Computational magnetic materials discovery

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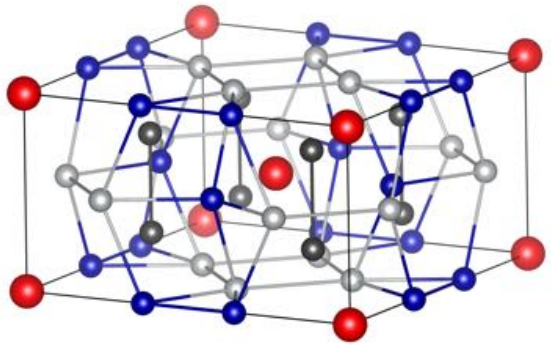
ESIC MM
Elements Strategy Initiative Center for Magnetic Materials

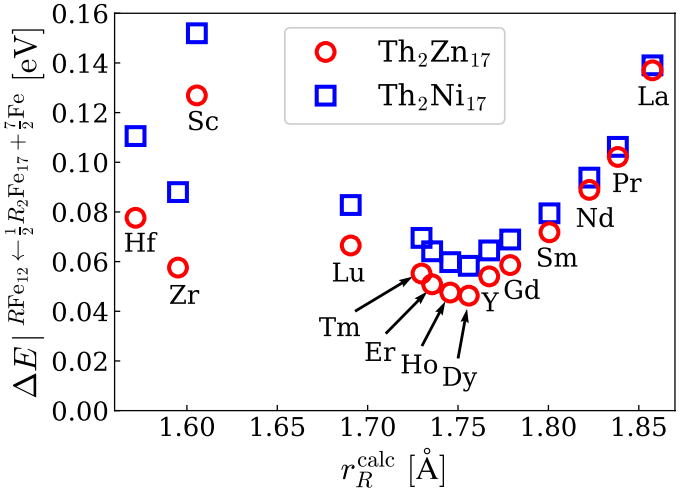
Mi²i

CdMSi

- ✓ Superconductor having highest T_c?
- ✓ Harder material than diamond ?
- ✓ Strong magnet ?

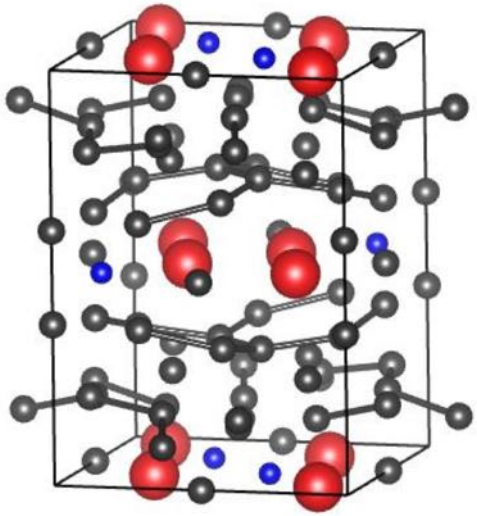
...





Atomic species + Composition + Structure

History of permanent magnet



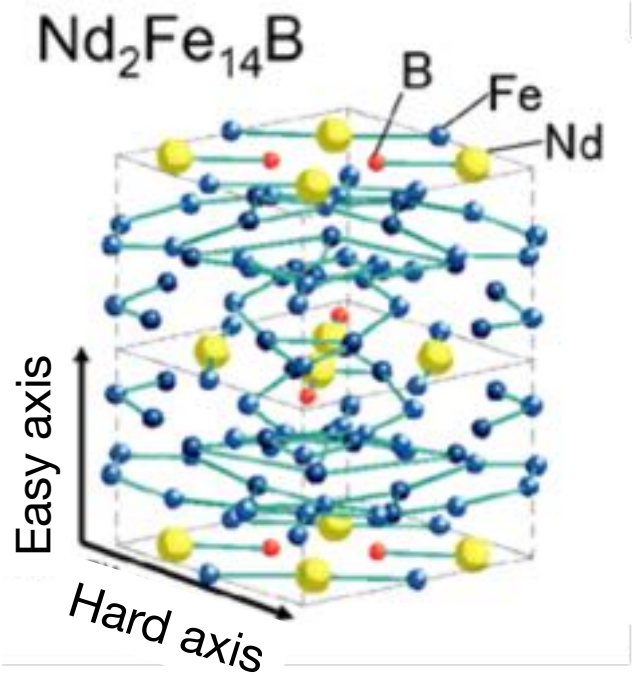
$Nd_2Fe_{14}B$

- ✓ High saturation magnetization
- ✓ High coercivity → High magnetocrystalline anisotropy
- ✓ High Curie temperature
- ✓ Phase stability

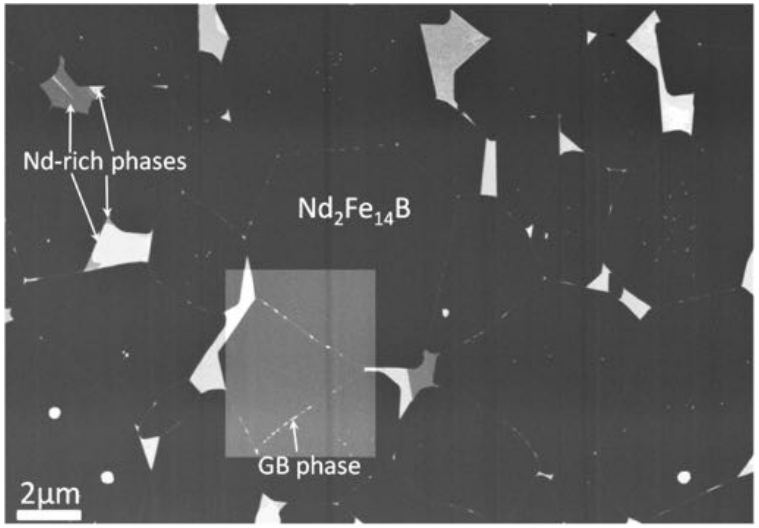
Magnet compound vs Magnet material

Intrinsic properties

Sample dependent
(microstructure, interfaces, …)



Nd-Fe-B magnet



K. Hono (NIMS)

- ✓ Saturation magnetization
- ✓ Magnetocrystalline anisotropy
- ✓ Curie temperature

- ✓ Coercivity
- ✓ Microstructure

Magnet compounds

Magnetic properties
- Saturation magnetization
- Magnetocrystalline anisotropy
- Curie temperature

