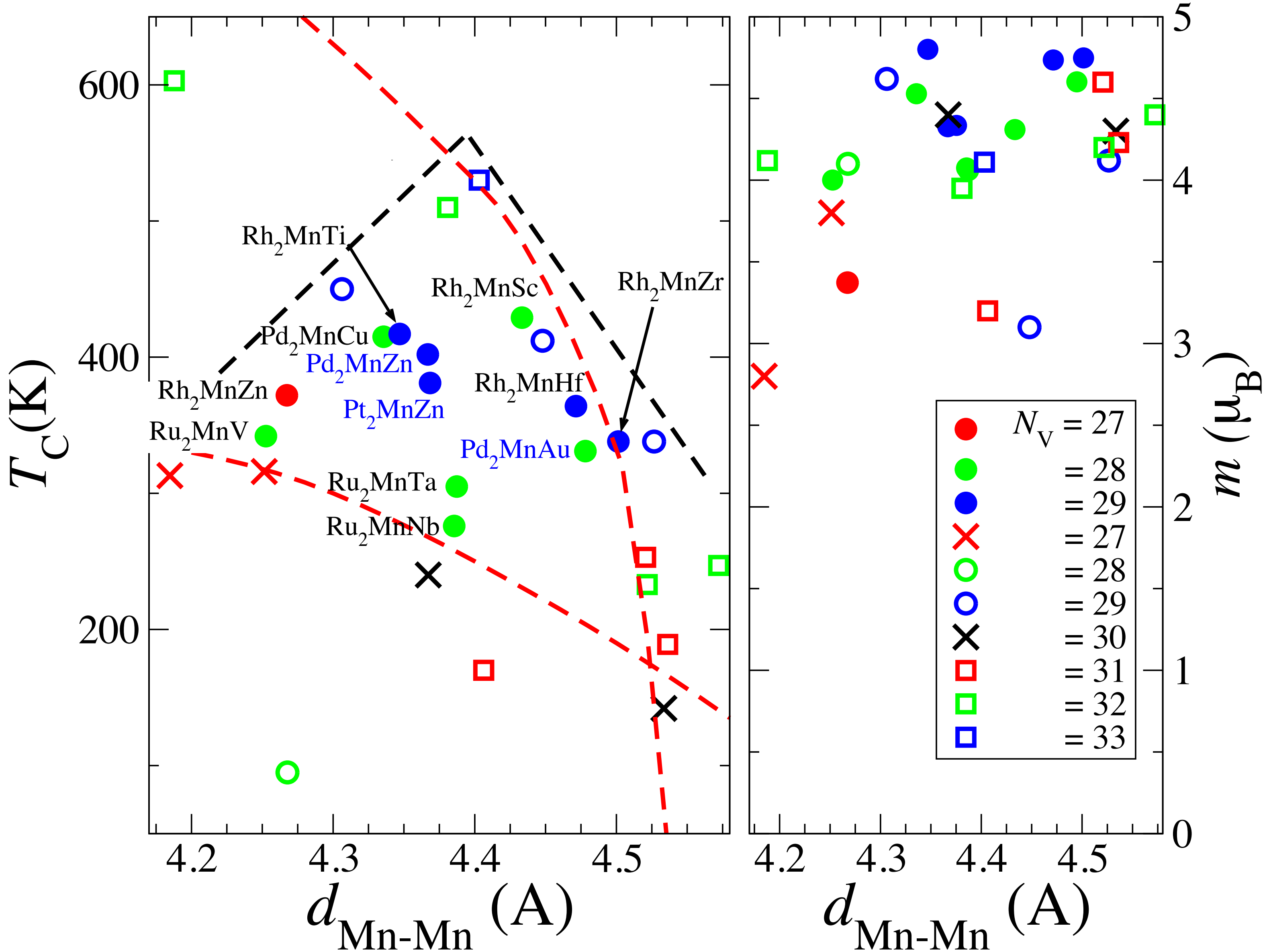
Slater-Pauling

$m_{X_2YZ} = N_V - 24$



X_2MnZ

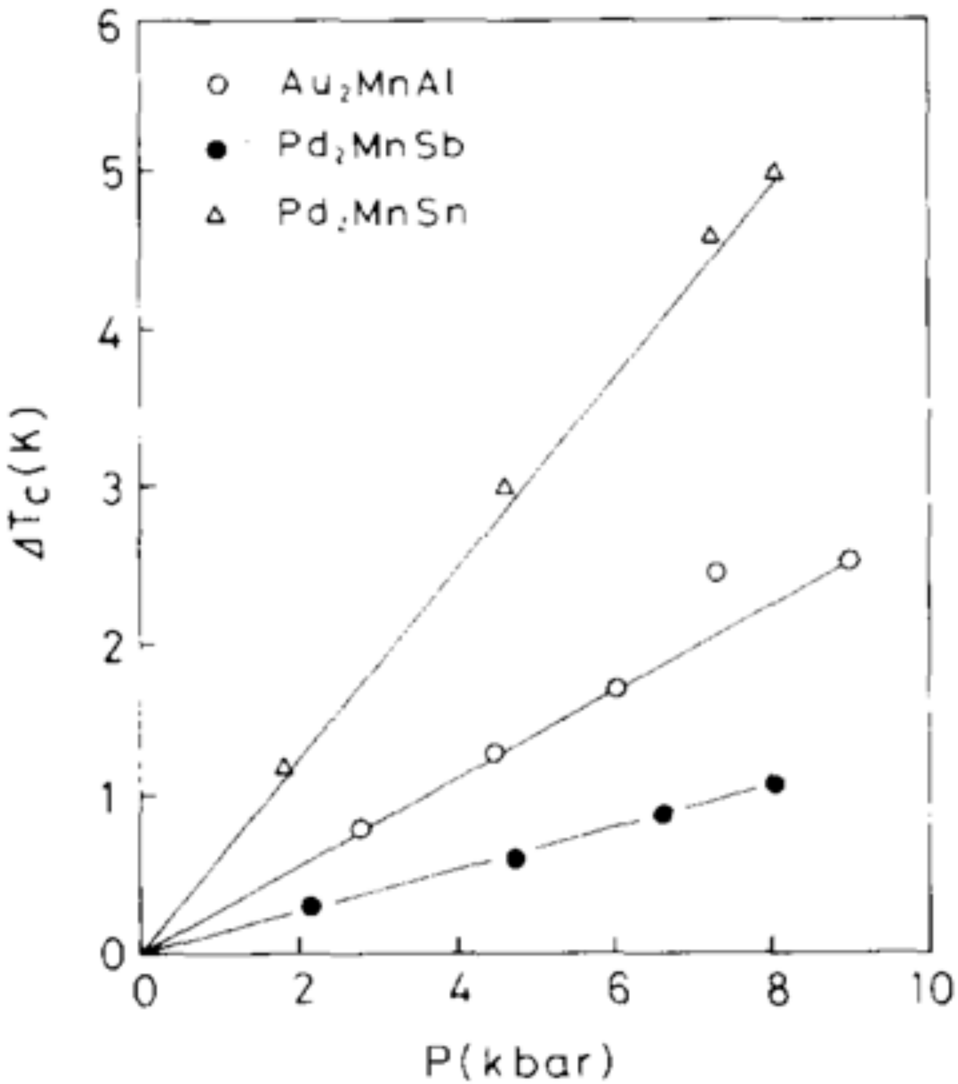
X_2MnZ



Castelliz-Kanomata curve

