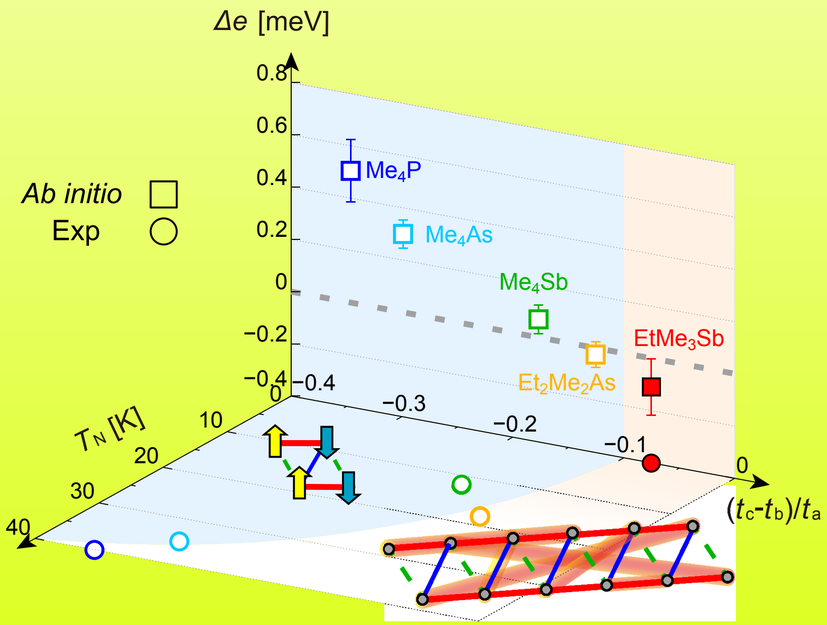
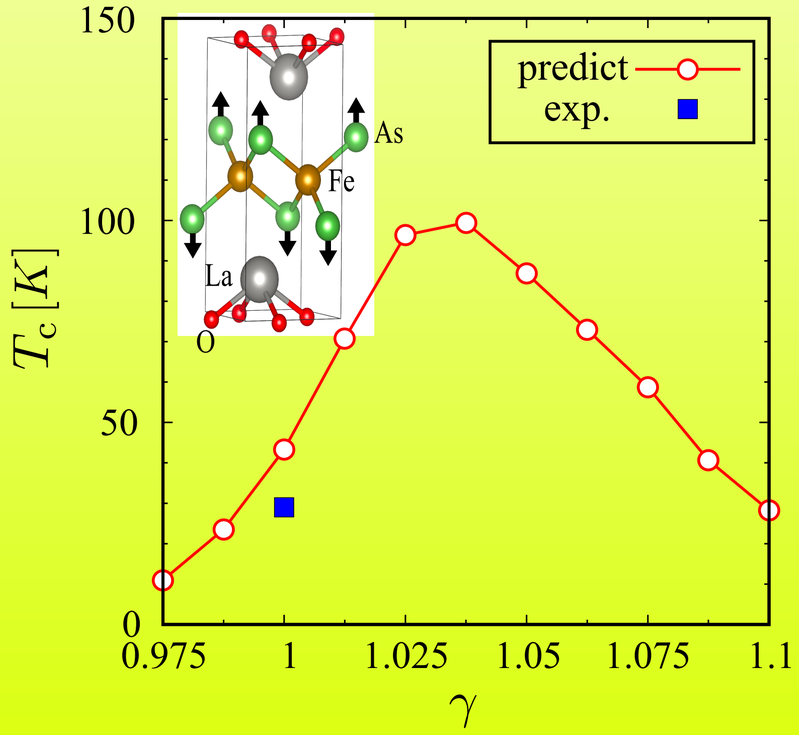
強相関電子系を取り扱うソフトウェア開発とその適用

Software development and application for analyzing strongly correlated electron systems

三澤 貴宏

東京大学物性研究所





Outline

1. Introduction: *Ab initio* method for correlated electron systems

2. Application to quantum spin liquid in β'-*X*[Pd(dmit)$_2$]$_2$

T. Misawa, K. Yoshimi, and T. Tsumuraya, Phys. Rev. Research **2**, 032072(R) (2020)
K. Yoshimi, T. Tsumuraya, and T. Misawa, Phys. Rev. Research **3**, 033224 (2021)
K . Ido, K. Yoshimi, T. Misawa, and M. Imada, npj Quantum Mater. **7**, 48 (2022)

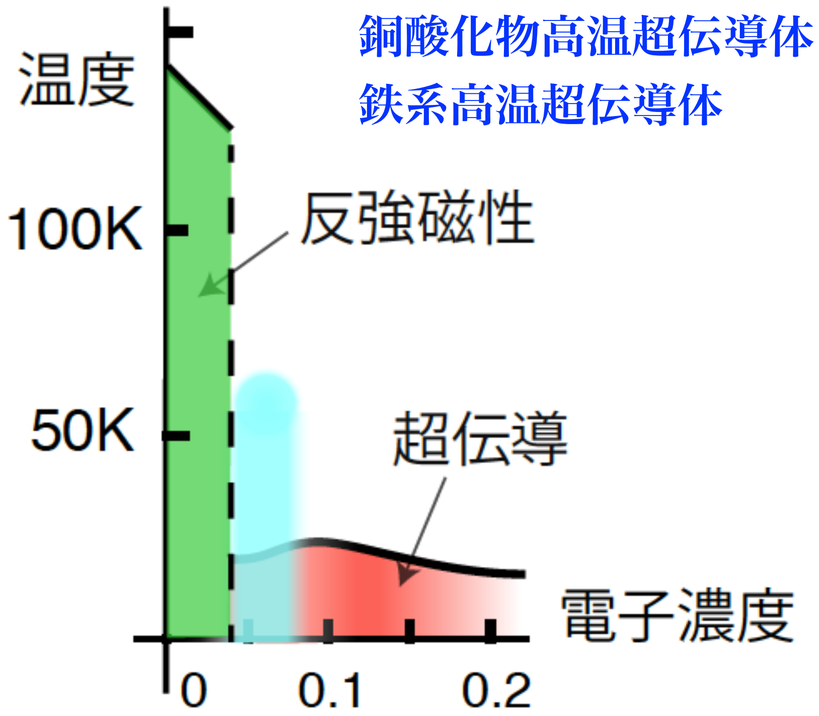
3. Application to iron-based high-Tc superconductors

K. Ido, Y. Motoyama, K. Yoshimi, and T. Misawa, J. Phys. Soc. Jpn **92**, 064702 (2023)

4. Summary

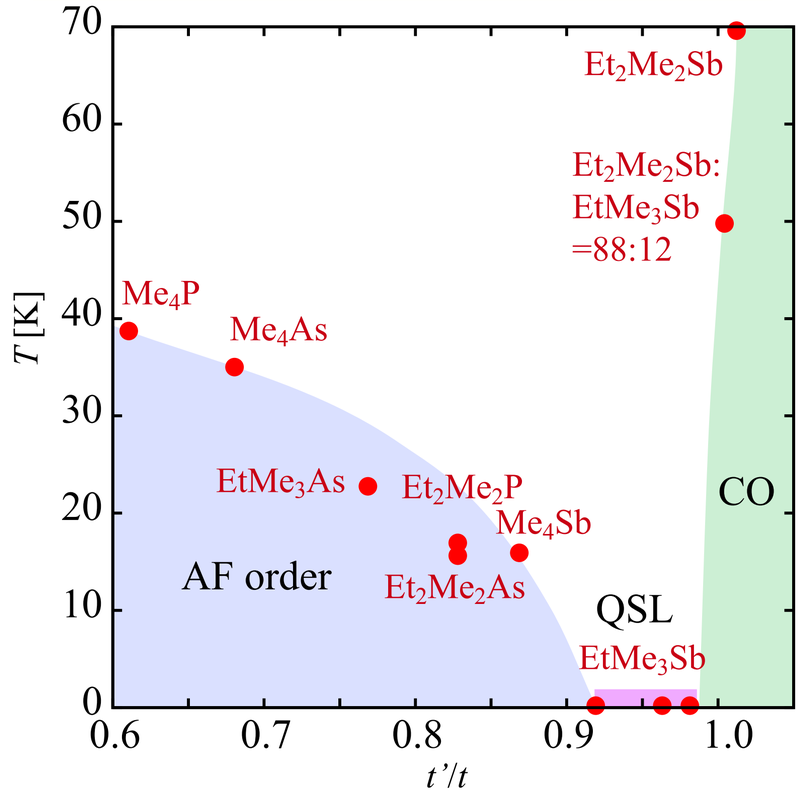
強相関電子系における新現象・新物質発見

電子がひしめき合う
強相関電子系ゆえの機能性



Y. Kamihara *et al.*, JACS 130, 3296 (2008)

新しい物質の状態の出現
(例)量子スピン液体



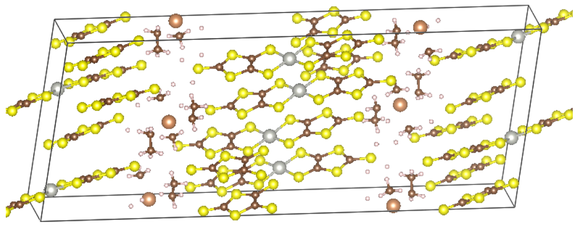
Read data from Fig. 17 in "K. Kanoda and R. Kato, Annu. Rev. Condens. Matter Phys. 2, 167-188 (2011)" and replotted

