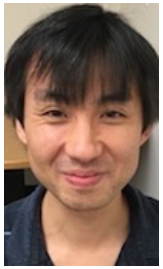
強磁性物質データベースの構築



物性研 福島グループ

櫻井誠大 (Masahiro Sakurai)　sakurai@issp.u-tokyo.ac.jp

2015年4月–2020年3月　米国 テキサス大 ポスドク*

TEXAS
The University of Texas at Austin

- 第一原理計算
  - 磁性クラスター（実空間 DFT）
  - 結晶磁気異方性（平面波 DFT）
  - バンドギャップ（GW法）、光吸収スペクトル（GW+BSE法）
- 強磁性体データベース
  - 機械学習向け基礎データ

* supported by the National Science Foundation (NSF)

Contents

*Most recent paper:

B. Bala., M. Sakurai, C.-Z. Wang, X. Xu, K.-M.Ho, J. R. Chelikowsky, and D. J. Sellmyer: "Synergistic Computational and Experimental Discovery of Novel Magnetic Materials" Molecular Systems Design & Engineering, 5, 1098-1117 (2020). https://doi.org/10.1039/D0ME00050G

NSF-DMREF project (2014–2017–2020)

"Collaborative Research for the Design and Synthesis of Novel Magnetic Materials"

The University of Texas at Austin [Theory]

James R. Chelikowsky

Iowa State University (Ames Lab.) [Theory]

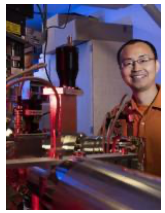
Kai-Ming Ho

Cai-Zhuang Wang

University of Nebraska–Lincoln [Experiment]

David J. Sellmyer

Xiaoshan Xu



RE-free magnetic materials

high magnetization (M_s)
high magnetic anisotropy (K)
high Curie temperature (T_c)

Methods

DFT (PARSEC, QE, VASP)
Adaptive genetic algorithm
LMTO-ASA-GF
"Non-equilibrium" synthesis

Joint work

joint papers
monthly Skype meeting

Permanent magnet materials

Permanent magnet

解説記事 「磁石の秘密」 赤井久純
日本物理学会誌 71 (2016) 277-281.

Key properties:

(i) Saturation magnetization J_S

(ii) Coercivity H_c (~ magnetic anisotropy K_1)

(iii) Curie temperature T_C

母物質(固有特性)
材料(界面、欠陥)
磁石(焼結)

These rely strongly on rare-earth elements (Nd, Sm, Dy, ...) ← *potential supply risk!*

Theoretical approach

pseudopotential DFT plus machine-learning technique

Basic inputs:

Composition: $A_xB_yC_z$

Space group (symmetry)

Lattice parameters: $\boldsymbol{a}$, $\boldsymbol{b}$, $\boldsymbol{c}$

Atomic positions: $\{\boldsymbol{r}_i\}$

→

Site-specific quantities:

Magnetic moments: m_j

Spin-orbit coupling: E_{soc}

Exchange interaction: J_{ij}

→

Key properties:

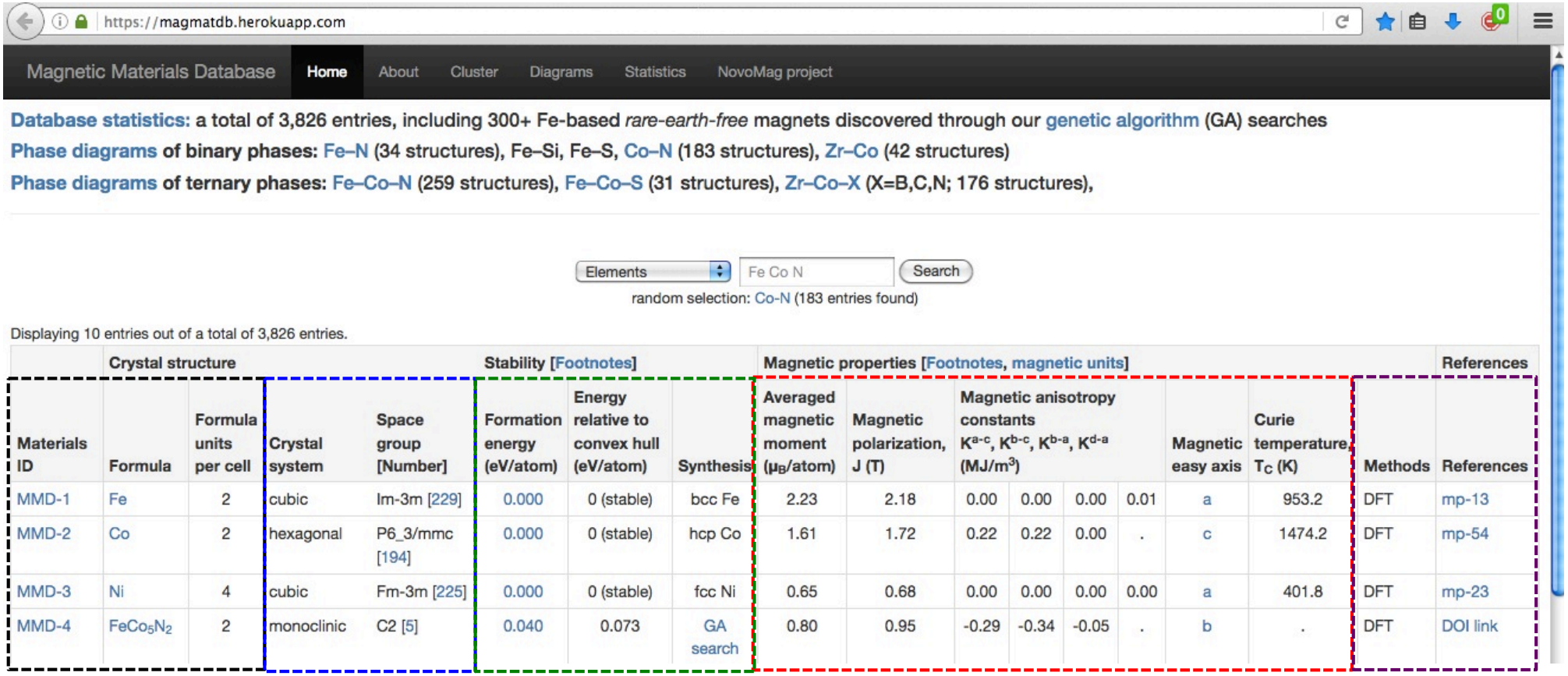
J_S

K_1

T_C

New materials with desired properties

Magnetic Materials Database at https://magmatdb.herokuapp.com



https://magmatdb.herokuapp.com

Magnetic Materials Database | Home | About | Cluster | Diagrams | Statistics | NovoMag project

Database statistics: a total of 3,826 entries, including 300+ Fe-based *rare-earth-free* magnets discovered through our genetic algorithm (GA) searches

Phase diagrams of binary phases: Fe–N (34 structures), Fe–Si, Fe–S, Co–N (183 structures), Zr–Co (42 structures)

Phase diagrams of ternary phases: Fe–Co–N (259 structures), Fe–Co–S (31 structures), Zr–Co–X (X=B,C,N; 176 structures),

Elements | Fe Co N | Search

random selection: Co-N (183 entries found)

Displaying 10 entries out of a total of 3,826 entries.