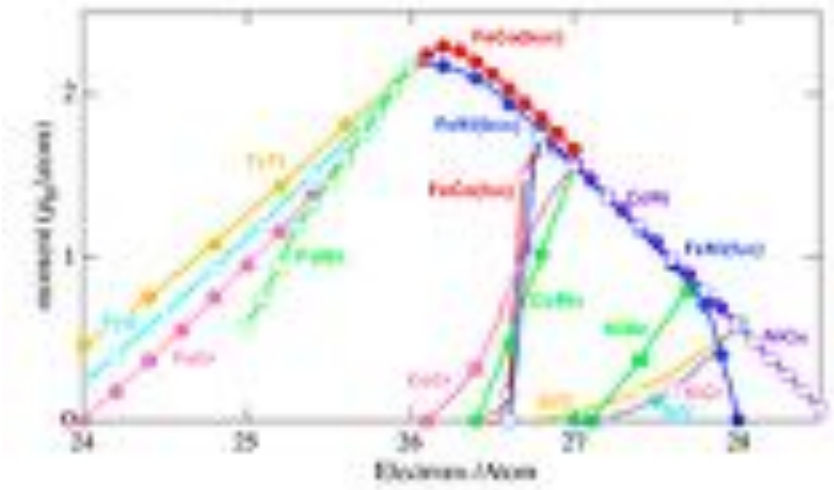
Structural properties
- Phase stability

Fe, Co, Mn, …

Nd, Sm, …

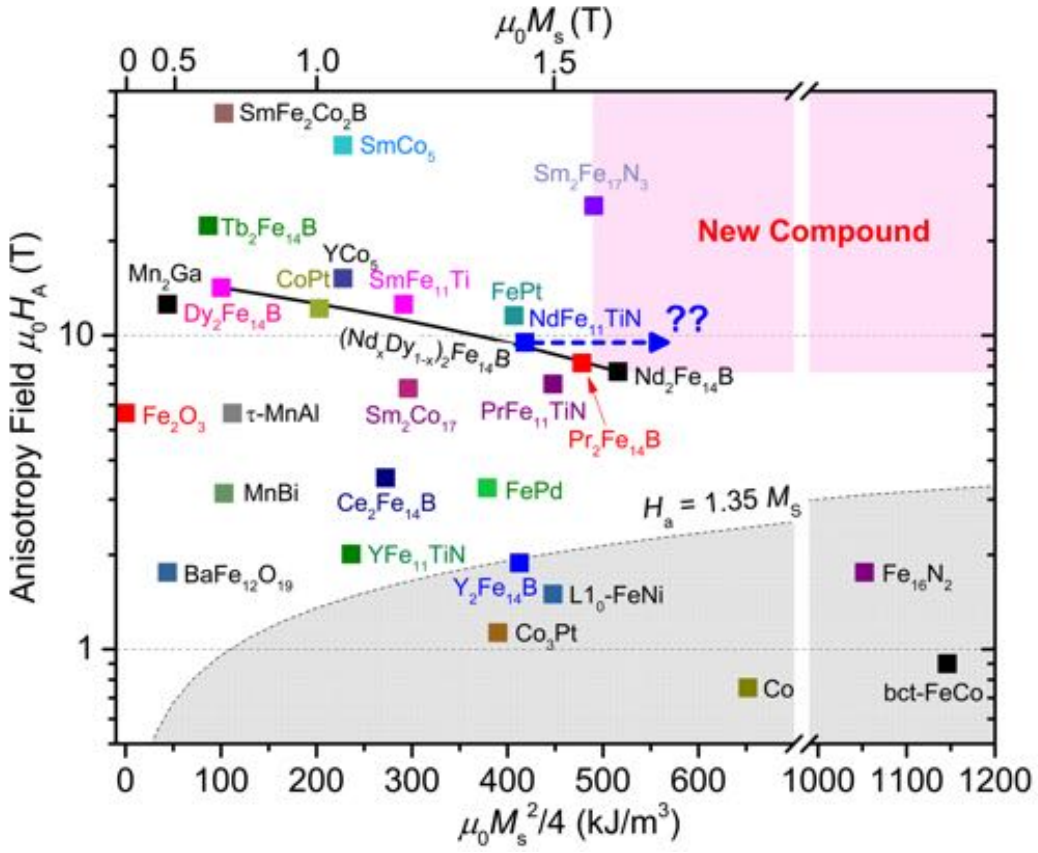
B, C, N, …

Other elements ???



Slater-Pauling curve
Calculation by H. Akai

Hard-magnetic compounds



Hirayama, Miyake and Hono, JOM **67**, 1344 (2015)

2:14:1 type
$Nd_2Fe_{14}B$

1:5 type (m=1, n=0)
$SmCo_5$, YCo_5

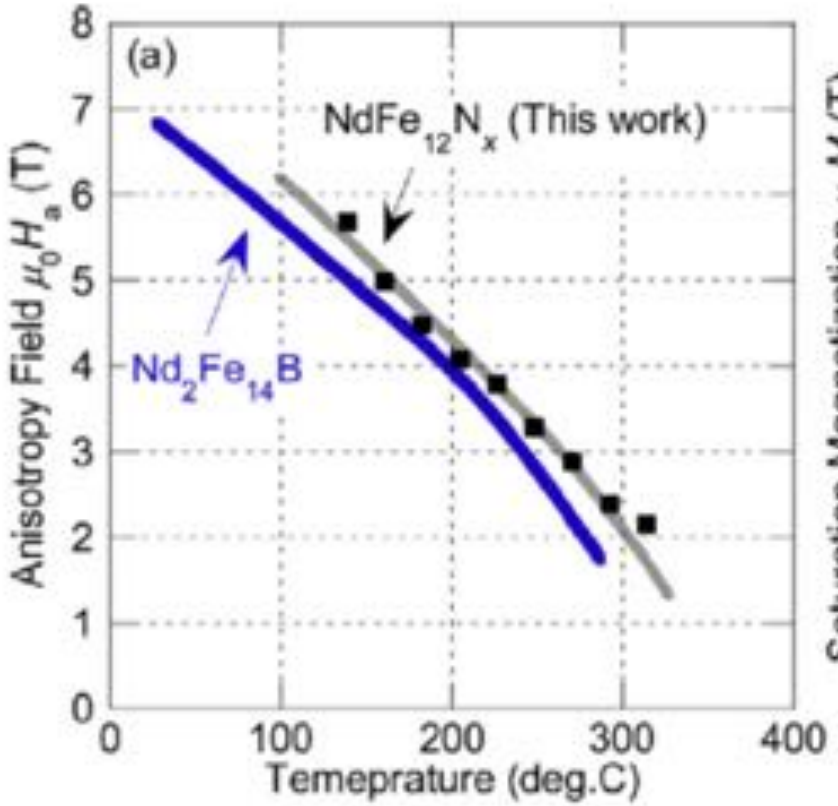
2:17, 2:17:3 type (m=3, n=1)
Sm_2Co_{17}, $Sm_2Fe_{17}N_3$

