Quantum Computation for Predicting Solids-state Material Properties

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Abstract

Quantum chemistry is one of the most promising near-term applications of quantum computers.

Quantum algorithms such as variational quantum eigen solver (VQE) and variational quantum

deflation (VQD) algorithms have been mainly applied for molecular systems and there is a need

to implement such methods for periodic solids. Using Wannier tight-binding Hamiltonian

(WTBH) approaches, we demonstrate the application of VQE and VQD to accurately predict both

electronic and phonon bandstructure properties of several elemental as well as multi-component

solid-state materials. We apply VQE-VQD calculations for 307 spin-orbit coupling based

electronic WTBHs and 933 finite-difference based phonon WTBHs. We establish a workflow for

using VQD with lattice Green's function that can be used for solving dynamical mean-field theory

problems. The WTBH model solvers can be used for testing other quantum algorithms also.

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Introduction

Quantum chemistry is one of the most attractive applications for quantum computations¹. Quantum computers with a few qubits can potentially exceed classical computers because the size of Hilbert space exponentially increases with the number of orbitals^{2,3}. Predicting the energy levels of a Hamiltonian is a key problem in quantum chemistry. In the last decade, there has been an significant effort in estimating energies of Hamiltonians^{4,5} using various quantum algorithms such as quantum phase estimation (QPE)¹, variational quantum eigen solver (VQE)⁶, variational quantum deflation eigen solver (VQD)⁷, quantum subspace expansion (QSE),⁸ quantum equation of motion (QEOM)⁹, quantum amplitude estimation (QAE)¹⁰, witness-assisted variational eigenspectra solver (WAVES)11, quantum approximate optimization algorithm (QAOA)12 and quantum annealing (QA)¹³. These algorithms have been used for predicting both ground state and excited states of the Hamiltonians. However, their applications are mainly limited to molecular systems such as H₂, LiH, BeH₂^{6,14-16} molecules and there is a lack of research to apply and evaluate these algorithms for solid-state materials. While the molecular systems have promising applications in drug-discovery process, the solid-state simulations can accelerate design of superconductors, low-dimensional and topological materials which can improve Noisy Intermediate-Scale Quantum (NISQ) technology ¹⁷⁻¹⁹.

Variational quantum eigen solver (VQE) is one of the most celebrated methods for predicting an approximate ground state of a Hamiltonian on a quantum computer following the variational principles of quantum mechanics. VQE is developed as an alternate algorithm of quantum phase estimation (QPE) which solves an eigenvalue of a matrix from a state vector. VQE is a hybrid classical-quantum algorithm because utilizes both the classical and quantum hardware and can circumvent the limited coherence time of currently available quantum circuits. For VQE, an ansatz

with tunable parameters is constructed and a quantum circuit capable of representing this ansatz is designed. The tunable parameters are variationally adjusted until they minimize the expectation value of the molecular Hamiltonian to arbitrary accuracy. Classical computers are used to setup the Hamiltonian terms (such as decomposing Hamiltonian into Pauli matrices) and update the tunable parameters during circuit optimization. Quantum computers are used to prepare a quantum state based on a set of ansatz parameters and perform measurements of interaction terms. More details about VQE can be found in Ref.²⁰ Significant research work has been done to expand VQE algorithm to evaluated energy levels of Hamiltonian beyond ground state^{7,8,21,22}. One of the common methods to estimate high energy levels is Variation Quantum Deflation (VQD), which shows that overlap estimation can be used to deflate eigenstates once they are found, enabling the calculation of excited state energies and their degeneracies. VQD requires the same number of qubits as VQE and is robust for controlling errors⁷. VQD has been successfully used to obtain higher energy levels for several molecular systems^{7,23}

In this work, we show that eigen energies of Hamiltonians for solid state materials can be accurately estimated using Wannier tight-binding Hamiltonian approach and variational quantum deflation eigen solver. WFs are a complete orthonormalized basis set that acts as a bridge between a delocalized plane wave representation commonly used in electronic structure calculations and a localized atomic orbital basis that more naturally describes chemical bonds²⁴⁻²⁶. One of the most common ways of obtaining Wannier tight-binding Hamiltonians (WTBH)²⁷⁻²⁹ is by using density functional theory (DFT) calculations. All major DFT codes support generation WTBHs for a material. WTBHs have been proven successful for accurately predicting both electron, phonon, and electron-phonon coupling and dynamical properties of solids. One of the most popular basis sets for simulating solids is plane-wave basis, but the Hamiltonians using such basis are quite large

