Sparse modeling of large-scale quantum impurity models with low symmetries

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Quantum embedding theories provide a feasible route for obtaining quantitative descriptions of correlated materials. However, a critical challenge is solving an effective impurity model of correlated orbitals embedded in an electron bath. Several advanced impurity solvers require the approximation of a bath continuum using a finite number of bath levels, producing a highly nonconvex, ill-conditioned inverse problem. To address this drawback, this study proposes an efficient fitting algorithm for matrix-valued hybridization functions based on a data-science approach, sparse modeling, and a compact representation of Matsubara Green’s functions. The efficiency of the proposed method is demonstrated by fitting random hybridization functions with large off-diagonal elements as well as those of a 20-orbital impurity model for a high-$T_c$ compound, LaAsFeO, at low temperatures. The results indicate that multi-orbital-atom cluster embedding calculations of materials may be feasible using existing computational resources.

Simulating correlated materials is one of the major challenges in the field of condensed matter physics. Local density approximation (LDA) based on the density functional theory has achieved significant success in describing the ground-state properties of many weakly correlated materials. However, LDA fails to describe correlated materials such as Mott insulators and high-$T_c$ superconductors. A naive direct simulation of the first-principles Hamiltonians of correlated materials is not feasible, owing to the exponential scaling of its required computational resources.

In recent times, extensive efforts have been made to use Greens-function-based quantum embedding theories to simulate correlated materials. Quantum embedding theories circumvent the need for exponential scaling by mapping the entire computationally intractable system onto an auxiliary impurity model of correlated orbitals embedded in a bath of noninteracting electrons. Examples of this methodology are the dynamical mean-field theory (DMFT)\textsuperscript{1}, GW+DMFT\textsuperscript{2}, nonlocal extensions of DMFT\textsuperscript{3,4}, and the self-energy embedding theory\textsuperscript{5,6}.

The limitation of these methodologies is in solving impurity models. Quantitative descriptions of correlated materials, such as predicting $T_c$, will require the solution of an impurity model with several spin orbitals ($N_{SO} \gg 10$) and low symmetries. Over the past few years, various sophisticated impurity solvers have been developed. Some examples include impurity solvers based on truncated exact diagonalization\textsuperscript{7,8}, configuration interaction\textsuperscript{9,10}, coupled-cluster theory\textsuperscript{11,12}, matrix product states\textsuperscript{13,14} and tensor networks\textsuperscript{15}. Now, these state-of-the-art algorithms even allow the handling of a few correlated atoms\textsuperscript{13}.

All the above-mentioned impurity solvers rely on approximating a bath continuum with a finite number

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(Color online) (a) Quantum impurity model. The bath is represented by a hybridization function $\Delta(\omega_n)$. (b) Two-step optimization procedure of the present algorithm. (c) Highest-order IR basis functions $V_{i-1}(\omega)$ (red solid curve) and the positions of the initial $\epsilon_b$ (bold and thin solid vertical lines) for $\beta = 100$, $\omega_{\text{max}} = 10$, $N_t = 72$ and $N_{\text{div}} = 5$ [see (b) and the text]. (d) Isosurface of the group LASSO regularization term in Eq. 6.}
\end{figure}

($N_{\text{bath}}$) of bath levels. Self-consistent calculations to determine a bath are stably performed in Matsubara frequencies. However, fitting a hybridization function, which represents a bath, is an ill-conditioned and non-convex inverse problem\textsuperscript{16–19}. This becomes more severe for multiple spin orbitals and low symmetries. Thus,

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an approach to discretize a hybridization function for $N_{SO} \gg 10$ and the size of $N_{bath}$ required for an accurate approximation are yet to be clarified. As a result, advanced impurity solvers cannot be fully utilized.

It is noted that another promising route to solving much larger-scale quantum impurity models is quantum computing [20, 22]. Very recently, preliminary calculations for a single-orbital impurity model have been performed using an IBM quantum computer without fault tolerance [23]. As the number of available qubits increases and noise levels decrease, quantum algorithms may begin to compete with or supersede classical algorithms.

This letter proposes the use of a data-science-based approach, sparse modeling [21, 25], to achieve efficient discretization of large-scale impurity models with low symmetries. Its novelty lies in (i) compactification of discretized models through automatic selection of relevant bath levels by sparse modeling, and (ii) projection to a recently proposed compact basis in Matsubara frequencies, the intermediate representation (IR) basis [26–29]. The efficiency of the proposed algorithm is demonstrated with random hybridization functions for $N_{SO} \gg 10$ with low symmetries. It is shown that for a fixed fitting tolerance, $N_{bath} \propto N_{SO} \ln(\beta W)$, where $W$ is the spectral width and $\beta$ is inverse temperature. Additionally, a realistic five-orbital $2 \times 2$ cluster impurity model for a Fe-based high-$T_c$ superconductor LaFeAsO is analyzed, and $N_{bath}$ is estimated.