1:12 type (m=2, n=1)
$SmFe_{11}Ti$, $NdFe_{11}TiN$

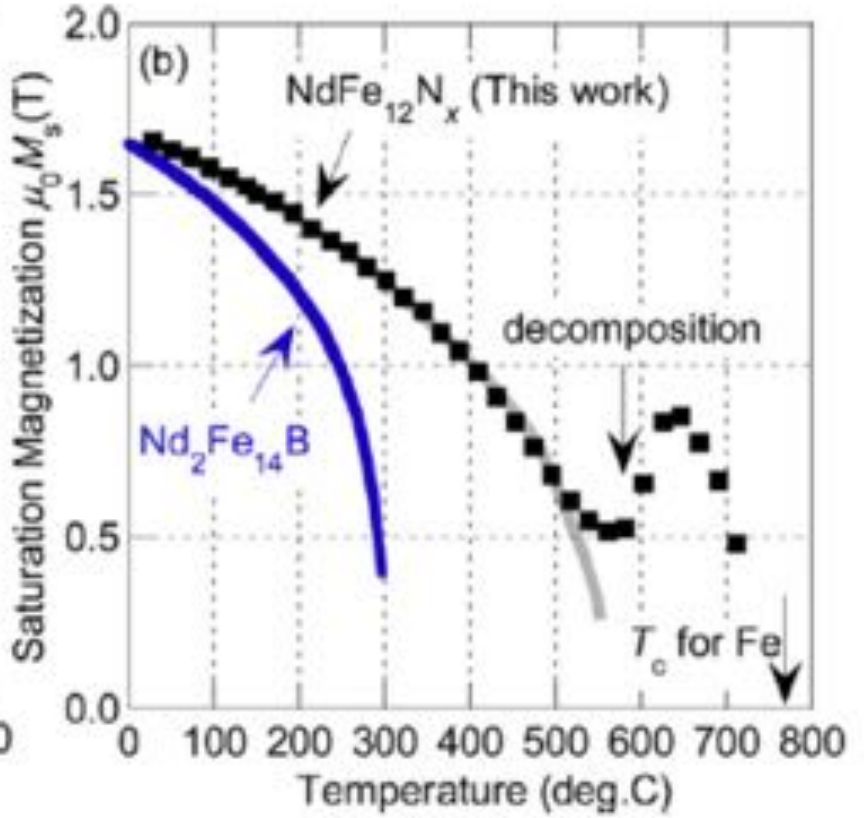
$$R_{m\text{-}n}T_{5m+2n}$$

$NdFe_{12}N$ film

Magnetic anisotropy



Magnetization



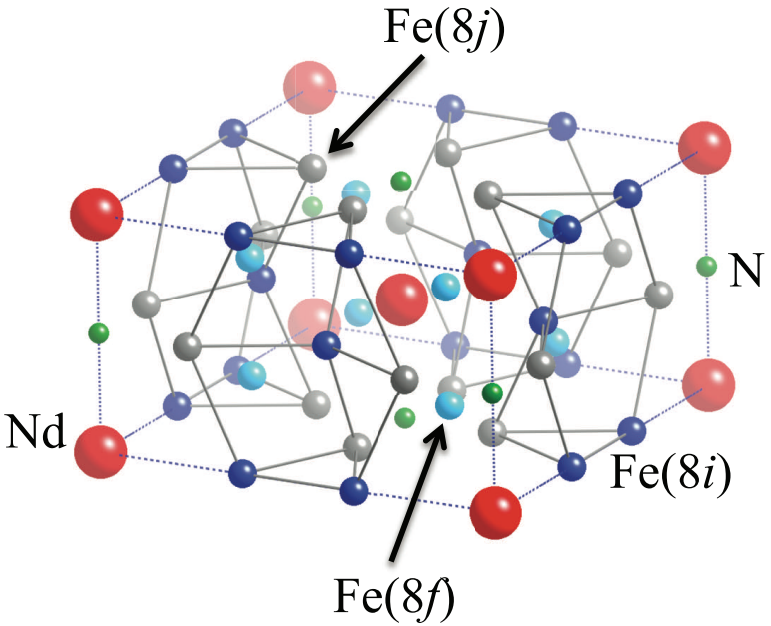
Y. Hirayama et al., Scripta Materialia **95**, 70 (2015)

Ce, Pr, Nd, Sm, …

B, C, N

$$R\,(\mathrm{Fe}_{1-\alpha}T_{\alpha})_{12}X$$

Ti, Cr, V, Mn, Co, Ni, …



- Magnetization
- Magnetocrystalline anisotropy
- Curie temperature
- Formation energy

High-throughput computational screening

Direct screening of 1,280 compounds by first-principles calculation

System	$(BH)_{max}^{EST}$ [kJ/m³]	$\mu_0 M^{ASA}$ [T]	K_1^{ASA} [MJ/m³]	H_a^{ASA} [T]	K_1^{other} [MJ/m³]	$\mu_0 M^{exp}$ [T]	H_a^{exp} [T]
$NdFe_{12}$*	636	1.99	3	3	−2.2[e]		
$NdFe_{12}B$	611	1.95	45	58			
$NdFe_{12}C$	617	1.96	47	60			
$NdFe_{12}N$	686	2.06	47	57	9.91[e]	1.66[d]	8[d]
$NdFe_{11}Ti$	438	1.65	4	7	−0.58[e] 1.70[a]	1.70[a]	2.0[a]
$NdFe_{11}TiB$	432	1.64	48	72	−0.70[f]		
$NdFe_{11}TiC$	432	1.64	50	76	2.6[f]		
$NdFe_{11}TiN$	487	1.74	49	71	11.3[e] 10.6[f]	1.48[d]	≥7[a],
$CeFe_{12}$*	586	1.91	4	5			
$CeFe_{12}B/C/N$	556/568/630	1.86/1.88/1.98	127/137/139	170/182/175			
$CeFe_{11}Ti$	396	1.57	11	18		1.19[c] 1.55[a]	2.96[c] 2.3[a]
$CeFe_{11}TiB/C/N$	396/391/443	1.57/1.56/1.66	134/145/148	213/232/222			
$SmFe_{12}$*	538	1.83	−5	−6	2.4[e]		
$SmFe_{11}Ti$	357	1.49	−8	−13	−0.52[e]		
$SmFe_{11}TiN$	401	1.58	−73	−115	−20.4[e]		
$SmFe_{12}N$	580	1.90	−71	−93	−18.1[e]		
$CeFe_{11}Co_1B/N$	536/605	1.83/1.91	129/142	176/183			
$CeFe_8Co_4B/C/N$	464/464/521	1.70/1.70/1.80	116/141/146	168/208/203			
$CeFe_8Ni_4N$	417	1.61	167	260			
$NdFe_{11}Co_1B/C/N$	586/586/661	1.91/1.91/2.03	46/48/48	60/63/59			
$NdFe_8Co_4B/C/N$	520/505/574	1.80/1.77/1.89	41/49/50	57/69/67			

Körner, Krugel and Elsässer, Sci. Rep. **6**, 24686 (2016)

Topological materials

T. Zhang et al., arXiv: 1807.08756

c.f. F. Tang et al., arXiv:1807.09774
M.G. Vergniory et al., arXiv:1807.10271

Structure data
from database

Electronic structure
by first-principles calc.

39,519 materials from ICSD

8,056 are topological

5,005 topological semimetals
1,814 topological insulators
1,237 topological crystalline insulators

Exhaustive search ??

Material data
from database

$\{ x_i \}$ material

↓

Candidate compounds
by chemical substitution

$\{ x'_i \}$ material

↓

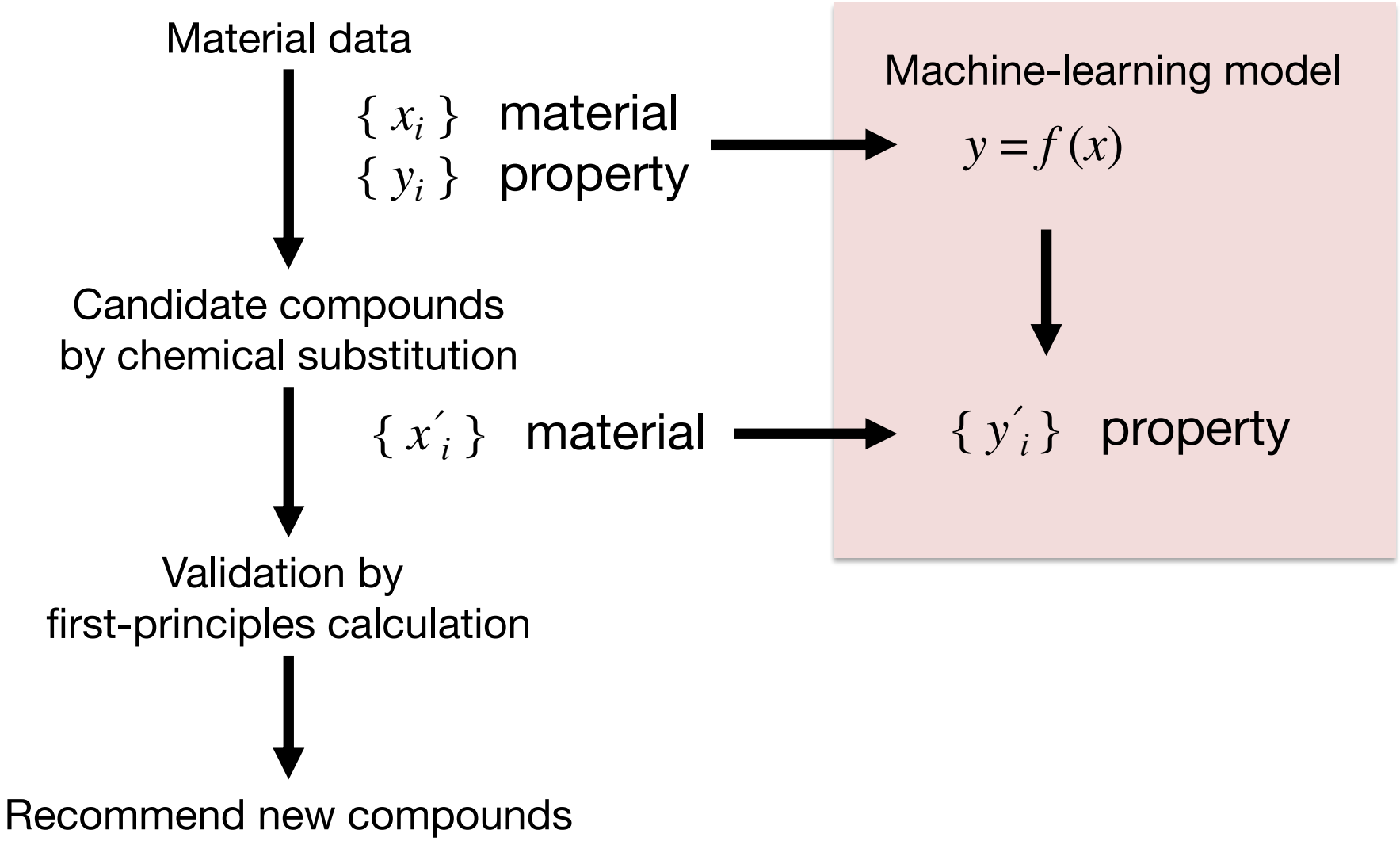
Evaluate properties by
first-principles calculation