	Crystal structure				Stability [Footnotes]			Magnetic properties [Footnotes, magnetic units]								References	
Materials ID	Formula	Formula units per cell	Crystal system	Space group [Number]	Formation energy (eV/atom)	Energy relative to convex hull (eV/atom)	Synthesis	Averaged magnetic moment (μ_B/atom)	Magnetic polarization, J (T)	Magnetic anisotropy constants K^{a-c}, K^{b-c}, K^{b-a}, K^{d-a} (MJ/m^3)				Magnetic easy axis	Curie temperature, T_C (K)	Methods	References
MMD-1	Fe	2	cubic	Im-3m [229]	0.000	0 (stable)	bcc Fe	2.23	2.18	0.00	0.00	0.00	0.01	a	953.2	DFT	mp-13
MMD-2	Co	2	hexagonal	P6_3/mmc [194]	0.000	0 (stable)	hcp Co	1.61	1.72	0.22	0.22	0.00	.	c	1474.2	DFT	mp-54
MMD-3	Ni	4	cubic	Fm-3m [225]	0.000	0 (stable)	fcc Ni	0.65	0.68	0.00	0.00	0.00	0.00	a	401.8	DFT	mp-23
MMD-4	$FeCo_5N_2$	2	monoclinic	C2 [5]	0.040	0.073	GA search	0.80	0.95	-0.29	-0.34	-0.05	.	b	.	DFT	DOI link

ID
Formula

Space group

Formation energy

Key magnetic properties:
(1) magnetic moments
(2) magnetic anisotropy energy
(3) Curie temperature

Methods
References

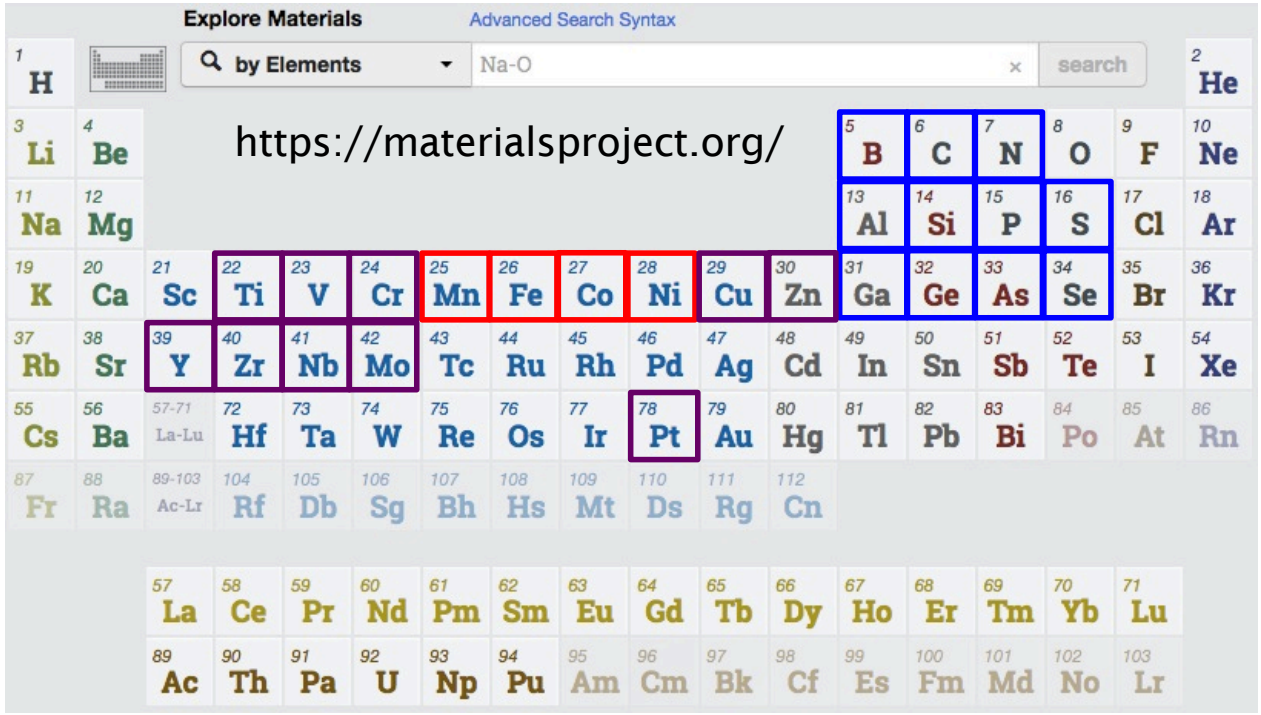
Search filters: elements, formula, space group, easy axis, ...

Available files: CIF, POSCAR, LAMMPS, JSON, YAML

Open-access Materials Database	**[1] Materials Project**	**[2] Open Quantum Materials Database**	**[3] Magnetic Materials Database**	**[4] Topological Material Database**
Team	UC Berkeley	Northwestern University	Texas, Iowa, Nebraska	Europe
Materials data				
# of entries	124,515	637,644	3,826	24,825
crystallography	✔	✔	✔	✔
phase stability	✔	✔	✔	–
Magnetism data				
magnetic moment	✔	–	✔	–
magnetic anisotropy	–	–	✔	–
Curie temperature	–	–	✔	–
Topological data				
Z indices, etc.	–	–	–	✔

[1] https://materialsproject.org
[2] http://oqmd.org
[3] https://magmatdb.herokuapp.com
[4] https://www.topologicalquantumchemistry.org