**General impurity model**— A general impurity model is defined by the action

$$S_{imp} = \int_0^\beta d\tau H_{loc}(\tau) + \int_0^\beta d\tau d\tau' \sum_{i,j=1}^{N_{SO}} c_i^\dagger(\tau) \Delta_{ij}(\tau - \tau') c_j(\tau'),$$

(1)

where $c_i$ and $c_i^\dagger$ are Grassmann variables for the $i$-th spin orbital in the impurity. The local action $H_{loc}$ acts on the impurity, while all the bath/environment information is encoded in the matrix-valued hybridization function $\Delta_{ij}(\tau)$. If $H_{loc}$ is instantaneous in $\tau$, this action can be regarded as the result of integrating out auxiliary degrees of freedom from a Hamiltonian model,

$$H_{imp} = H_{loc} + \sum_{b=1}^{N_{bath}} \epsilon_b \hat{a}_b^\dagger \hat{a}_b + \sum_{i=1}^{N_{SO}} \sum_{b=1}^{N_{bath}} (V_b \hat{a}_i^\dagger c_b + V_b^* c_i^\dagger \hat{a}_b),$$

(2)

where $\hat{c}$ and $\hat{d}$ are annihilation operators acting on spin orbitals and bath levels, respectively, and $V_b$ and $\epsilon_b$ are bath parameters. The bath levels are assumed to be sufficient to precisely satisfy the equality condition $\Delta_{ij}(\omega_n) = \sum_b V_{b}^* V_b (\omega_n - \epsilon_b)$. Each bath level is hybridized with all the spin orbitals in the impurity [see Fig. 1(b)]. Note that this transformation is not unique.

**Number of parameters required to represent a bath**— We expand a hybridization function as

$$\Delta_{ij}(\omega_n) = \sum_{l=0}^{\infty} \Delta_{ij}(l) U_{ij}^F(\omega_n)$$

(3)

where fermionic IR basis functions $U_{ij}^F(\omega_n)$ depend on $\beta$ and a cutoff frequency $\omega_{max}$ for spectral functions [24] (the notation used in Ref. 28 is utilized herein). $S_F$ denote the singular values of the kernel in the Lehmann representation, being system independent. Because $S_F$ decay super-exponentially, the summation is truncated at $S_F^0/\Delta = 10^{-15}$. $N_{i} = \exp(\ln(\beta W_{loc})/\beta)$ grows only logarithmically with $\beta \omega_{max}$ ($N_i = 40, 72, 104$ for $\beta \omega_{max} = 10^2, 10^3, 10^4$, respectively). This implies that any bath can be represented using $N_{SO}^2 N_i \times N_{SO}^2 \ln(\beta W_{loc})$ parameters alone, regardless of the number of physical degrees of freedom encoded in it. On the other hand, a discretized model involves $N_{bath} + N_{SO} N_{bath}$ parameters. This implies that $N_{bath}$ must scale at least as $O(N_{SO} \ln(\beta W_{loc}))$.

**Efficient discretization algorithm**— The hybridization fitting is a highly non-convex and ill-conditioned inverse problem. To alleviate this, we consider the regularized cost function

$$f(x) = \sum_{n=-\infty}^{\infty} \sum_{i,j=1}^{N_{SO}} |\Delta_{ij}(\omega_n) - \sum_{b=1}^{N_{bath}} V_{ib} V_{bj}^* |\omega_n - \epsilon_b| + \alpha \sum_{b=1}^{N_{bath}} \|v_b\|^2$$

(4)

$$= \sum_{l=0}^{N_{i}-1} \sum_{b=1}^{N_{bath}} |\Delta_{ij}(l)| + \sum_{b=1}^{N_{bath}} V_{ib} V_{bj}^* S_F^0 V_l^F(\epsilon_b) + \alpha \sum_{b=1}^{N_{bath}} \|v_b\|^2$$

(5)

where $N_{bath}$ is the number of initial bath levels, and $\|\cdot\|$ denotes the Frobenius norm, $v_b \equiv (V_{b1}, \cdots, V_{bN_{SO}})^T$, $\alpha > 0$. The fitting parameters are $\epsilon = (\epsilon_1, \cdots)$ and $V = (V_{11}, \cdots, V_{N_{SO} N_{SO}})$. The second term serves to prune redundant bath levels, as will be explained further on. In Eq. (5), the truncation error in Matsubara frequencies is eliminated by transforming the cost function to the IR basis. $V_l^F(\omega)$ is IR basis functions defined in $[-\omega_{max}, \omega_{max}]$. Hereinafter, only cases where $V_{b}^\dagger V_{b}$ are real are considered.