$\{ y'_i \}$ property

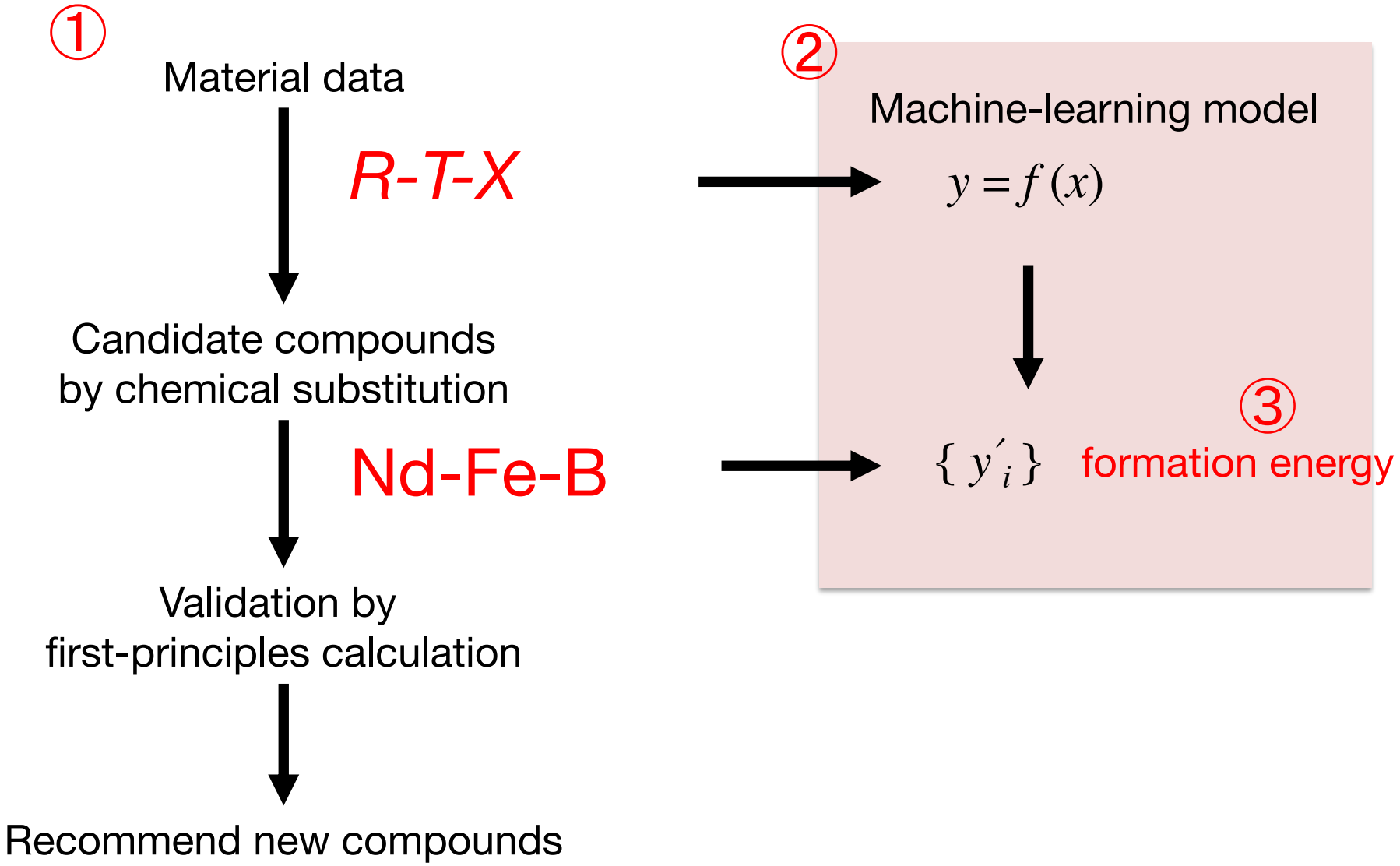
↓

Recommend new compounds

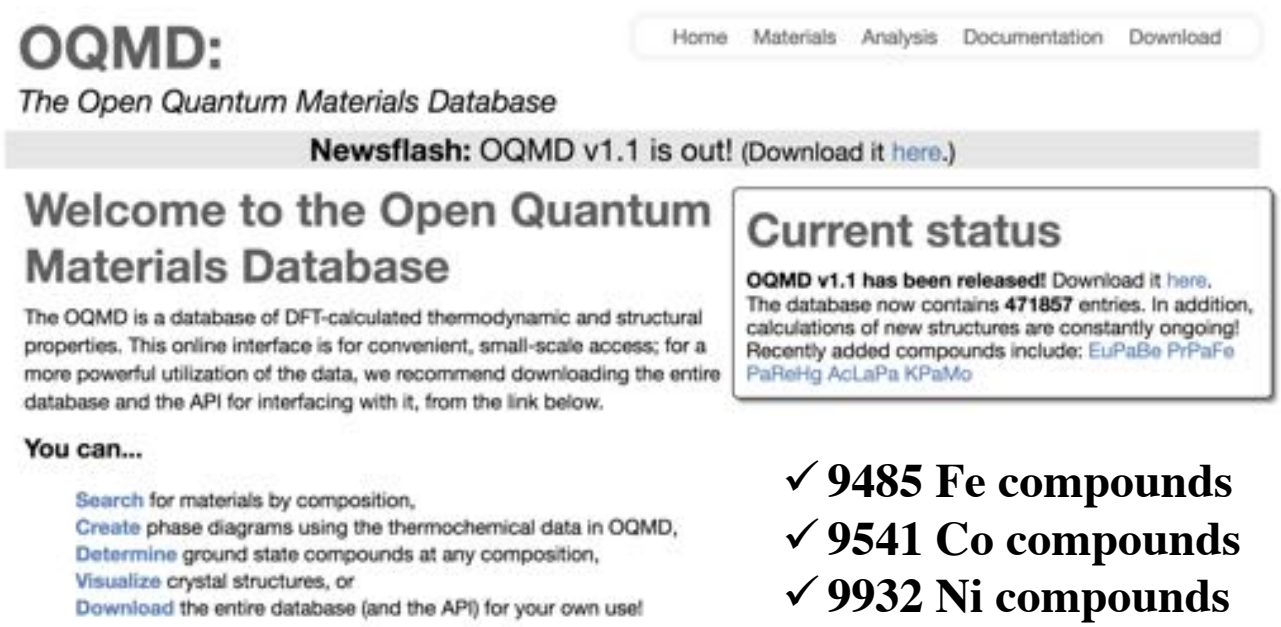
Virtual screening



Virtual screening



① Computational database



✓ **9485 Fe compounds**
✓ **9541 Co compounds**
✓ **9932 Ni compounds**

RTX materials

$T_{4d\text{-}5d}$ = ["Y", "Zr", "Nb", "Mo", "Tc", "Ru", "Rh", "Pd", "Ag", "Cd", "Hf", "Ta", "W", "Re", "Os", "Ir", "Pt", "Au", "Hg"]

