Data analysis on \textit{ab initio} effective Hamiltonians of iron-based superconductors

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High-temperature superconductivity occurs in strongly correlated materials such as copper oxides and iron-based superconductors. Numerous experimental and theoretical works have been done to identify the key parameters that induce high-temperature superconductivity. However, the key parameters governing the high-temperature superconductivity remain still unclear, which hamper the prediction of superconducting critical temperatures ($T_c$) of strongly correlated materials. Here by using data-science techniques, we clarified how the microscopic parameters in the \textit{ab initio} effective Hamiltonians correlate with the experimental $T_c$s in iron-based superconductors. We showed that a combination of microscopic parameters can characterize the compound-dependence of $T_c$ using the principal component analysis. We also constructed a linear regression model that reproduces the experimental $T_c$ from the microscopic parameters. Based on the regression model, we showed a way for increasing $T_c$ by changing the lattice parameters. The developed methodology opens a new field of materials informatics for strongly correlated electron systems.

I. INTRODUCTION

The discovery of the high-$T_c$ superconductivity in the copper oxides has inspired a huge amount of studies to clarify the relation between electronic correlations and high-$T_c$ superconductivity\cite{1}. In 2008, the discovery of another high-$T_c$ superconductivity in iron-based superconductors renewed the interest in the superconductivity induced by the electronic correlations\cite{2}. Theoretical and experimental studies of these high-$T_c$ superconductors have revealed that electronic correlations such as Coulomb interactions are key factors that stabilize the high-$T_c$ superconductivity. However, it remains unclear how the electronic correlations correlate with $T_c$.

Recent theoretical progress enables us to determine the microscopic parameters in the low-energy effective Hamiltonians of solids in an \textit{ab initio} way. The method is often referred to as the \textit{ab initio} downfolding method\cite{3}. Through this method, based on the band structures obtained from the density functional theory (DFT) calculations, screened interactions (e.g., Coulomb interactions) in the low-energy effective Hamiltonians are calculated using the constrained random phase approximation (cRPA) method\cite{4}. By solving the low-energy effective Hamiltonians, we can evaluate the physical properties of strongly correlated materials in a fully \textit{ab initio} way beyond the DFT calculations. It has been shown that the scheme can reproduce the ground-state phase diagrams of the high-$T_c$ superconductors\cite{5,6}.

The combination of an \textit{ab initio} downfolding scheme and an accurate analysis of the derived Hamiltonians is useful for clarifying the mechanism of the high-$T_c$ superconductivity. However, application of this method has been limited to a few compounds due to the high numerical cost associated with the accurate analysis of the effective Hamiltonians. The difficulty of solving the effective Hamiltonians prevents us from gaining a deep understanding of the high-$T_c$ superconductivity and designing new high-$T_c$ superconductors. Therefore, to predict new superconductors based on the theoretical calculations, development of an alternative and efficient method without solving \textit{ab initio} Hamiltonians is desirable.

Data-driven approaches, such as high-throughput screening, regression analysis, and multivariate statistics, have been used for discovering and designing new compounds with high functionality\cite{7}. Stimulated by the recent progress of data-science, data-driven analyses focused on superconductors have been performed\cite{8–15}. For example, in Ref.\cite{11}, utilizing the existing database, the authors constructed regression models for predicting $T_c$ based on the information of the chemical-composition data of the compounds and the DFT calculations. Their analysis offers several important insights into the relation between the chemical composition and $T_c$. However, the effects of electronic correlations in the compounds have not been directly considered in the existing database. Electronic correlations are key factors that stabilize the high-$T_c$ superconductivity and hence the analysis of datasets that explicitly include the information about the electronic correlations is desirable.

In this paper, to elucidate the relation between the compound dependence of $T_c$ and the microscopic parameters, we derive the low-energy effective Hamiltonians for 32 iron-based superconductors and 4 related compounds. These superconductors offer an ideal platform for examining the relation between $T_c$ and the correlation effects because $T_c$ and the strength of the correlations largely depend on the compounds.

Based on the obtained low-energy effective Hamiltonians, rather than directly solving the Hamiltonians, we analyze the relations between the experimentally observed $T_c$ and the microscopic parameters comprising these Hamiltonians. This analysis is performed using the data-science techniques such as principal component analysis (PCA) and construction of a linear regression model. Through the PCA, we find that the compound dependence is well characterized by the first and second principal components and that the first (second) comp-
ment consists mainly of the Coulomb interactions (hopping parameters). For example, we find that 1111 compounds have a similar first component and the difference in these compounds is characterized by the second component. We also show that the compound dependence of $T_c$ with respect to the second principal component is manifested as a dome structure. This result indicates that the microscopic parameters comprising the effective Hamiltonians provide sufficient information for describing the compound dependence.

We also construct a linear regression model, which reproduces the experimentally observed $T_c$ from the microscopic parameters comprising the effective Hamiltonians. The obtained model succeeds in reproducing the materials dependence of $T_c$, as evidenced by a high coefficient of determination ($R^2 \sim 0.92$). We show that our regression model can predict a way to enhance the $T_c$ of LaFeAsO by changing the structure of the material.

This paper is organized as follows: In Sec. II we denote the basics of the target materials and explain the ab initio downfolding method. In Sec. III A we show the compound dependence of the microscopic parameters including the electronic correlations in the obtained low-energy effective Hamiltonians. In Sec. III B we perform PCA aimed at identifying the main parameters that describe the iron-based superconductors using the obtained Hamiltonians. We also construct a regression model that reproduces $T_c$ from the microscopic parameters. Based on this model, we show a way to optimize $T_c$ of the iron-based superconductors. A summary and issues that will be addressed in future work are presented in Sec. IV.

II. METHOD

In this section, we briefly explain the numerical methods we employed, namely the ab initio downfolding method, the PCA, and the construction of a linear regression model.

