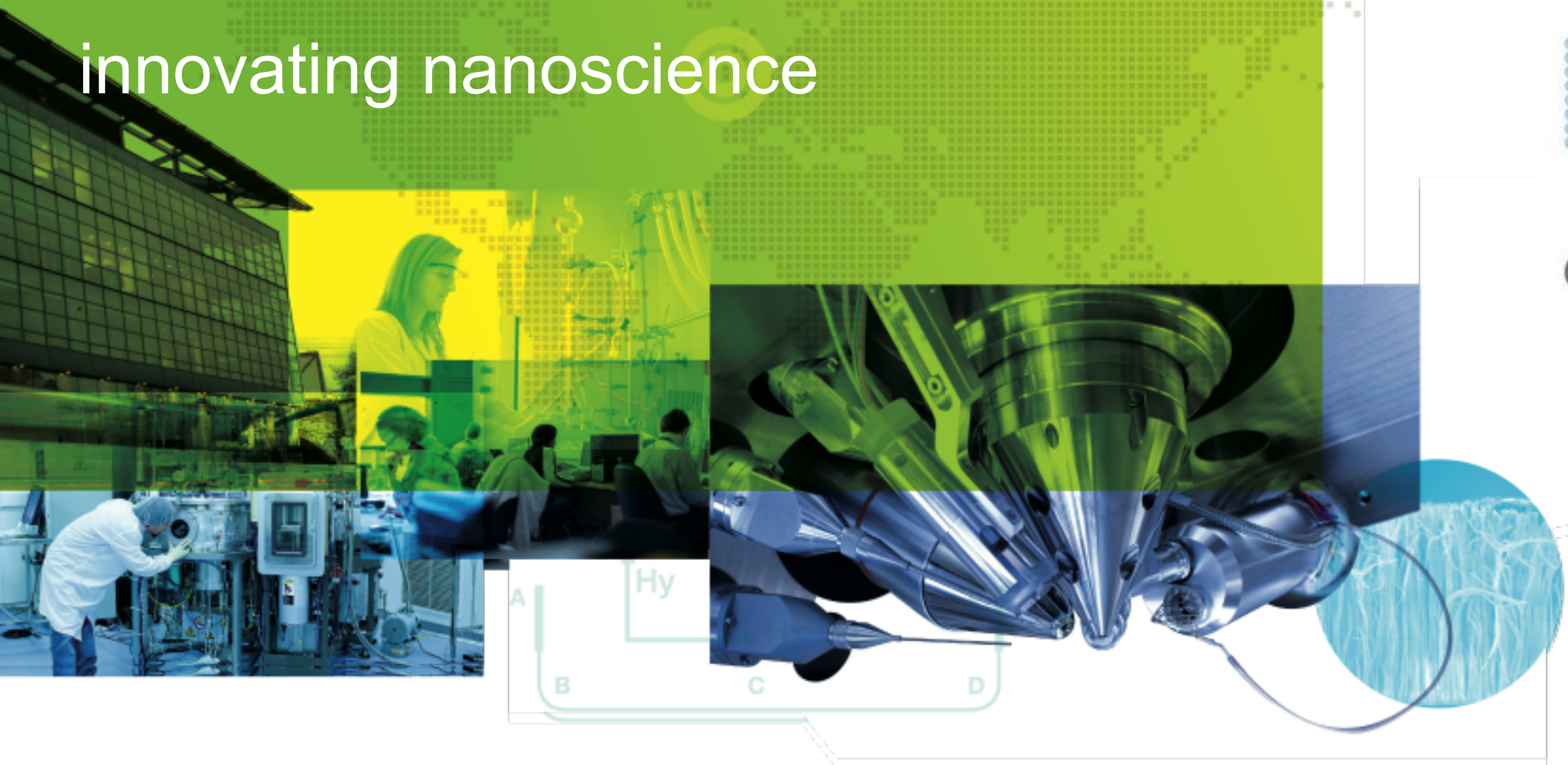
SPICE (Mainz) February 2023



CRANN

Machine learning as a tool to accelerate magnetic materials discovery

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Trinity College The University of Dublin

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COMPUTATIONAL SPINTRONICS

SANVITO RESEARCH GROUP
TRINITY COLLEGE, DUBLIN

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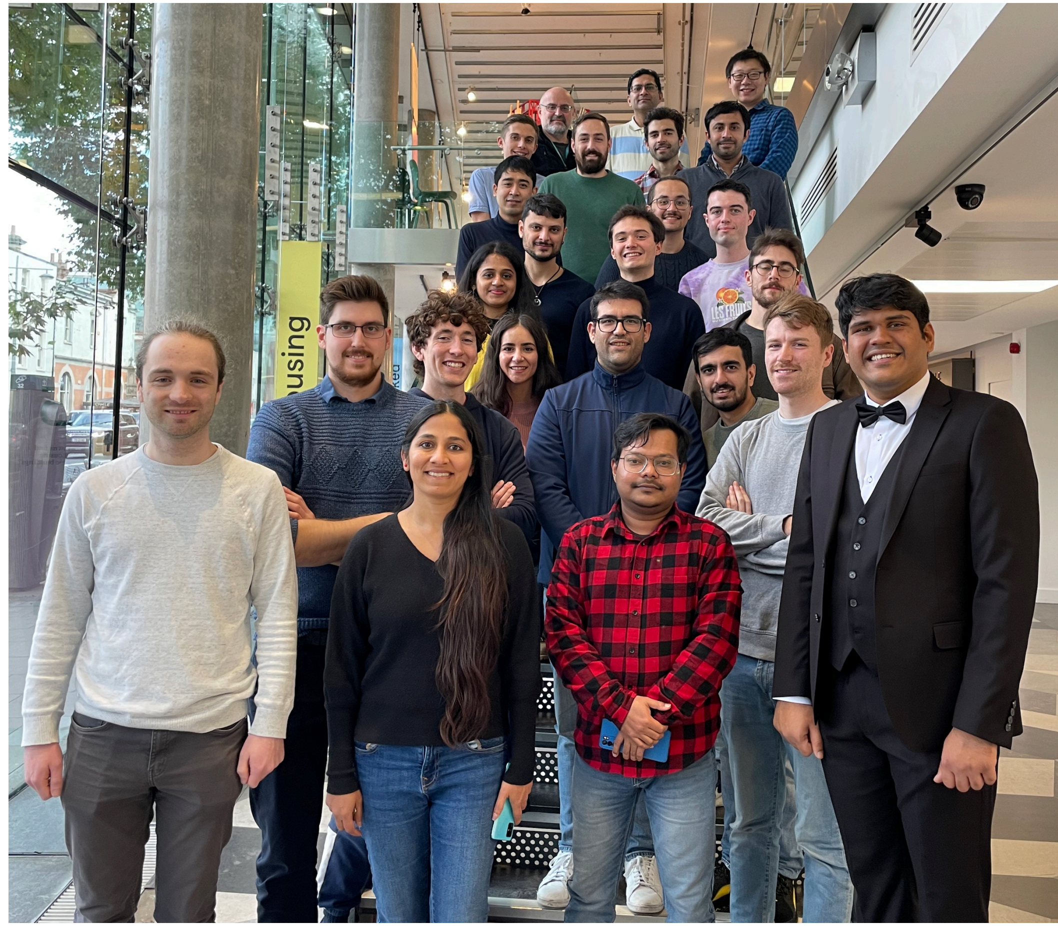
HORIZON 2020

KAUST
King Abdullah University of Science and Technology

SEVENTH FRAMEWORK PROGRAMME



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Unlocking human potential.

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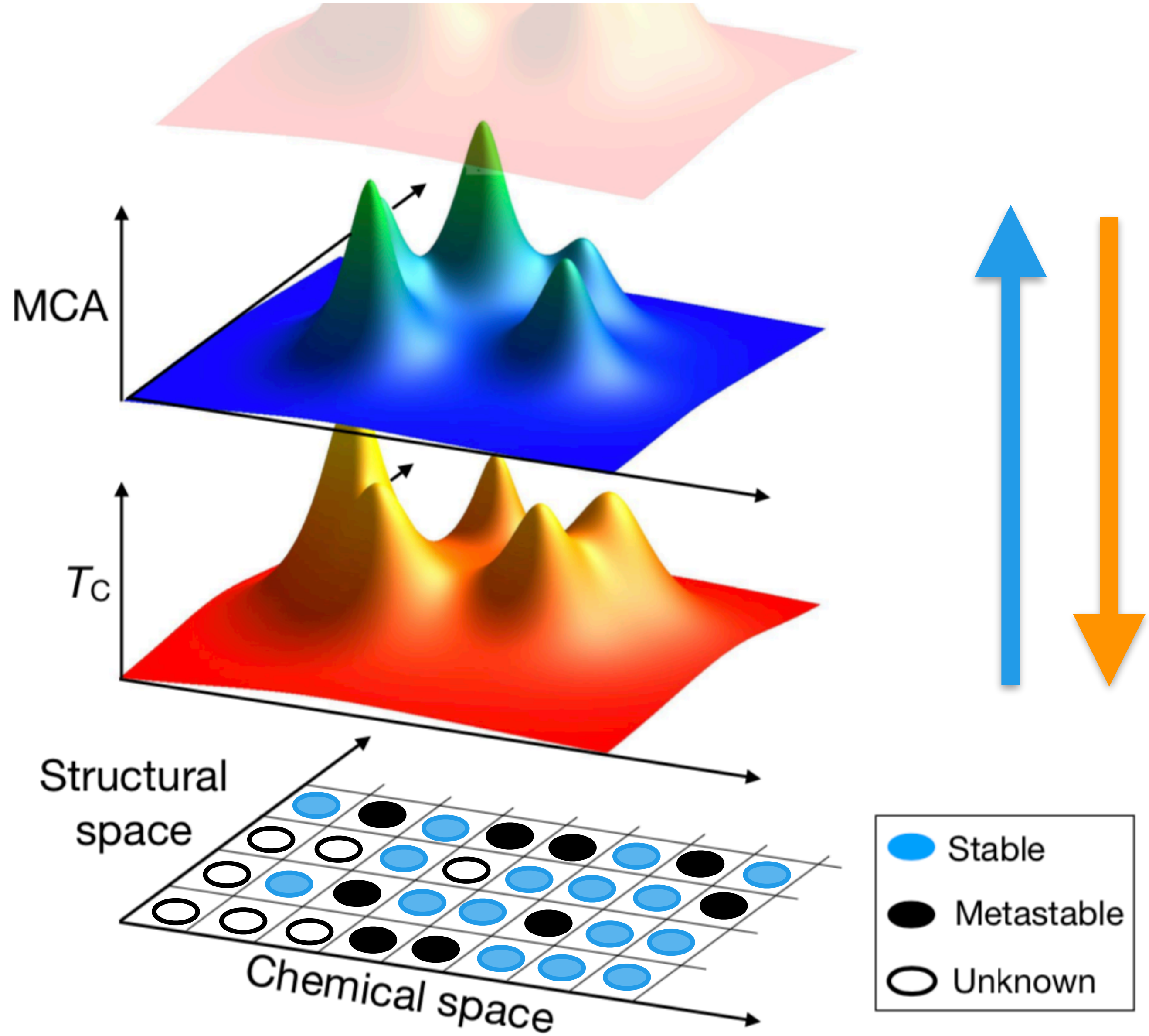
IRISH RESEARCH COUNCIL
An Chomhairle um Thaighde in Éirinn

Direct vs Inverse problem



The problems of making a roadmap

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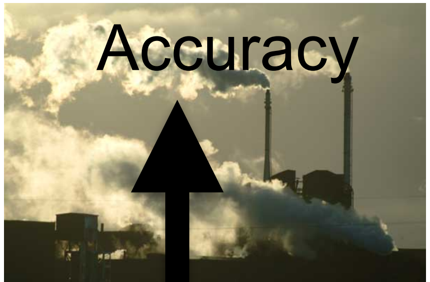
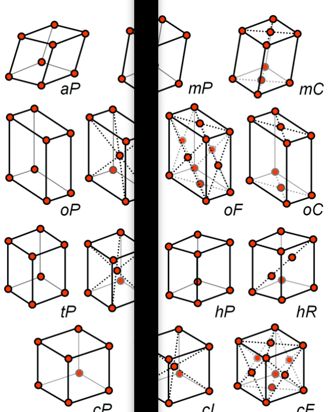