T_{3d} = ["Ti", "V", "Cr", "Mn", "Fe", "Co", "Ni", "Cu", "Zn"]

R = ["La", "Ce", "Pr", "Nd", "Pm", "Sm", "Eu", "Gd", "Tb", "Dy", "Ho", "Er", "Tm", "Yb", "Lu"]

X = ["B", "C", "N", "O"]

Optimized structure
Total energy
Magnetic moment
etc.

http://oqmd.org

Kernel regression

$$f(\vec{x}) = \sum_{i=1}^{N} \alpha_i k(\vec{x}^{(i)}, \vec{x})$$

Gaussian kernel

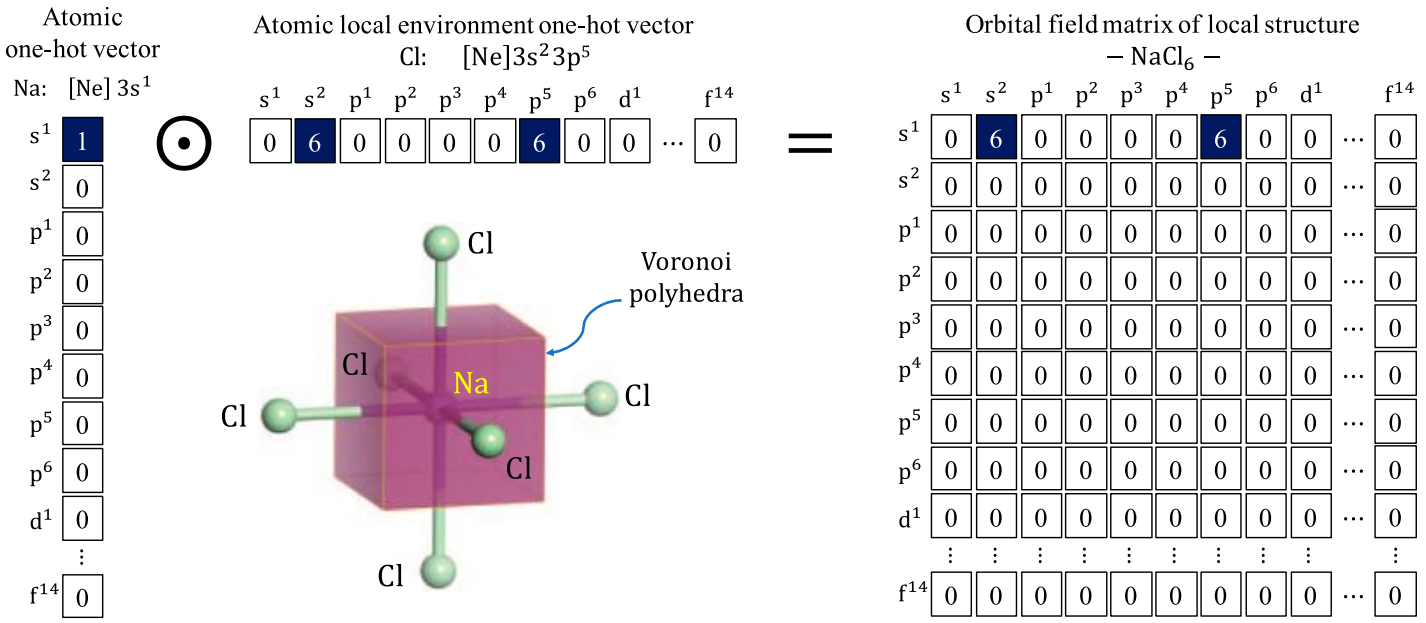
$$k(\vec{x}, \vec{x'}) = C \exp(-\beta ||\vec{x} - \vec{x'}||^2)$$

Laplacian kernel

$$k(\vec{x}, \vec{x'}) = C \exp(-\beta ||\vec{x} - \vec{x'}||)$$

Orbital Field Matrix (OFM)

Pham et al., STAM **18**, 756 (2017)

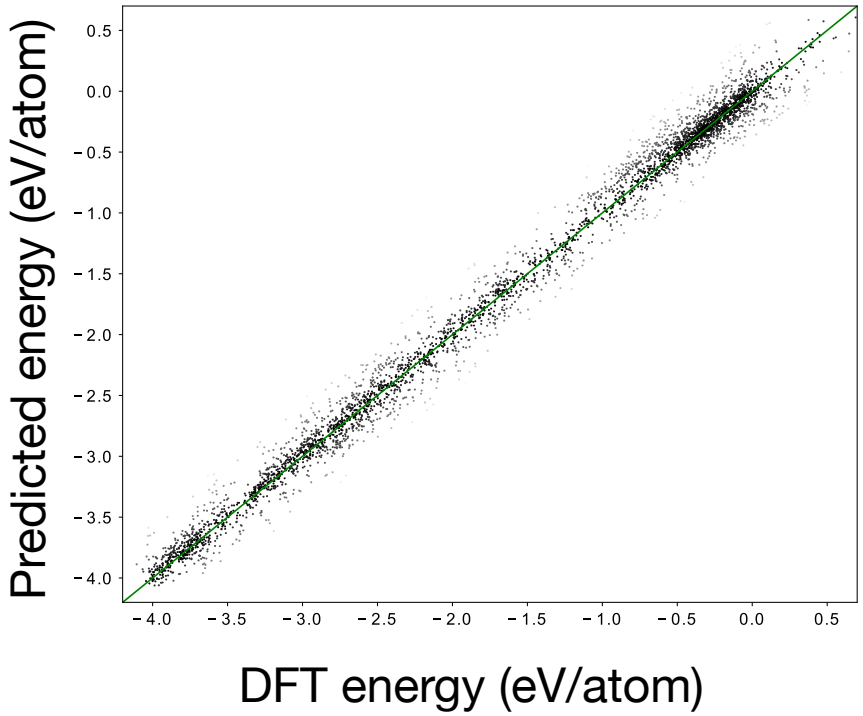


$$X^p = \sum_{k=1}^{n_p} \vec{O}p^T \times \vec{O}_k \longrightarrow X^p = \sum_{k=1}^{n_p} \vec{O}p^T \times \vec{O}_k \times w_k$$

Generalization

② Orbital Field Matrix (OFM)

Pham et al., STAM **18**, 756 (2017)