Elements and Stats

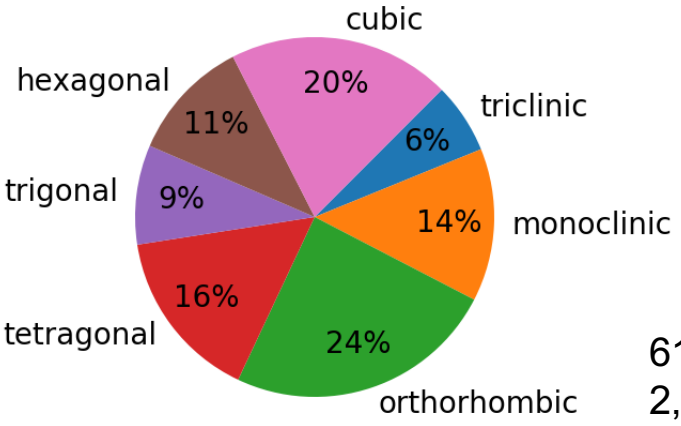


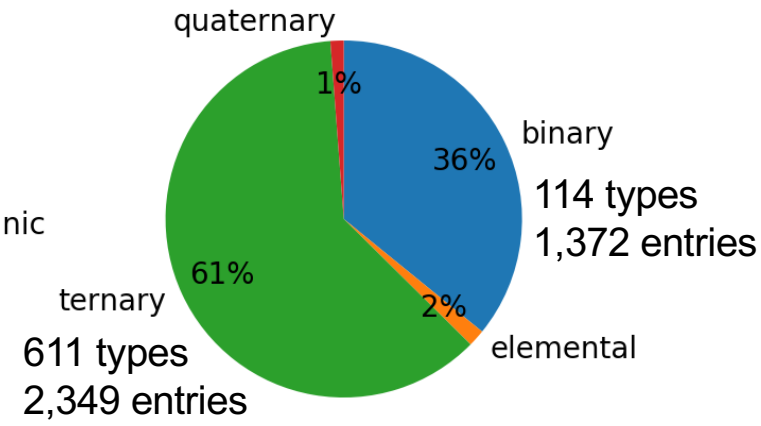
M = Mn, Fe, Co, Ni

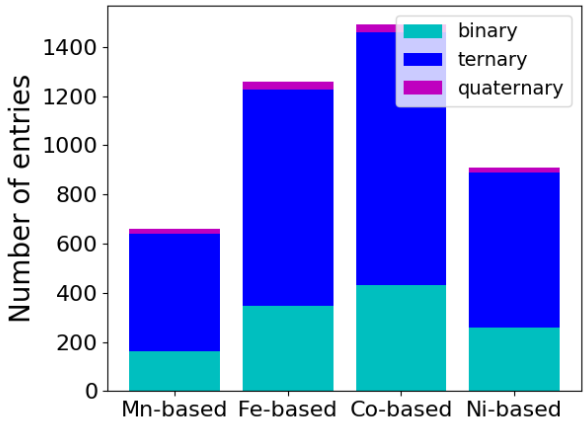
X = B, C, N, ~~O~~;
Al, Si, P, S;
Ga, Ge, As, Se

X = Ti, V, Cr, Cu, Zn;
Y, Zr, Nb, Mo;
Pt

https://magmatdb.herokuapp.com/stats







Adaptive Genetic Algorithm (AGA)

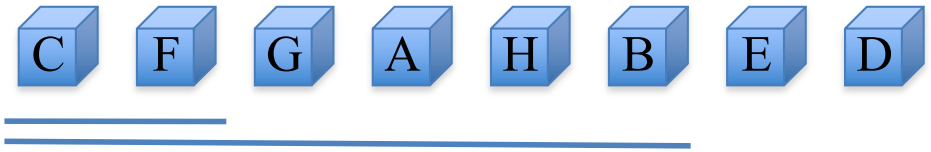
How AGA works:

Step 1: initial seeds

8 slots in a pool

Step 2: relax with classical potentials $\phi(r)$

sort by energy ("rough guess")



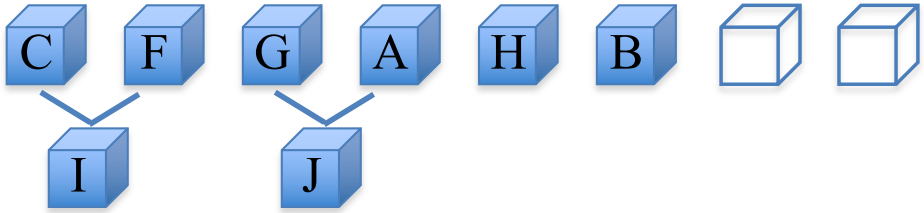
Step 3: refine with DFT ("adaptive")

update classical potentials

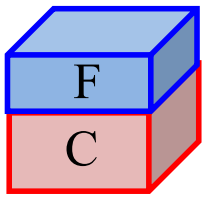
lowest 2 structures:

Energy E, Forces $\{\boldsymbol{F}_i\}$, Stress $\underline{S}$ $\rightarrow$ $\phi^{\text{new}}(r)$

Step 4: new generation



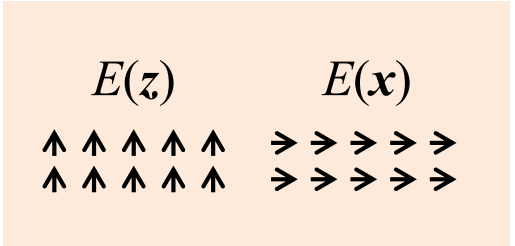
"cut-and-paste" mixing



Pseudopotential DFT

Magnetization & formation energy

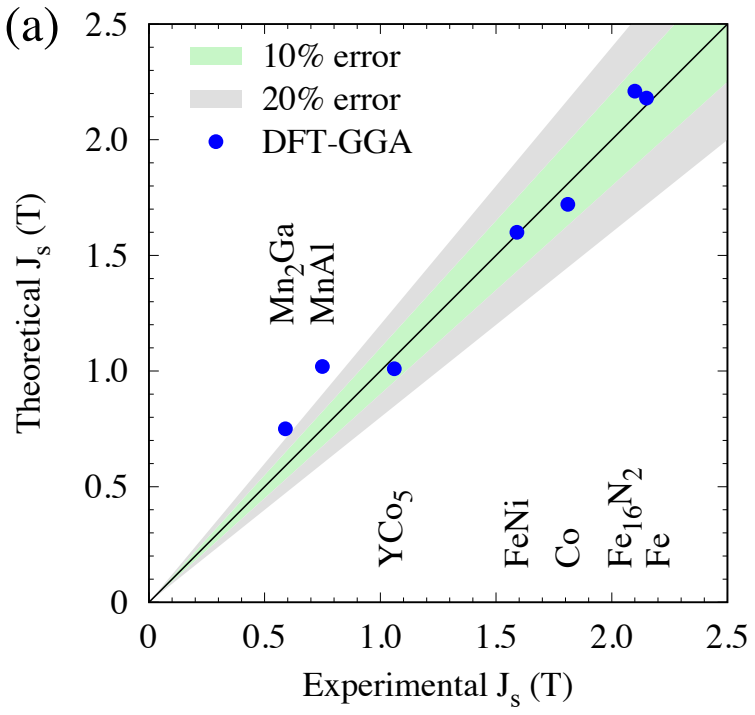
Spin-polarized calculations: $\rho_{up} - \rho_{dn} \rightarrow$ moments, magnetization