-強相関電子系は機能性材料の宝庫

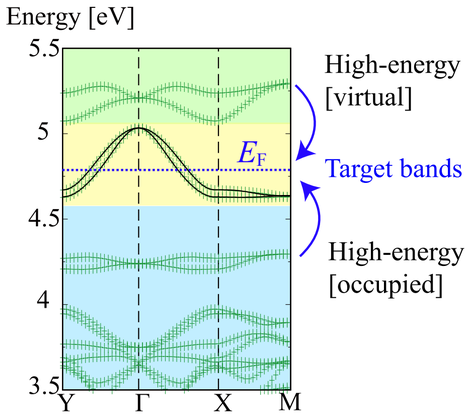
-電子相関の効果を取り扱える計算手法を開発することで、現象の起源解明を行い、予測・制御につなげることで、新奇現象・新物質の産業利用の可能性

Ab *initio* method for correlated electron systems

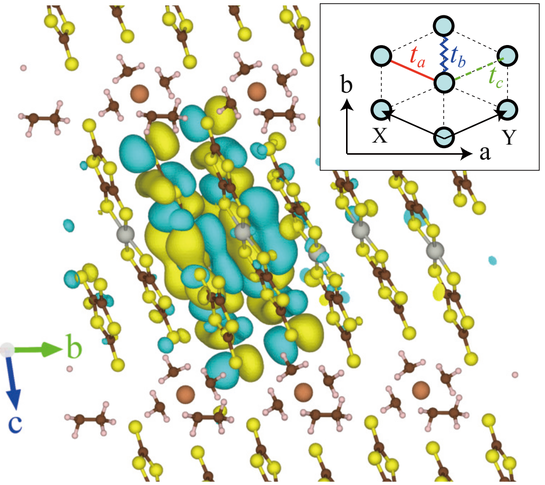
✓ lattice structure



✓ band calc.



✓Wannier func.



RESPACK
第一原理多体摂動計算

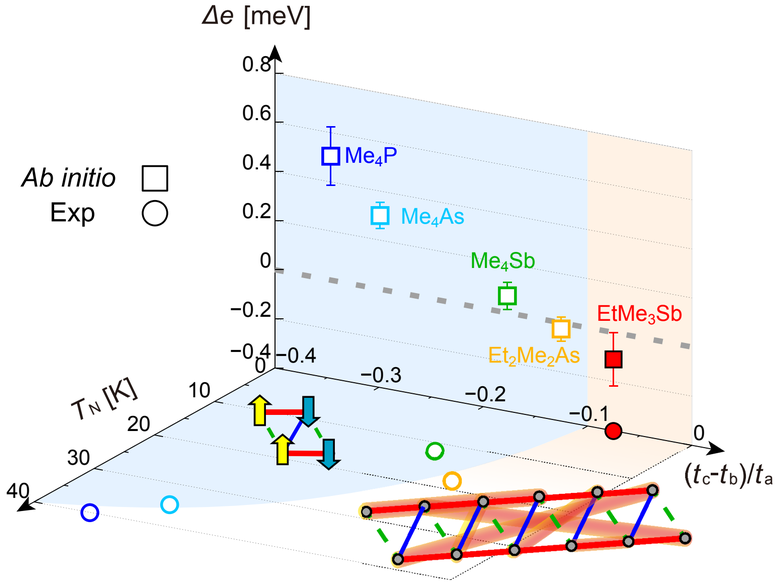
ex-Hubbard type Hamiltonians

Max-loc. Wannier +cRPA

$$H = \sum_{ij,\sigma} t_{ij}(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.c.})$$

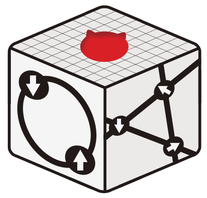
$$+ U \sum_{i} n_{i\uparrow}n_{i\downarrow} + \sum_{ij} V_{ij} N_i N_j$$

✓Clarifying electronic structures



HΦ

mVMC



Not only transfers, but also interaction parameters (*t*, *U*, *V*) are determined in an *ab initio* way

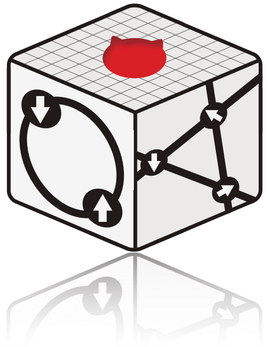
Open-source software packages

HΦ: exact diagonalization

-Exact calc. for small system sizes (~40 sites)
-Cutting-edge theoretical and mathematical method
-Applications to QSL

https://github.com/issp-center-dev/HPhi

mVMC: many-variable variational MC



-Applicable for large system sizes (~1000 sites)
-Highly accurate and flexible method
-# of variational parameters > 10^4
-Applications to high-Tc SCs & QSL

https://github.com/issp-center-dev/mVMC

RESPACK

RESPACK
第一原理多体摂動計算

-Derivation of low-energy effective Hamiltonians

-Using wan2respack, user can use wannier functions obtained by Wannier90 as input for RESPACK

Seamless combination with the *ab initio* derivation of the low-energy effective models
→ Systematic & comprehensive calculations for SCES are now possible
(eg. Applications to molecular solids)

RESPACK: https://sites.google.com/view/kazuma7k6r
wan2respack: https://github.com/respack-dev/wan2respack

A part of development is supported by PASUMS@ ISSP.

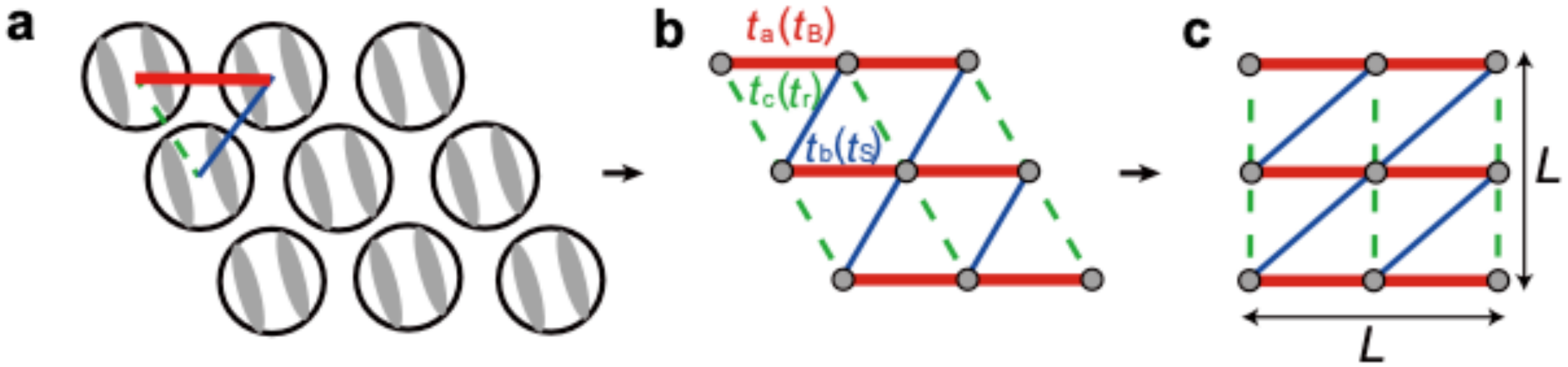
PASUMS
Project for advancement of software usability in materials science