- Data: 4,220 *R-T-X* compounds
- DFT: VASP+GGA
- Machine learning: KRR + OFM

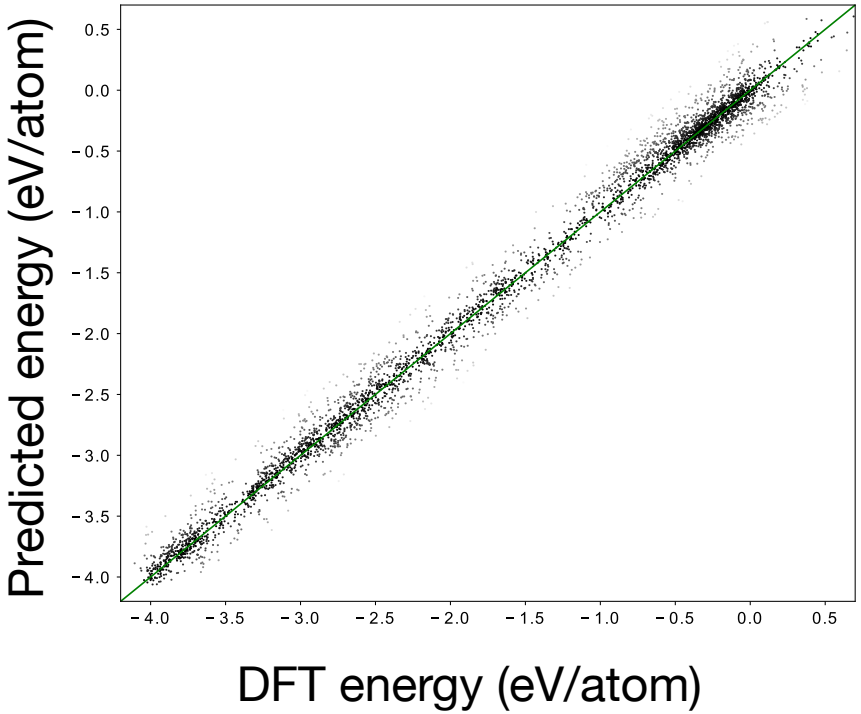
RMSE = 0.190 eV/atom

MAE = 0.112 eV/atom

R^2 = 0.98

Pham et al., STAM **18**, 756 (2017)

Formation energy



- Data: 4,220 *R-T-X* compounds
- DFT: VASP+GGA
- Machine learning: KRR + OFM

RMSE = 0.190 eV/atom

MAE = 0.112 eV/atom

R^2 = 0.98

OFM1 + Manhattan distance + Laplace kernel regressor

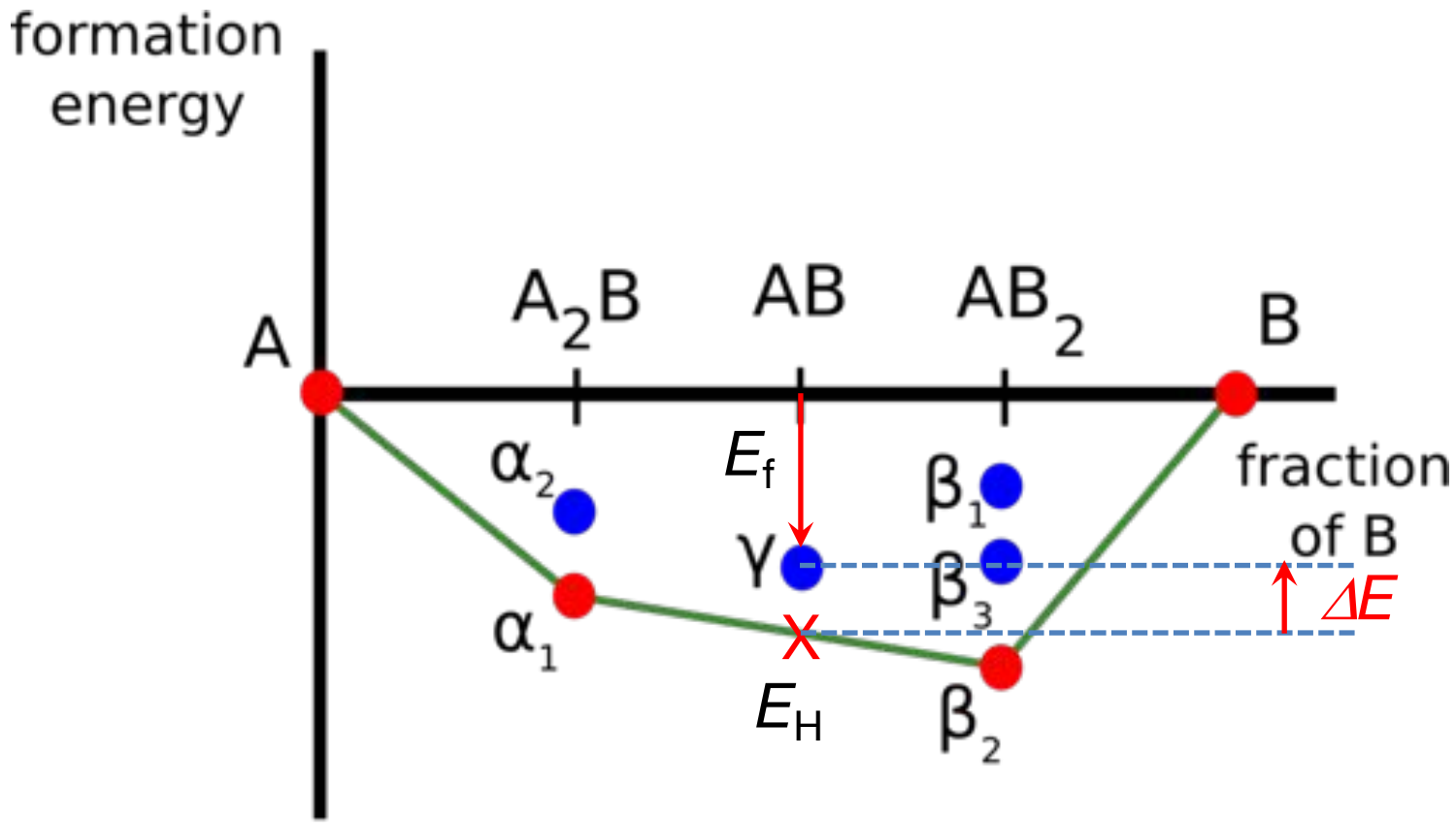
RMSE = 0.110 eV/atom

MAE = 0.067 eV/atom

R^2 = 0.994

Pham et al., JCP **148**, 204106 (2018)