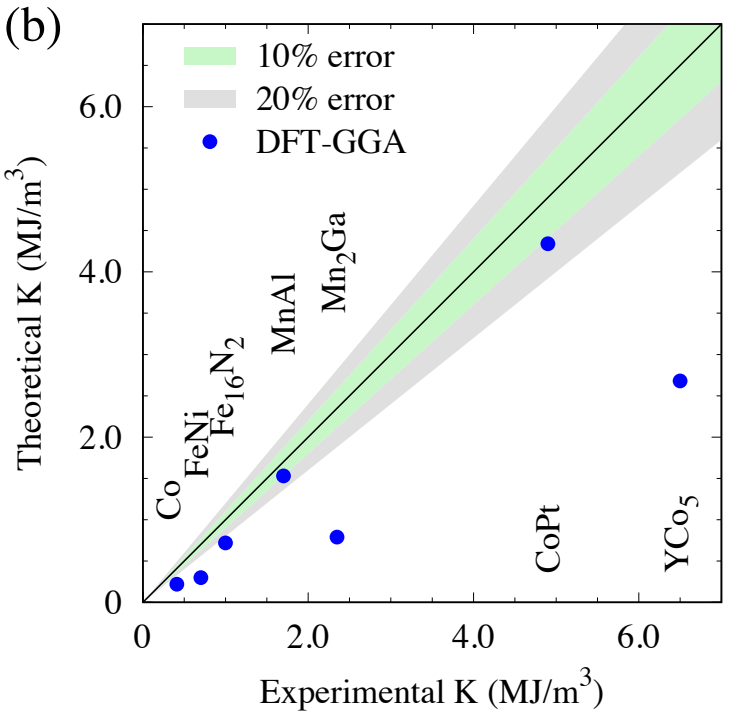


Magnetic anisotropy energy (constant)

$$E_A = K_1 V \sin^2\theta + K_2 V \sin^4\theta + \ldots = E_{tot}(\boldsymbol{x}) - E_{tot}(\boldsymbol{z}) \quad [\sim 1-10 \text{ meV per formula unit}]$$

$$K_1 = E_A / V \quad [\sim 1-10 \text{ MJ/m}^3]$$



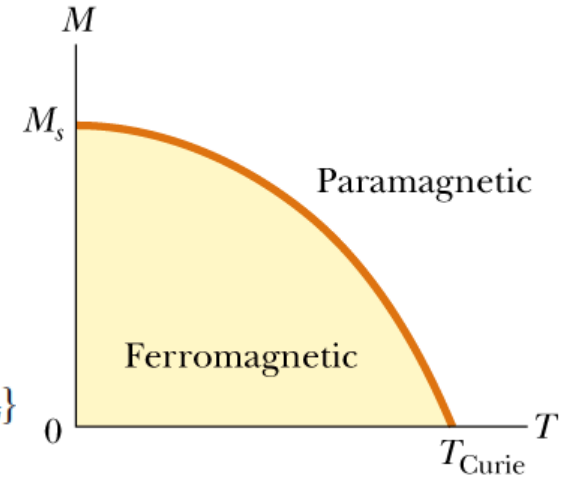


Curie temperature T_C

Classical Heisenberg model $H = -\Sigma_{ij} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$

Mean-field approximation $T_C = 2/3 \, \Sigma_{ij} J_{ij}$

Exchange interactions $J_{ij} = \frac{1}{2\pi} \int^{\epsilon_F} d\epsilon \times \mathrm{Im}\,\mathrm{Tr}_L\{\delta P_i (T_{ij}^{\uparrow} T_{ji}^{\downarrow} + T_{ij}^{\downarrow} T_{ji}^{\uparrow}) \delta P_j\}$