Minimizing Eq. (5) with $\alpha = 0$ would yield an unfavorable solution in which all bath levels would be strongly or weakly coupled to the impurity as $\|v_b\| \neq 0$. Thus, we prefer a sparse solution, in which $\|v_b\| = 0$ for irrelevant bath levels. This is achieved by regularization, which is based on the so-called group least absolute shrinkage and selection operator (group LASSO) [30].

To understand how the group LASSO works, a general underdetermined linear regression problem $y = Ax$ is considered, in which $x$ (data to be fitted) and $y$ (fitting parameters) are vectors of $N$- and $M$-dimensions, respectively. $A$ is an $M \times N$ coefficient matrix ($M < N$). This system has an infinite number of solutions. Its degener-
acy is lifted using a group LASSO regularization term,

\[ \mathbf{x}^* = \arg\min_{\mathbf{x}} \| \mathbf{y} - \mathbf{A}\mathbf{x} \|^2 + \alpha \left( \sqrt{x_1^2 + x_2^2 + x_3^2} \right), \]

where \( N = 3 \) and \( \mathbf{x} = (x_1, x_2, x_3)^T \) are taken for simplicity. Note that \( x_1 \) and \( x_2 \) are grouped, while \( x_3 \) forms another group on its own. This regularization term is the sum of Frobenius norms of vectors consisting of fitting parameters in each group. As shown in Fig. 1(d), an iso-surface of this term has sharp corners and edges, where grouped fitting parameters are either entirely zero or entirely nonzero. The solution of Eq. (6) represents the contact point(s) between an iso-surface and the plane \( \mathbf{y} = \mathbf{A}\mathbf{x} \). Because these solutions lie somewhere on the sharp corners and edges, the group LASSO removes irrelevant fitting parameters in a grouped manner. In the present study, the group LASSO decouples an irrelevant bath level \( b \) by individually removing all \( N_{SO} \) coupling constants \( V_{\text{bath}} \).

Following the above argument on the scaling of \( N_{\text{bath}} \), we prepare \( \mathcal{O}(N_{SO} \ln N) \) poles \( \epsilon_{b} \) based on the distribution of the roots of the highest-order basis function \( V_{F}^{N_{\text{SO}}-1}(\omega) \). \( V_{F}^{l}(\omega) \) has \( l \) roots in the interval of \( [-\omega_{\text{max}}, \omega_{\text{max}}] \), and they are nonuniformly distributed among real frequencies [see Fig. 1(c)]. The cost function (5) depends on \( \epsilon_{b} \) through the IR basis functions alone. If \( \epsilon_{b} \) lies between the wide interval between two neighboring roots, particularly for high frequencies, the value of the cost function is insensitive to a shift of \( \epsilon_{b} \) in this interval, which is a source of the ill-condition. To alleviate this problem, \( N_{\text{bath}}^{b} \) and \( \epsilon_{b} \) are chosen as follows. First, a grid consisting of the boundary points \( \pm \omega_{\text{max}} \) and the roots is constructed. The middle points of two neighboring grid points define the coarser grid shown in Fig. 1(c). \( \epsilon_{b} \) are initialized to these coarse grid points and \( N_{\text{div}} \) equal division points of each interval of the coarse grid [31]. \( N_{\text{div}} = c N_{SO} (c=2–10) \) is taken such that \( N^{b}_{\text{bath}} \gg N^{b}_{\text{bath}} \) after optimization.

The bath parameters \( V_{\text{b}} \) and \( \epsilon_{\text{b}} \) are optimized using the two-step procedure [Fig. 1(b)]. First, only \( V_{\text{b}} \) is optimized with \( \epsilon_{\text{b}} \) fixed at the initial estimate. Although this is still a non-convex optimization, we found that this is empirically stable for \( \alpha > 0 \). The regularization term, based on the so-called group LASSO [31], suppresses \( \| V_{\text{b}} \| \) of some of the bath levels to zero, while maintaining the finiteness of \( \| \epsilon_{b} \| \) of the rest. After convergence, the bath levels that are almost decoupled from the impurity are eliminated. In the second step, \( V_{\text{b}} \) and \( \epsilon_{\text{b}} \) are simultaneously optimized, which generally reduces the value of the cost function less significantly than the first step. Finally, redundant bath levels are removed again. A quasi-Newton method is used for optimization [32]. The algorithm is detailed in Supplemental Material.