X_2MnZ

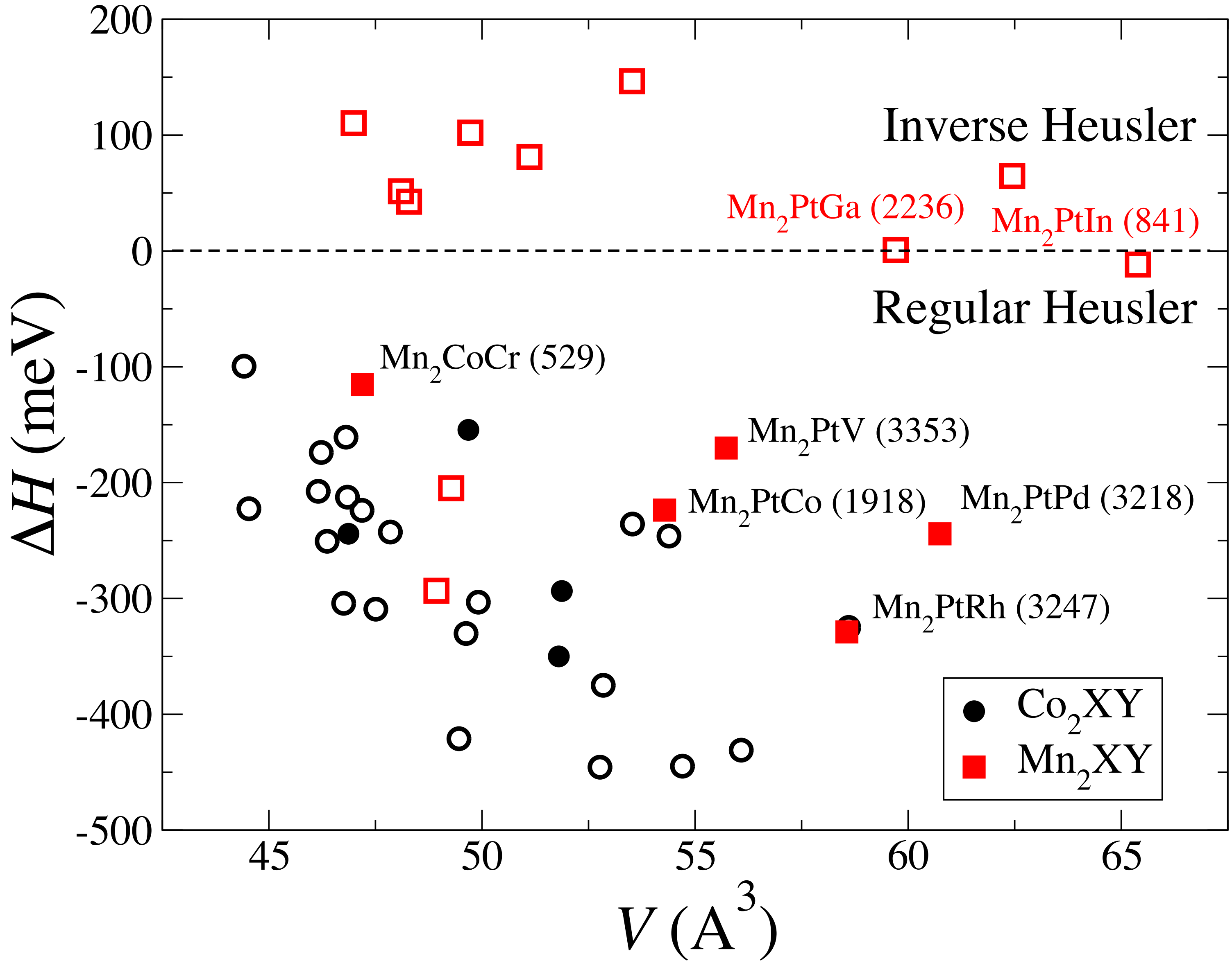
X_2MnZ



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

Mn_2YZ

Mn_2YZ



200
100
0
-100
-200
-300
-400
-500
45
50
55
60
65
ΔH (meV)
V (A^3)
Inverse Heusler
Regular Heusler
Mn2PtGa (2236)
Mn2PtIn (841)
Mn2CoCr (529)
Mn2PtV (3353)
Mn2PtCo (1918)
Mn2PtPd (3218)
Mn2PtRh (3247)
Co2XY
Mn2XY