A. Ab initio downfolding method

The DFT calculations of the target materials are performed using QUANTUM ESPRESSO [10]. As the pseudopotential set, we use sg15 library [17], which includes the optimized norm-conserving Vanderbilt pseudopotentials (ONCVPSP) [15]. We replace the pseudopotentials for Gd, Nd, Pr, Ce, and Sm, and Tb with that of La to eliminate the $f$ metallic bands around the Fermi energy. We use a $12 \times 12 \times 12$ k-mesh in the first Brillouin zone. The optimized tetrahedron method is employed for the DFT calculations [15]. An energy cutoff of 120 (480) Ry is set for plane waves (charge densities).

To derive effective Hamiltonians for our target materials, we use RESPACK [20]. In this work, we construct effective models that are the ten-orbital models consisting of 3d orbitals associated with transition metals (TMs), e.g., Fe, Mn, and Ni. The Hamiltonians are given as follows.

$$ H = H_{1body} + H_{2body}, $$

$$ H_{1body} = \sum_{i,j} t_{i,j,\sigma} c_{i\sigma}^\dagger c_{j\sigma}, $$

$$ H_{2body} = \frac{1}{2} \sum_{i,j,\sigma,\rho} \left( V_{ij} c_{i\sigma}^\dagger t_{jp} c_{j\rho} c_{i\sigma} + c_{i\rho}^\dagger t_{jp} c_{j\sigma} c_{i\sigma} \right), $$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) denotes the creation (annihilation) operator of an electron with spin $\sigma$ at the $i$-th maximally localized Wannier function (MLWF) [21] [22]. The indices of the MLWFs include the position of the unit cell $R$, the orbital degrees of freedom $\mu$ and the index of the TM $n$, i.e., $i = (R, \mu, n)$. Note that the unit cell contains two TMs. $t_{i,j,\sigma}$ in the one-body part of the effective Hamiltonians ($H_{1body}$) represents the hopping parameters of an electron with spin $\sigma$ between the $i$-th and $j$-th MLWFs. When we construct the MLWFs, the $x$-$y$ axis is rotated by 45° from the $a$-$b$ axis of the unit cell. For this condition, the $x$-$y$ axis is parallel to the nearest TM-TM directions. Two-body interactions such as Coulomb interactions ($V_{ij}$) and the exchange interactions ($J_{ij}$) are obtained via cRPA calculations [4]. We set an energy cutoff of 20 Ry for the dielectric function and calculate dielectric functions using more than 100 bands. In both calculations, we use $6 \times 6 \times 6$ k points for the 122 systems and $8 \times 8 \times 4$ k points for the others.

For the 122 family, an ab initio model including $d$ orbitals of alkaline earth metals such as Ba is more appropriate (than other models) because these bands are located above but near the Fermi level [23]. Nevertheless, using 10 $d$ orbitals in TM atoms, we can construct the MLWFs that reproduce the DFT band structure below the energy associated with the $d$ band of alkaline earth metals.

Using the derived microscopic parameters, we employ the following typical parameters as descriptors for the data analysis,

$$ t_{p,q} = g_p \left( t_{(0,\mu,0),(0,\nu,1)} \cdot \Delta_q(\mu, \nu) \right), $$

$$ t'_{p,q} = g_p \left( t'_{(0,\mu,0),(R,\nu,0)} \cdot \Delta_q(\mu, \nu) \right), $$

$$ U_{p,q} = g_p \left( V_{(0,\mu,0),(0,\nu,0)} \cdot \Delta_q(\mu, \nu) \right), $$

$$ V_{p,q} = g_p \left( V_{(0,\mu,0),(0,\nu,1)} \cdot \Delta_q(\mu, \nu) \right), $$

$$ V'_{p,q} = g_p \left( V'_{(0,\mu,0),(R,\nu,0)} \cdot \Delta_q(\mu, \nu) \right), $$

$$ J'_{p,q} = g_p \left( J'_{(0,\mu,0),(0,\nu,1)} \cdot \Delta_q(\mu, \nu) \right), $$

where $R_a$ is a primitive translation vector along the $a$ axis. $\Delta_q(\mu, \nu)$ specifies orbital information; $\Delta_{q_{\text{off}}}(\mu, \nu) = 1$, $\Delta_{q_{\text{off}}}(\mu, \nu) = \delta_{\mu, \nu}$, and $\Delta_{q_{\text{off}}}(\mu, \nu) = 1 - \delta_{\mu, \nu}$, $g_p$ is a function for extracting a feature of the derived microscopic parameters among all combinations of $\mu$ and $\nu$. We use three types of $g_p$: a function that returns the maximum value ($p = \text{max}$), a function that returns the mean
value (p=mean), and a function that returns the minimum value (p=min). We exclude $t_{\text{min}}$ and $t'_{\text{min}}$ because these are zero. Throughout this paper, $t_{\text{max}} = t_{\text{max}, \text{all}}$, $U = U_{\text{mean}, \text{diag}}$, $V = V_{\text{mean}, \text{diag}}$, and $J = J_{\text{mean}, \text{offdiag}}$ for simplicity.

B. PCA

It is expected that the difference in the parameters of the derived effective Hamiltonians can characterize various target materials. To verify this hypothesis, we visualize the relationship between the parameters and extract the main parameters that govern the materials dependence via PCA. The PCA, which has been applied to a dataset obtained for superconductors\textsuperscript{24}, leads to a reduction in the dimensionality of a dataset, thereby providing information on the principal components of the target dataset.

Here, we briefly explain basics of the PCA \textsuperscript{25}. Using the descriptor vector $x^{(n)}$ of the $n$-th data (where $n$ corresponds to the index of materials), we construct the covariance matrix $C$, which is defined as

$$C_{ij} = \frac{1}{N-1} \sum_{n} x^{(n)}_i x^{(n)}_j,$$

where $N$ is the total number of the materials. We note that the average value of the descriptors over the materials is set to 0 ($\sum_n x^{(n)}_i = 0$) and its standard deviation $\sqrt{\sum_n \left(x^{(n)}_i\right)^2}/N$ is set to 1. By diagonalizing $C$, we obtain the principal vectors $v^m$ with the principal values $\lambda_m$ (where $\lambda_m$'s are sorted in descending order). The first principal vector $v^1$ represents the direction associated with the most diverse data. Using the principal vectors $v^m$, we can project the descriptor vectors onto the principal vectors and obtain the $m$th principal value $y^{(n)}_m$ as follows,

$$y^{(n)}_m = \sum_i x^{(n)}_i v^m_i. \tag{11}$$

In Sec. III B we show that the materials dependence can be characterized by the principal values.