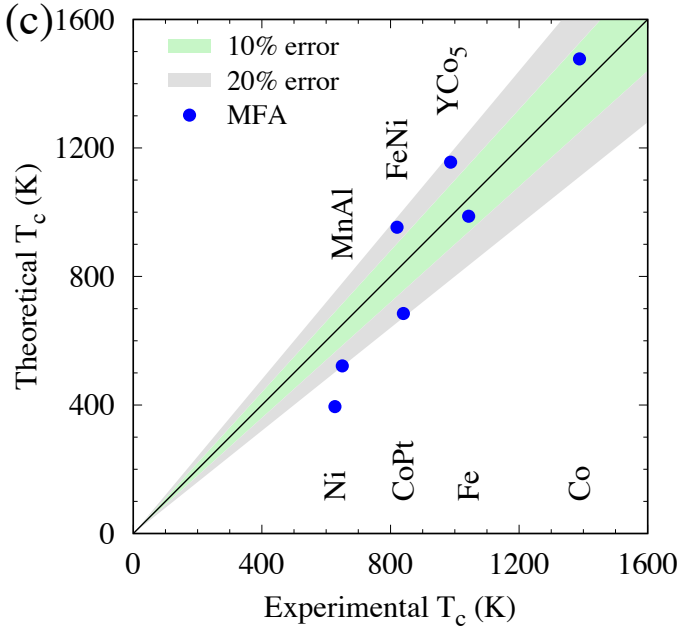


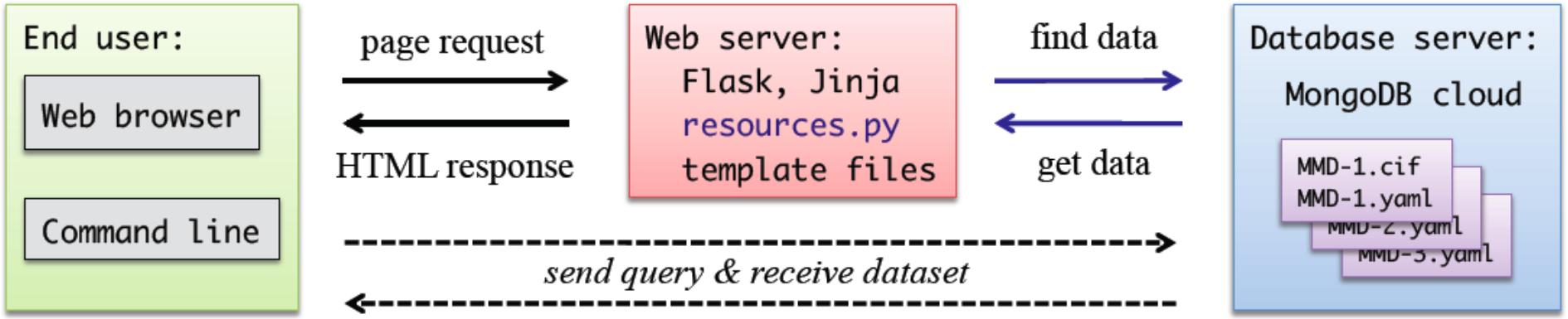
Linear muffin-tin method (LMTO)
Green's function (GF) approach

J. Appl. Phys. 85, 4827 (1999)

QUESTALL (M. van Schilfgaarde)
https://www.questaal.org/

Works well to capture trends!





Software	Role	URL
Database		
MongoDB	Database cloud	www.mongodb.com
Flask, Jinja	Web applications	palletsprojects.com/p/flask
Data entries		
Pymatgen	Crystallographic data (CIF) Computed data (YAML)	pymatgen.org
Adaptive GA	Crystal structure search	doi.org/10.1088/0953-8984/26/3/035402
Materials Project	Known structures	materialsproject.org
First-principles calculations		
QE, VASP	DFT for ΔE_{hull}, M_S, and K_1	quantum-espresso.org + vasp
PARSEC	DFT for clusters	real-space.org
Qustaal	LMTO-GF for T_C and J_{ij}	questaal.org

Data structure

← bcc_Fe.yaml

```
- '@class': ComputedStructureEntry
  '@module': pymatgen.entries.computed_entries
  composition: !!python/object/apply:collections.defaultdict
        dictitems: {Fe: 2.0}
  data:
    critical_temperature:
        Curie_temperature: {unit: K, value: 953.2}
    formation_energy:
        above_hull: {unit: meV/atom, value: 0.0}
        decomposition: {unit: meV/atom, value: 0.0}
    magnetic_anisotropy:
        constant: {a-c: null, b-a: null, b-c: null, d-a: 0.009, unit: MJ/m^3}
        easy_axis: a
        energy: {a-c: null, b-a: null, b-c: null, d-a: 0.001, unit: meV/cell}
        parameters:
            kappa: 0.049
            kgrid: [22, 22, 22]
    magnetic_moment:
        magnetic_polarization: {unit: Tesla, value: 2.18}
        ordering: Ferromagnetic
        total_magnetic_moment: {unit: Bohr_magneton/atom, value: 2.21}
    :
    :
    :
```

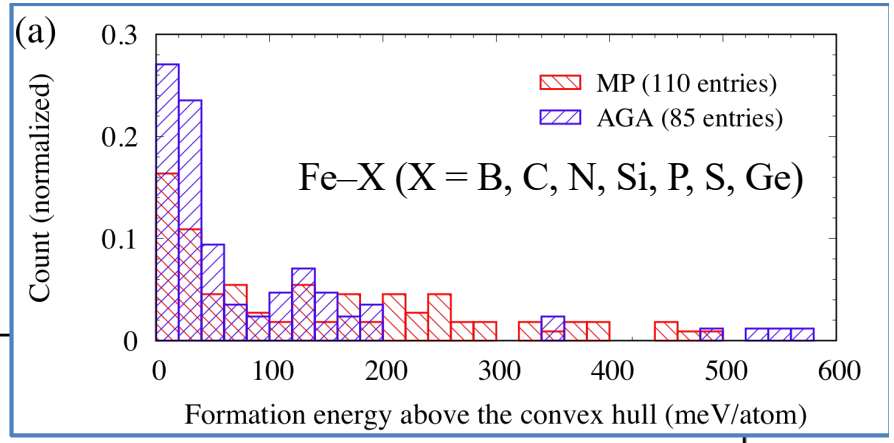
Data structure (continued)