Mn_3Ga

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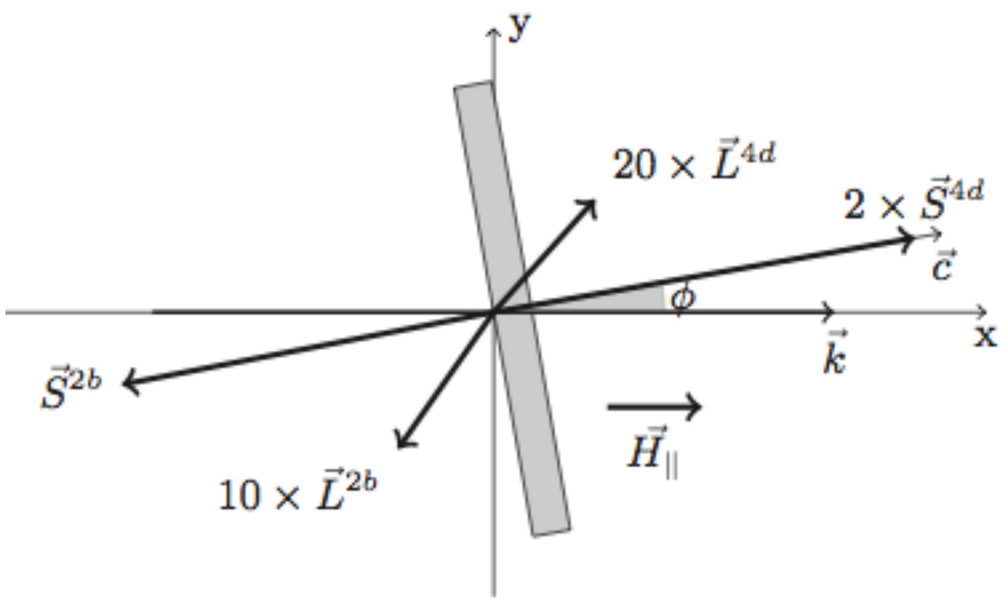
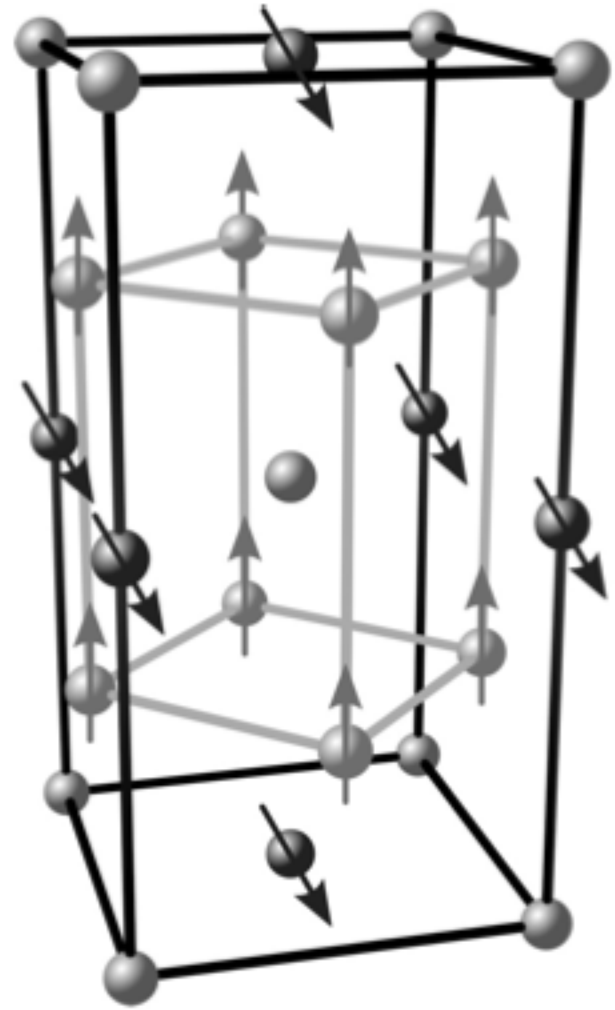
Mn_3Ga





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

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

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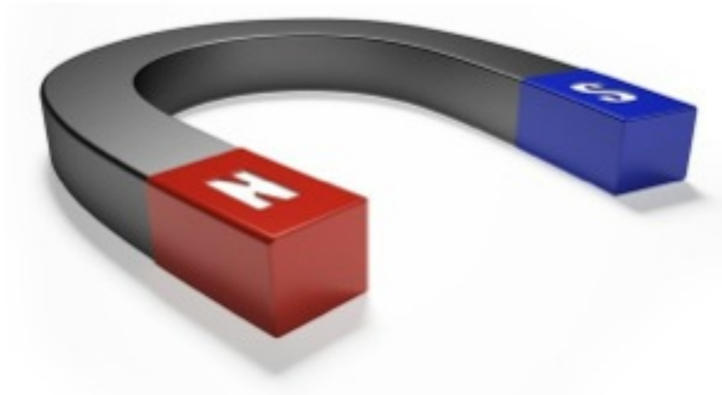
OK, but does all that work?