Quantum spin liquid in β'-*X*[Pd(dmit)$_2$]$_2$

T. Misawa, K. Yoshimi, and T. Tsumuraya, Phys. Rev. Research 2, 032072(R) (2020)
K. Yoshimi, T. Tsumuraya, and T. Misawa, Phys. Rev. Research 3, 033224 (2021)
K. Ido, K. Yoshimi, T. Misawa, and M. Imada, npj Quantum Mater. 7, 48 (2022)

Derivation of low-energy effective Hamiltonians

Mapping to the equivalent anisotropic triangular lattice



Ex. Hubbard Hamiltonians on anisotropic triangular lattice

9 compounds at room temperatures
5 compounds at low temperatures

$$H = \sum_{ij,\sigma} t_{ij}(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.c.})$$

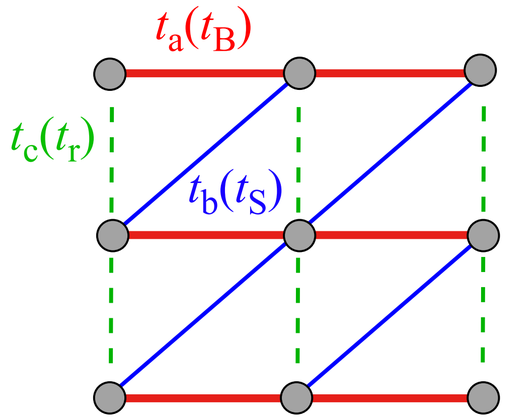
$$+ U\sum_{i} n_{i\uparrow}n_{i\downarrow} + \sum_{ij} V_{ij}N_iN_j$$

Not only transfers, but also interaction parameters (*t*, *U*, *V*) are determined in an *ab initio* way

Effects of dimensional downfolding [3D → 2D]
***U*=*U*-Δ,*V*=*V*-Δ, Δ=0.18 eV**
[cf. K.Nakamura *et al.*, PRB 86,205117 (2012)]

Derivation of low-energy effective Hamiltonians

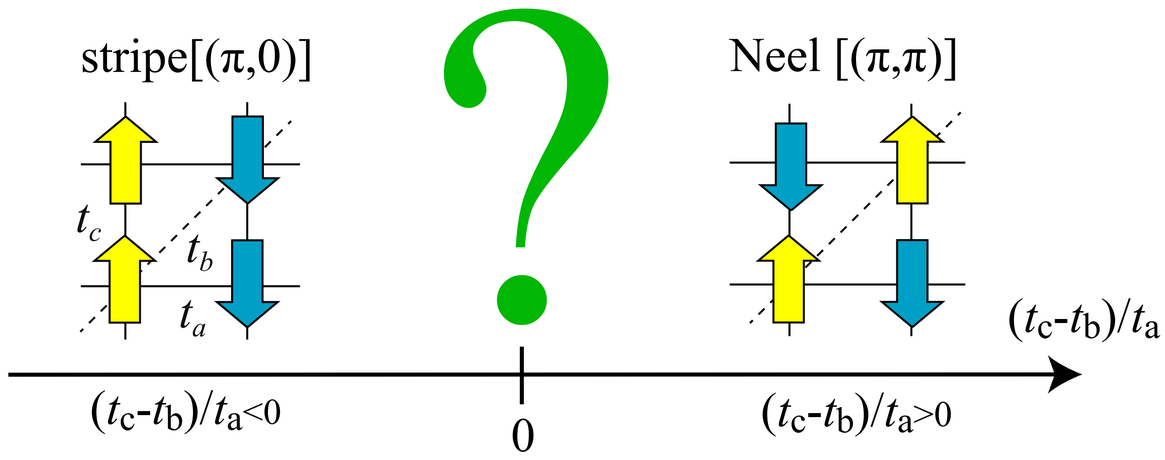
Ex. Hubbard Hamiltonians on anisotropic triangular lattice



$$H = \sum_{ij,\sigma} t_{ij}(c^{\dagger}_{i\sigma}c_{j\sigma} + \mathrm{H.c.})$$

$$+ U\sum_{i} n_{i\uparrow}n_{i\downarrow} + \sum_{ij} V_{ij}N_iN_j$$

Expected phase diagram



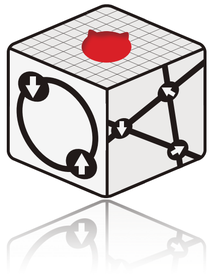
For low-temperature structures, all compounds show $(t_c\text{-}t_b)/t_a<0$

Around $(t_c\text{-}t_b)/t_a\sim0$, it is expected that magnetic order is melted and QSL appears

mVMC analysis

$$|\psi\rangle = \mathcal{P}_J \mathcal{P}_G \mathcal{L}^S |\phi_{\rm pair}\rangle$$

https://github.com/issp-center-dev/mVMC



Pair-product part

$$|\phi_{\rm pair}\rangle = \left[\sum_{I,J} F_{IJ} c_I^\dagger c_J^\dagger\right]^{N_{\rm e}/2} |0\rangle$$

Projected BCS wave functions, which can describe

- Correlated metals
- AF, CO order
- Superconducting state & QSL

Correlation factors

Gutzwiller-Jastrow factors

$$\mathcal{P}_G = \exp\left[\sum_i g_i n_{i\uparrow} n_{i\downarrow}\right], \mathcal{P}_J = \exp\left[\sum_{i,j} v_{ij} N_i N_j\right]$$

Quantum number projection

Total spin: $\mathcal{L}_S$

Point-group, momentum projections are also possible

Optimization (SR method)

Mimimizing energy $E_{\boldsymbol{\alpha}}$ S. Sorella PRB (2001)

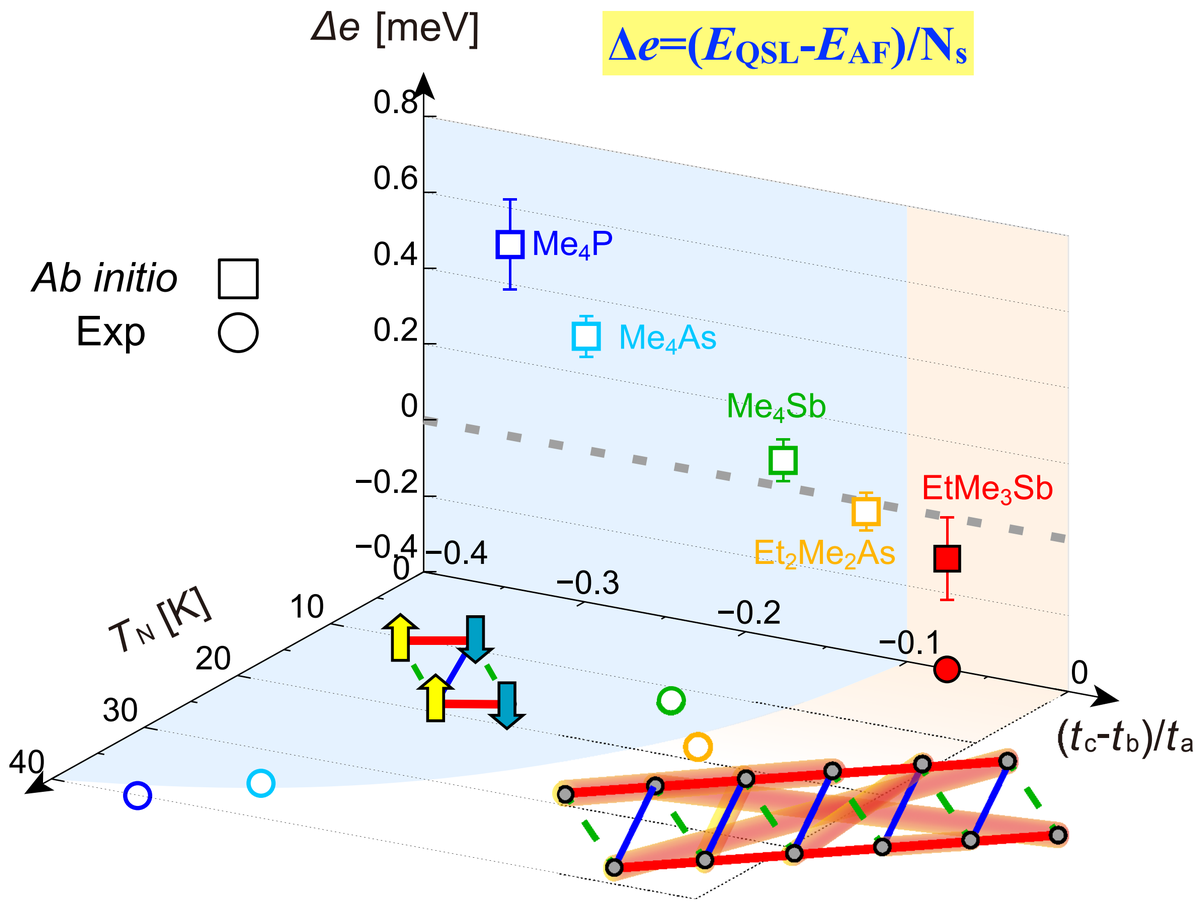
$$\boldsymbol{\alpha}_{\rm new} - \boldsymbol{\alpha}_{\rm old} = X^{-1}\boldsymbol{g}$$