C. Construction of regression model

We construct a linear regression model $f$ for reproducing $T_c$ from the parameters of the effective Hamiltonian,

$$f(x; w) = w_0 + \sum_{i=1}^{N_s} x_i w_i. \tag{12}$$

To introduce non-linearity, we inserted the quadratic terms and the ratio terms (such as $t_{\text{max}} U$ and $V/U_{\text{max}}$) between the original parameters. (please see Appendix A for further details of the descriptors).

The number of samples in our dataset is comparable to that of the descriptors, and hence we should carefully control the number of the descriptors in the regression models to avoid overfitting. Therefore, we prepared models where some descriptors were dropped in a systematic way — for example, one model has only hopping parameters as descriptors, another one has hopping and onsite-Coulomb interactions, and yet another one has hopping, onsite-Coulomb and the cross term between these parameters (for details, see Appendix A). To identify the optimal model among these models, we calculate the scores associated with each model. One of the simplest ways to estimate score is the hold-out validation method [Fig. 1 (a)]. For this method, we first split the original dataset (white boxes in Fig. 1) into two parts, the training dataset (blue boxes) and the validation dataset (red boxes). The model is optimized by using the training dataset and a training algorithm with the hyperparameter $\alpha^{(m)}$. The score of the model $m$ is calculated as follows,

$$S^{(m)} = \sum_{n \in D_{\text{validation}}} \frac{\left| T_c^{(n)} - f(x^{(n)}; w^{(m)}) \right|^2}{N_{\text{validation}}}, \tag{13}$$

where $D_{\text{validation}}$ is the validation dataset, $T_c^{(n)}$ is the experimental $T_c$ of the material labeled with $n$ and $N_{\text{validation}}$ is the number of the validation data. The model with a minimum score is chosen as the optimal model. While the hold-out validation method has a bias from how to split the dataset, the cross-validation (CV) method reduces it. In the CV method [Fig. 1 (b)], we split the dataset into $k$ blocks, and designate $k$ pairs of the training data ($k-1$ blocks) and the validation data (one block). The final score $S^{(m)}$ is calculated by averaging scores of all the pairs, $S^{(m)} = \frac{1}{k} \sum_{i=1}^{k} S^{(m)}$. Our dataset is small, and therefore we adopt $k = N$ (so called leave-one-out CV, LOOCV).

In training, we optimize the weights of each model using LASSO (least absolute shrinkage and selection operator) to reproduce the compound dependence of $T_c$. LASSO has one hyperparameter $\alpha$, which is a weight for $L_1$ regularization. In order to optimize the $\alpha$ value of each model and thereby lower the score, we use the LOOCV method again [nested CV \textsuperscript{26}, Fig. 1 (c)]. In this (inner) LOOCV, the training dataset with $N - 1$ samples is split further into $N - 2$ samples for training and one sample for validation. We perform LASSO using scikit-learn\textsuperscript{27}. In addition, we use Optuna\textsuperscript{28} to optimize $\alpha$ by means of the Bayesian optimization method.
FIG. 1. (color online) Schematic picture of the validation methods. (a) Hold-out validation, which is a basic method for validating. In this method, we split the original data (white rectangle) into the training data (blue) and the validation data (red). Each model $m$ is optimized by the training dataset and the training algorithm with the hyperparameter $\alpha^{(m)}$. Once the model is optimized, the score of the model $S^{(m)}$ is calculated by using the validation data. (b) $k$-fold cross-validation (CV), which reduces the bias associated with splitting the data using the hold-out validation method. (c) Nested CV, where the method used in the present study. The training algorithm of a model from a training dataset has a hyperparameter, $\alpha^{(m)}$; we fix the value of this parameter. To choose the optimal value, we again apply the CV method (the inner CV) for each training dataset.

III. RESULTS

A. Ab initio results

We obtain the low-energy effective Hamiltonians for the 11-family (e.g., FeSe), 111-family (e.g., LaFeAsO), 11-family (e.g., LiFeAs), 122-family (e.g., BaFe$_2$As$_2$), and 42622-family (e.g., Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$). For comparison with the iron-based superconductors, we have also obtained the low-energy effective Hamiltonians for Mn (e.g., BaMnAsF) and Ni (e.g., LaNiAsO) analogs of the iron-based superconductors. Each target material is categorized as one of the above-mentioned families based on the chemical composition of the materials.

The typical microscopic parameters (such as the maximum values of the hopping parameters and the averaged onsite-Coulomb interactions) comprising the low-energy effective Hamiltonians and the families of our target compounds are listed in Table I. The numerical conditions of the $ab$ initio calculations are detailed in Sec. II A. The crystal structures of all the target materials except for Ca$_4$Al$_2$O$_6$Fe$_2$P$_2$, GdFePO and NdFePO are obtained from the experiments. Most of them are obtained from the Inorganic Crystal Structure Database (ICSD) and SpringerMaterials. We note that we employ a tetragonal crystal phase at ambient pressure. For calculations of Ca$_4$Al$_2$O$_6$Fe$_2$P$_2$ (GdFePO and NdFePO), the fractional coordinates of the atoms are used as the same as those of Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$(PrFePO), and the lattice constant is employed from the experiment.