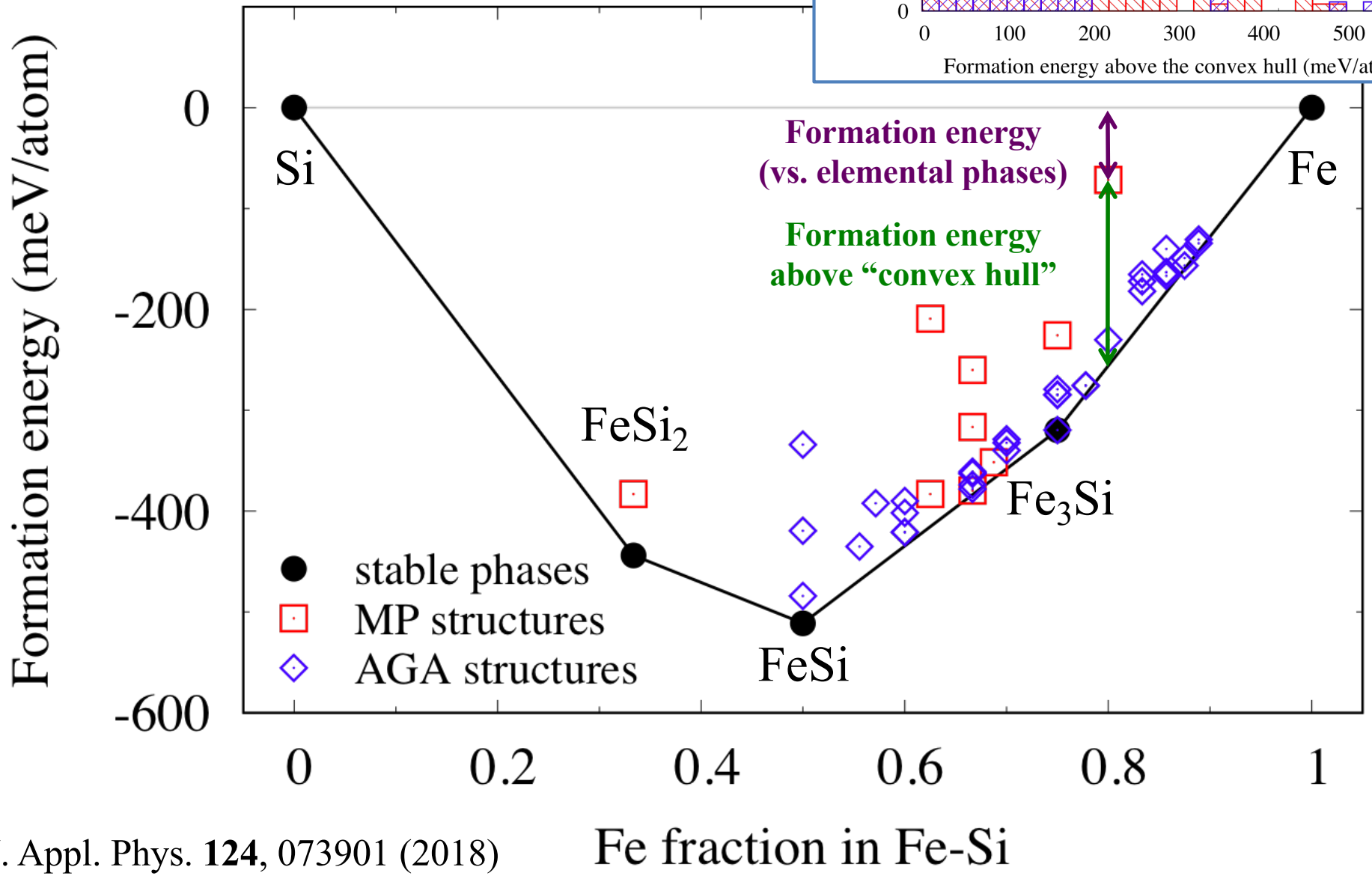
```
structure:
    '@class': Structure
    '@module': pymatgen.core.structure
    lattice:
        a: 2.86944
        alpha: 90.0
        b: 2.86944
        beta: 90.0
        c: 2.86944
        gamma: 90.0
    volume: 23.626067707920384
    sites:
    - abc: [0.0, 0.0, 0.0]
      label: Fe
      properties: {}
      species:
      - {element: Fe, occu: 1.0}
      xyz: [0.0, 0.0, 0.0]
    - abc: [0.5, 0.5, 0.5]
      label: Fe
      properties: {}
      species:
      - {element: Fe, occu: 1.0}
      xyz: [1.43472, 1.43472, 1.43472]
```

← bcc_Fe.cif

```
Site-specific data:
    magmom: 2.2628      (on-site)
    soc: null           (on-site)
    jij: [null, null]   (pairwise)
```