③ Phase stability

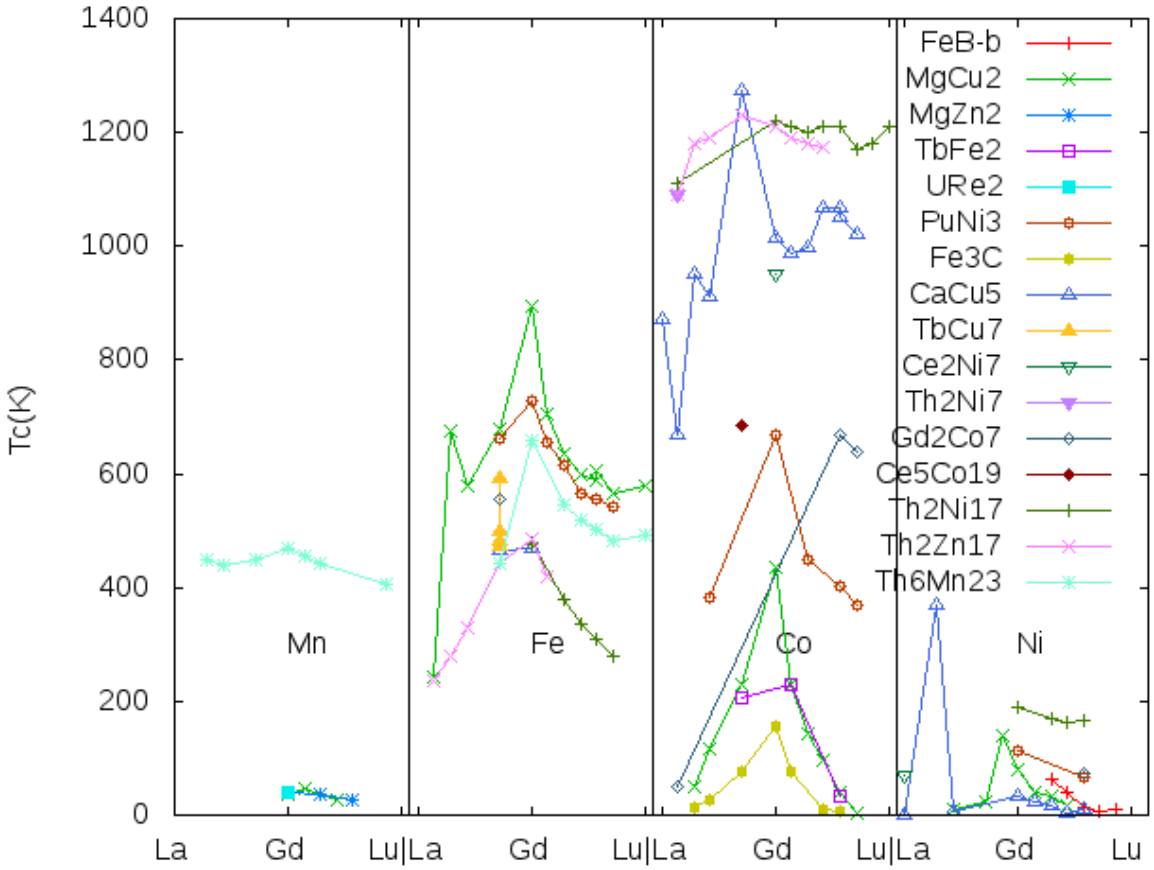


$\Delta E = E_f - E_H$

$\Delta E < 0$: Stable

https://materialsproject.org/wiki/images/6/6f/Convexhull.png

Curie temperature of *R*-*T* bimetals



Collect experimental T_C's of 101 *R*-*T* alloys from database (AtomWork)

H. Kino (NIMS)

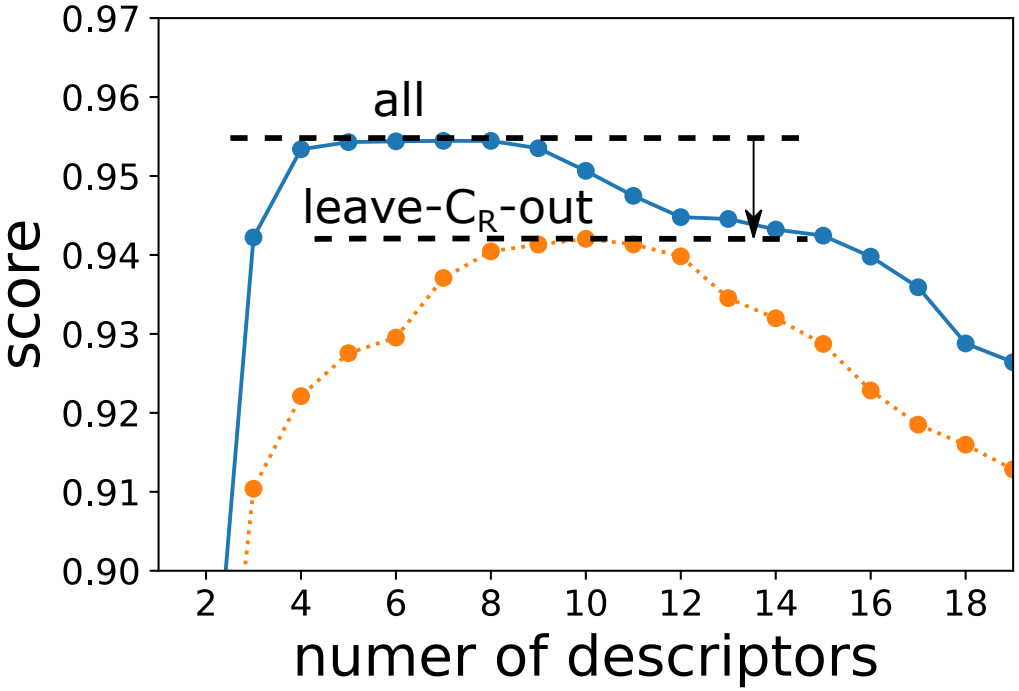
Descriptors

- ✓ Represent a compound by a vector $\boldsymbol{x}$
- ✓ 27 primary descriptors characterizing crystal constituent elements and structure

Category	Descriptors
Atomic properties of transition metals (T)	Z_T, r_T, r_T^{cv}, IP_T, χ_T, S_{3d}, L_{3d}, J_{3d}
Atomic properties of rare-earth metals (R)	Z_R, r_R, r_R^{cv}, IP_R, χ_R, S_{4f}, L_{4f}, J_{4f}, g_J, $J_{4f}g_J$, $J_{4f}(1-g_J)$
Structural information (S)	C_T, C_R, d_{T-T}, d_{T-R}, d_{R-R}, N_{T-R}, N_{R-R}, N_{R-T}

$$\boldsymbol{x} = (Z_T, r_T, \ldots, Z_R, r_R, \ldots, C_T, C_R, \ldots)$$

- ✓ Select a subset of 27 primary descriptors that maximizes prediction accuracy (Exhaustive search for 2^{27}-1 combinations)
- ✓ Gaussian kernel-ridge regression
- ✓ 10-fold cross validation

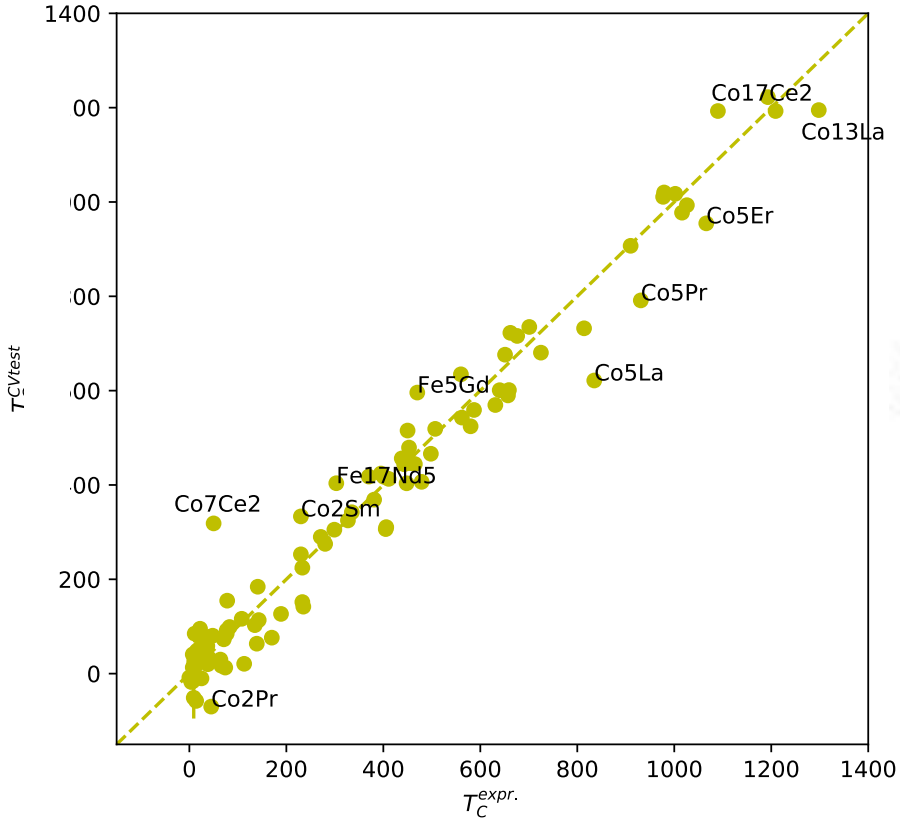


n	score	descriptor(s)
2	0.870153	C_R,Z_T
3	0.942222	C_R,Z_R,Z_T
4	0.953386	J_{3d},C_R,Z_R,Z_T
5	0.954294	L_{3d},J_{3d},C_R,Z_R,Z_T
6	0.954391	L_{3d},J_{3d},χ_T,C_R,Z_R,Z_T
7	0.954452	L_{3d},J_{3d},χ_T,C_R,Z_R,Z_T,r_T^{cv}
8	0.954448	L_{3d},J_{3d},χ_T,IP_T,C_R,Z_R,Z_T,r_T^{cv}