We find that the amplitudes of the hopping parameters and the Coulomb interactions exhibit a strong dependence on the compounds. For example, $U/t_{\text{max}}$ of FeSe ($U/t_{\text{max}} = 10.11$) is approximately twice that of GdFePO ($U/t_{\text{max}} = 5.15$). This significant variation indicates that iron-based superconductors offer an ideal platform for examining the relation between $T_c$ and the microscopic parameters. We note that the overall tendency of the obtained parameters is consistent with the tendency revealed by previous calculations. In particular, the averaged Coulomb interaction $U$ values agree (to within 10%) with those obtained in the previous studies, except for BaFe$_2$As$_2$. The $U$ value of BaFe$_2$As$_2$ is ~20% smaller than that reported in the previous study. This deviation may have resulted from the difference between the entanglement treatment for the Ba $d$ band near the Fermi level. We neglect clarification of this difference because (in the present study) we focus on the relation between the $T_c$ and overall compound dependence of the microscopic parameters.

In Fig. 2 (a-d), we plot the compound dependence of the typical microscopic parameters. We find that the effective onsite interaction $U/t_{\text{max}}$ value of the target materials varies significantly (from five to ten). A significant variation was also noted in Ref. 32 where six iron-based superconductors were analyzed. The Hund coupling $J$ is also dependent on the materials, although its range is narrower than that of $U$. From the results obtained for the 1111 family, the compounds can be classified into three categories, namely 1111 compounds with P (1111-P), with As (1111-As), and with F or H (1111-F,H). We find that stronger effective Coulomb interactions $U/t_{\text{max}}$ occur in 111-F,H than in the other 1111 compounds, owing to the larger $U$ compared with those of 1111-P and 1111-As. However, the similarity between the $U/t_{\text{max}}$ values of 1111-P and 1111-As stems from transfer integrals associated with the
### Table I. Compound-dependence of \textit{ab initio} parameters for target materials.

$t_{\text{max}}$ denotes the maximum value of the hopping parameters between the nearest-neighbor transition metals (TMs). $\overline{U}$ and $\overline{J}$ denote the averaged values of the diagonal onsite Coulomb interactions and the onsite Hund coupling over the TM $d$ orbitals, respectively. $\overline{V}$ is the average value of the diagonal density-density interactions between the nearest-neighbor TM sites. The unit of the interaction and hopping parameters is given in eV. The effective interactions $\overline{U}/t_{\text{max}}$, $\overline{V}/t_{\text{max}}$, and $\overline{J}/t_{\text{max}}$ are also listed. Ref shows the reference of the crystal structures we employed.

| Material family | $t_{\text{max}}$ (eV) | $\overline{U}$ (eV) | $\overline{V}$ (eV) | $\overline{J}$ (eV) | $\overline{U}/t_{\text{max}}$ | $\overline{V}/t_{\text{max}}$ | $\overline{J}/t_{\text{max}}$ | Ref |
|-----------------|------------------------|----------------------|----------------------|------------------------|------------------------|------------------------|------------------------|-----|
| FeS | 11 | 0.46 | 4.53 | 1.20 | 0.55 | 9.91 | 2.63 | 1.21 | [34] |
| FeSe | 11 | 0.41 | 4.14 | 1.04 | 0.55 | 10.11 | 2.53 | 1.33 | [35] |
| FeTe | 11 | 0.37 | 3.15 | 0.73 | 0.50 | 8.57 | 1.98 | 1.37 | [36] |
| CaFeAsH | 1111 | 0.37 | 2.71 | 0.70 | 0.44 | 7.26 | 1.88 | 1.19 | [37] |
| SrFeAsF | 1111 | 0.37 | 3.13 | 0.88 | 0.47 | 8.44 | 2.38 | 1.26 | [38] |
| CaFeAsF | 1111 | 0.38 | 3.01 | 0.87 | 0.40 | 7.96 | 1.98 | 1.37 | [39] |
| LaFePO | 1111 | 0.43 | 2.40 | 0.65 | 0.40 | 5.59 | 1.51 | 0.92 | [40] |
| PrFePO | 1111 | 0.43 | 2.44 | 0.65 | 0.40 | 5.62 | 1.51 | 0.93 | [41] |
| NdFePO | 1111 | 0.44 | 2.44 | 0.66 | 0.40 | 5.51 | 1.49 | 0.90 | [41] |
| SmFePO | 1111 | 0.45 | 2.44 | 0.66 | 0.40 | 5.45 | 1.47 | 0.89 | [41] |
| GdFePO | 1111 | 0.47 | 2.40 | 0.66 | 0.39 | 5.15 | 1.41 | 0.84 | [41] |
| LaFeAsO | 1111 | 0.36 | 2.46 | 0.61 | 0.43 | 6.90 | 1.72 | 1.19 | [2] |
| CeFeAsO | 1111 | 0.36 | 2.49 | 0.62 | 0.43 | 6.88 | 1.72 | 1.18 | [42] |
| PrFeAsO | 1111 | 0.36 | 2.50 | 0.62 | 0.43 | 6.91 | 1.72 | 1.19 | [43] |
| NdFeAsO | 1111 | 0.36 | 2.51 | 0.62 | 0.44 | 6.95 | 1.71 | 1.22 | [43] |
| SmFeAsO | 1111 | 0.37 | 2.51 | 0.63 | 0.43 | 6.84 | 1.71 | 1.17 | [43] |
| GdFeAsO | 1111 | 0.37 | 2.52 | 0.63 | 0.44 | 6.73 | 1.67 | 1.16 | [44] |
| TbFeAsO | 1111 | 0.37 | 2.52 | 0.63 | 0.43 | 6.76 | 1.76 | 1.19 | [43] |
| LiFeP | 111 | 0.50 | 3.09 | 0.90 | 0.45 | 6.23 | 1.82 | 0.90 | [45] |
| LiFeAs | 111 | 0.42 | 3.02 | 0.78 | 0.47 | 7.17 | 1.86 | 1.12 | [46] |
| NaFeAs | 111 | 0.39 | 3.06 | 0.79 | 0.48 | 7.86 | 2.02 | 1.23 | [47] |
| KFe$_2$P$_2$ | 122 | 0.34 | 2.43 | 0.61 | 0.45 | 7.20 | 1.81 | 1.32 | [48] |
| SrFe$_2$P$_2$ | 122 | 0.46 | 2.72 | 0.74 | 0.43 | 5.86 | 1.59 | 0.93 | [49] |
| CaFe$_2$P$_2$ | 122 | 0.42 | 2.81 | 0.60 | 0.52 | 6.75 | 1.44 | 1.24 | [50] |
| BaFe$_2$P$_2$ | 122 | 0.45 | 2.51 | 0.64 | 0.43 | 5.56 | 1.42 | 0.95 | [51] |
| KFe$_2$As$_2$ | 122 | 0.47 | 3.07 | 0.94 | 0.46 | 6.59 | 2.03 | 0.98 | [52] |
| SrFe$_2$As$_2$ | 122 | 0.37 | 2.62 | 0.64 | 0.44 | 7.09 | 1.74 | 1.20 | [50] |
| CaFe$_2$As$_2$ | 122 | 0.37 | 2.38 | 0.52 | 0.44 | 6.36 | 1.39 | 1.18 | [53] |
| BaFe$_2$As$_2$ | 122 | 0.36 | 2.40 | 0.55 | 0.45 | 6.59 | 1.50 | 1.22 | [54] |
| Sr$_4$Sc$_2$O$_6$Fe$_2$P$_2$ | 42622 | 0.41 | 2.99 | 0.86 | 0.45 | 7.31 | 2.11 | 1.09 | [55] |
| Ca$_4$Al$_2$O$_6$Fe$_2$P$_2$ | 42622 | 0.42 | 3.00 | 0.93 | 0.45 | 7.10 | 2.21 | 1.07 | [56] |
| Ca$_4$Al$_2$O$_6$Fe$_2$As$_2$ | 42622 | 0.41 | 2.95 | 0.90 | 0.45 | 7.12 | 2.16 | 1.09 | [56] |
| BaMnAsF | Mn | 0.29 | 2.75 | 0.77 | 0.45 | 9.43 | 2.64 | 1.55 | [57] |
| BaMn$_2$As$_2$ | Mn | 0.31 | 2.33 | 0.57 | 0.43 | 7.47 | 1.82 | 1.36 | [53] |
| BaNi$_2$As$_2$ | Ni | 0.42 | 2.81 | 0.48 | 0.47 | 6.64 | 1.15 | 1.11 | [59] |
| LaNiAsO | Ni | 0.39 | 2.81 | 0.61 | 0.44 | 7.11 | 1.55 | 1.11 | [60] |