$$\boldsymbol{g} = \partial E_{\boldsymbol{\alpha}} / \partial \boldsymbol{\alpha} \quad X = \langle \bar{\psi}_\alpha | \bar{\psi}_\beta \rangle$$

Equivalent to natural gradient method
S.-I. Amari Neural Comp. (1998)

By optimizing many variational parameters (#>=10^4), highly accurate calculations are possible!

Phase diagram



-Solving Hamiltonians for available low-temperature structures

-Spin Jastrow factors, Composite-fermion pairing, RBM factors are included

-Initial state of the QSL is projected BCS wave functions
→ 1D anisotropic QSL after optimization

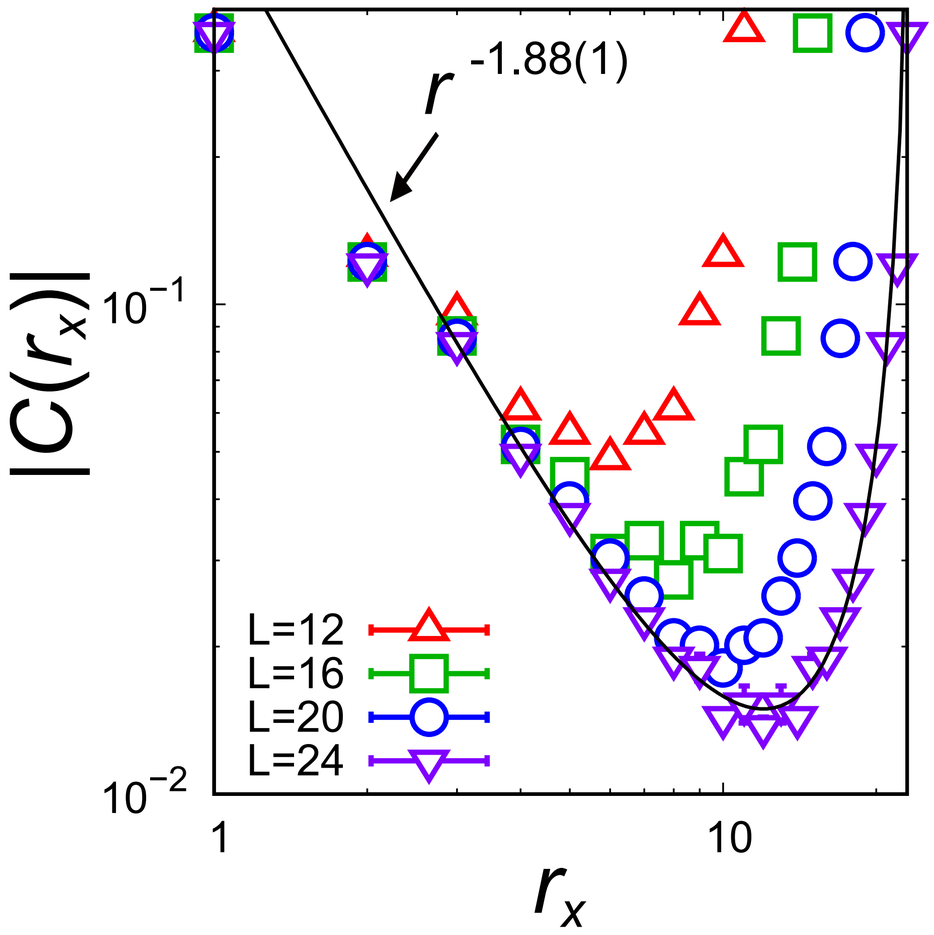
-Eigen-energies (zero-variance limit) are estimated by variance extrapolation

K. Ido *et al.*, npj Quantum Mater. 7, 48 (2022)

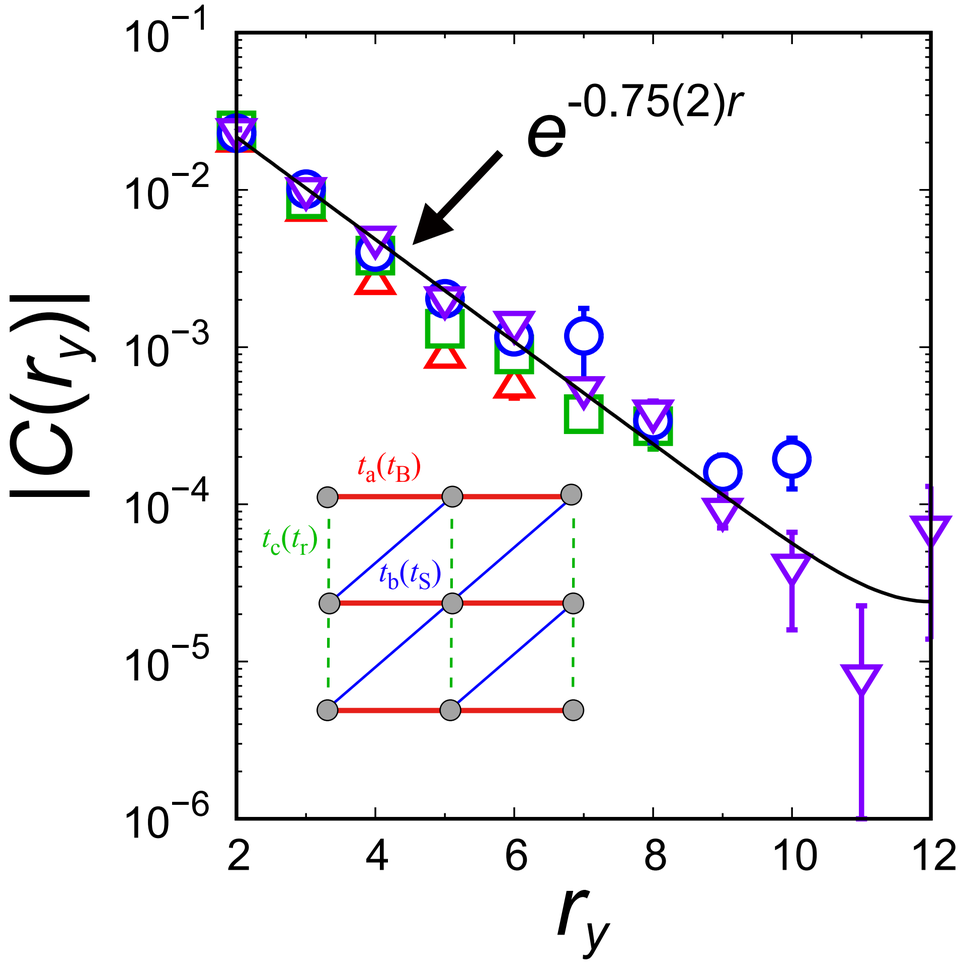
-Reproducing the trend of stability of the AF order:By changing cation X from Me_4P to $EtMe_3Sb$, the AF order becomes unstable
-Consistent w/ experimental Neel temperature T_N
-QSL becomes stable in $EtMe_3Sb$!