LAB

Environment

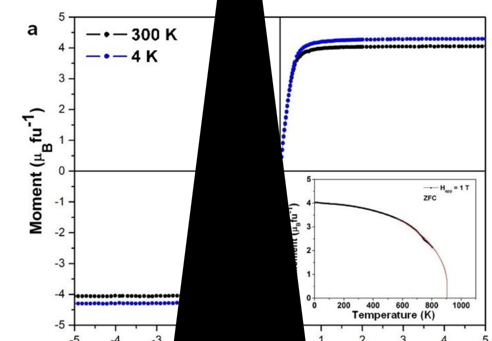
Properties

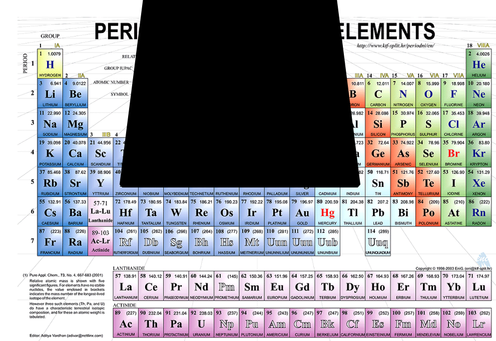
Explore structural space

and

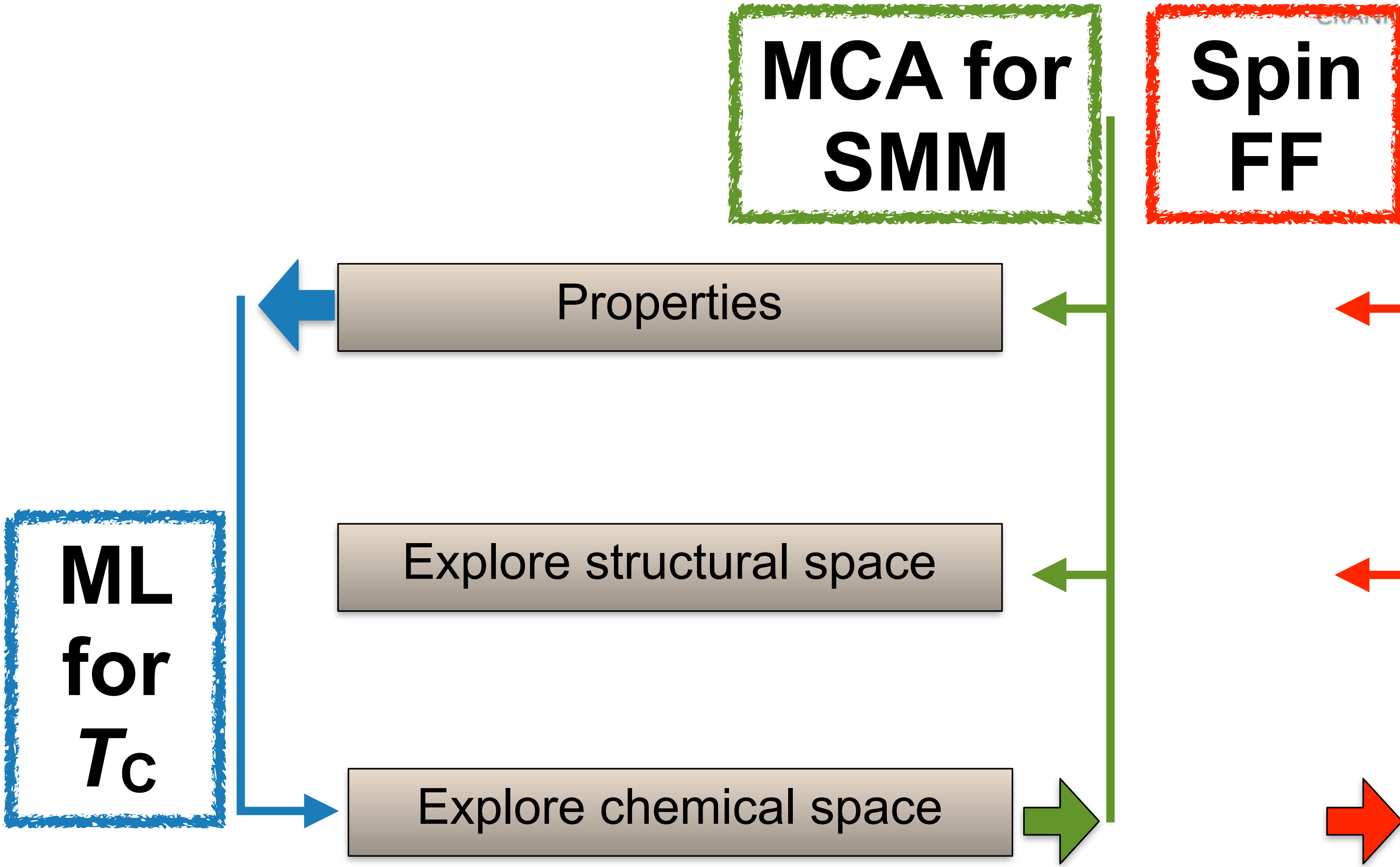
Explore chemical space

Materials





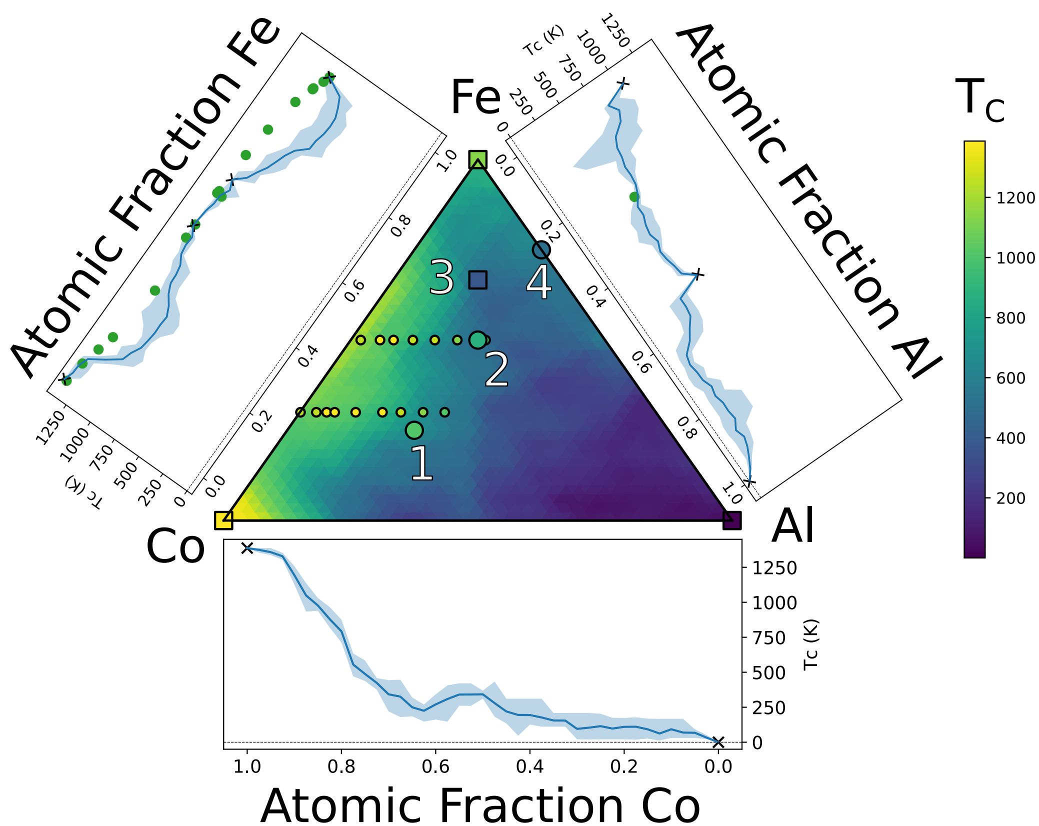
Outline

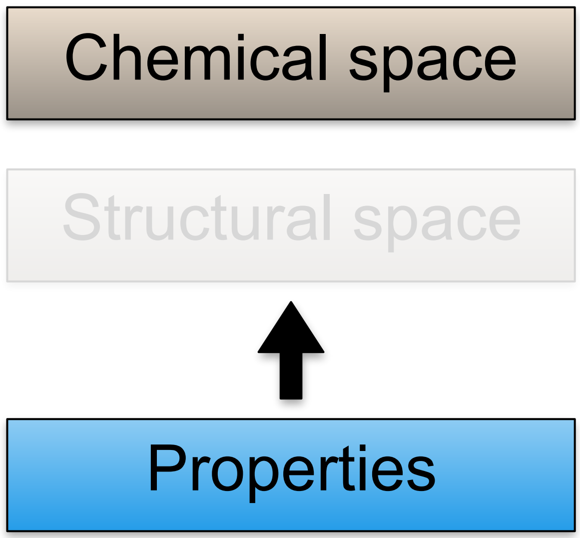

MCA for SMM
Spin FF
Properties
Explore structural space
ML for T_c
Explore chemical space

CRANN

James Nelson

Data to predictions: T_C of ferromagnets



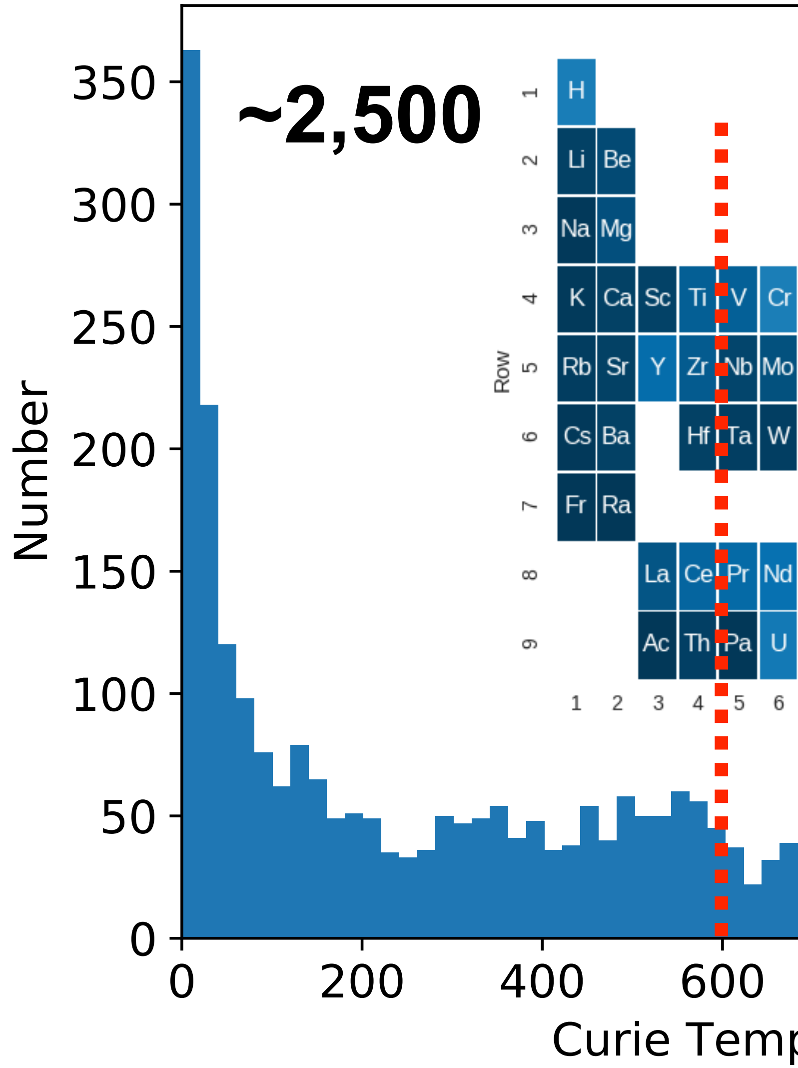


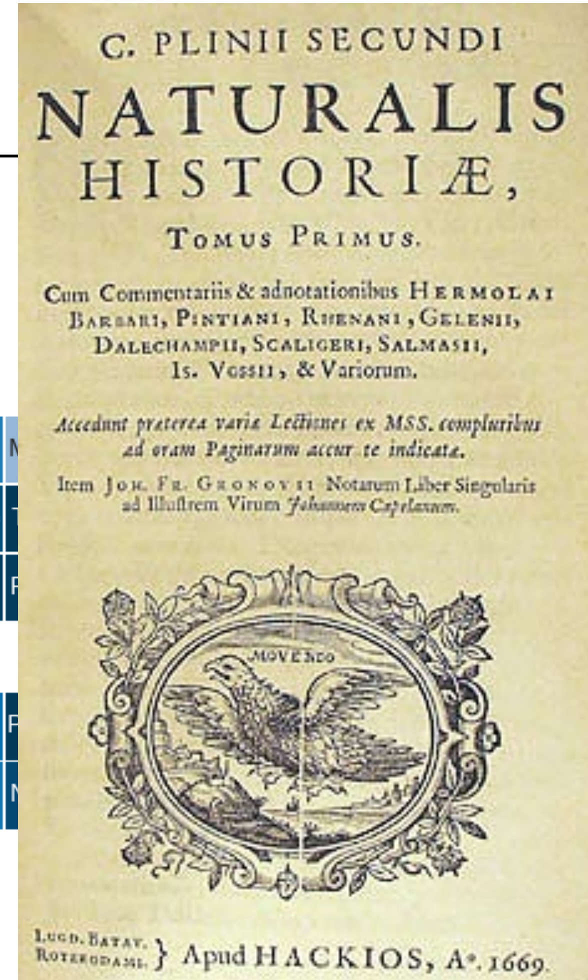
J. Nelson and SS, Phys. Rev. Mater **3**, 104405 (2019)