to simulate on current quantum hardware. Instead, Wannier based approaches provide smaller basis sizes (tens to hundreds of orbitals) which can be simulated on current quantum hardware with tens to hundreds of qubits. Additionally, WTBHs provide dense interpolation on Brillouin zone which is difficult for plane waves even on classical computers. We use the WTBHs for electrons databases³⁰ for 3D and 2D materials, which was recently populated for thousands of materials using density functional theory approaches. WTBHs can also be used to obtain many other solid-state properties such as optical conductivity, Berry phase, and Chern number³¹. The WTBHs for phonons³² can be obtained with finite-difference (FD)³³ and density functional perturbation theories (DFPT)³⁴. Such databases have also been populated for thousands of materials, especially for accurate gamma-point phonons^{33,34}. Both electron and phonon WTBHs are publicly available in the Joint Automated Repository for Various Integrated Simulations (JARVIS) infrastructure (https://jarvis.nist.gov/) of databases and tools³⁵. We integrate these databases with VQD algorithms and simulate thousands of materials using quantum algorithms using JARVIS-tools and Qiskit software^{36,37}. In addition to providing an interface to Qiskit, we provide integration with other software such as Tequila³⁸, and Pennylane³⁹.

First, we show a detailed analysis of example material, Aluminum and then use the workflow for several materials for statistical analysis. Although this work deals with single-particle picture, we believe our work can pave the way for solving interacting Hamiltonians (such as dynamical meanfield theory (DMFT)⁴⁰ and Green's function and screened Coulomb (GW)⁴¹), which can be more suitable to simulate on quantum computers than classical computers. We provide a preliminary workflow that can be used to integrate the VQD algorithms with DMFT based solving of lattice Green's function. All the databases and tools from this work are made publicly available so that researchers can apply their own algorithms as well as reproduce our work independently.

Results and discussion

As a first example of applying a quantum algorithm using Wannier tight-binding Hamiltonian for solids, we compare classical and quantum algorithm-based electronic bandstructure predictions of face-centered cubic Aluminum (JVASP-816). Choosing Al's s, px, py, pz orbitals we obtain an 8x8 Hermitian matrix at any k-point in the Brillouin zone. This matrix can be easily diagonalized on a classical computer and will act as a reference when comparing with a quantum algorithm. As discussed earlier, the Hermitian matrix is transformed into a sum of Pauli matrices using unitary transformation. The first step in getting the eigenvalues at each k-point is to get the ground using the variational quantum eigensolver (VQE) algorithm. VQE is a hybrid classical-quantum algorithm and has been applied to many molecular systems. It is important to note that unlike classical computers, a quantum circuit needs to be made before running quantum algorithms. These circuits are made with the Pauli matrices here. Quantum circuits can be thought of as the instructions of the quantum system holding all of the quantum operations. Then the coefficients of these matrices were optimized. In a circuit, the qubits are put in order, with qubit zero at the top and qubit two at the bottom and they are read left to right. There are several backends in Qiskit for running the quantum algorithms. We use EffieicntSU2 circuit as available in Qiskit package for ansatz. This circuit has been applied for several VQE problems before. An example of EfficientSU2 circuit for solving ground state at X point [0.5,0,0.5] is shown in Fig. 1a with RX and RZ gates. Corresponding measurement probabilities on an actual IBM quantum computer ibm q athens which a five-qubit processor is shown in Fig. 1b. We optimize the circuit parameters using classical optimizers such as constrained optimization by linear approximation (COBYLA), limited-memory Broyden-Fletcher-Goldfarb-Shanno bound (L_BFGS_B), sequential least squares programming (SLSQP), conjugate gradient (CG), and simultaneous

perturbation stochastic approximation (SPSA). These are local optimizers that attempt to find an optimal value within the neighboring set of a candidate solution. We monitor the convergence iterations for several optimizers in Fig. 2a. For the particular case, we observe that COBYLA converges fastest and CG the slowest. SLSQP is another suitable optimizer which converges comparably to COBYLA.

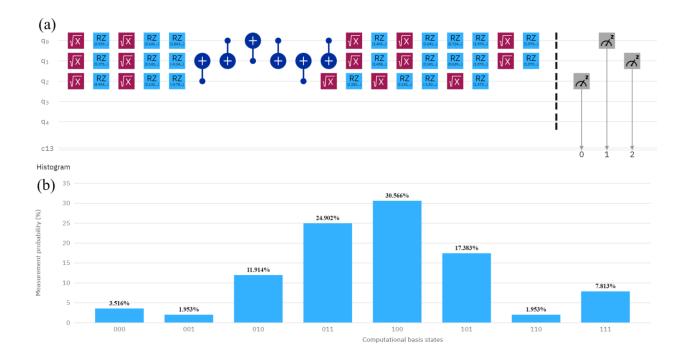


Fig. 1 Circuit model example for calculating ground state energy level for Al (JVASP-816) at X k-point [0.5,0.5,0] using VQE. a) EfficientSU2 circuit for 3 cubits with RY and RZ rotations. b) measurement probabilities of circuits.