A similar tendency is observed for the 111 family.

The \textit{ab initio} parameters of the relevant compounds are similar to those of iron-based superconductors. Mn compounds have strong effective interactions $\overline{U}/t_{\text{max}}$, which arise from the smaller hopping integrals than those of other compounds. We note that their occupation numbers in one TM atom are different from those of iron-based superconductors.

Before performing the data-science analyses such as PCA, we examine the influence of the lattice structure on the materials dependence of the microscopic parameters. The importance of the anion [chalcogen (Ch) or pnictide (Pn)] position is pointed out in the previous
FIG. 2. (color online) Ab initio parameters for iron-based superconductors and the related compounds. These definitions are explained in the caption of Table I. (a-d) Compound-dependence of ab initio parameters for (a) $U/t_{\text{max}}$, (b) $U$, (c) $t_{\text{max}}$, and (d) $\tilde{J}$. Colors represent the family name: red bars are the results for the 11 family, orange bars are the results for the 1111 family, light-green bars are the results for the 111 family, green bars are the results for the 42622 family, light-blue bars are the results for the 122 family, blue bars are the results for the Mn compounds, and purple bars are the results for the Ni compounds. (e-h) Ab initio parameters as a function of the distance of TM from Ch/Pn. Legends denote the kinds of TM and Ch/Pn of the target materials. Filled, open and cross symbols represent the results of Fe, Ni and Mn compounds, respectively. For the classification of the family name, the same colors of the symbols are employed as those in the panels (a-d). $l$ is the distance between TM and Ch/Pn, which is schematically shown in the inset in panel (g). Gray thick lines in panels (e) and (g) are guides for the eye.

studies [61, 62], and hence we plot the typical microscopic parameters as a function of the distance between Ch/Pn and TM [see Fig. 2(e-h)]. As shown in Fig. 2(e), the magnitude of the effective onsite Coulomb interaction
\( \overline{T}/t_{\text{max}} \) increases with increasing distance. As shown in Fig. 2 (f) and (g), we find that this trend is mainly governed by \( t_{\text{max}} \), i.e., \( t_{\text{max}} \) decreases almost monotonically with increasing the distance whereas \( \overline{T} \) is almost independent of the distance. This result indicates that the difference between the distances controls \( t_{\text{max}} \) mainly through the hybridization between TM \( d \) orbitals and Ch/Pn \( p \) orbitals. We note that \( \overline{T} \) exhibits no clear distance dependence.

B. Data analysis

By using the effective Hamiltonians reported in the previous section, we perform the PCA and the regression analysis for \( T_c \) of iron-based superconductors. We focus on the superconductivity of the iron-based superconductors in this study, since the \( T_c \) values of Ni compounds are very low, and Mn compounds exhibit no superconductivity. The results obtained from our data analysis are summarized in Table III. We provide details of the PCA analysis and the construction of the regression model in subsequent sections.

1. PCA

For performing the PCA, we employ 36 microscopic parameters as the descriptors, which consist of the Coulomb interactions \( (V_{ij}) \), the Hund couplings \( (J_{ij}) \), and the hopping parameters \( (t_{ij}) \). We obtain \( \lambda_1 = 25.06, \lambda_2 = 7.94, \lambda_3 = 2.12, \lambda_4 = 1.20, \) and \( \lambda_5 = 0.37 \) as the five dominant principal values. The first and the second principal values are large compared with the other principal values, and hence it is expected that the first and the second principal vectors \( (\mathbf{v}^1_i \) and \( \mathbf{v}^2_i \)) accurately reflect the material dependence. In Fig. 3(a), we plot the absolute value of each component comprising these vectors. We find that the interaction parameters (such as onsite and offsite Coulomb interactions) govern the first principal vector while the hopping parameters and Hund couplings govern the second principal vector.