Example: T_C of ferromagnets

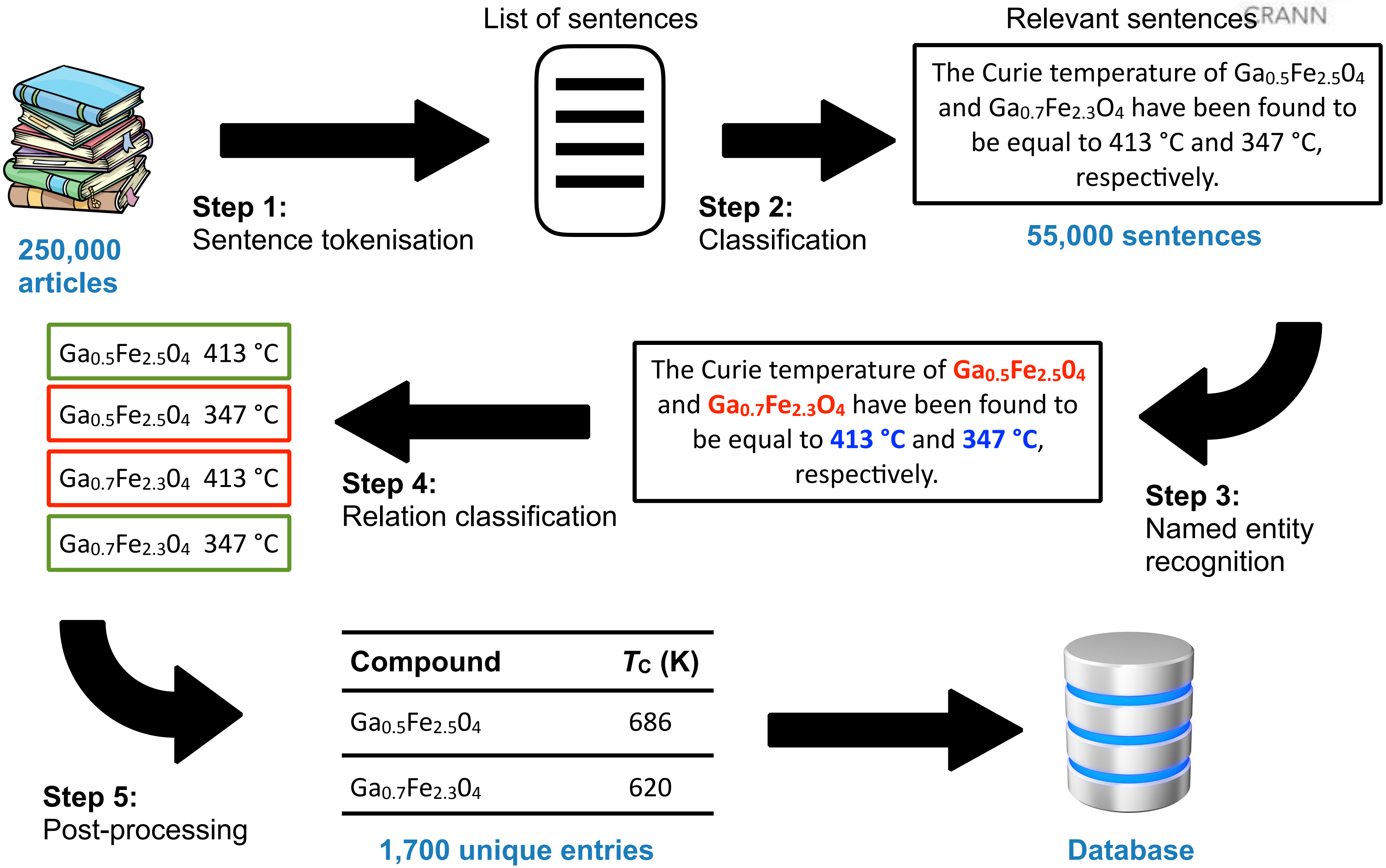
Goal: predict T_C of ferromagnets

Start from experimental data





Automated Extraction



Compound	T_C **(K)**
$Ga_{0.5}Fe_{2.5}O_4$	686
$Ga_{0.7}Fe_{2.3}O_4$	620

L.P.J. Gilligan *et al.*, arXiv: 2301.11689 (2023)

Example: T_C of ferromagnets

CRANN

Goal: predict T_C of ferromagnets

"Standard way"

Density functional theory → Mapping on Heisenberg model → Classical Monte Carlo → T_C

Approximation
Cell size
Broken symmetry
…

Non-magnons
Interaction range
….

Quantum effects
….

Often if we do not know the answer we cannot 'predict'

Example: T_C of ferromagnets

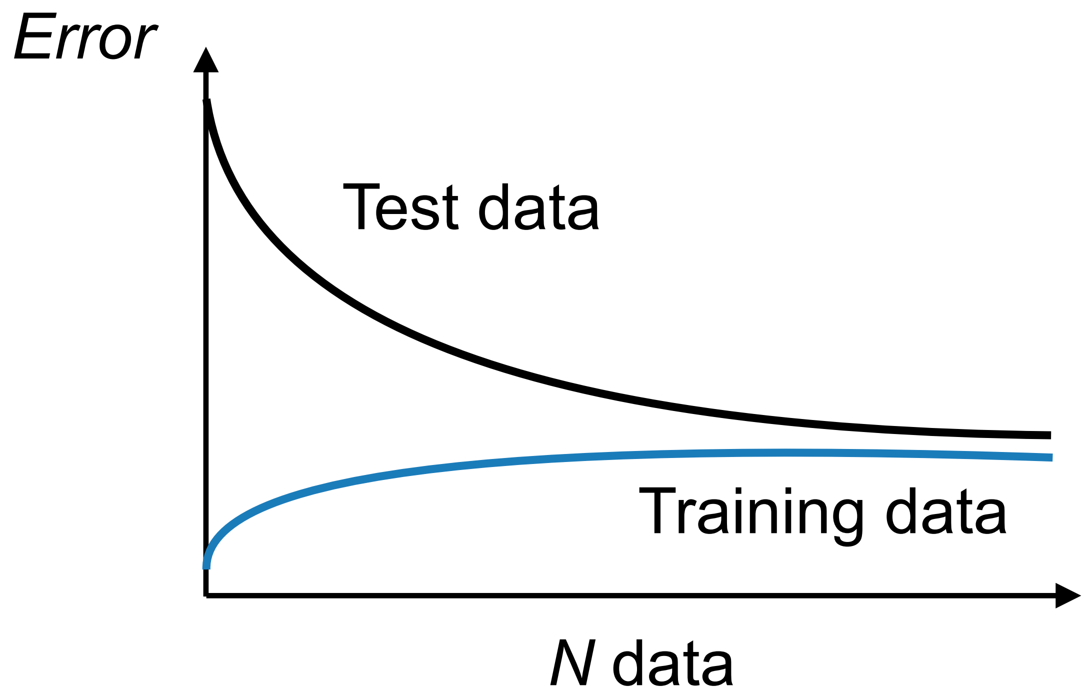
CRANN

O=8 Fe=26 ML algorithm

$Fe_3O_4 \Rightarrow (\ldots, 0, 4/7, 0, \ldots, 0, 3/7, 0\ldots) \Rightarrow T_C$

1818 Training

746 Test



Example: T_C of ferromagnets

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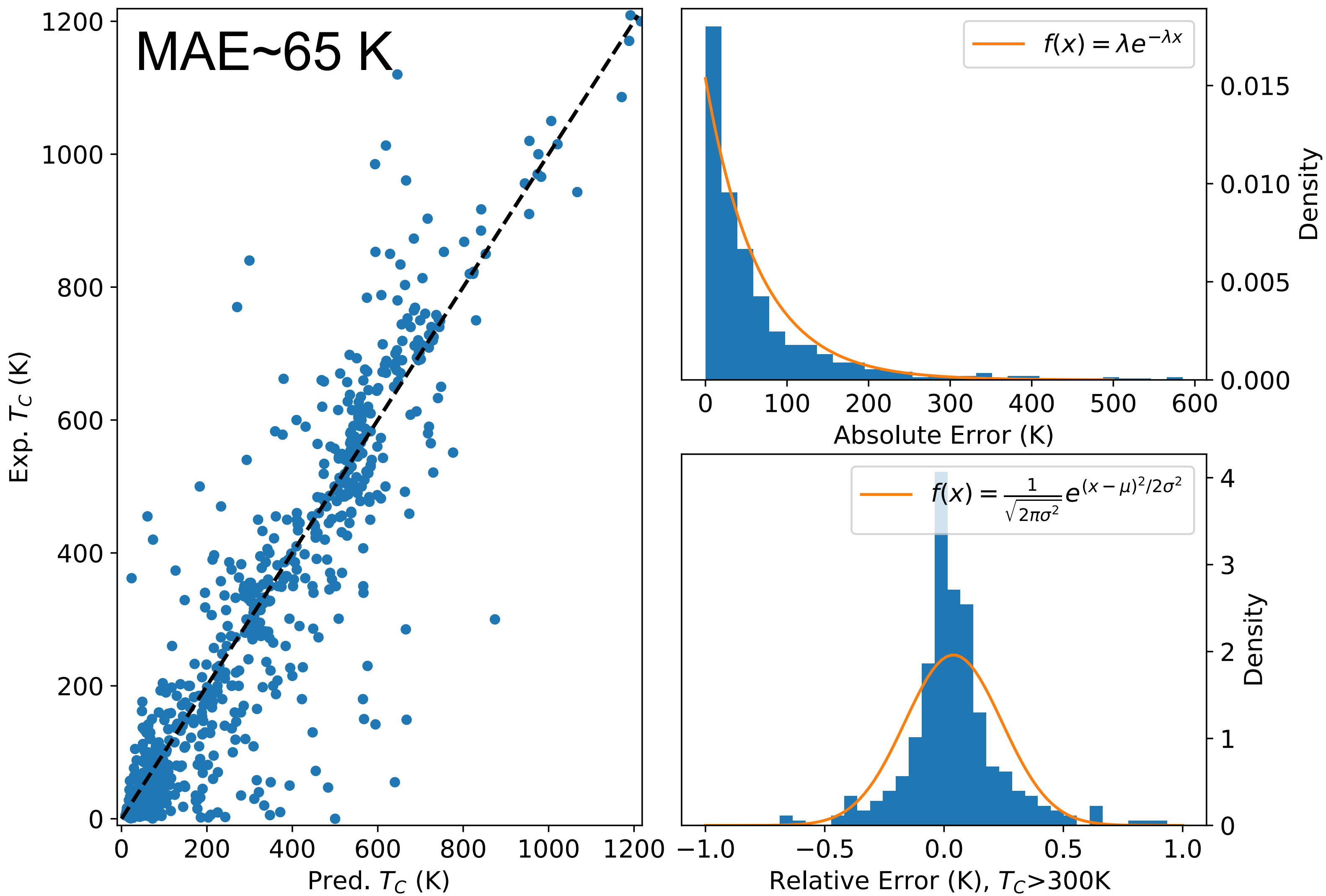
R^2	All	C10	C20	C40	C80	P10	P20	P40	P80
Ridge	0.50	0.48	0.50	0.51	0.50	0.27	0.30	0.32	0.37
KRR	0.70	0.72	0.76	0.74	0.71	0.70	0.71	0.70	0.69
NN	0.75	0.74	0.77	0.77	0.75	0.71	0.74	0.75	0.73
RF	0.83	0.78	0.80	0.80	0.81	0.73	0.75	0.74	0.73

Ridge = Ridge Regression
KRR = Kernel Ridge Reg.
NN = Neural Network
RF = Random Forest