Results of random models with low symmetries—First, the present algorithm was benchmarked for an ensemble of random matrix-valued hybridization functions generated as \( \Delta_{ij}(\omega_n) = \frac{1}{\sqrt{N_{\text{b}}}} \sum_{b=1}^{N_{\text{b}}} \tilde{V}_{b}\delta_{ij} V_{\text{bath}} \), where \( \tilde{V}_{ij} \) are drawn from a uniform distribution on \([-1/2, 1/2] \), while \( \tilde{v}_{ij} \) are uniformly and densely distributed in the interval of \([-W/2, W/2] \) with the full spectral width \( W = 2 \). \( N_{\text{b}} \) was taken to be sufficiently large, \( N_{\text{b}} = 10 N_{SO}^{2} \). This model represents gapless and quasi-continuous baths with low symmetries. We took \( \omega_{\text{max}} = 10 (\gg W/2) \) and intro-
The regularization removes the majority of the redundant bath levels in $W/2 (=1) < |\epsilon_b| < \omega_{\text{max}} (=10)$.

Following this, the quality of the fits were assessed. As seen in Fig. 3(b), the fitted model reproduces the complex structures of $\Delta_{\text{IJ}}(l)$ up to $l \approx 8$. Figure 3(a) shows that in Matsubara frequencies, the hybridization function is well-fitted from low to high frequencies, and it does not exhibit overfitting.

Then, the $N_{\text{SO}}$ and $\beta$ dependencies were investigated. By changing $\alpha$, we can estimate the minimum number of bath levels $N_{\text{bath}}$ required to reach the threshold $\delta_{\text{th}} = 10^{-3}$ for the relative residual norm. Ten samples were taken at each parameter. Figure 3(a) shows that the required $N_{\text{bath}}$ grows approximately linearly with $N_{\text{SO}}$ for a fixed $\beta$, whereas it grows slowly with $\beta$. The scaling plot in Fig. 3(b) strongly supports the expected scaling relation

$$N_{\text{bath}}(\delta_{\text{th}}) \sim cN_{\text{SO}} \ln \beta W$$  \hspace{1cm} (7)$$

with $c \approx 2.5$. Figure 3(c) plots the data points for all values of $\alpha$, $\beta$, and $N_{\text{SO}}$. The error vanishes exponentially with $N_{\text{bath}}$. This also indicates the logarithmic dependence of $c$ on $\delta_{\text{th}}$ in Eq. (7).

**Results of realistic model for Fe-based superconductors**—The present algorithm was benchmarked for a realistic five-orbital 2x2 cluster impurity model for LaAsO FeO. The crystal structure is shown in Fig. 4(a). A unit cell contains one Fe atom. Here, the tight-binding model constructed in Ref. [33], where each Fe atom has five 3$d$ orbitals, was adopted. Both the orbital and short-ranged antiferromagnetic correlations play an essential role in Fe-based high-$T_c$ compounds [34]; therefore, a quantitative prediction of the future $T_c$ may require at least cluster DMFT calculations using a $2 \times 2$ supercell in the $ab$ plane. They require the solution of a 20-orbital impurity model (the number of spin orbitals is 40).

A spin-diagonal matrix-valued hybridization function in the noninteracting limit was constructed using the standard procedure of cluster DMFT at $\beta = 500$ eV$^{-1}$ ($T \approx 24$ K $< T_c \approx 26$ K). Each spin sector of the hybridization function is a $20 \times 20$ matrix-valued function at each Matsubara frequency. It was transformed into the IR basis ($\omega_{\text{max}} = 20$) and fitted via the procedure used for the random models.

Figure 4(c) plots $N_{\text{bath}}$ per spin versus the relative residual norm of the fit. The residual decays quickly with respect to $N_{\text{bath}}$. The quality of the fit for $N_{\text{bath}} = 332$ is assessed by plotting the hybridization function in Fig. 4(d). For both inter-atom and intra-atom components, the discretized model approximately fits the hybridization with four significant digits. Considering the spin degrees of freedom, for $N_{\text{bath}} = 332$, solve a discretized model with a total of 40+$N_{\text{bath}}$=372 spin orbitals must be solved. A recently developed exact diagonalization solver with the truncation of the Hilbert space can handle $N_{\text{bath}} > 300$ [37]. Although this solver is limited to zero $T$, an extension to finite $T$ may be possible.
The group LASSO can be applied to bath fitting in the real-frequency formalism. Comparisons with nonuniform meshes used in the numerical renormalization group (NRG) \cite{[35]} and an efficient exact-diagonalization solver \cite{[7]} may produce interesting results. Recently, the complexity of quantum impurity models was analyzed \cite{[35]}. It was proven that $N_{\text{bath}} \sim N_{\text{SO}}$ suffices to compute exact ground-state energies based on removing redundant bath degrees of freedom using basis rotation and bath levels. In contrast, the present method does not mix spin orbitals and bath levels, thus does not generate nonlocal Coulomb interactions. This is preferable for some impurity solvers. Another bath compression method has also been proposed \cite{[19]}. Because the method requires the construction of a discrete model in advance, combining it with the algorithm proposed in the present study may be interesting.