We plot the first and the second principal components \( (y^{(1)}_i \) and \( y^{(2)}_i \)) in Fig. 3(b). For 1111 and 111 compounds, we find that the second component characterizes the differences within families, i.e., the same family has a similar first component, but each compound in the family has a different second component. For example, the first component of the 1111 family is approximately four, but the second component varies from -3.2 to 6.0. In contrast, we find that 122 and 42622 compounds exhibit opposite tendencies: the same family has a similar second component, but each compound has a different first component. We note that the first component changes significantly by changing the total electron density for 1111 and 122 families.

The 11 family exhibits exceptional behavior, i.e., both the first and the second components exhibit considerable compound dependence. This exceptional behavior may be related to exotic phenomena such as the absence of antiferromagnetic order and the high-\( T_c \) superconductivity.

FIG. 3. (color online) PCA analysis of the \( ab \text{ initio} \) parameters associated with the iron-based superconductors. (a) Each component of the 1st and the 2nd principal vector. The descriptors are sorted in descending order of the absolute value corresponding to the components of the first principal vector, \( |\mathbf{v}^1_i| \). The interaction parameters (onsite and offsite Coulomb interactions) govern the 1st principal vector, while the hopping parameters and Hund couplings govern the 2nd principal vector. The descriptors in the yellow, green, and white regions are related to the Coulomb interactions, the Hund coupling, and the transfer parameters, respectively. See Appendix A for the features of the descriptors for the PCA and the details of the index \( i \). (b) Compound-dependence of the 1st and the 2nd principal component. Notations are the same as in panels (c) and (d). Colored ellipses are guides for the eye. (c) and (d) \( T_c \) as functions of the 1st and the 2nd principal component. Gray thick curves are guides for the eye.
TABLE II. Superconducting transition temperature $T_c$ values of target materials. *Ab initio* effective Hamiltonians are used as descriptors for the construction of the regression model. $T_c^{\exp.}$ and $T_c^{\text{pred.}}$ denote onset $T_c$ observed in experiments and $T_c$ predicted by our regression model, respectively. $y_1$ and $y_2$ are the values of the 1st and 2nd principal components in the PCA, respectively. $(H_A, H_B)$ denotes the linear interpolation of the parameters between $H_A$ and $H_B$, whose ratio corresponds to the stoichiometric ratio of a target material. The dataset contains 29 materials.

| Material          | $T_c^{\exp.}$ (K) | $T_c^{\text{pred.}}$ (K) | $y_1$   | $y_2$   | Hamiltonian                  |
|-------------------|-------------------|--------------------------|---------|---------|-----------------------------|
| FeS               | 11                | 51                       | 6.2     | -14.89  | 2.07                        |
| FeSe              | 11                | 13.5                      | 13.4    | -10.51  | -2.79                       |
| FeSeO$_{0.4}$Te$_{0.6}$ | 11         | 23.5                      | 24.3    | -4.56   | -4.63                       |
| Ca$_{0.77}$La$_{0.23}$FeAsH | 1111     | 47.4                      | 55.1    | 0.98    | -1.33                       |
| Sr$_{0.5}$Sm$_{0.5}$FeAsF | 1111     | 56                        | 51.1    | -3.33   | -2.68                       |
| Ca$_{0.4}$Pr$_{0.6}$FeAsF | 1111    | 56                        | 53.6    | -3.33   | -1.43                       |
| LaFePO            | 1111              | 6.6                       | 6.8     | 3.2     | 3.97                        |
| PrFePO            | 1111              | 3.2                       | 6.8     | 8.2     | 3.54                        |
| NdFePO            | 1111              | 3.1                       | 6.8     | 4.6     | 3.46                        |
| SmFePO            | 1111              | 3.6                       | 6.9     | 6.4     | 3.39                        |
| GdFePO            | 1111              | 6.1                       | 7.0     | 2.5     | 3.33                        |
| LaFeAsO$_{0.92}$H$_{0.08}$ | 1111    | 29                        | 71      | 4.33    | 4.60                        |
| CeFeAsO$_{0.75}$H$_{0.25}$ | 1111   | 47                        | 72      | 46.1    | 4.15                        |
| PrFeAsO$_{0.89}$F$_{0.11}$ | 1111    | 52                        | 73      | 48.4    | 4.05                        |
| NdFeAsO$_{0.8}$F$_{0.2}$ | 1111    | 52.8                      | 74      | 55.2    | 3.85                        |
| SmFeAsO$_{0.91}$F$_{0.093}$ | 1111  | 55.6                      | 75      | 48.1    | 3.61                        |
| GdFeAsO$_{0.85}$ | 1111              | 53.7                      | 76      | 49.5    | 3.26                        |
| TbFeAsO$_{0.9}$F$_{0.1}$ | 1111     | 50                        | 77      | 46.9    | 3.23                        |
| LiFeP             | 1111              | 6                        | 54      | 5.7     | -5.44                       |
| NaFeAs            | 111               | 26                        | 78      | 24.1    | -1.89                       |
| Li$_{0.8}$FeAs    | 111               | 18                        | 79      | 28.1    | -2.60                       |
| SrFe$_2$As$_{1.3}$P$_{0.7}$ | 122       | 27                        | 80      | 31.0    | 2.15                        |
| BaFe$_2$As$_{1.48}$P$_{0.52}$ | 122    | 31                        | 81      | 33.0    | 4.73                        |
| K$_{0.5}$Sr$_{0.5}$Fe$_2$As$_2$ | 122    | 37                        | 82      | 26.1    | -1.17                       |
| Ca$_{0.83}$La$_{0.17}$Fe$_2$As$_{1.48}$P$_{0.12}$ | 122  | 41                        | 83      | 49.0    | 6.17                        |
| Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ | 122    | 38.7                      | 84      | 29.2    | 1.03                        |
| Ca$_{4}$Al$_2$O$_{80}$Fe$_2$P$_2$ | 42622 | 17.1                       | 56      | 19.2    | -5.29                       |
| Sr$_4$Sc$_{2}$O$_{6}$Fe$_2$P$_2$ | 42622 | 17                        | 55      | 18.8    | -2.11                       |
| Ca$_{4}$Al$_2$O$_{65}$Fe$_2$As$_2$ | 42622 | 28.3                      | 56      | 26.9    | -4.37                       |

$^a$ $T_c^{\exp.}$ is estimated from a figure where the temperature dependence of the electronic specific heat is shown.