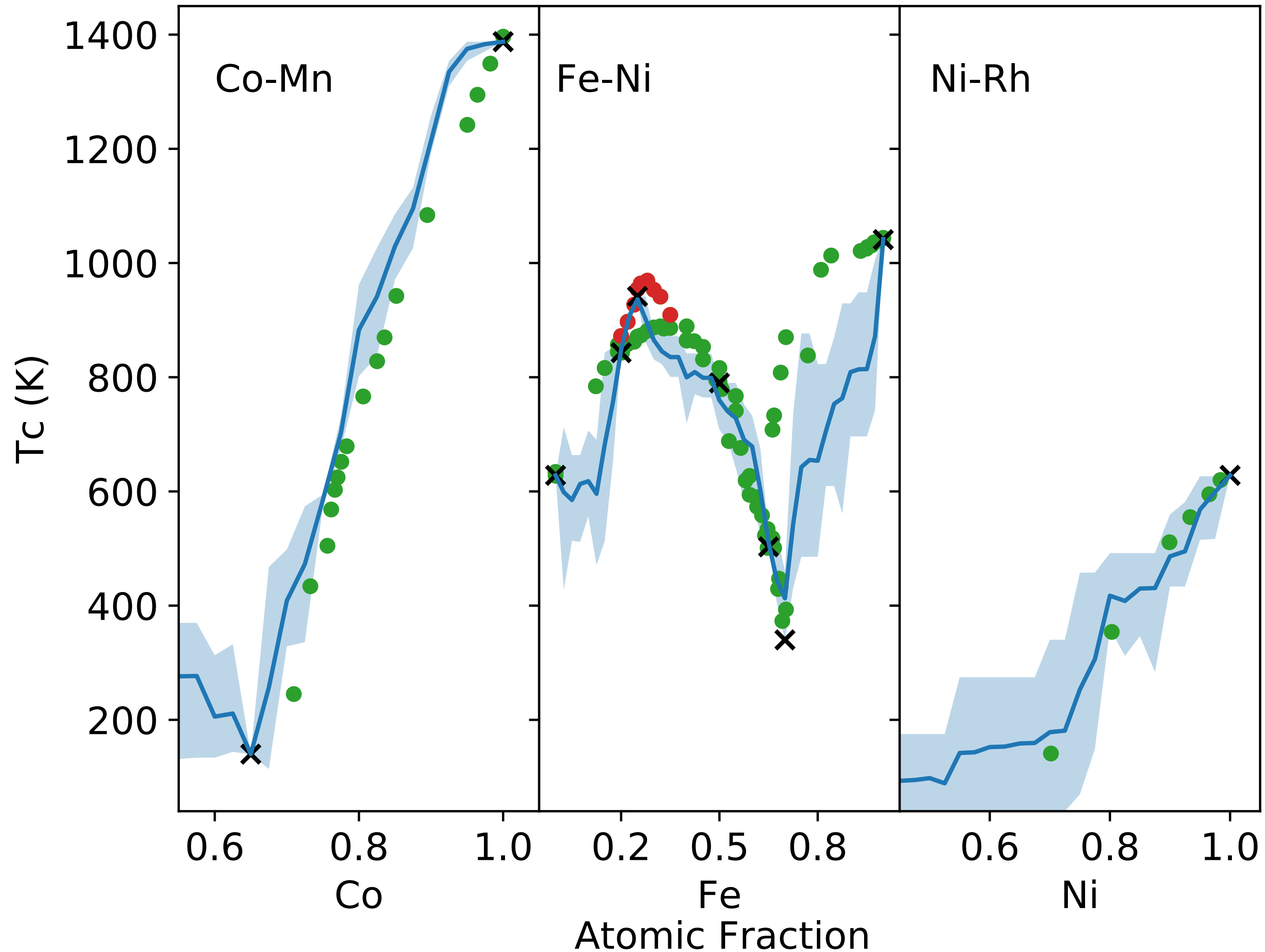
$$R^2 = 1 - \frac{\sum_i [y^{(i)} - f(\mathbf{x}^{(i)})]^2}{\sum_i [y^{(i)} - \mu]^2}$$

Example: T_C of ferromagnets



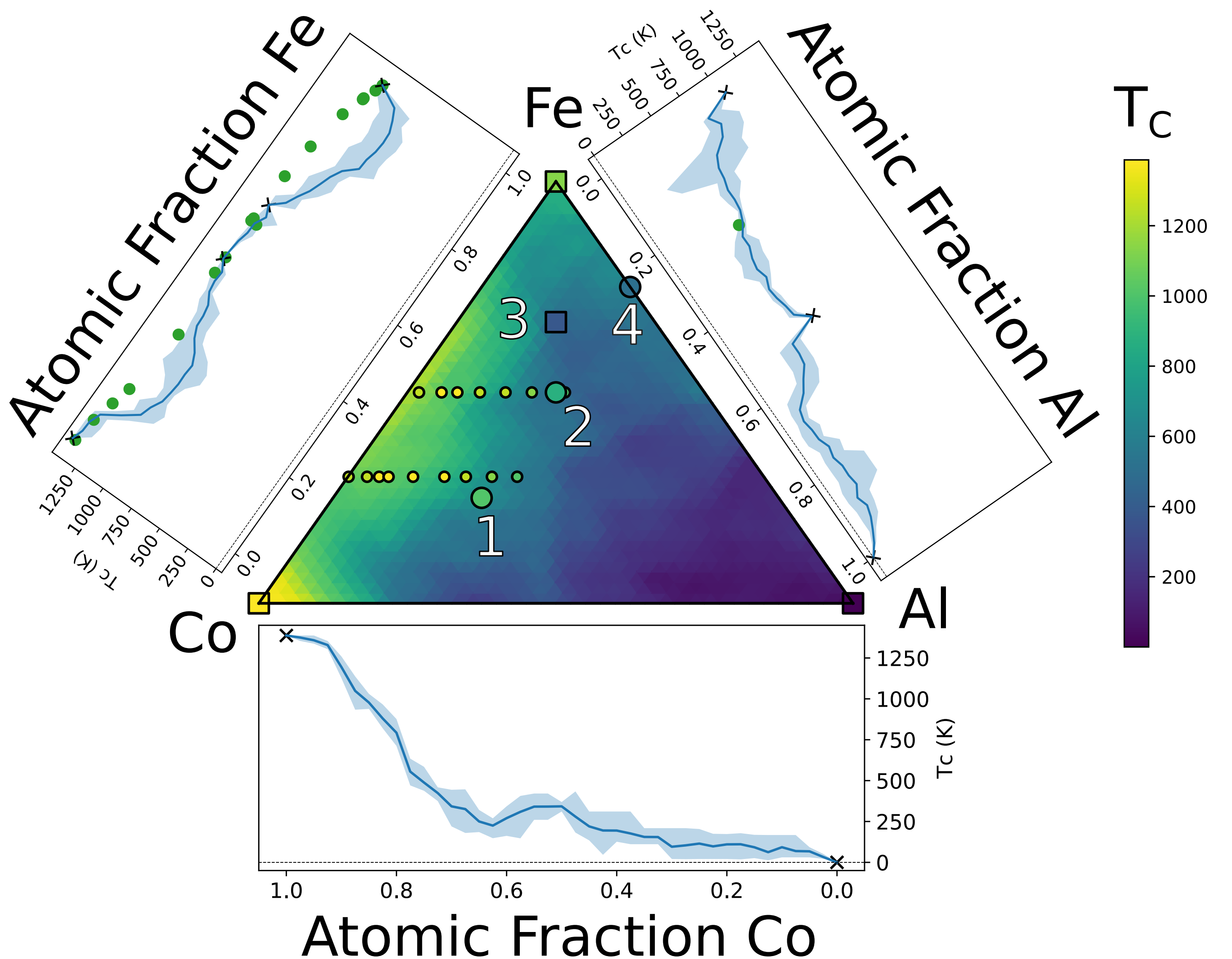
J. Nelson and S. Sanvito, Phys. Rev. Mater **3**, 104405 (2019)

Example: T_C of ferromagnets



J. Nelson and S. Sanvito, Phys. Rev. Mater **3**, 104405 (2019)

Example: T_C of ferromagnets



J. Nelson and S. Sanvito, Phys. Rev. Mater **3**, 104405 (2019)

What we have learnt ?

Data can be used to construct models !

Useful for an initial exploration

Extremely high throughput

Only experimental data are needed

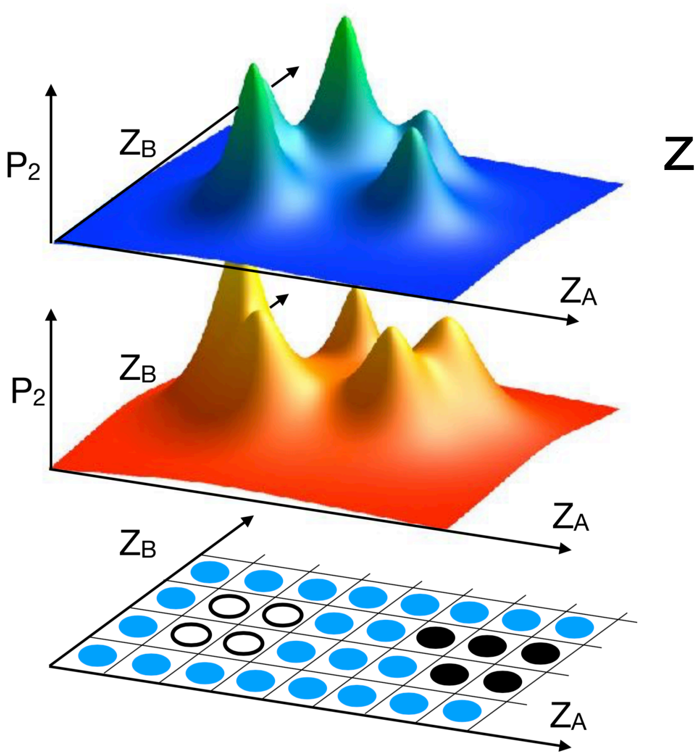
Need to harvest more data: automation possible now!!

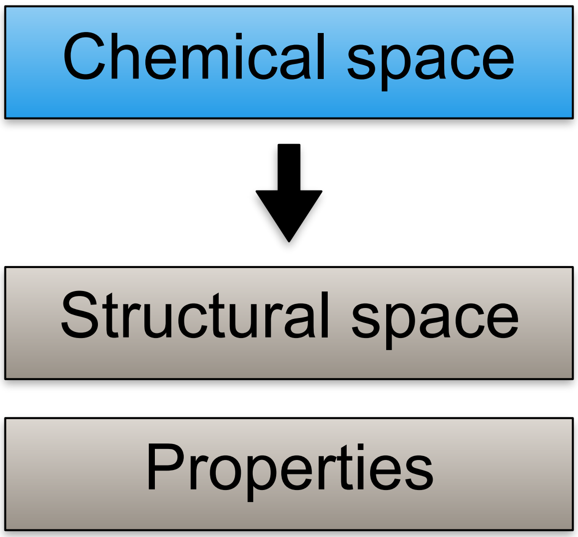
Structure difficult to introduce

J. Nelson and S. Sanvito, Phys. Rev. Mater **3**, 104405 (2019)

Alessandro Lunghi, Yanhui Zhang, Michelangelo Domina

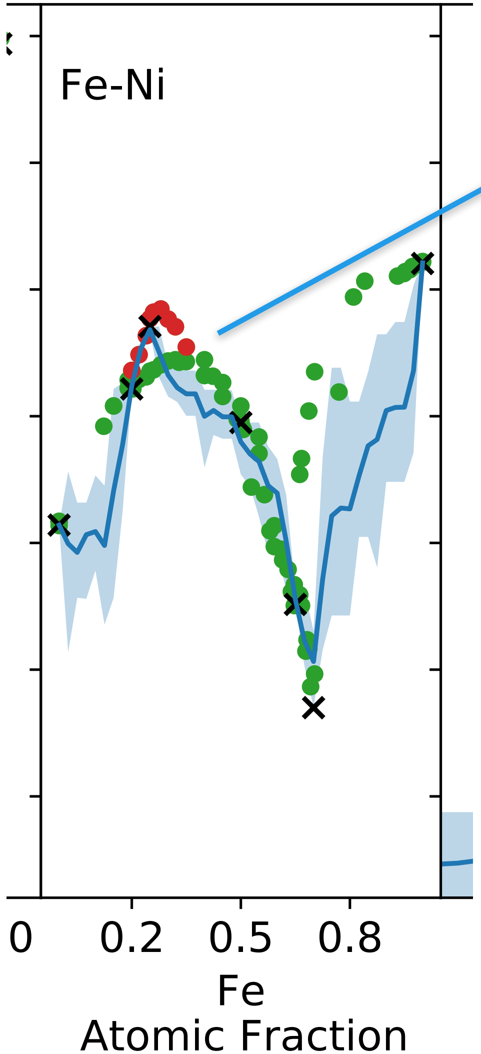
Continuous properties representation





A. Lunghi and SS, J. Chem. Phys. C **124**, 5802 (2019)

Representing atomic distributions

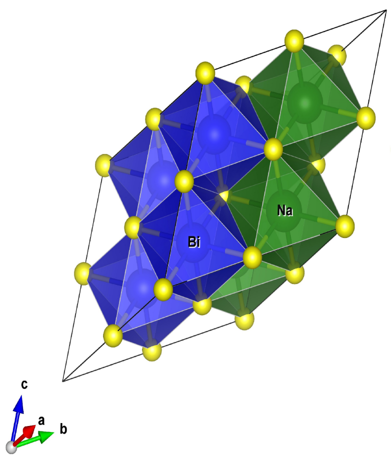


We need to distinguish different structures

This is not trivial:

Space groups ?