After obtaining the ground state eigenvalue and state using VQE, we obtain high energy levels using Variational Quantum Deflation (VQD) algorithm. The same procedure was repeated for all the k-points leading to the electronic and phonon bandstructures for Al as shown in Fig. 2a and 2d. These simulations are carried out on IBM's statevector simulator. We compare the

Numpy⁴² based eigenvalue solver also to compare with VQE-VQD results. In both of the bandstructure predictions, we observe excellent agreement among them suggesting that WTBH can successfully be used to predict bandstructures with high accuracy.

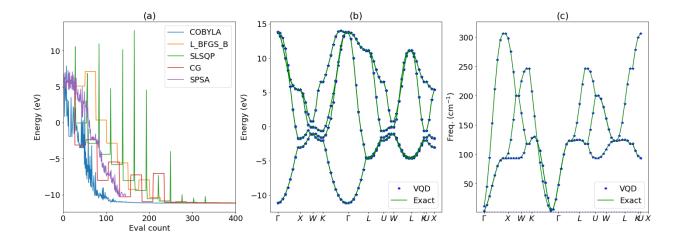


Fig.2 a) Monitoring VQE optimization progress with several local optimizers such COBYLA, L_BFGS_B , SLSQP, CG, and SPSA for Al electronic WTBH and at Γ -point. b) electronic bandstructure calculated from classical diagonalization (Numpy-based exact solution) and VQD algorithm for Al. b) phonon bandstructure for Al.

To further check the validity of the workflow, we screen materials for which the number of orbitals was less than 32 and calculate the minimum and maximum allowed energy level with VQE for both phonons and electrons. We compare these values with ground state energy from classical Numpy⁴² eigensolvers and the results are shown in Fig. 3. We apply VQE-VQD calculations for 307 spin-orbit coupling based electronic and 933 finite-difference based WTBHs from JARVIS-DFT database. Electronic WTBHs have 16 % unary, 53.36 % binary, 29 % ternary and 2 % quaternary compounds while phonon WTBHs have 0.76 % unary, 31.97 % binary, 66.50 % ternary and 0.76 % ternary compounds suggesting a reasonable vast chemical space. These WTBHs along with the scripts to run the quantum algorithms are made available using JARVIS-Tools. We find excellent agreement (r^2 =0.999) between the quantum algorithms and classical predictions for all

four cases suggesting that quantum algorithms are applicable to multicomponent solids as well. While electronic bandstructures are key properties for solids, several other key electronic properties can be predicted using the WTBHs and quantum algorithms such as surface bandstrucrurs, Chern number, Berry curvature, optical conductivity, thermoelectric coefficients³¹. Also, the WTBHs used here are single-particle description of solids but many body picture can be constructed for methods such as Wannier-based dynamical mean-field theory (DMFT)⁴⁰, GW⁴¹, and time-dependent Wannier functions⁴³.

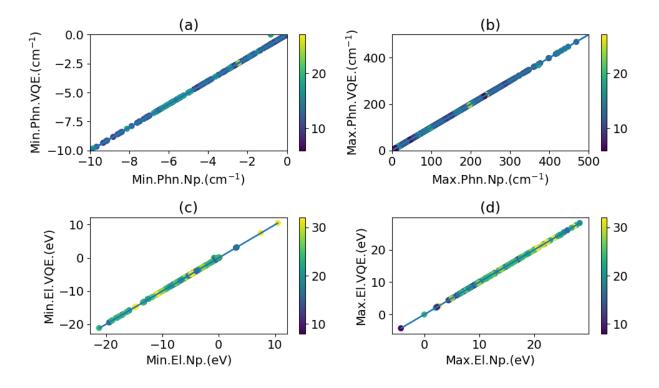


Fig. 3 Comparison of minimum (Min.) and maximum (Max.) energy levels at Γ -point for electronic and phonon WTBH using classical eigenvalue routine in Numpy (Np.) and VQE solver. a) and b) comparison of phonon (Phn.) minimum and maximum energy levels for 933 materials, c) and d) comparison of electronic (El.) minimum and maximum energy levels for 307 materials. The colorbar represents the number of Wannier orbitals.