$^b$ LaFeAsO$_{1-x}$H$_x$ has the two-dome $T_c$ structure. $T_c^{\exp.}$ is used in the maximum $T_c$ of the dome in the low-doping region.

$^c$ It is not fully clarified whether the system shows the bulk superconductivity.

in a monolayer occurring in FeSe$^{[55][57]}$.

In Fig. 3(c) and (d), we plot $T_c$ as a function of the first and the second principal components. We find that $T_c$ is better correlated with the second component than the first component. In particular, we find that the dependence of $T_c$ on the second component is manifested as a dome structure reminiscent of Lee’s plot $^{[61][62]}$. This result indicates that the difference in the microscopic parameters associated with the effective Hamiltonians provides sufficient information for explaining the compound dependence of $T_c$. By constructing the regression model for reproducing $T_c$, we show that these parameters allow quantitative reproduction of this dependence.

2. Regression model for estimating $T_c$

After determining the best regression model within the nested CV approach explained in Sec. II, we obtain our predictor using all the $T_c$ data obtained in the experiments. Figure 4(a) shows the accuracy of the obtained regression model. We find that this model reproduces the experimental $T_c$ data, as indicated by the high coefficient of determination ($R^2 = 0.92$). This result confirms that our regression model can predict the $T_c$ of associated with several different families for iron-based superconductors.

We apply our predictor to low-energy Hamiltonians
for a hypothetical compound, whose lattice structure is systematically changed from that of LaFeAsO. We vary the fractional coordinates of two As atoms as \((0.5, 0.0, \gamma_f As)\) and \((0.0, 0.5, 1-\gamma_f As)\); \(f_{As}\) is the fractional coordinate of As for the LaFeAsO and the parameter \(\gamma\) indicates changes in the height of As from the Fe plane. A schematic showing this lattice distortion is presented in the inset of Fig. 4(b).

We obtain the low-energy effective Hamiltonians for the hypothetical compounds. From the microscopic parameters in the Hamiltonians, we calculate the \(T_c\) of each compound using the regression model. In Fig. 4(b), we show the \(\gamma\) dependence of \(T_c\) for hypothetical LaFeAsO materials. We find that \(T_c\) increases with increasing \(\gamma\) and vice versa. At \(\gamma \sim 1.03\), \(T_c\) reaches \(\sim 100\)K. The applicability range of the regression model is limited to the parameter space around the existing compounds, and therefore the role of lattice distortion in inducing high-\(T_c\) superconductivity \((T_c \approx 100\)K) remains unclear. Nevertheless, the qualitative behavior of \(T_c\) may be correctly predicted by the regression model. This result indicates that our regression model captures the essence of Lee’s plot \([61, 62]\), which suggests that the height of As from the Fe plane plays a key role in determining the compound dependence of \(T_c\).

We also consider the possibility of realizing hypothetical materials. In our \textit{ab initio} calculations of such materials, we do not perform the structure optimization and simulations for phonon’s properties. Therefore, our proposed materials may be unattainable for the conventional bulk system at ambient pressure. Stable materials may be used to obtain an interface structure. Such interface or thin-film structures induce drastic changes in the electronic states of target materials and may enhance \(T_c\). Successful experiments applying this approach have already been reported for high-\(T_c\) superconductors such as La\(_2\)CuO\(_4\) and FeSe \([87, 88]\). The material structure can also be controlled via laser irradiation, which has been successfully applied to FeSe \([89]\). In that work, strong laser irradiation led to changes in the height of Se atoms from the Fe-Fe plane. This results from the displacive excitation of coherent phonon mechanism \([90]\), which governs the excitation of \(A_{1g}\) Raman active modes. These modes also occur in LaFeAsO \([91]\), and hence we expect that our predicted structure would be realized via laser irradiation.

![FIG. 4. (color online) (a) Experimental \(T_c\) vs. predicted \(T_c\) obtained from our regression model. Notations are the same as those of Fig. 2. The black thin line represents \(T_c^{\text{predict}} = T_c^{\exp.}\). The coefficient determination \(R^2\) is about 0.92. Data is also shown in Table II. Note that SrFe\(_2\)As\(_{1.3}\)P\(_{0.7}\) and BaFe\(_2\)As\(_{1.4}P_{0.52}\) are categorized as Fe – As, and Fe\(_{0.4}S\(_{0.6}\) is categorized as Fe – Te. (b) Prediction of \(T_c\) for LaFeAsO with hypothetical structures with different c-axis components of fractional coordinates corresponding to As, \(f_{As}\). \(\gamma\) corresponds to the ratio of \(f_{As}\) associated with the original LaFeAsO \([2]\) to that of the hypothetical materials. Note that only the material for \(\gamma = 1\) has been found in the experiments; the corresponding \(T_c\) is plotted as the blue square. Inset represents the structure of the original LaFeAsO, which is depicted using VESTA \([92]\). Black thick arrows denote the displacement pattern of the Raman-active \(A_{1g}\) phonon.]