No. 225



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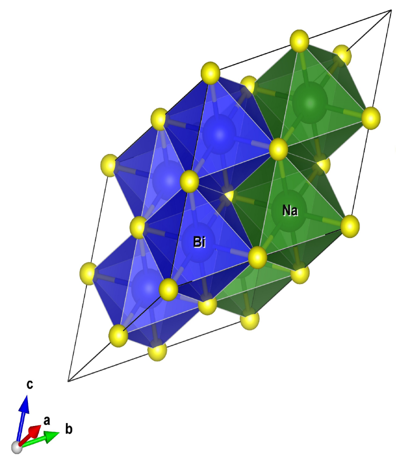
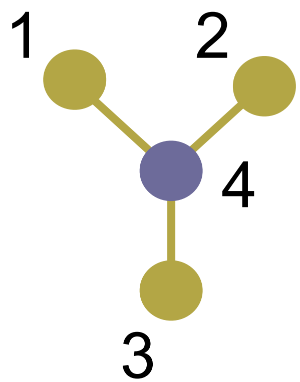


Representing atomic distributions

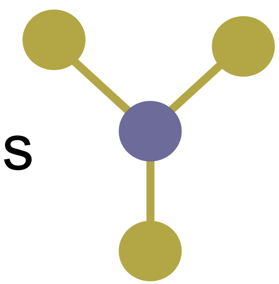


Cartesian coordinates

$$\vec{v} = (\vec{x_1}, \vec{x_2}, \vec{x_3}, \vec{x_4})$$

$$\vec{v} + \vec{w}$$

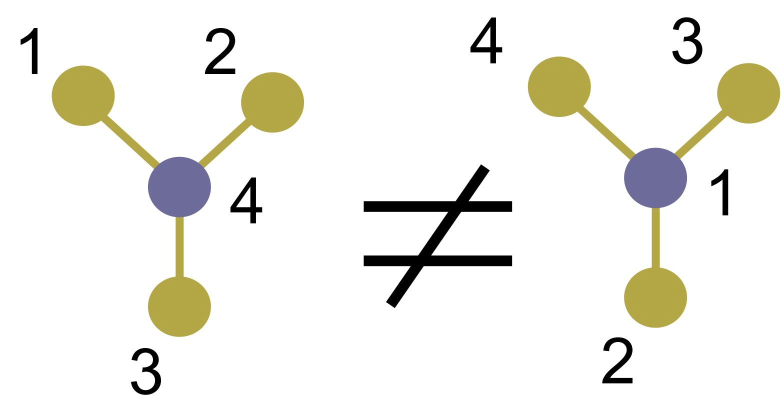
Invariant by translations



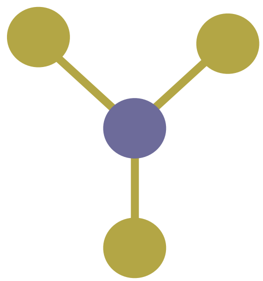
Locality fixes the problem

$$Q_{\mathrm{local}}(\mathbf{r}^N) = \sum_{i=1}^{N} Q_i(\mathbf{r}'_i)$$

Locality fixes also invariance by permutation



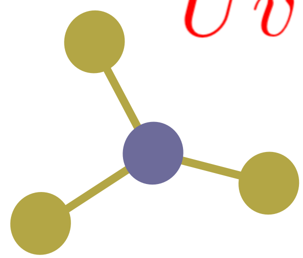
Representing atomic distributions



$\vec{v}$

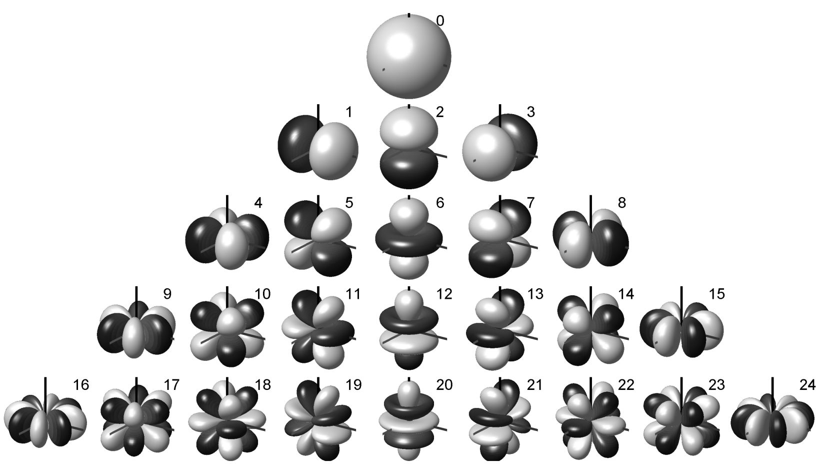
$U\vec{v}$

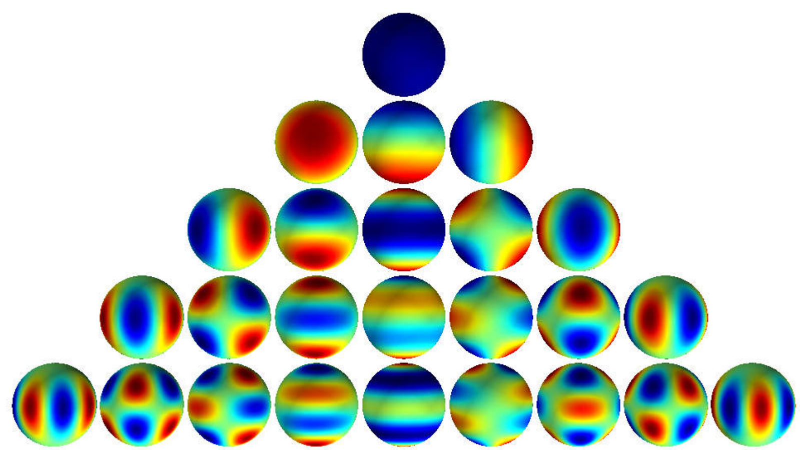
Invariant by rotations



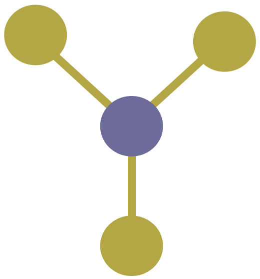
Needs to be built in

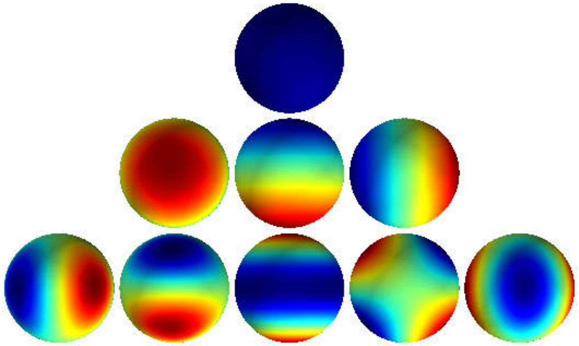
Usually spherical harmonics expansion





Representing atomic distributions





$$\rho(\mathbf{r}) = \sum_{\mathbf{i}} \alpha_{\mathbf{i}}\delta(\mathbf{r} - \mathbf{r}_{\mathbf{i}})$$

$$|\rho\rangle = \sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\sum_{m=-l}^{l} c_{nlm}\,|nlm\rangle$$

This is a rotational invariant (*power spectrum*)

$$p_{nl} = \sum_{m=-l}^{l} |c_{nlm}|^2$$

Need continuous representation

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Spectral Neighbour Analysis Potential (SNAP)

Clever representation of the local chemical environment

$$\rho_i(\mathbf{r}) = \delta(\mathbf{r}) + \sum_{r_{ii'} < R_{cut}} f_c(r_{ii'}) w_{i'} \delta(\mathbf{r} - \mathbf{r}_{ii'})$$

$$\rho(\mathbf{r}) = \sum_{j=0,\frac{1}{2},\ldots}^{\infty} \sum_{m=-j}^{j} \sum_{m'=-j}^{j} u^j_{m,m'} U^j_{m,m'}(\theta_0, \theta, \phi)$$

Rotationally invariant *descriptors*

$$B_{j_1,j_2,j} = \sum_{m_1,m_1'=-j_1}^{j_1} \sum_{m_2,m_2'=-j_2}^{j_2} \sum_{m,m'=-j}^{j} (u^j_{m,m'})^* H^{jmm'}_{\substack{j_1 m_1 m_1' \\ j_2 m_2 m_2'}} u^{j_1}_{m_1,m_1'} u^{j_2}_{m_2,m_2'}$$

Total energy linear in the *descriptors*

$$\left.\begin{aligned} E(\mathbf{r}^N) &= E_{ref}(\mathbf{r}^N) + E_{local}(\mathbf{r}^N) \\ E_{local}(\mathbf{r}^N) &= \sum_{i=1}^{N} E_i(\mathbf{q}_i) \end{aligned}\right\}$$

$$\boxed{E^i_{SNAP}(\mathbf{B}^i) = \beta_0^{\alpha_i} + \sum_{k=1}^{K} \beta_k^{\alpha_i} B_k^i = \beta_0^{\alpha_i} + \boldsymbol{\beta}^{\alpha_i} \cdot \mathbf{B}^i}$$

PRL **104**, 136403 (2010); J. Comp. Phys. **285**, 316 (2015)

Total energy: $[Co(pdms)_2]^{2-}$

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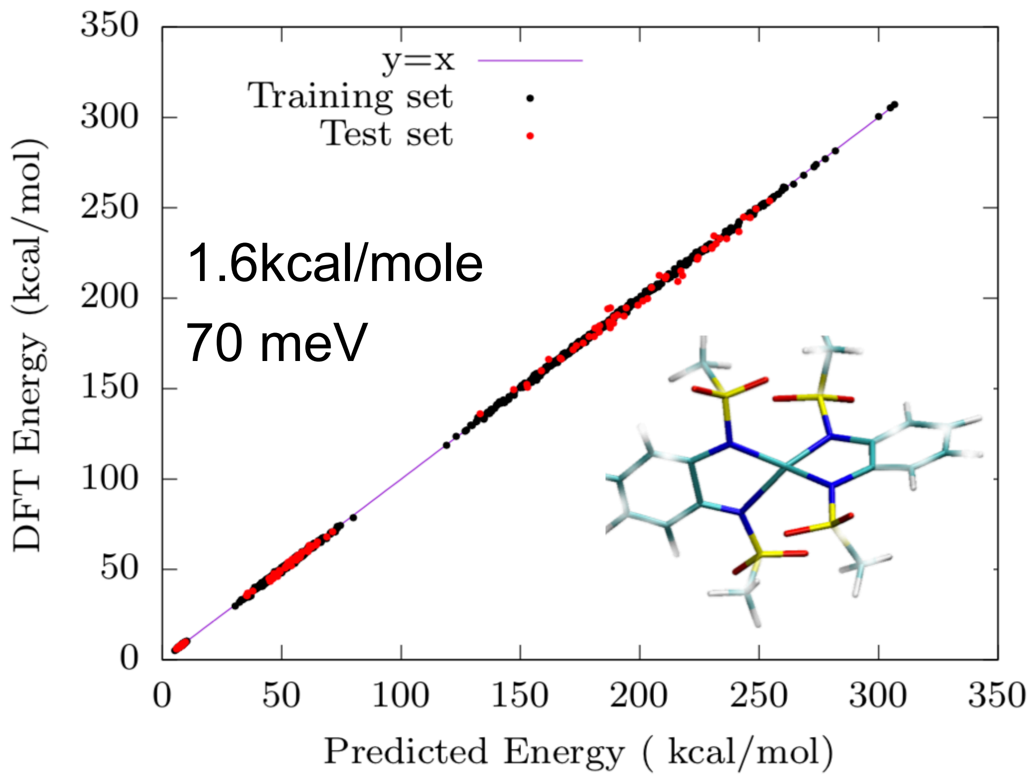
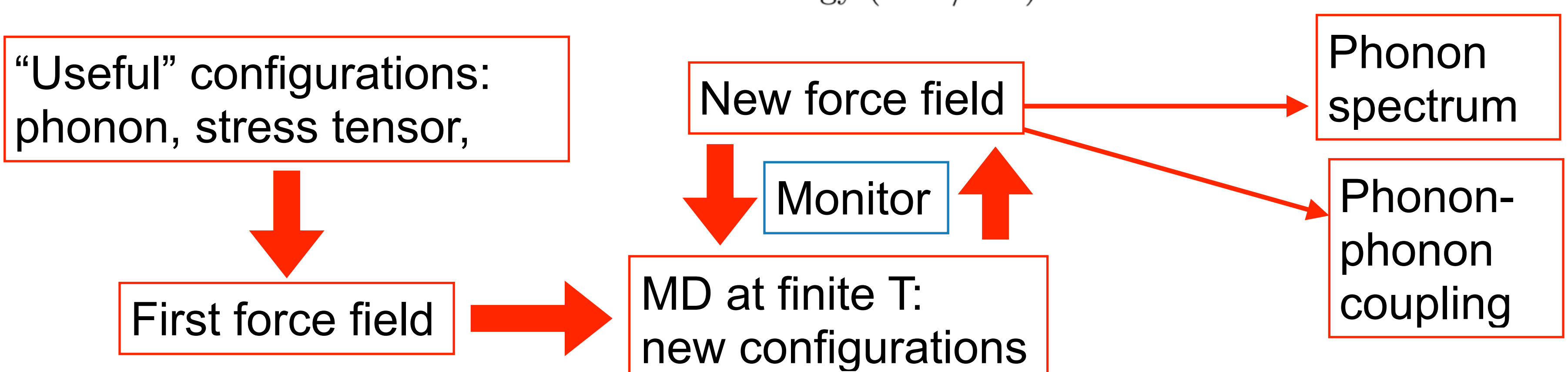
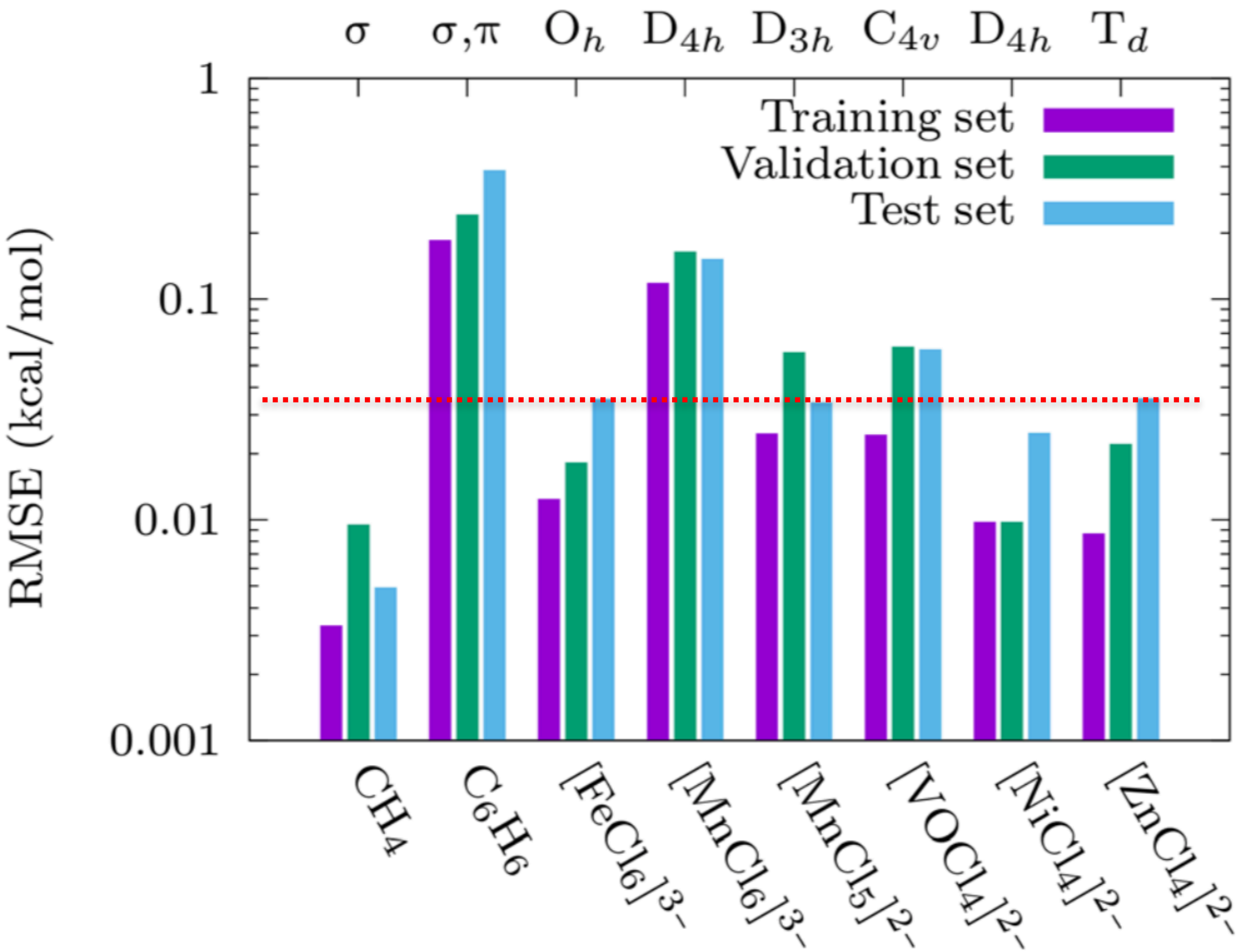