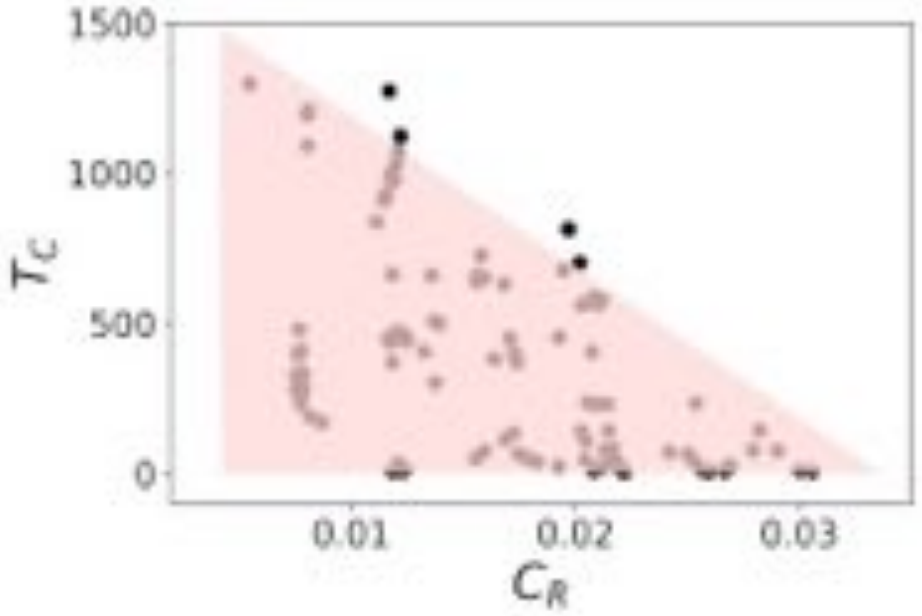
Dam et al., J. Phys. Soc. Jpn. (accepted)

T_C of *R*-*T* bimetals

Prediction



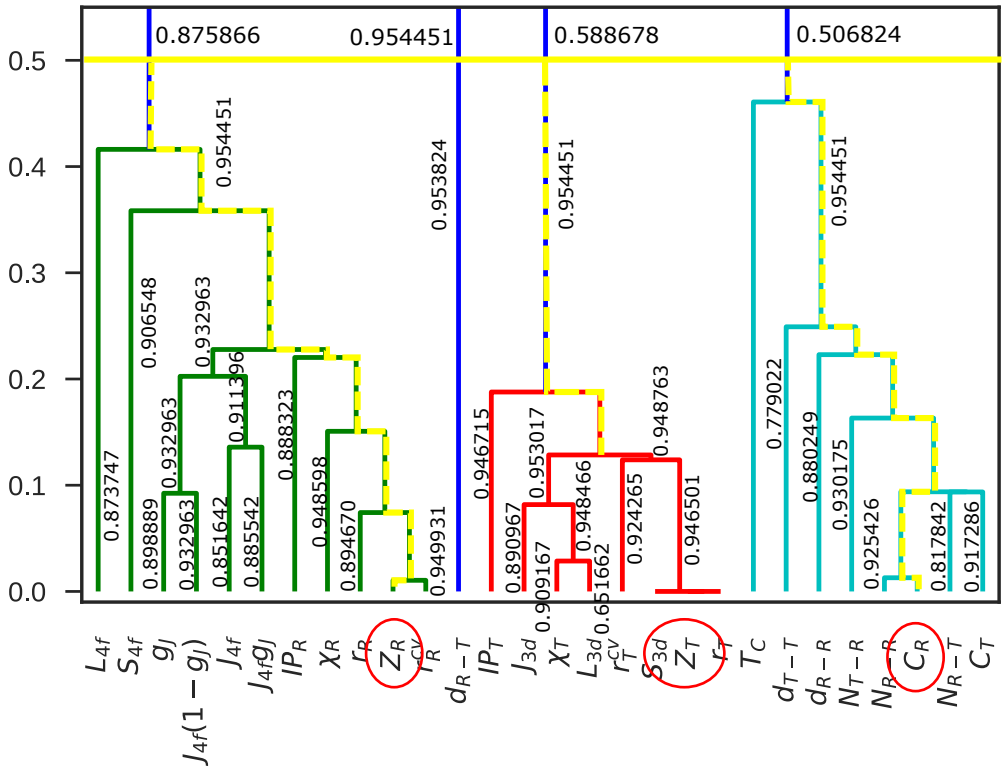
Understanding?



Subgroup relevance analysis

Dam et al., J. Phys. Soc. Jpn. (accepted)

$1 - |\rho|$



Pearson's correlation coefficient

$$\rho_{X,Y} = \frac{\mathrm{cov}(X,Y)}{\sigma_X \sigma_Y}$$

covariance

$$\mathrm{cov}(X,Y) = \langle X - \langle X \rangle \rangle \langle Y - \langle Y \rangle \rangle$$

Subgroup relevance analysis

Dam et al., J. Phys. Soc. Jpn. (accepted)

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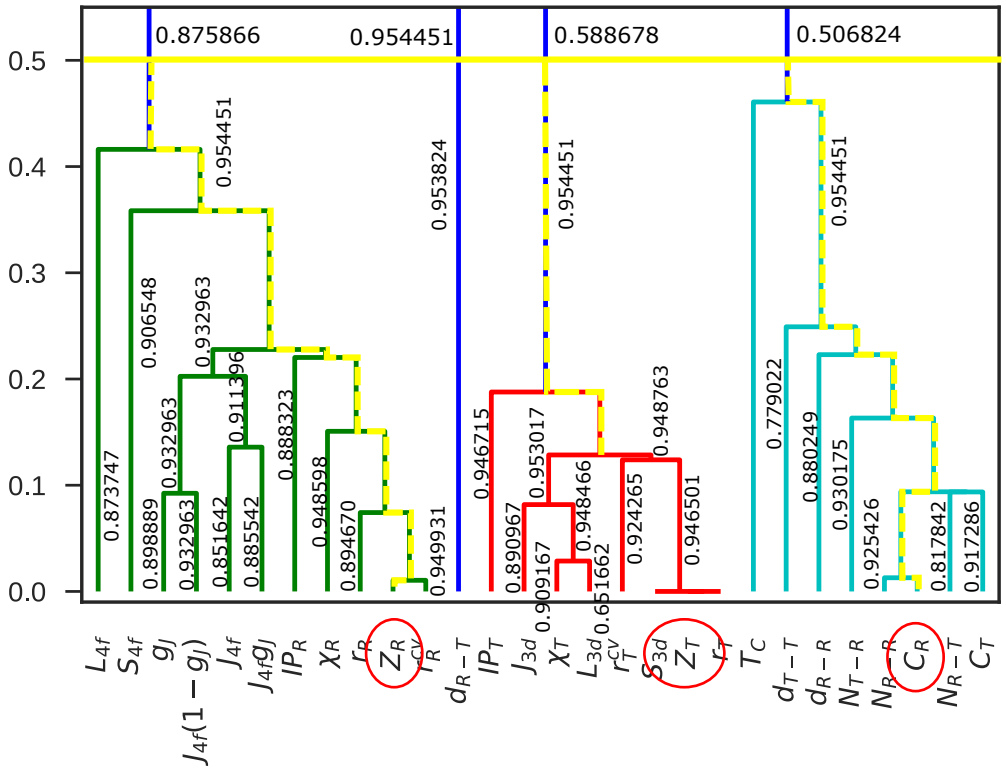

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Result by exhaustive search

Summary

- High-throughput screening by a combined computational and machine-learning techniques
- Virtual screening of Nd-Fe-B compounds using kernel-ridge regression and orbital-field-matrix
- Kernel-ridge regression of Curie temperature: important descriptors and descriptor groups by subgroup relevance analysis