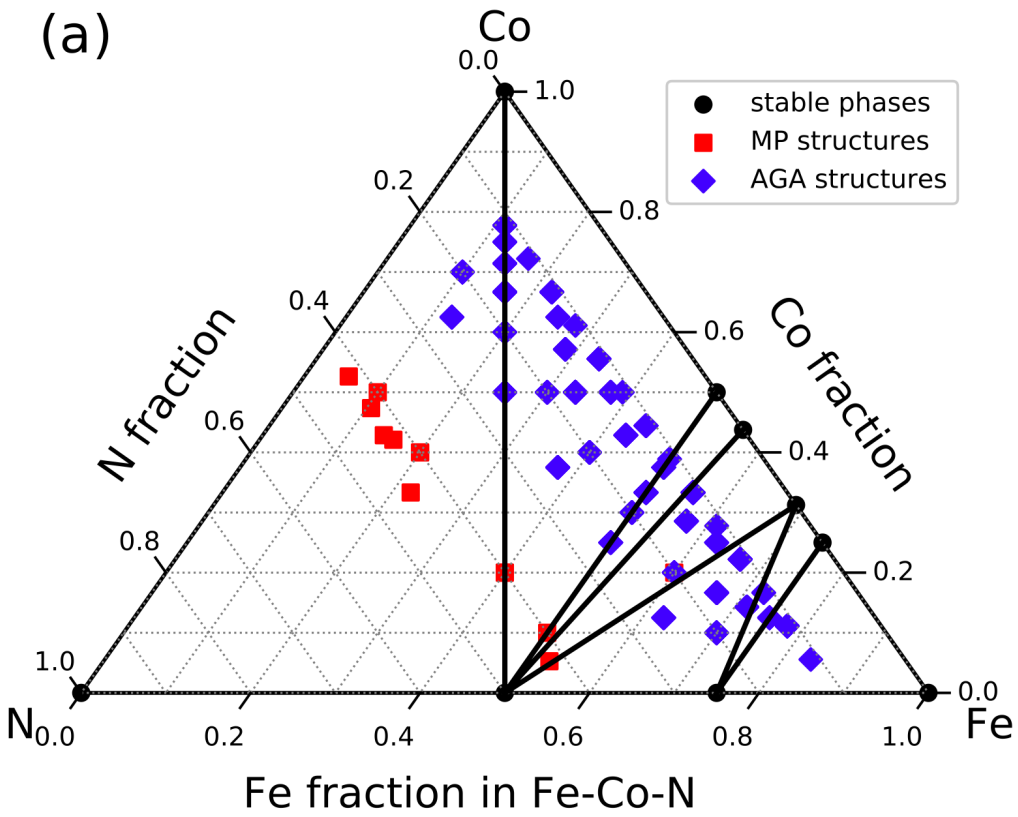
Metastable structures from AGA search



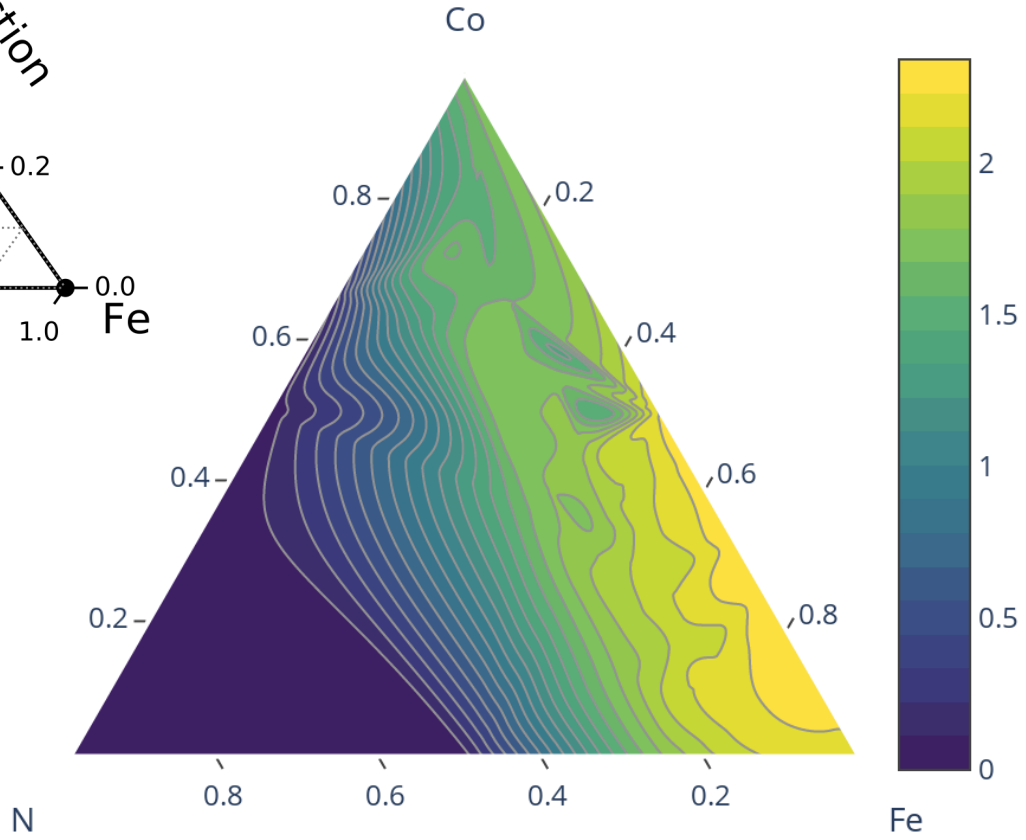


J. Appl. Phys. **124**, 073901 (2018)

Metastable structures from AGA search

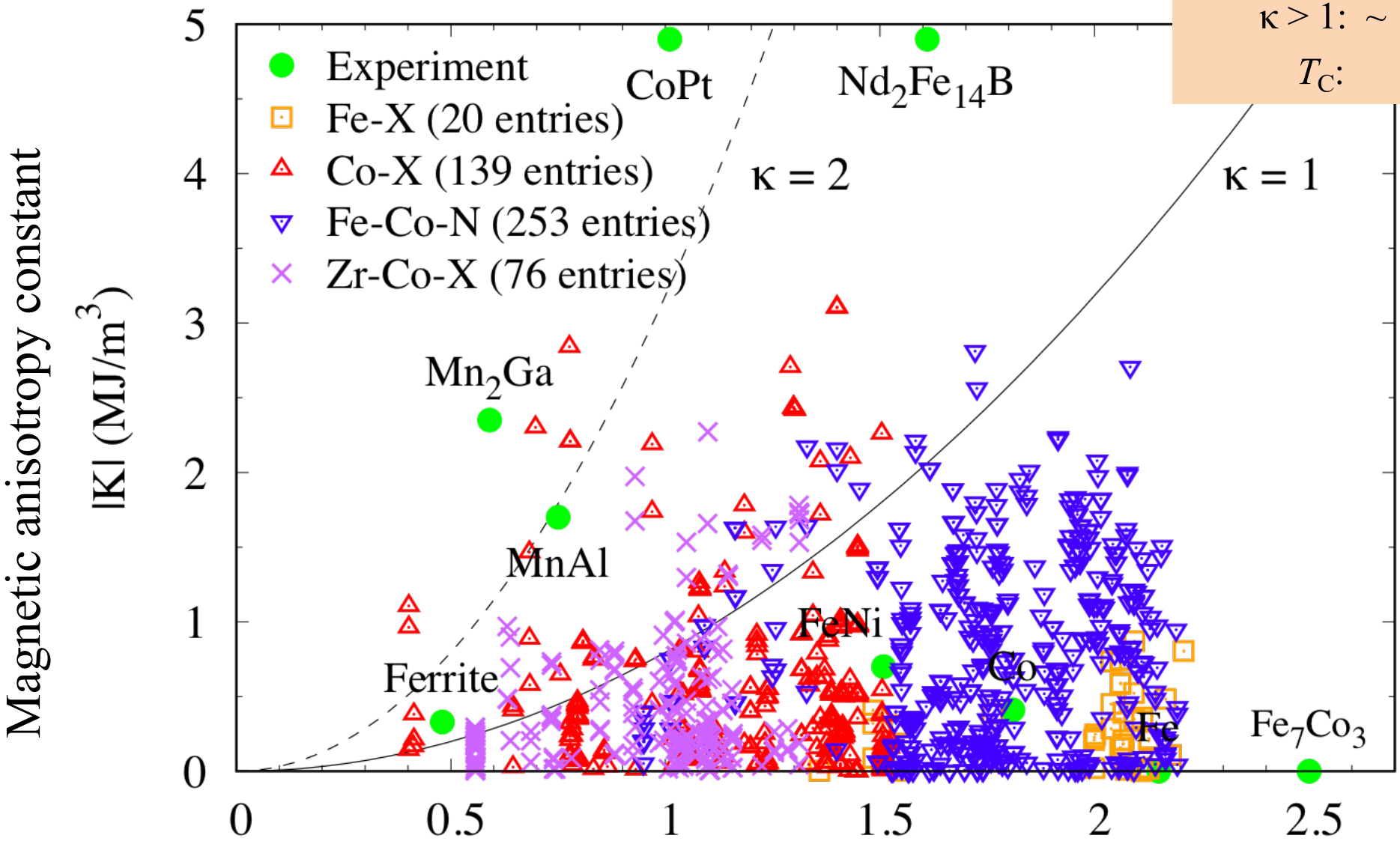


Saturation magnetic polarization J_s

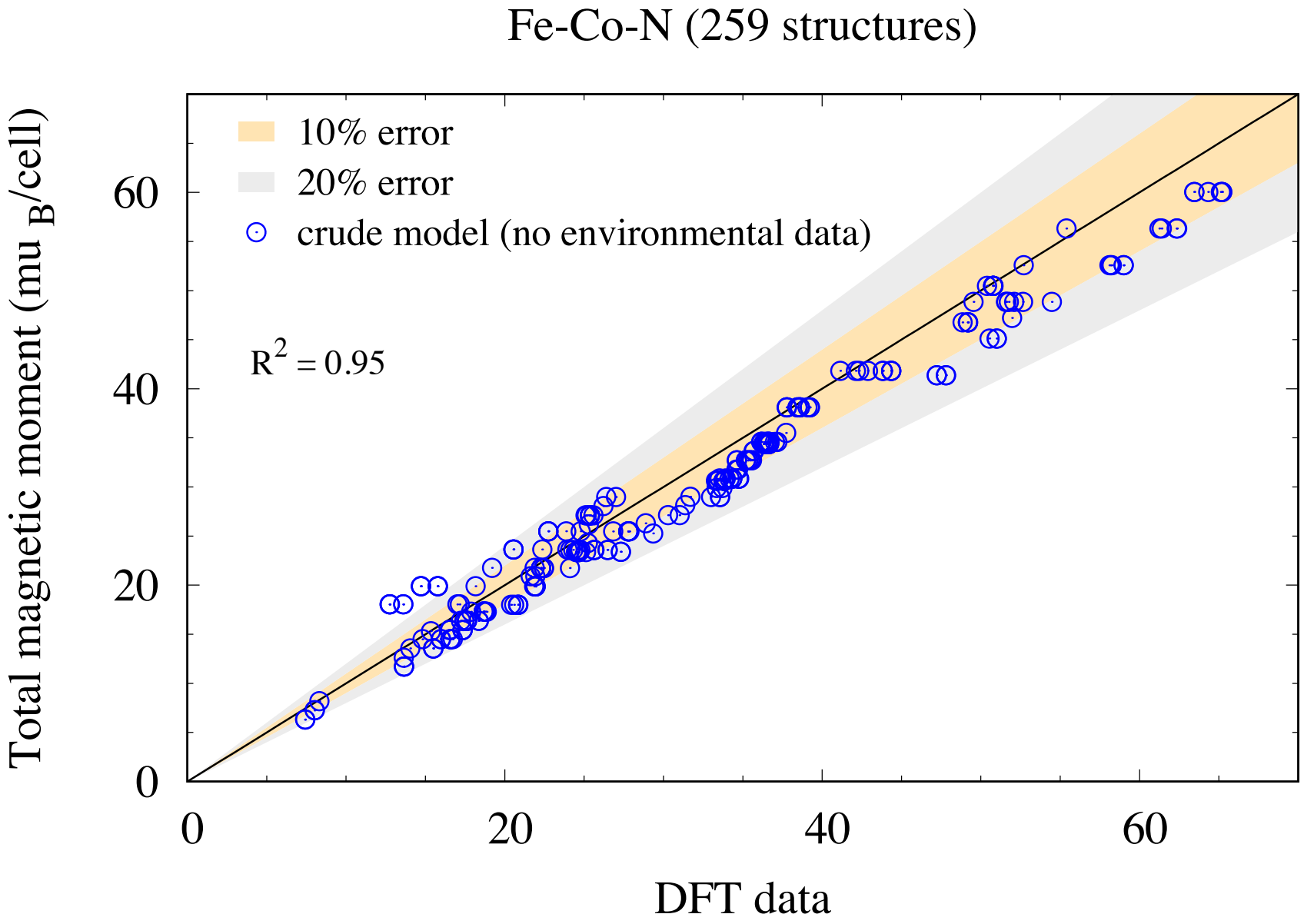


Magnetic properties of AGA structures