Total energy: agnostic of the bond nature

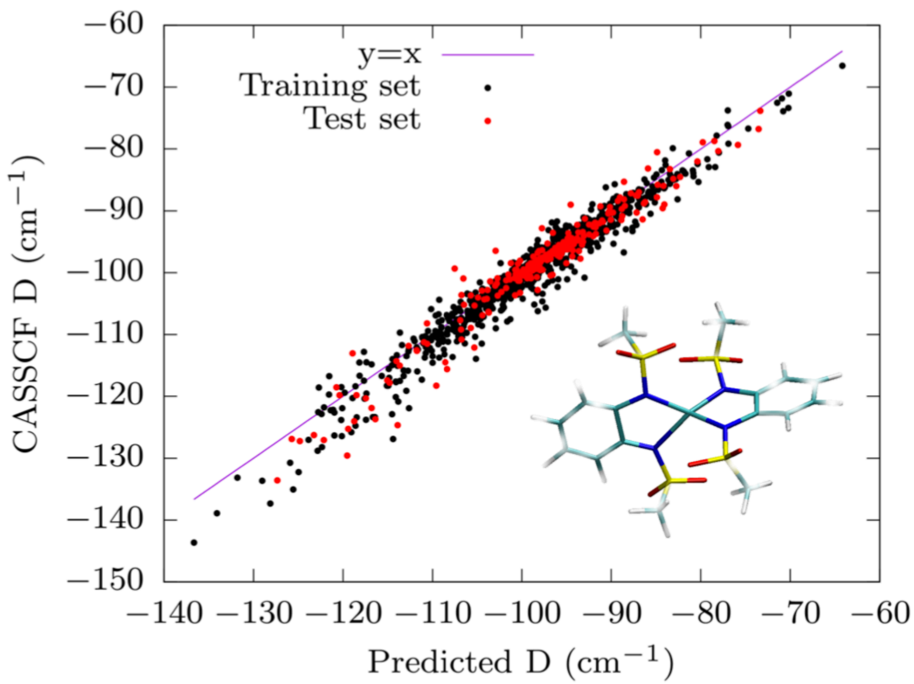


1kcal/mol = 40meV

A. Lunghi and SS, Science Adv. **5**, eaaw2210 (2019)

Not just for total energy

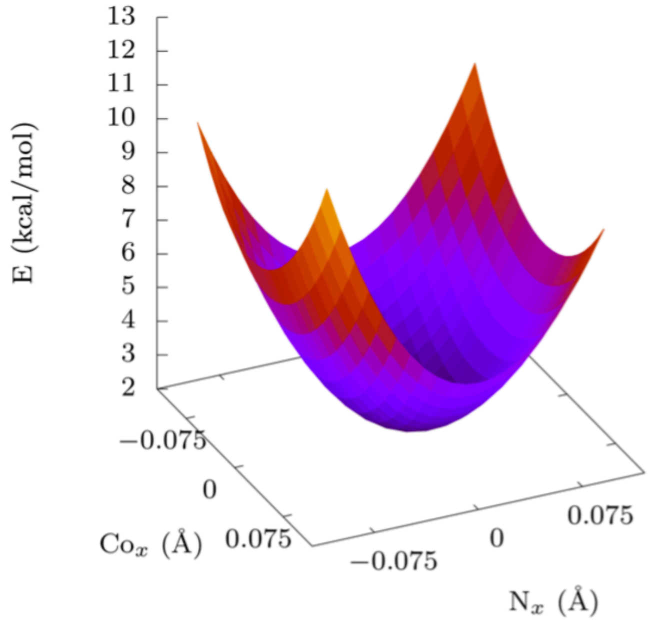
$$\hat{H}_S = D\hat{S}_z$$

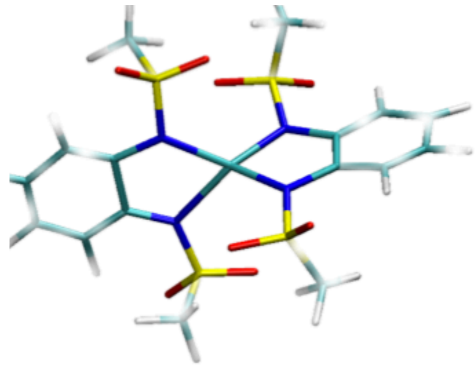


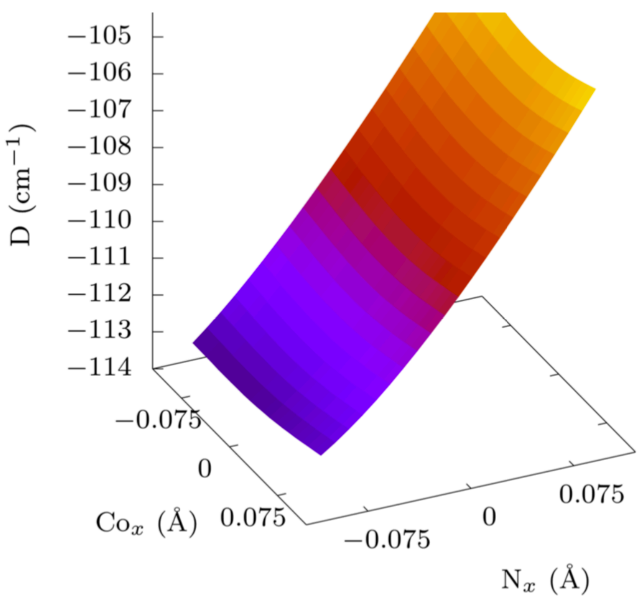
2.2 cm^{-1}

A. Lunghi and SS, Science Adv. **5**, eaaw2210 (2019); J. Chem. Phys. C **124**, 5802 (2019).

You can search in continuous surfaces!







5% reduction of: $d_{Co\text{-}N}$
N-Co-N

50% increase of *D*

$$H(q_\alpha) = E(q_\alpha) + \lambda D(q_\alpha)$$

What is this useful for?

CRANN

Very accurate atomic models possible

Useful for finite temperature

Mainly energy but other quantities possible

Only moderately large training set needed

Much better for molecules than solids

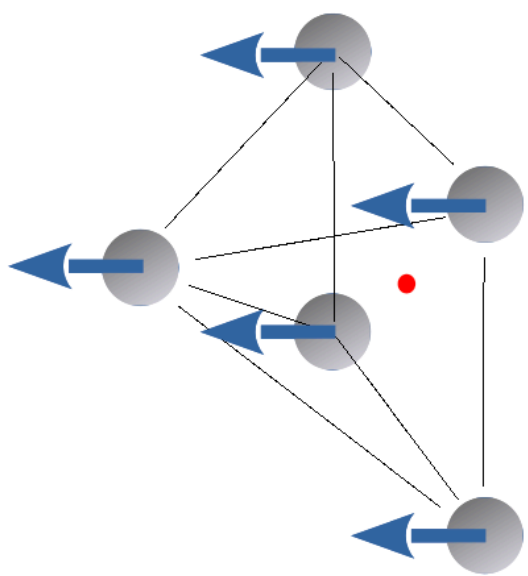
High risk of overfitting

Not really interpretable

Michelangelo Domina, Matteo Cobelli

CRANN

ML potential for vector fields



Chemical space

↓

Structural space

Properties

M. Domina, M. Cobelli and SS, Phys. Rev. B **105**, 214439 (2022)

Extension to spin

The idea is to construct a force-field, which depends on a vector field

$$\boldsymbol{\rho}(\mathbf{r}) = \sum_i \alpha_i \delta(\mathbf{r} - \mathbf{r}_i)\mathbf{s}_i$$

Then expand

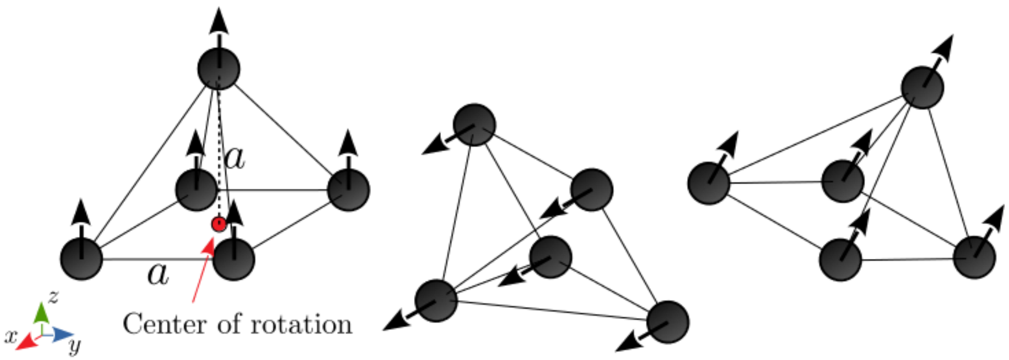
$$|\boldsymbol{\rho}\rangle = \sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}\sum_{q=-1}^{1} c_{nlmq}\,|n\underbrace{lm}q\rangle$$