IV. SUMMARY

In summary, we have derived the low-energy effective Hamiltonians for 32 iron-based compounds and four related compounds using the \textit{ab initio} downfolding method. We have found that microscopic parameters such as the hopping parameters and the Coulomb interaction are associated with a wide range of iron-based superconductors. To systematically characterize the compound dependence of the microscopic parameters, we perform the PCA. As a result, we find that the first principal vector consists of mainly interaction terms,
while the second consists of mainly hopping terms. We show that the first principal values characterize the difference within the 42622 and 122 families while the second principal values characterize the difference within the 111 and 111 families. Moreover, we show that the compound dependence of $T_c$ with respect to the second principal value is manifested as a dome structure reminiscent of Lee’s plot \[61, 62\]. This result suggests that the obtained microscopic parameters adequately reflect the compound dependence of $T_c$.

Afterward, we construct a regression model for reproducing the compound dependence of $T_c$ from the microscopic parameters of the \textit{ab initio} Hamiltonians. We succeed in reproducing experimental $T_c$ values of the iron-based superconductors ($R^2 \approx 0.92$). Using the regression model, we show that the $T_c$ of LaFeAsO can be enhanced by properly changing the As height from the Fe plane. Such a hypothetical structure would be realized in experiments including laser irradiation processes. Applying pressure can also control the anion height from the Fe plane, and this is accompanied by changes in the lattice constants\[93, 94\]. Future studies will determine whether our regression model reproduces $T_c$ of iron-based superconductors under pressure. Using cutting-edge numerical packages (such as mVMC\[95\]) to directly solve the low-energy effective Hamiltonians in order to determine whether $T_c$ is really enhanced for the hypothetical structures would be an intriguing challenge. However, the numerical cost of such calculations is rather high and hence this analysis will be considered in future studies.

The present work shows that the data-science techniques help us to bypass a difficult problem–solving the \textit{Hamiltonians for quantum many-body systems}– and to clarify the effect of microscopic parameters on superconductivity. Further applications of the developed method to other exotic phenomena (such as the correlated topological materials) may be considered in future work.

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In constructing the regression models, we prepare 72 components of the first principal vector, \(|e_1^2|\).

In Table IV, we list the descriptors used in PCA in descending order of the absolute value corresponding to the components of the first principal vector, \(|e_1^2|\).

In Table V, we list the descriptors used in PCA in descending order of the absolute value corresponding to the components of the first principal vector, \(|e_1^2|\).

Appendix A: Features of Descriptors

Here, we summarize the features of the descriptors used in constructing the regression models and the PCA. In constructing the regression models, we prepare 72 models based on the seven choices shown in Table III.

In addition, all the models include \(t_p, u_l(p=\text{mean}, \text{max})\) and \(U_{p, \text{diag}} (p=\text{mean}, \text{min})\). We standardized these descriptors for the construction of the regression model.
TABLE III. Choices of descriptors used in the regression model. Our choices when constructing the best regression model are shown in the second column.

| Choices of Descriptors | Best model |
|------------------------|------------|
| Normalizing all the descriptors except for $t_{\text{max,all}}$ by $t_{\text{max,all}}$ | True |
| Using square of descriptors (e.g., $U_{\text{mean,diag}}^2$) | True |
| Using cross terms (e.g., $U_{\text{mean,diag}} \times t_{\text{max,all}}$) | True |
| Including $V_{\text{max,diag}}, V_{\text{min,diag}}, V_{\text{mean,diag}}$ | False |
| Including $J_{\text{max,offdiag}}, J_{\text{min,offdiag}}, J_{\text{mean,offdiag}}$ | False |
| Including $J_{\text{mean,diag}}/U_{\text{max,diag}}$ when $V_{\text{mean,diag}}$ is used as the descriptor | – |
| Including $J_{\text{mean,offdiag}}/U_{\text{max,diag}}$ when $J_{\text{mean,offdiag}}$ is used as the descriptor | – |
TABLE IV. Descriptors used in PCA

| $i$ | descriptor                  |
|-----|-----------------------------|
| 0   | $V_{\text{min},\text{diag}}$ |
| 1   | $V_{\text{mean},\text{offdiag}}$ |
| 2   | $V_{\text{mean},\text{diag}}$ |
| 3   | $V_{\text{mean},\text{all}}$ |
| 4   | $V_{\text{max},\text{offdiag}}$ |
| 5   | $V_{\text{max},\text{diag}}$ |
| 6   | $V_{\text{mean},\text{all}}$ |
| 7   | $V'_{\text{min},\text{diag}}$ |
| 8   | $V'_{\text{min},\text{offdiag}}$ |
| 9   | $V'_{\text{mean},\text{diag}}$ |
| 10  | $V'_{\text{mean},\text{all}}$ |
| 11  | $V'_{\text{max},\text{offdiag}}$ |
| 12  | $V'_{\text{max},\text{diag}}$ |
| 13  | $U_{\text{min},\text{offdiag}}$ |
| 14  | $U_{\text{mean},\text{offdiag}}$ |
| 15  | $U_{\text{max},\text{offdiag}}$ |
| 16  | $U_{\text{mean},\text{diag}}$ |
| 17  | $U_{\text{max},\text{diag}}$ |
| 18  | $U_{\text{mean},\text{all}}$ |
| 19  | $U'_{\text{min},\text{diag}}$ |
| 20  | $U'_{\text{mean},\text{diag}}$ |
| 21  | $J_{\text{mean},\text{offdiag}}$ |
| 22  | $J_{\text{max},\text{offdiag}}$ |
| 23  | $t_{\text{mean},\text{offdiag}}$ |
| 24  | $t_{\text{max},\text{diag}}$ |
| 25  | $J_{\text{min},\text{offdiag}}$ |
| 26  | $t_{\text{min},\text{diag}}$ |
| 27  | $t'_{\text{mean},\text{all}}$ |
| 28  | $t'_{\text{max},\text{offdiag}}$ |
| 29  | $t'_{\text{max},\text{diag}}$ |
| 30  | $t'_{\text{mean},\text{offdiag}}$ |
| 31  | $t'_{\text{mean},\text{diag}}$ |
| 32  | $t'_{\text{max},\text{all}}$ |
| 33  | $t'_{\text{mean},\text{all}}$ |
| 34  | $t'_{\text{mean},\text{diag}}$ |

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