In this letter, an efficient and stable discretization algorithm for large-scale impurity models with low symmetries is proposed. The proposed algorithm uses a regularization term based on group LASSO, a sparse-modeling technique, as well as a compact representation of Matsubara Green’s function. Its efficiency was demonstrated for random models with several spin orbitals $N_{\text{SO}} \gg 10$. Additionally, the number of required bath levels for a 20-impurity model for LaAsFeO was estimated.

The results of the present study are encouraging because they indicate that multi-orbital-atom cluster embedding calculations of correlated materials may be within reach even for a classical computer.

HS thanks Markus Wallerberger for the critical reading of the manuscript and useful comments. Part of the calculations was run on the facilities of the Supercomputer Center at the Institute for Solid State Physics, University of Tokyo. This research was conducted using the Fujitsu PRIMERGY CX400M1/CX2550M5 (Oakbridge-CX) in the Information Technology Center, University of Tokyo. We used the \texttt{irbasis} library \cite{[28]} for computing IR basis functions. We used DCore \cite{[37]} based on TRIQS \cite{[38]} and TRIQS/DFTTools \cite{[39]} for computing the hybridization function for LaFeAsO. HS was supported by JSPS KAKENHI Grant Nos. 18H01158 and 16K17735. YN was partially supported by JSPS KAKENHI Grant Numbers 18K11345. We used VESTA \cite{[40]} for visualizing the crystal structure.

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III. ROBUSTNESS AGAINST NOISE

To test the robustness of the present algorithm against noise in the hybridization function, we add Gaussian noise to the hybridization function, used to procedure the data in Fig. 2 of the main text. The standard deviation of the noise is chosen to be $10^{-5}$. We fit the hybridization function with the noise using exactly the same procedure. A remarkable different from the result without noise is the existence of an overfitting regime ($\alpha < 10^{-3}$), which is signaled a flat region of $\delta(\alpha)$ and the jump in $N_{\text{bath}}(\alpha)$. The algorithm works efficiently down to $\alpha = 10^{-3}$.

II. OPTIMIZATION ALGORITHM

Algorithm 1 shows a pseudocode for the optimization algorithm. In the pseudocode, we use $\mathbf{\theta} = (\theta_1, \ldots, \theta_{N_0})$, and $\mathbf{\epsilon} = (\epsilon_1, \ldots, \epsilon_{N_0})$ and $V = (V_{11}, \ldots, V_{1N_0}, \ldots)$.

Algorithm 1 Optimization algorithm

```
\epsilon \leftarrow \text{Non-uniform grid points from IR basis}
\theta_b \leftarrow \text{cos}^{-1}(g_{lb}/\omega_{\text{max}})
V \leftarrow \text{Random numbers from normalized Gaussian distribution}
Optimize \ V \ \text{using L-BFGS method}
Optimize \ V \ \text{and} \ \theta \ \text{using L-BFGS method}
Compute \ |\epsilon_b| \ \text{and sort the bath levels in descending order}
\begin{align*}
\text{r}_0 & \leftarrow \text{relative residual norm} \\
\text{for} \ \kappa = 1, 2, \ldots & \ \text{do} \\
\text{r} & \leftarrow \text{relative residual norm without the last bath level} \\
& \text{if} \ \text{r} > 1.1 \times \text{r}_0 \ \text{then} \\
& \ \ \ \ \text{Exit loop} \\
& \ \text{end if} \\
& \ \text{Remove the last bath level}
\end{align*}
end for
```
FIG. S1. (Color online) Results of one sample for $N_{SO} = 10$ and $\beta = 10$ and the noise level of $10^{-5}$. The hybridization function and the fitted results are compared in Matsubara frequency [(a)] and in the IR basis [(b)], respectively. In (a) and (b), the fitting error is also shown. (c) Number of bath levels $N_{\text{bath}}$ and relative residual norm $\delta$. (d) Strength of the coupling to the impurity $\|v_b\|$ in the decreasing order. (e) Positions of $\epsilon_b$ and $\|v_b\|$.