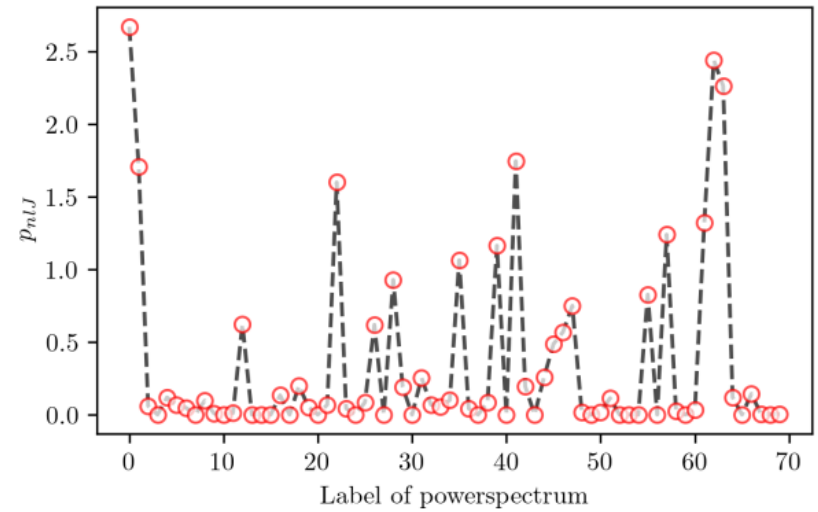
In position representation: $g_n(r)$ $\quad Y_l^m(\hat{\mathbf{r}})$ $\quad \hat{\mathbf{e}}_q$

$$\boxed{p_{nlJ} = \sum_M |u_{nlJM}|^2}$$

Extension to spin

$$p_{nlJ} = \sum_{M} \left| u_{nlJM} \right|^2$$ is fully rotational invariant



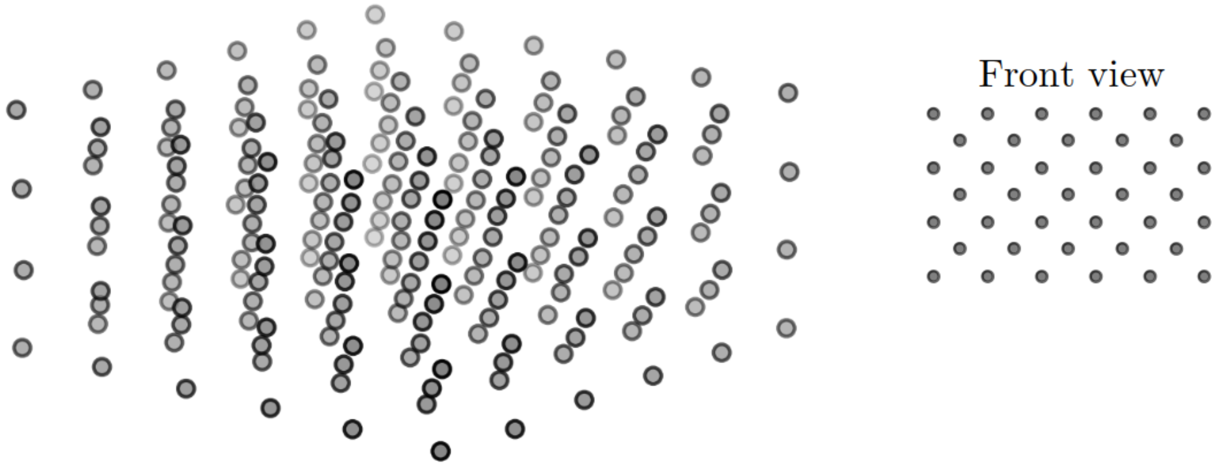


The *l*=0 term is the Heisenberg model

$$p_{n0J} \propto \sum_{ab} \alpha_a \alpha_b g_{n0}(r_a) g_{n0}(r_b) \mathbf{s_a} \cdot \mathbf{s_a}$$

Test against simple models

$$E \simeq \boldsymbol{\theta} \cdot \sum_i \boldsymbol{p}^{(i)} = \sum_{nlJ} \theta_{nlJ} \sum_i p_{nlJ}^{(i)}$$



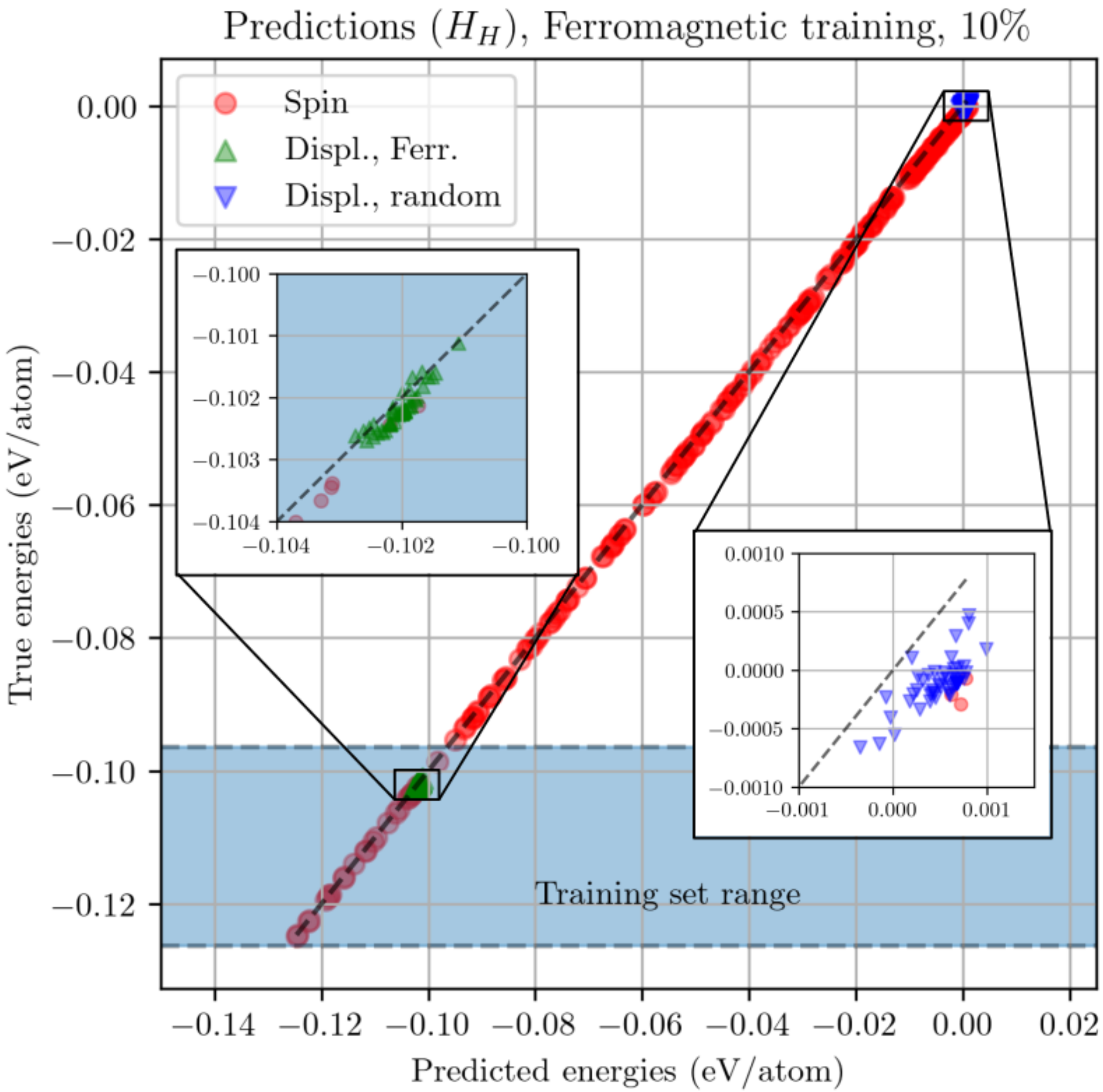
$$H_{\mathrm{H}} = -\frac{1}{2} \sum_{\langle i,j \rangle} J_{ij}(r_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j$$ → Transverse excitations

$$H_{\mathrm{L}} = \sum_i \left(A S_i^2 + B S_i^4 + C S_i^6 \right)$$ → Longitudinal excitations