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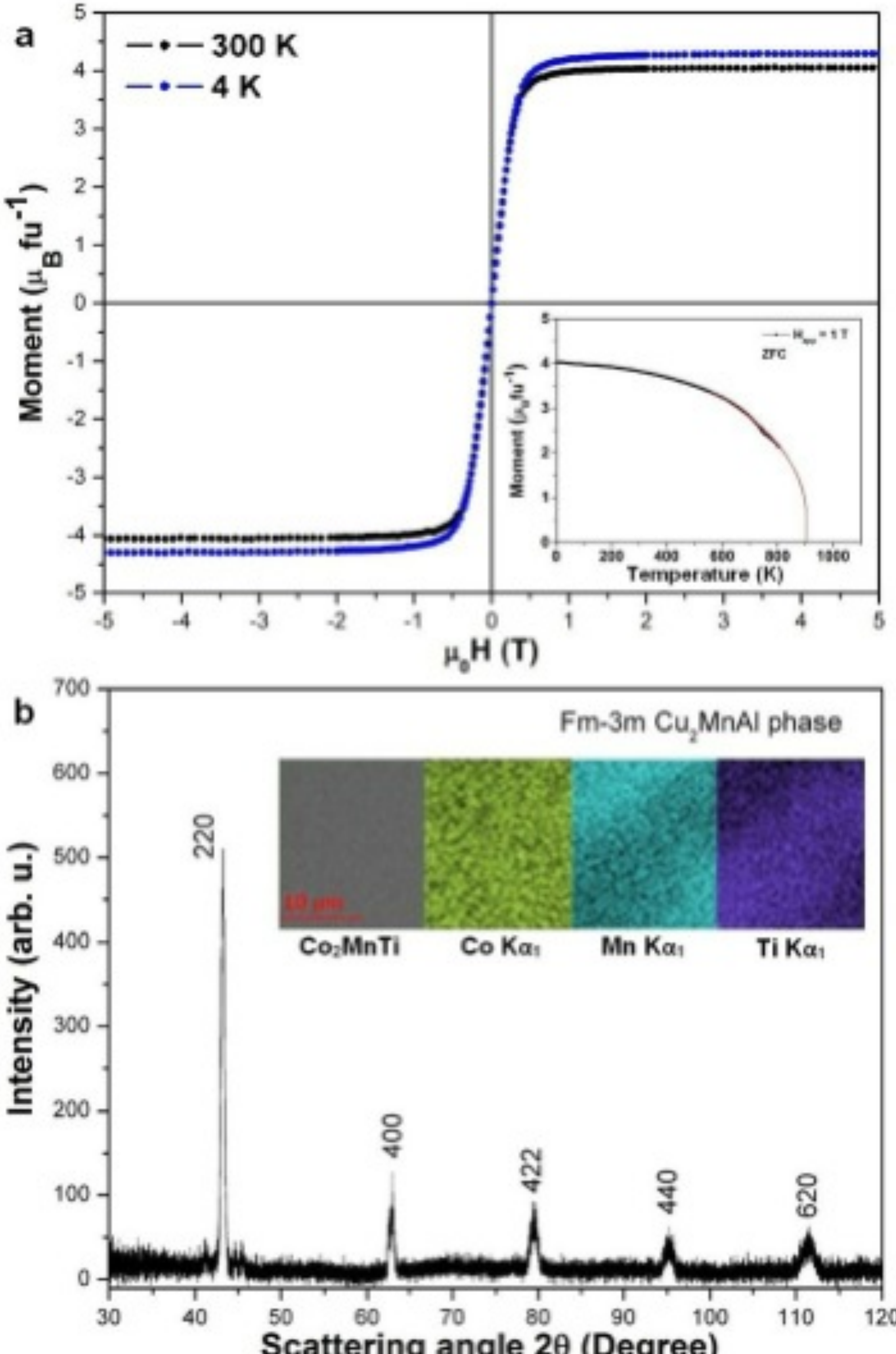