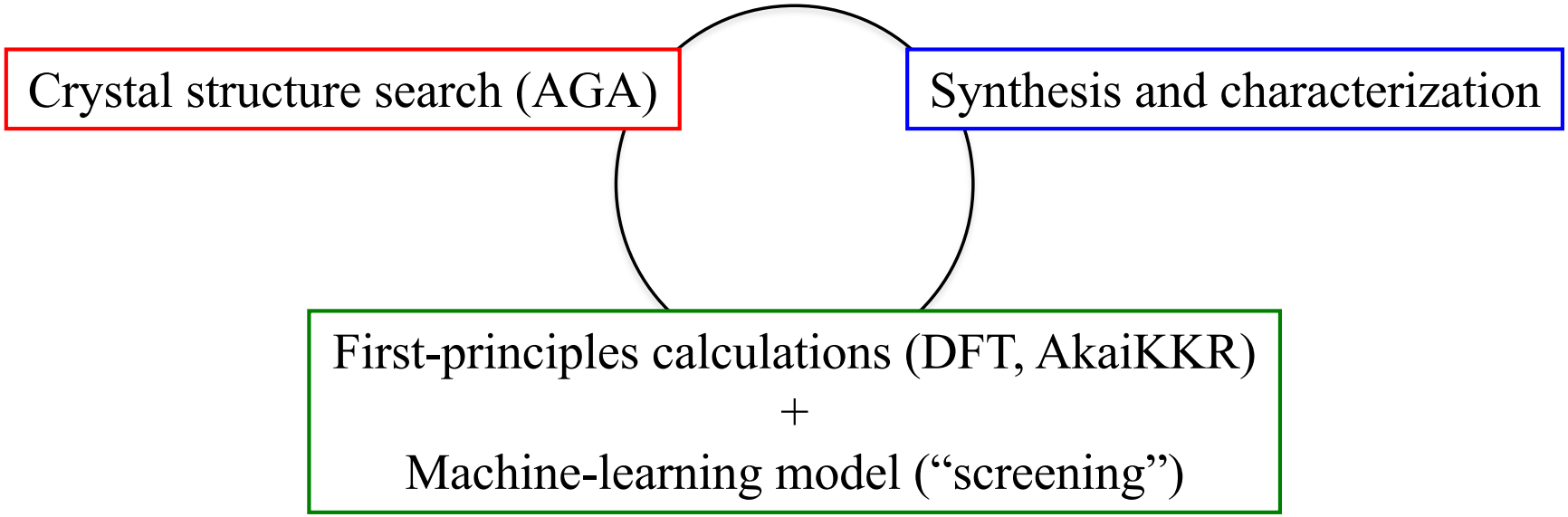
Total: 351 entries
$K > 1$: ~ 33%
$\kappa > 1$: ~ 10%
T_C: ?



“Prediction” from site-specific data



Data-driven approaches for new magnets



Targets:

(i) Nd–Fe–B, Sm–Fe–N

(ii) rare-earth-*lean*

(iii) rare-earth-free: Fe–Co–X, Fe–Mn–X, ...

Concerns:

f-electron

poor properties?

massive calculations?