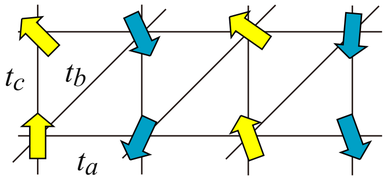
Spin correlations in $EtMe_3Sb$

***x* direction**



***y* direction**





***x* direction → power low decay [r^{-p}, p~1.88 (1)] (gapless)**
***y* direction → exponential decay (gapped)**

鉄系超伝導体の有効模型導出
+データ科学的解析

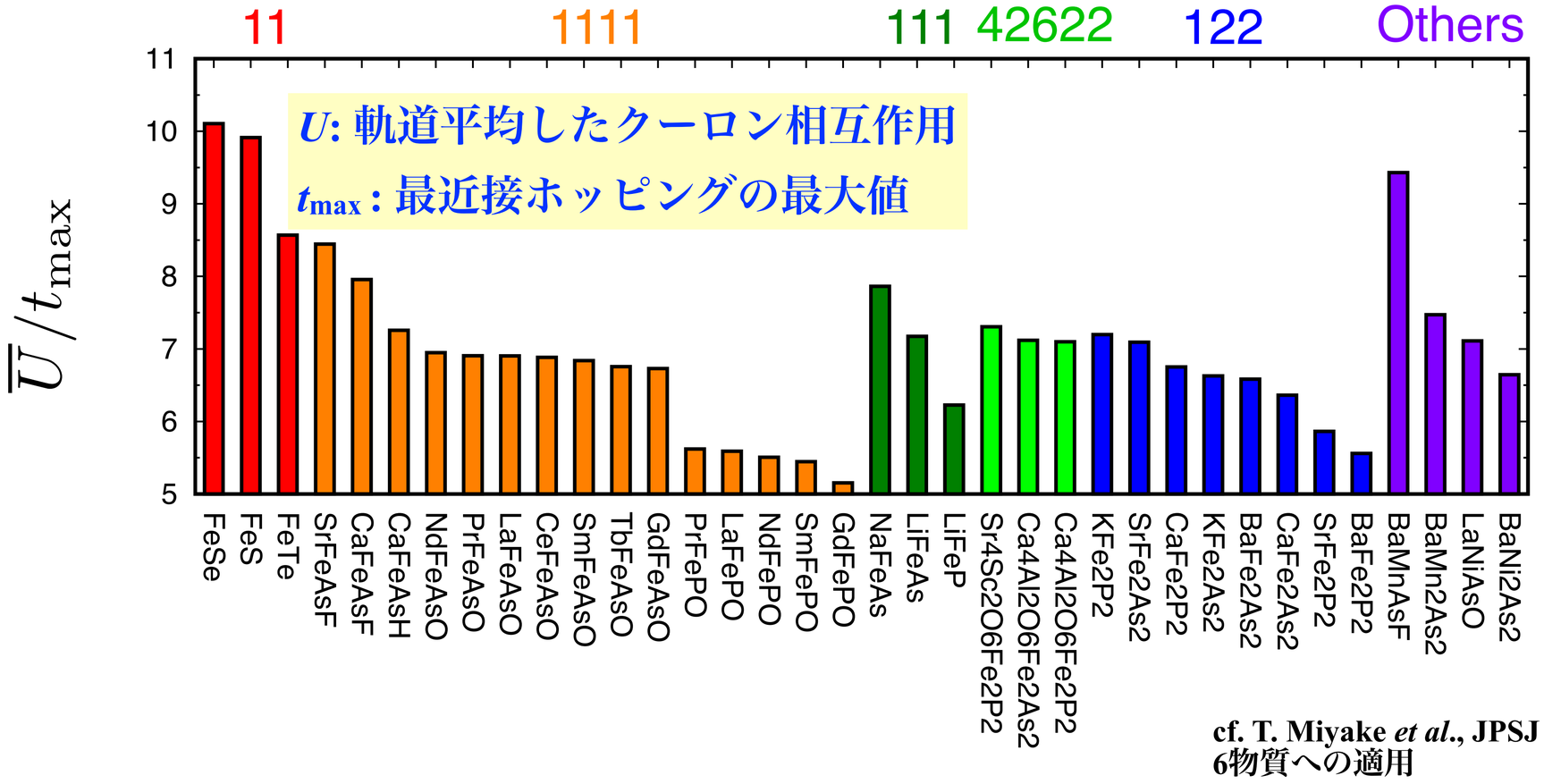
K. Ido, Y. Motoyama, K. Yoshimi, and T. Misawa,
J. Phys. Soc. Jpn. 92, 064702 (2023)

鉄系超伝導体の網羅的な有効模型導出

鉄系高温超伝導体への網羅計算[36物質]

K. Ido, Y. Motoyama, K. Yoshimi, and T. Misawa,
J. Phys. Soc. Jpn. 92, 064702 (2023)
一部はトヨタ自動車株式会社の支援を受けて実施

$$\mathcal{H} = \sum_{\sigma}\sum_{RR'}\sum_{nm} t_{mRnR'} a_{nR}^{\sigma\dagger} a_{mR'}^{\sigma} \quad \rightarrow \text{Hopping Term}$$
$$+\frac{1}{2}\sum_{\sigma\rho}\sum_{RR'}\sum_{nm}\Big\{ U_{mRnR'} a_{nR}^{\sigma\dagger} a_{mR'}^{\rho\dagger} a_{mR'}^{\rho} a_{nR}^{\sigma} \rightarrow \text{Coulomb Term}$$
$$+J_{mRnR'}\left(a_{nR}^{\sigma\dagger} a_{mR}^{\rho\dagger} a_{nR}^{\rho} a_{mR'}^{\sigma} + a_{nR}^{\sigma\dagger} a_{nR}^{\rho\dagger} a_{mR}^{\rho} a_{mR'}^{\sigma}\right)\Big\} \rightarrow \text{Exchange Term}$$



cf. T. Miyake *et al.*, JPSJ (2010).
6物質への適用

- 相互作用が物質によって2倍程度変化
- 超伝導転移温度の実験値と有効模型のパラメータの間に関係は？

Low-energy effective Hamiltonians for iron-based SCs

5-orbital extended Hubbard models

$$\mathcal{H} = \sum_{\sigma}\sum_{\boldsymbol{R}\boldsymbol{R}'}\sum_{nm} t_{m\mathbf{R}n\mathbf{R}'} a^{\sigma\dagger}_{n\boldsymbol{R}} a^{\sigma}_{m\boldsymbol{R}'} \quad \rightarrow \text{Hopping Term}$$

$$+\frac{1}{2}\sum_{\sigma\rho}\sum_{\boldsymbol{R}\boldsymbol{R}'}\sum_{nm}\Big\{ U_{m\mathbf{R}n\mathbf{R}'} a^{\sigma\dagger}_{n\boldsymbol{R}} a^{\rho\dagger}_{m\boldsymbol{R}'} a^{\rho}_{m\boldsymbol{R}'} a^{\sigma}_{n\boldsymbol{R}} \rightarrow \text{Coulomb Term}$$