Dynamical mean-field theory (DMFT)^{40,44} is one for the commonly used techniques for solving predicting electronic structure of correlated systems using impurity solver models. DMFT maps a many-body lattice problem to a many-body local problem with impurity models. In DMFT one of the central quantities of interest is the Green's function such as: $G(k, \omega_n) = [\omega_n + \mu - H(k) - \mu]$ $\Sigma(\omega_n)]^{-1}$, where $\omega_n=(2n+1)\pi T$ is a fermionic Matsubara frequency at temperature T,μ is the chemical potential, H(k) is a diagonal matrix containing the electronic WTBH eigenvalues (obtained with VQD algorithm mentioned above) at k-point k, and Σ is non-diagonal part and represents the self-energy. As an initial guess, we set $\Sigma = 0$ and calculate local Green's function for k-points on a dense k-point grid (31x31x31) and frequency grid (-20 eV to 5 eV with 0.25 eV step-size). Now the spectral function (A) and DMFT hybridization function (Δ) is calculates as: $A(\omega) = -\frac{1}{\pi} \sum_{k} Im(G(\omega + i\delta))$ and $\Delta(\omega + i\delta) = \omega - (G)^{-1}$ with $\delta \to 0$. Many quantum Monty Carlo (QMC) impurity solvers^{45,46} take the hybridization function as the input to calculate selfenergy. The Green's function is then solved iteratively until the impurity Green's function coincides with that of local lattice Green's function. The final self-energy found after the selfconsistency cycle can be used to predict the desired spectral function. An example for the imaginary part of Al's DMFT hybridization function for a few components considering zero selfenergy is shown below in Fig. 4.

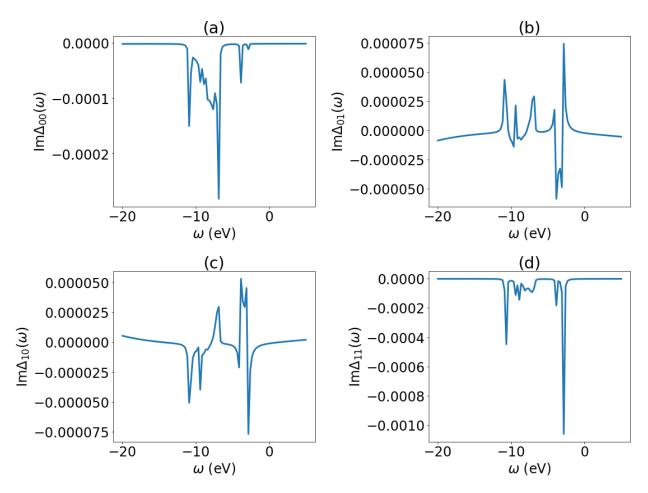


Fig. 4 Imaginary part of Al's DMFT hybridization function for a few components considering zero self-energy. $a)\Delta_{00}$, $b)\Delta_{01}$, $c)\Delta_{10}$, $d)\Delta_{11}$.

Conclusions

In conclusion, we demonstrate the applications of VQE and VQD to accurately predict both electronic and phonon properties of several solid-state materials using Wannier tight-binding Hamiltonian approach. The WTBH models can act as the test-bed for many other quantum algorithms. Although the present work has been used with WTBH, our workflow can be extended for solving Hermitian matrices from plane-wave DFT codes as well, given a larger number of qubits. Also, as Wannier based approaches have already been used to solve time dependent and

many-body problem on classical computers, WTBH with quantum algorithms can be useful to tackle problems which are intractable for classical computers.

Methods

Density functional theory calculations for generating electronic and phonon WTBHs were carried out using the Vienna Ab-initio simulation package (VASP)^{47,48} software using the workflow given on our JARVIS-Tools³⁵ GitHub page (https://github.com/usnistgov/jarvis). We use the OptB88vdW functional⁴⁹, which gives accurate lattice parameters for both vdW and non-vdW (3D-bulk) solids⁵⁰. We optimize the crystal-structures of the bulk and monolayer phases using VASP with OptB88vdW. The crystal structure was optimized until the forces on the ions were less than 0.01 eV/Å and energy less than 10^{-6} eV.