Test against simple models: H_H

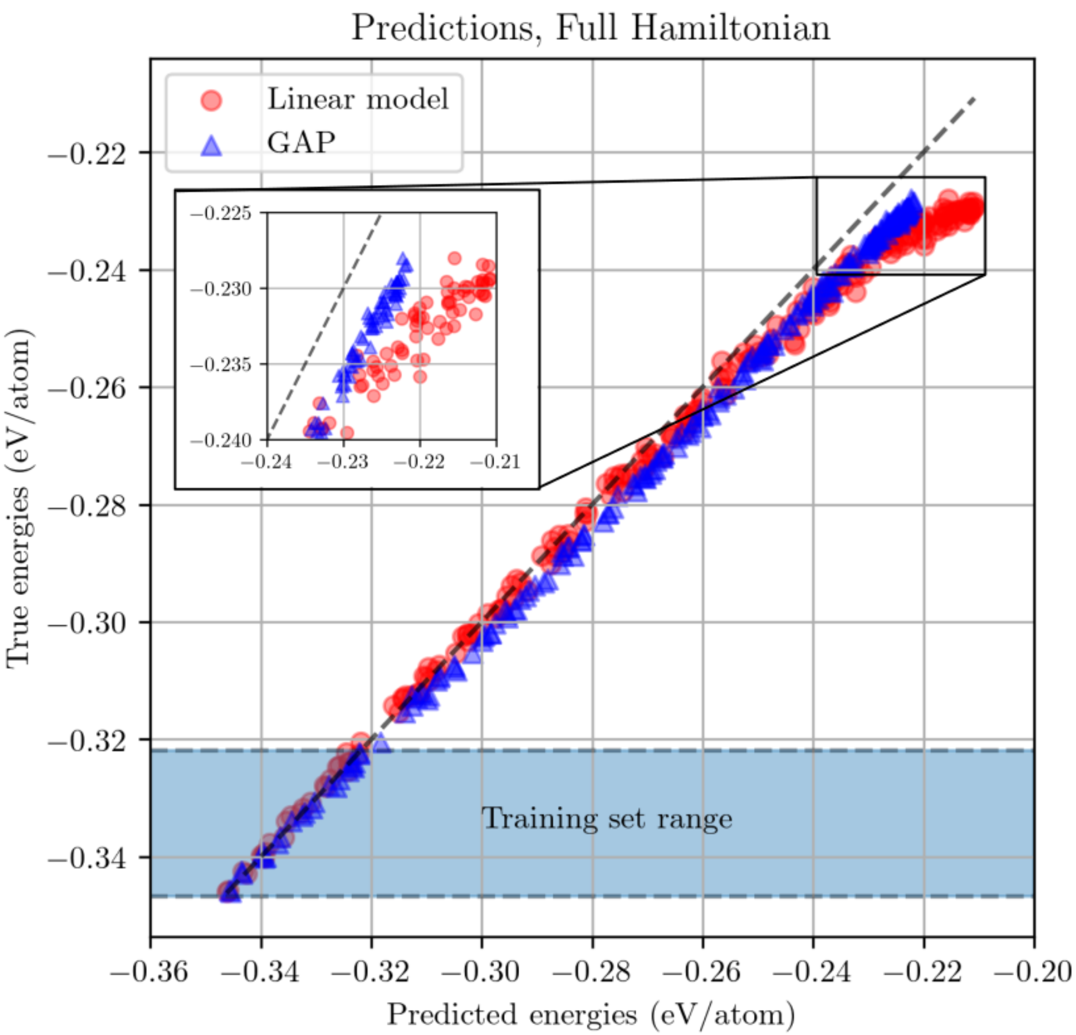
35 features

~200 training data



E error ~0.1%

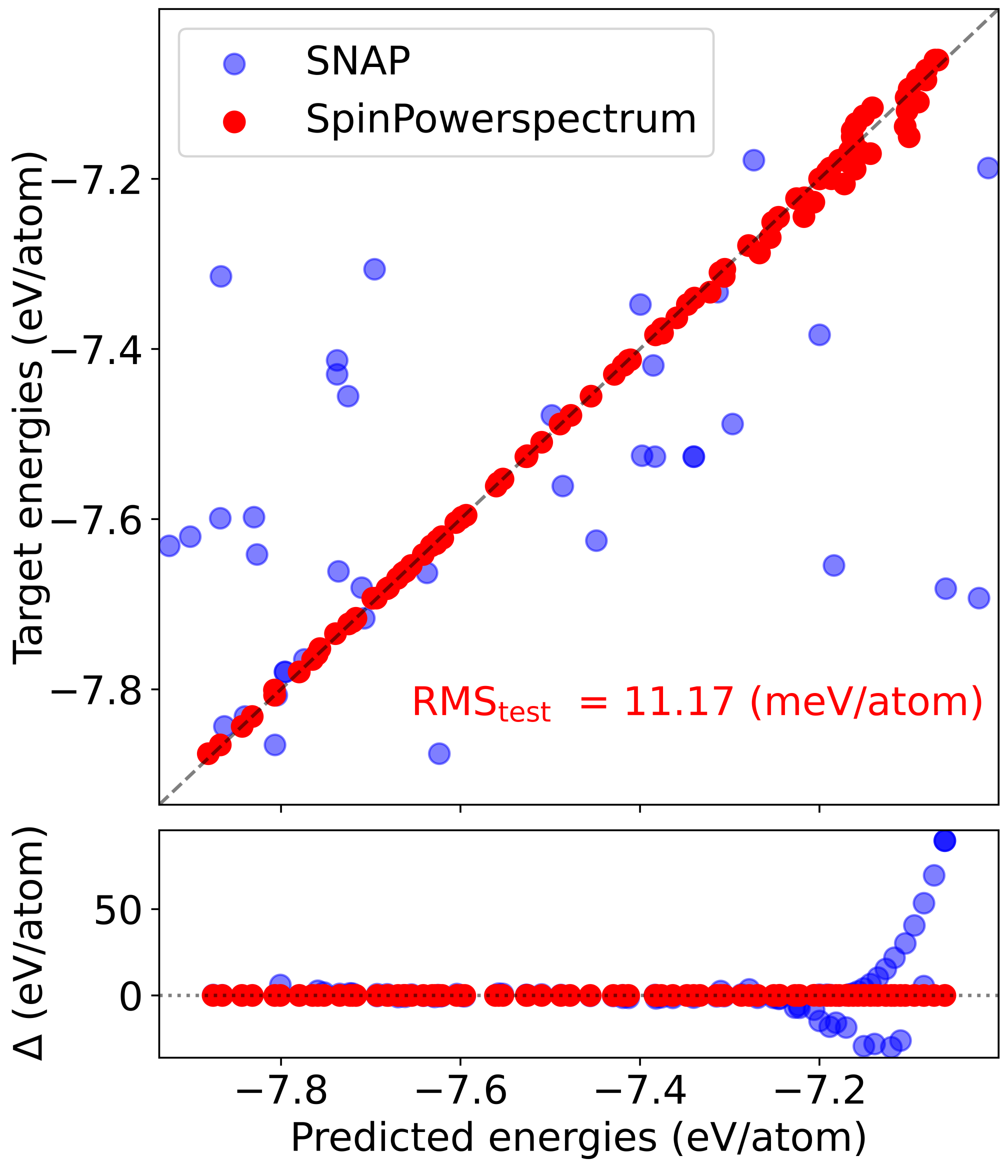
Test against simple models: $H_H + H_L$



GAP ML model

$$\varepsilon_i = \sum_{t=0}^{N_{\mathrm{train}}} \theta_t S(\boldsymbol{p}^i, \boldsymbol{p}^t) = \sum_t \theta_t e^{-\frac{1}{2\sigma}(\boldsymbol{p}^i - \boldsymbol{p}^t)^2}$$

Test against simple models



Now fitting ab initio data

α-(*bcc*)-ε(*hcp*)

Test against simple models

CRANN

Extension of FF to vector fields possible

Very general and agnostic of the PES

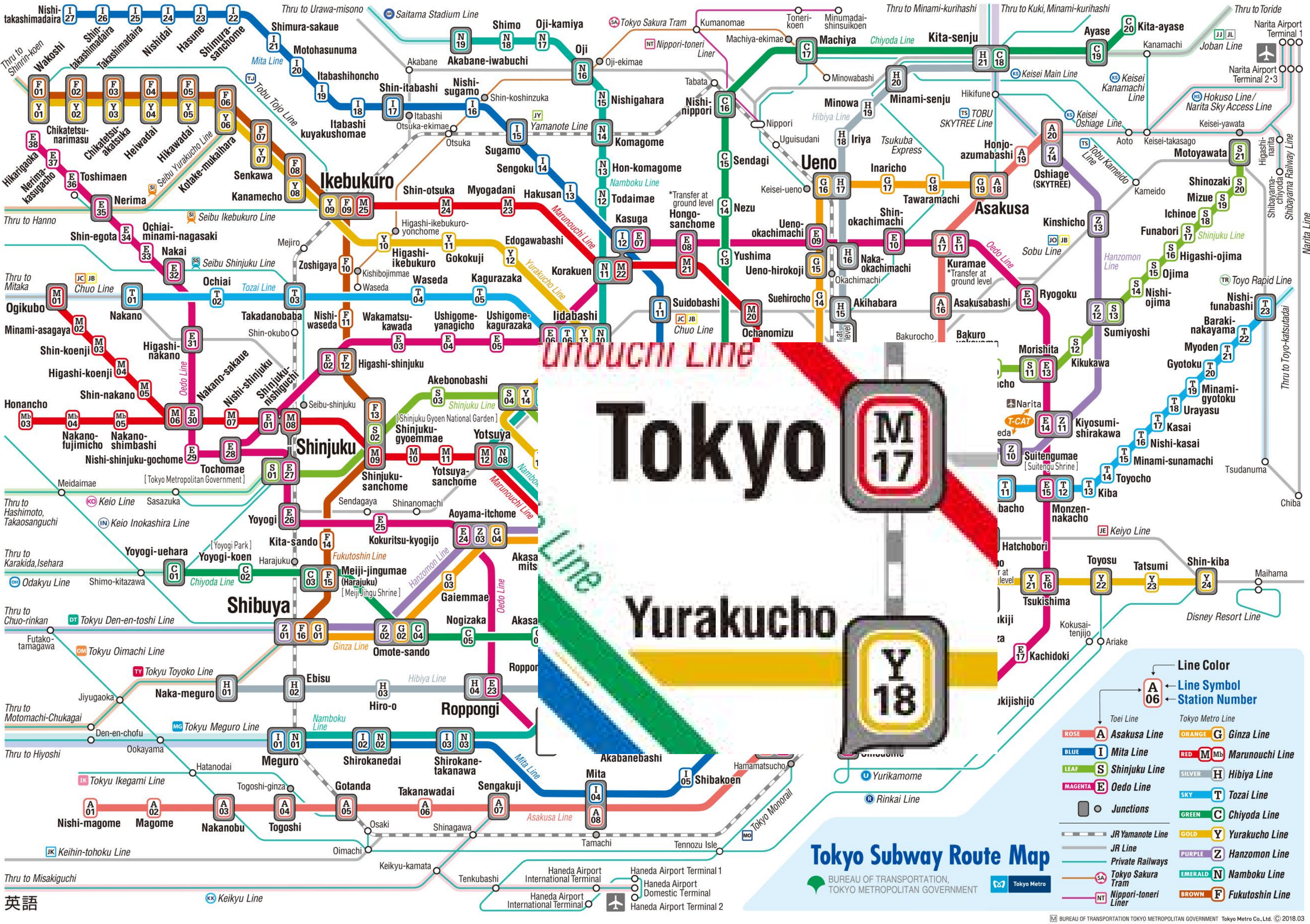
Formalism extendible to bi-spectrum

Small training sets needed (so far)

Concept extendable to other FF classes

Need non-linear model to go beyond Heisenberg

Constructing dataset is complicated


Tokyo Subway Route Map
BUREAU OF TRANSPORTATION, TOKYO METROPOLITAN GOVERNMENT
Tokyo Metro
Tokyo
M 17
Yurakucho
Y 18
Line Color
Line Symbol
Station Number
Toei Line
Tokyo Metro Line
ROSE A Asakusa Line
BLUE I Mita Line
LEAF S Shinjuku Line
MAGENTA E Oedo Line
Junctions
ORANGE G Ginza Line
RED M Mb Marunouchi Line
SILVER H Hibiya Line
SKY T Tozai Line
GREEN C Chiyoda Line
GOLD Y Yurakucho Line
PURPLE Z Hanzomon Line
EMERALD N Namboku Line
BROWN F Fukutoshin Line
JR Yamanote Line
JR Line
Private Railways
Tokyo Sakura Tram
Nippori-toneri Liner
Nishi-takashimadaira
Shin-takashimadaira
Takashimadaira
Nishidai
Hasune
Shimura-sanchome
Shimura-sakaue
Motohasunuma
Itabashihoncho
Shin-itabashi
Nishi-sugamo
Itabashi kuyakushomae
Sugamo
Sengoku
Hakusan
Kasuga
Thru to Urawa-misono
Saitama Stadium Line
Shimo
Oji-kamiya
Oji
Akabane-iwabuchi
Nishigahara
Komagome
Hon-komagome
Todaimae
Tokyo Sakura Tram
Nippori-toneri Liner
Kumanomae
Toneri-koen
Minumadai-shinsuikoen
Machiya
Kita-senju
Minami-senju
Minowa
Iriya
Ueno
Inaricho
Tawaramachi
Asakusa
Honjo-azumabashi
Oshiage (SKYTREE)
Thru to Minami-kurihashi
Thru to Kuki, Minami-kurihashi
Ayase
Kita-ayase
Thru to Toride
Joban Line
Narita Airport Terminal 1
Narita Airport Terminal 2·3
Keisei Main Line
Keisei Kanamachi Line
Hokuso Line / Narita Sky Access Line
Keisei Oshiage Line
Keisei-yawata
Keisei-takasago
Aoto
Motoyawata
Shibayama Railway Line
Narita Line
Thru to Shinrin-koen
Wakoshi
Chikatetsu-narimasu
Chikatetsu-akatsuka
Heiwadai
Hikawadai
Kotake-mukaihara
Senkawa
Kanamecho
Ikebukuro
Hikarigaoka
Nerima-kasugacho
Toshimaen
Nerima
Thru to Hanno
Seibu Ikebukuro Line
Seibu Yurakucho Line
Tobu Tojo Line
Mita Line
Namboku Line
Marunouchi Line
Shin-otsuka
Myogadani
Higashi-ikebukuro
Gokokuji
Edogawabashi
Kagurazaka
Korakuen
Shin-egota
Ochiai-minami-nagasaki
Nakai
Seibu Shinjuku Line
Ochiai
Thru to Mitaka
Chuo Line
Ogikubo
Nakano
Takadanobaba
Waseda
Iidabashi
Suidobashi
Ochanomizu
Hongo-sanchome
Ueno-okachimachi
Shin-okachimachi
Naka-okachimachi
Yushima
Ueno-hirokoji
Nezu
Sendagi
Nishi-nippori
Nippori
Suehirocho
Akihabara
Kuramae
Asakusabashi
Ryogoku
Kinshicho
Sumiyoshi
Bakurocho
Morishita
Kikukawa
Funabori
Ichinoe
Mizue
Shinozaki
Higashi-ojima
Ojima
Nishi-ojima
Toyo Rapid Line
Nishi-funabashi
Baraki-nakayama
Myoden
Gyotoku
Minami-gyotoku
Urayasu
Kasai
Nishi-kasai
Minami-sunamachi
Toyocho
Kiba
Monzen-nakacho
Kiyosumi-shirakawa
Suitengumae
Tsudanuma
Chiba
Thru to Toyo-katsutadai
Minami-asagaya
Shin-koenji
Higashi-koenji
Shin-nakano
Honancho
Nakano-fujimicho
Nakano-shimbashi
Nishi-shinjuku-gochome
Higashi-nakano
Nakano-sakaue
Nishi-shinjuku
Shinjuku-nishiguchi
Higashi-shinjuku
Wakamatsu-kawada
Ushigome-yanagicho
Ushigome-kagurazaka
Shinjuku
Tochomae
Shinjuku-sanchome
Shinjuku-gyoemmae
Yotsuya
Yotsuya-sanchome
Akebonobashi
Meidaimae
Keio Line
Sasazuka
Keio Inokashira Line
Yoyogi
Kita-sando
Kokuritsu-kyogijo
Aoyama-itchome
Yoyogi-uehara
Yoyogi-koen
Meiji-jingumae (Harajuku)
Gaiemmae
Nogizaka
Shibuya
Omote-sando
Odakyu Line
Shimo-kitazawa
Tokyu Den-en-toshi Line
Futako-tamagawa
Tokyu Oimachi Line
Tokyu Toyoko Line
Naka-meguro
Ebisu
Hiro-o
Roppongi
Jiyugaoka
Tokyu Meguro Line
Meguro
Shirokanedai
Shirokane-takanawa
Den-en-chofu
Ookayama
Hatanodai
Tokyu Ikegami Line
Togoshi-ginza
Gotanda
Takanawadai
Sengakuji
Mita
Shibakoen
Akabanebashi
Hamamatsucho
Nishi-magome
Magome
Nakanobu
Togoshi
Osaki
Shinagawa
Tamachi
Tennozu Isle
Tokyo Monorail
Keihin-tohoku Line
Oimachi
Keikyu-kamata
Tenkubashi
Haneda Airport International Terminal
Haneda Airport Terminal 1
Haneda Airport Domestic Terminal
Haneda Airport Terminal 2
Keikyu Line
Thru to Misakiguchi
Yurikamome
Rinkai Line
Toyosu
Tatsumi
Shin-kiba
Maihama
Disney Resort Line
Kokusai-tenjijo
Ariake
Tsukishima
Kachidoki
Keiyo Line
Hatchobori
Thru to Hashimoto, Takaosanguchi
Thru to Karakida, Isehara
Thru to Chuo-rinkan
Thru to Motomachi-Chukagai
Thru to Hiyoshi
英語
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