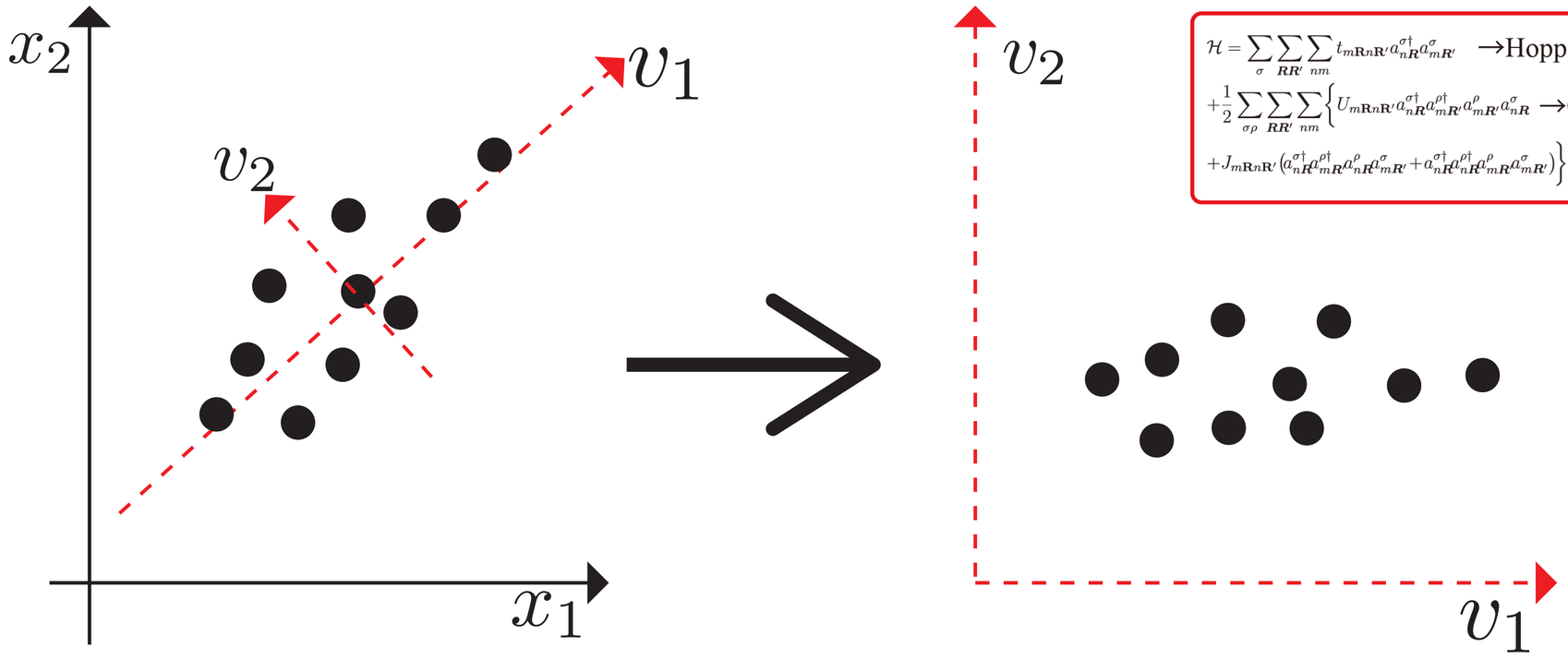
$$+ J_{m\mathbf{R}n\mathbf{R}'}\left(a^{\sigma\dagger}_{n\boldsymbol{R}} a^{\rho\dagger}_{m\boldsymbol{R}} a^{\rho}_{n\boldsymbol{R}} a^{\sigma}_{m\boldsymbol{R}'} + a^{\sigma\dagger}_{n\boldsymbol{R}} a^{\rho\dagger}_{n\boldsymbol{R}} a^{\rho}_{m\boldsymbol{R}} a^{\sigma}_{m\boldsymbol{R}'}\right)\Big\} \rightarrow \text{Exchange Term}$$

Microscopic parameters: Hopping t_{ij},
Coulomb interaction V_{ij}, Exchange terms J_{ij}

Main questions:
-How are there any relation between microscopic parameters and T_c?
→ Principle component analysis (PCA)

-If so, is it possible to predict Tc only from the microscopic parameters?
→ Construction of regression model

Principle Component Analysis (PCA)



descriptors: x_1, $x_{2,\ \ldots}$ = *U,V,t, J*..

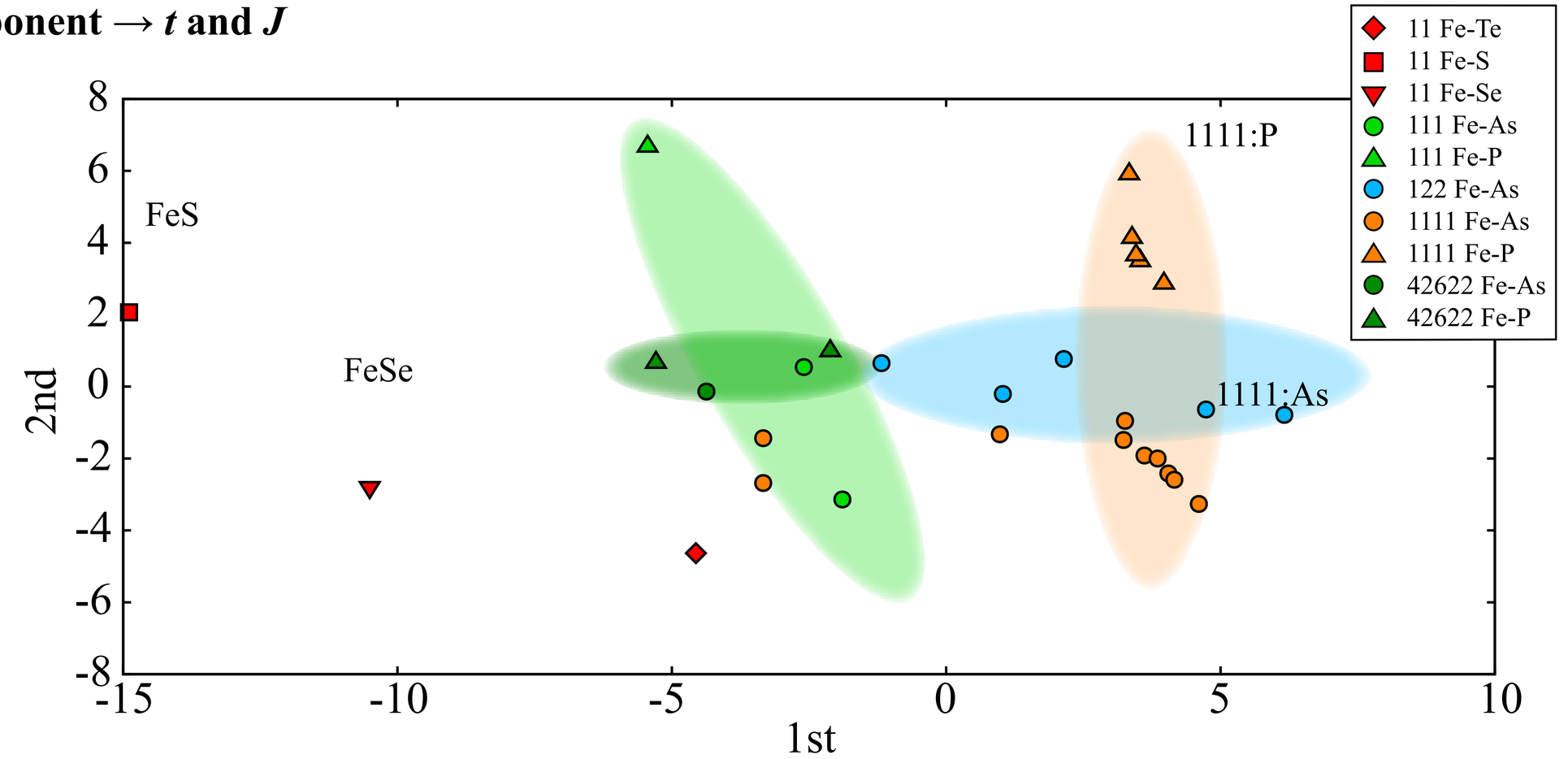
In this case, v_1 is a good descriptor for characterizing materials

By performing the orthogonal trans., we can obtain independent (uncorrelated) variables that describe the relevant features of parameter sets