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Co_2MnTi

$T_C^{measured}$ = 940K

$T_C^{predicted}$ = 938K

Prepared by arc melting in an Ar atmosphere

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

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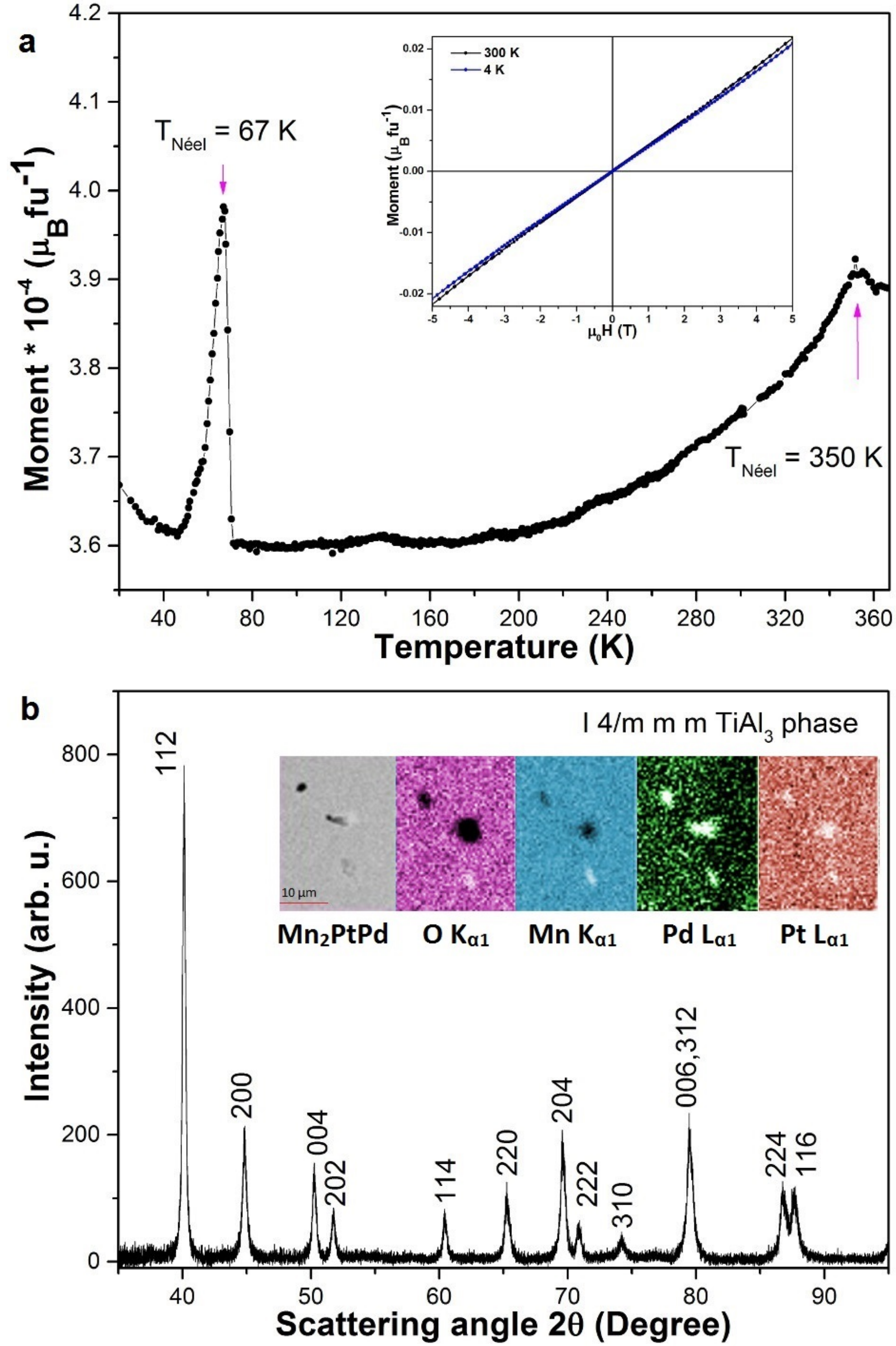
Mn_2PtPd



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

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

Complex antiferromagnetic order

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

Bottom line ….

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
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TRINITY COLLEGE, DUBLIN

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TCD Team:

Tom Archer, Anurag Tiwari, Mario Zic, James Nelson

Duke Team:

Stefano Curtarolo, Junkai Xue, Kevin Rasch, Corey Oses

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SEVENTH FRAMEWORK PROGRAMME

erc

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