The basic formalism of Wannierization is well-established³¹. Wannier functions for a cell **R** and band n for a given Bloch state $\psi_{n\mathbf{k}}$ and unit-cell volume V is given as: $|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3} \int_{BZ} d\mathbf{k} e^{-i\mathbf{k}\cdot\mathbf{R}} |\psi_{n\mathbf{k}}\rangle$. The tight-binding Hamiltonians of electrons and phonons are built with different basis. For electrons, basis orbitals are selected according to the constituent elements, type of pseudopotential chosen and the number of basis orbitals (n per atom). For example, four orbitals (s, p_x, p_y, p_z) are selected as the basis for Aluminum in a system. A full list of orbitals used for different elements can be found in Ref. ³⁰. The site-site coupling is then described by a n × n matrix and the Hamiltonian H_k by a nN × nN matrix (N atoms per unit cell) for k-point k. For phonons in a 3-dimensional space, the interatomic coupling is described by a 3 ×3 matrix and H_k has a fixed dimension of 3N ×3N. From the symmetry point of view, the basis orbitals are p_x,p_y,p_z³². Spin-orbit coupling was included in generating WTBHs for electrons but not for phonons. We use Wannier90⁵¹ to construct Maximally-Localized Wannier Functions (MLWF) based TB-

Hamiltonians for electrons. For the case of interest in this work, where we wish to describe both the valence and conduction bands near the Fermi level, it is necessary to first select a set of bands to Wannierize, which includes separating the conduction bands from the free-electron-like bands that generally overlap with them in energy⁶². We use the maximum energy difference between DFT and Wannier bands criteria to examine the accuracy of WTBHs obtained from VASP+Wannier90 calculations. For phonons, we use both finite-difference method for calculating force-constants of conventional cells which are then processed with Phonopy⁵² software. We make efficient python modules (such as HermitianSolver) available in JARVIS-Tools so that users can obtain electron or phonon WTBHs for given JARVIS-IDs (if available) and then easily run quantum algorithms on them. The input/output files used in generating the WTBHs are also made publicly available through Figshare API to enhance transparency.

The variational quantum deflation algorithm is based on the combination of variation quantum eigensolver (VQE) and deflation algorithm for finding eigenvalue of a matrix. VQD^{7,22} is one of the most straightforward ways to predict eigenstates of a Hamiltonian. In this algorithm, first, we transform the Hermitian Hamiltonian matrix into Pauli operators. The Hamiltonian is now transformed into the following form: $H = \sum_{P \in \{I,X,Y,Z\}} \otimes_n h_P P$ where I, X, Y, Z are single-qubit Pauli operators and $h_P \in \mathbb{R}$ are corresponding coefficients. One of the common ways to achieve this using Qiskit's qiskit.aqua.operators.weighted_pauli_operator. After transforming the Hamiltonian, we choose a parametrized quantum circuit $U(\theta)$ (shown later) and iteratively optimize the parameter θ so that energy expectation value for ground state $\langle 0|U^{\dagger}(\theta)HU(\theta)|0\rangle$ is minimized. Here, $|0\rangle$ denotes the initialized state of the quantum computer. After the minimization we an optimal parameter θ^* which gives the approximate eigenstate $|\psi(\theta_0^*)\rangle$. Considering the shape of the input Hamiltonian as $k \times k$ and setting j = 1, we define a

new Hamiltonian as $H_j = H + \sum_{i=0}^{j-1} \beta_i |\psi(\boldsymbol{\theta}_0^*)\rangle \langle \psi(\boldsymbol{\theta}_0^*)|$. Here $\{\beta_i\}$ is set to numbers defined as the difference between eigenvalues $\{\varepsilon_i\}$ and a large number. In this approach, we deflate the Hamiltonian, and perform VQE on the resultant Hamiltonian to find k high energy states.

Parametrized quantum circuits are essential for many quantum algorithms designed for NISQ devices. There are numerous ways to choose a quantum circuit as ansatz for running quantum algorithms. One should avoid superfluous parameters in the circuit resulting in unnecessary gate operations. At the same time, having an insufficient number of independent parameters in the circuit can lead the classical minimization algorithm to fall into false local minima. A careful design of the quantum circuit is therefore essential to make optimal use of current NISQ devices. We choose the ansatz as QISKIT's³⁶ EfficientSU2 two-local circuit as the ansatz which the QISKIT documentation proposes as "a heuristic pattern that can be used to prepare trial wave functions for variational quantum algorithms or classification circuit for machine learning." This circuit consists of N + 1 blocks of RY and RZ gates applied to every qubit. These blocks are interlaced with N blocks containing CNOT (q, q') gates for all q < q'. EfficientSU2 is considered as one of the most expressive and noise-resilient circuits used in several VQE problems⁵³⁻⁵⁵.

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Data Availability

The data and the code that support the results within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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