→ Hint for searching the descriptors

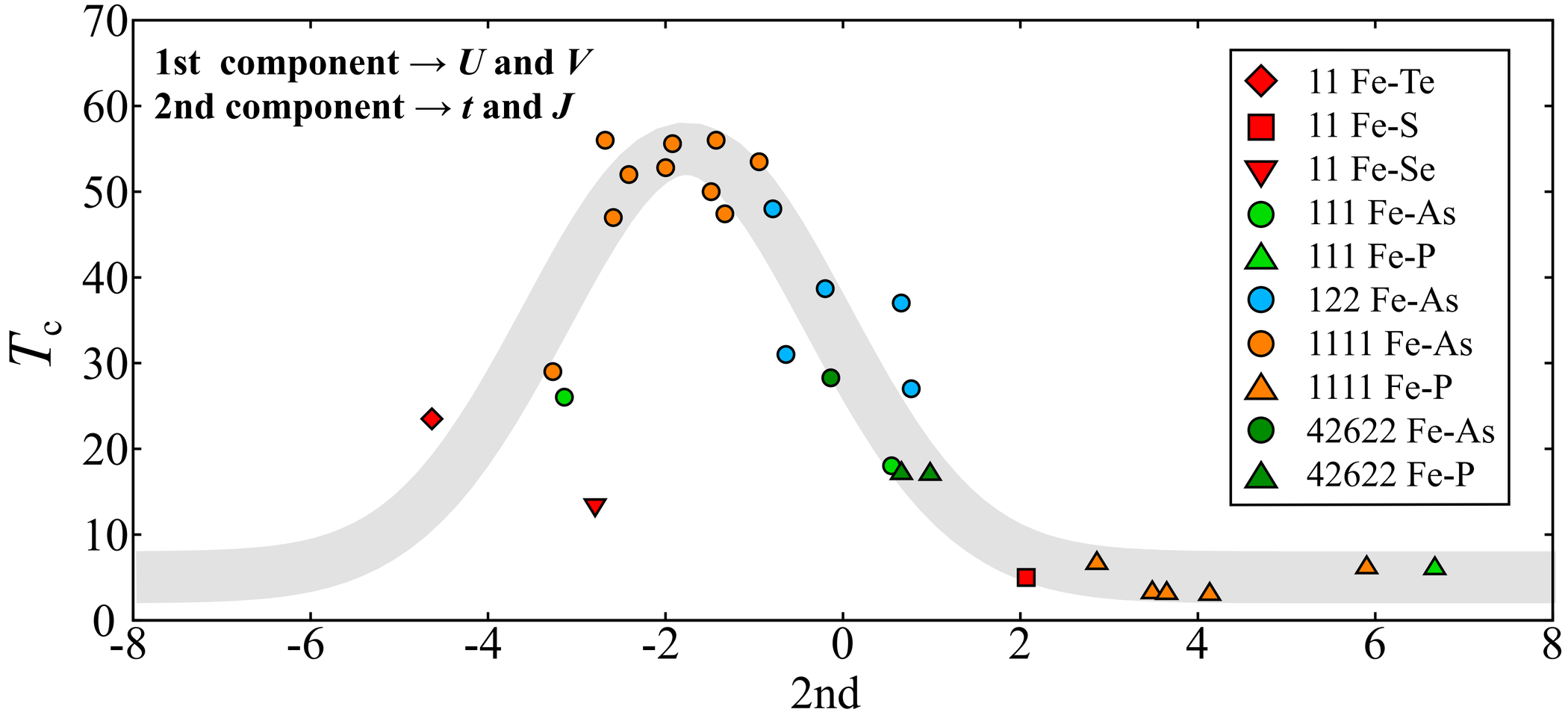
PCA for iron-based SCs

1st component → *U* and *V*
2nd component → *t* and *J*



Difference in intra- and inter families are characterized by 1st & 2nd components.
1111, 111 family has similar 1st comp. = U and V [Difference is characterized by t and J]
122, 4262 family has similar 2nd comp. = t and J [Difference is characterized by U and V]
11 family is exceptional (origin of exotic phenomena in FeSe ?)

T_c vs 2nd principal components



2nd comp. well describes mat. dep. of experimental Tc! [similar to Lee's plot]

✓ Hamiltonians have information for describing Tc

✓ PCA automatically detects a key parameter w/o a priori knowledge

超伝導転移温度を予測する回帰モデル

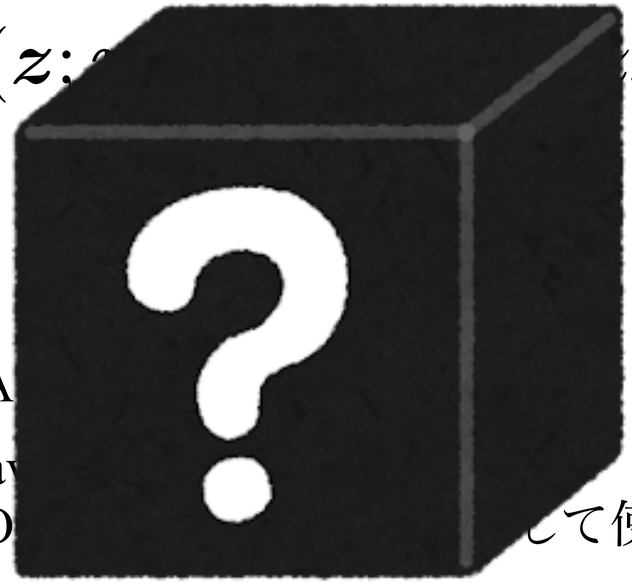
入力:微視的なパラメータ

U_{ij}, V_{ij}, t_{ij}, V_{ij}/U_{ii}, …

出力: 超伝導転移温度 T_c

回帰モデル

$f(z; \ldots) \ldots \sum_i w_i$



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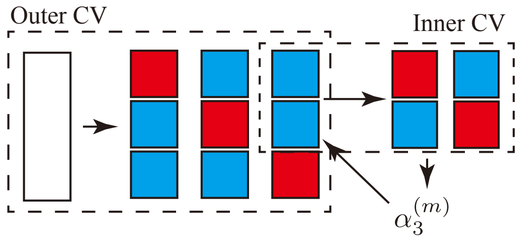


有効模型の微視的なパラメータの情報のみから

超伝導転移温度を再現・予測する回帰モデルの作成

超伝導転移温度を予測する回帰モデル

入力:微視的なパラメータ

U_{ij}, V_{ij}, t_{ij}, V_{ij}/U_{ii}, …

出力: 超伝導転移温度 T_c

回帰モデル

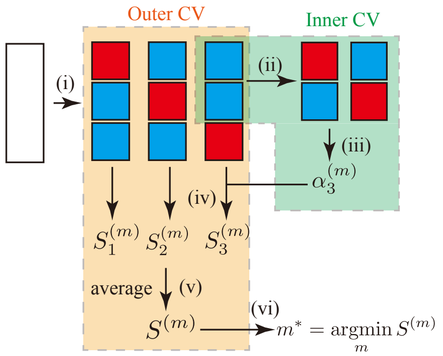
$$f(\boldsymbol{z};\boldsymbol{w}) = w_0 + \sum_i z_i w_i$$

z_i: 記述子 (U_{ij},t_{ij}, V_{ij}^2,$(V_{ij}/t)^2$…)

w_i: 重み

-LASSOによる重みの最適化

-leave-one-out cross validation (LOO-CV)を学習器の評価として使用



有効模型の微視的なパラメータの情報のみから

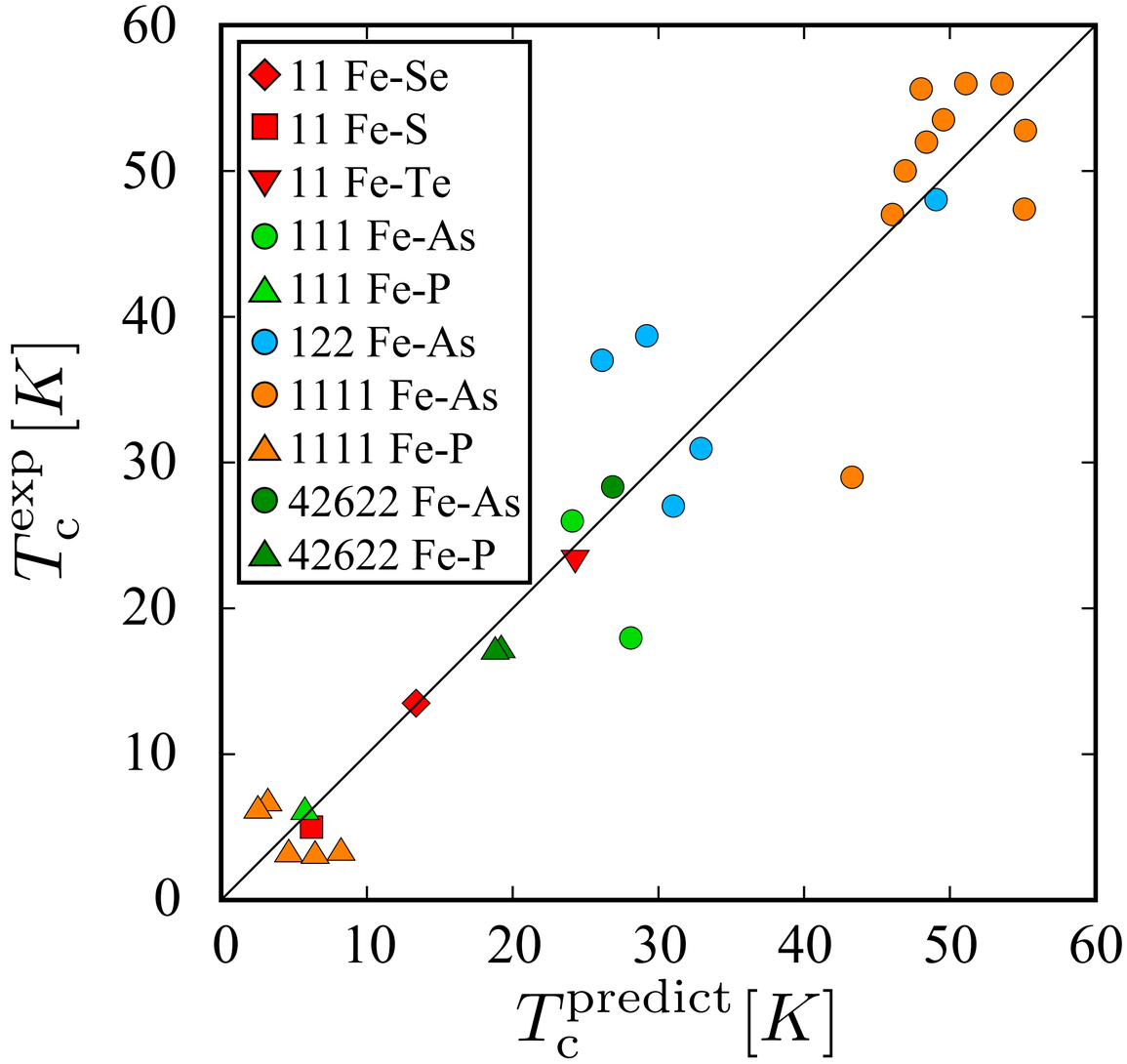
超伝導転移温度を再現・予測する回帰モデルの作成

超伝導転移温度を予測する回帰モデル

Tc予測の回帰モデル

[Tc = f(U, V, t, J, U/V…)]

$$f(\boldsymbol{z};\boldsymbol{w}) = w_0 + \sum_i z_i w_i$$



実験の超伝導転移温度をよく再現する回帰モデルの作成に成功

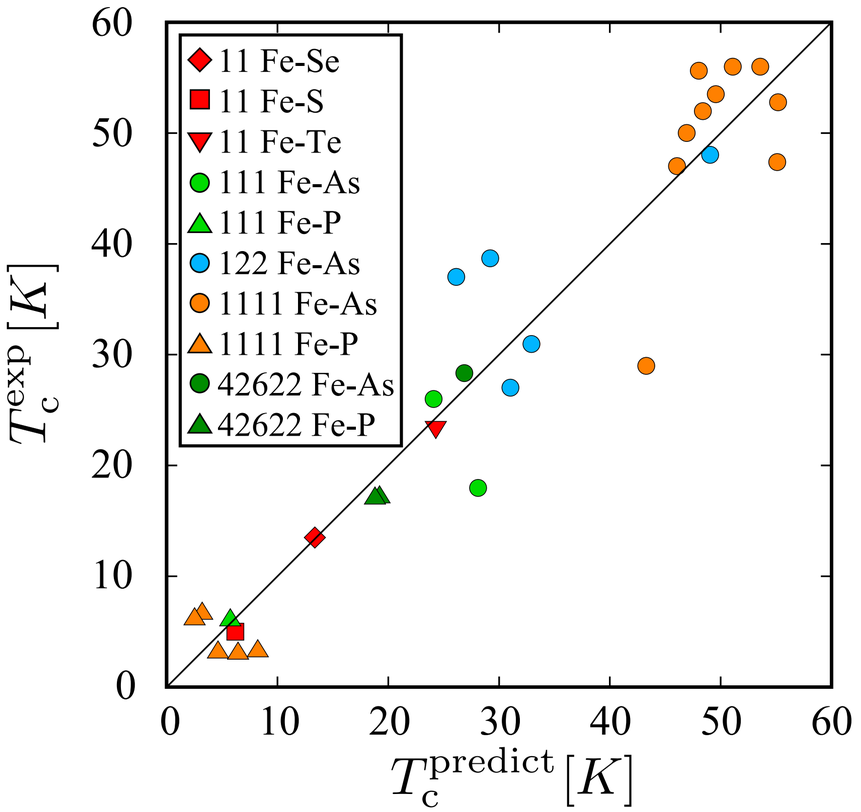
超伝導転移温度上昇の指針提示

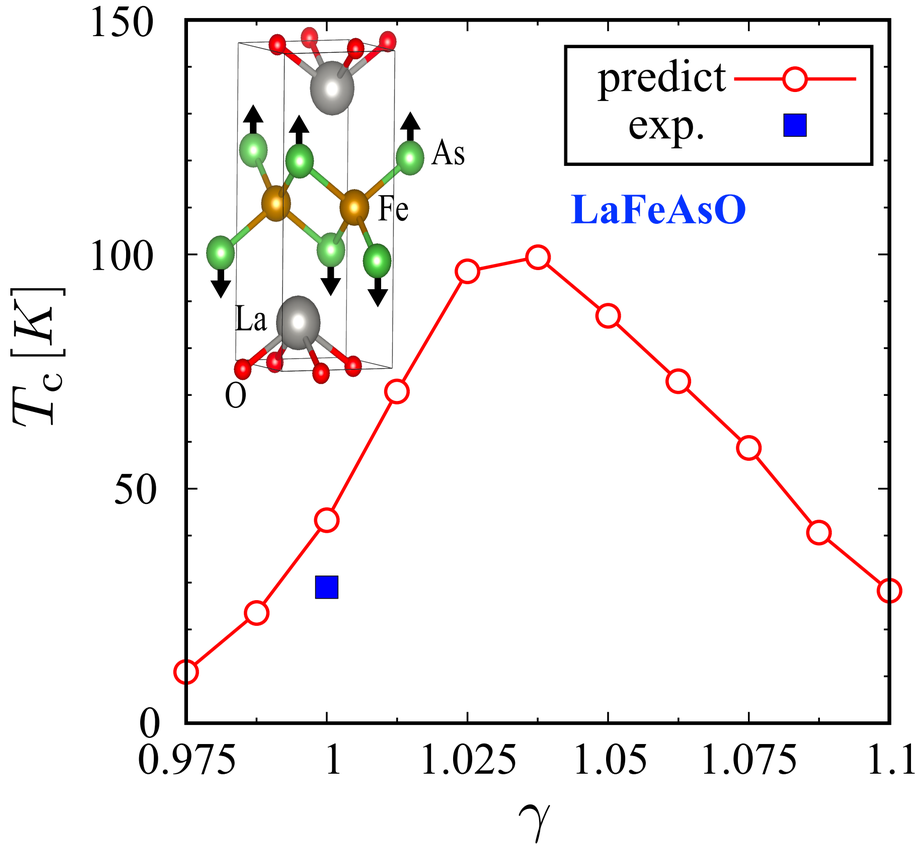
Tc予測の回帰モデル

[Tc = f(U, V, t, J, U/V…)]

仮想物質の有効模型導出

→回帰モデルを用いたTc予測





Tcを予測する学習器を用いて、転移温度をあげる指針を模索

→Asの高さを制御することで転移温度上昇の可能性

[レーザー照射での実現可能性]

K. Kuroki et al., PRB (2009)

FeSeへのレーザー照射

T. Suzuki et al., Commun. Phys. (2019)

Summary

強相関第一原理計算の適用例として、以下の例について述べた

- β’-*X*[Pd(admit)$_2$]$_2$ における量子スピン液体の解析

T. Misawa, K. Yoshimi, and T. Tsumuraya, Phys. Rev. Research **2**, 032072(R) (2020)
K. Yoshimi, T. Tsumuraya, and T. Misawa, Phys. Rev. Research **3**, 033224 (2021)
K. Ido, K. Yoshimi, T. Misawa, and M. Imada, npj Quantum Mater. **7**, 48 (2022)

-鉄系超伝導体の有効模型のパラメータと超伝導転移温度の関係の解析

K. Ido, Y. Motoyama, K. Yoshimi, and T. Misawa, J. Phys. Soc. Jpn. 92, 064702 (2023)

ソフトウェアの整備によって、物質の結晶構造(cifファイル)を
もとに、強相関第一原理計算を網羅的に行うことが可能に
なりつつある
→新奇現象の記述子を同定することで新材料提案へ!