# Electronic Structure Calculations and the Ising Hamiltonian

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ABSTRACT: Obtaining exact solutions to the Schrödinger equation for atoms, molecules, and extended systems continues to be a "Holy Grail" problem which the fields of theoretical chemistry and physics have been striving to solve since inception. Recent breakthroughs have been made in the development of hardware-efficient quantum optimizers and coherent Ising machines capable of simulating hundreds of interacting spins with an Ising-type Hamiltonian. One of the most vital questions pertaining to these new devices is, "Can these machines be used to perform electronic structure calculations?" Within this work, we review the general procedure used by these devices and prove that there is an exact mapping between the electronic structure Hamiltonian and the Ising Hamiltonian. Additionally, we provide simulation results of the transformed Ising Hamiltonian for H_2, He_2, HeH^+, and LiH molecules, which match the exact numerical calculations. This demonstrates that one can map the molecular Hamiltonian to an Ising-type Hamiltonian which could easily be implemented on currently available quantum hardware. This is an early step in developing generalized methods on such devices for chemical physics.
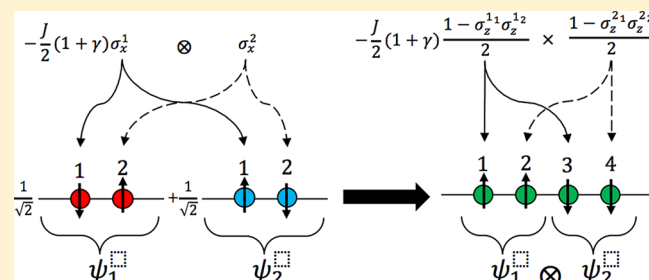
Determining solutions to the Schrödinger equation is a fundamentally challenging problem due to the exponential increase in the dimensionality of the corresponding Hilbert space with the number of particles in the system; this increase in computation system size requires a commensurate increase in computational resources. Modern quantum chemistry—hampered by the difficulty associated with solving the Schrödinger equation to chemical accuracy (~1 kcal/mol)—has largely become an endeavor to find approximate methods and corrections. A few products of this effort include *ab initio*, density functional, density matrix, algebraic, quantum Monte Carlo, and dimensional scaling methods.[1−8] All methods devised to date, however, face the insurmountable challenge of computational resource requirements as the calculation is extended either to higher accuracy or to larger systems. Computational complexity in electronic structure calculations suggests that these restrictions are a result of an inherent difficulty associated with simulating quantum systems.[9−11]

Electronic structure algorithms developed for quantum computers provide a promising new route to advance electronic structure calculations for large systems.[12−14] Recently, there has been an attempt at using an adiabatic quantum computing model—as implemented on the D-Wave machine—to perform electronic structure calculations.[15] Fundamentally, the approach behind the adiabatic quantum computing (AQC) method begins by defining a problem Hamiltonian, H_P, engineered to have its ground state encode the solution of a corresponding computational problem. The system is initialized in the ground state of a "beginning" Hamiltonian, H_B, which is easily solved by classical methods. The system is then allowed to evolve adiabatically as

$H(s) = (1 − s)H_B + sH_P$ (where s is a time evolution parameter, $s \in [0, 1]$). Adiabatic evolution is governed by the Schrödinger equation for the time-dependent Hamiltonian $H(s(t))$. Thus, permitting the system to evolve from H_B to H_P is a slow and smooth manner, such that the system remains in its ground eigenstate during the perturbative evolution.

Hitherto, the largest scale implementation of AQC is by D-Wave Systems.[16,17] In the case of the D-Wave device, the physical process acting as adiabatic evolution is more broadly called *quantum annealing* (QA). The quantum processors manufactured by D-Wave are essentially a transverse Ising model with tunable local fields and coupling coefficients: $H = \sum_i \Delta_i \sigma_x^i + \sum_i h_i \sigma_z^i + \sum_{i,j} J_{ij}\sigma_z^i\sigma_z^j$, where the parameters Δ_i, h_i, and J_{ij} are physically tunable. The qubits (quantum bits) are connected in a specified graph geometry; this design permits the embedding of arbitrary graphs. Zoller and co-workers presented a scalable architecture with full connectivity, yet it can only be implemented with local interactions.[18] The adiabatic evolution is initialized at $H_B = −h\sum_i \sigma_x^i$ and evolves into the problem Hamiltonian: $H_P = \sum_i h_i \sigma_z^i + \sum_{i,j} J_{ij}\sigma_z^i\sigma_z^j$. This equation describes a classical Ising model whose ground state is—in the worst case—NP-complete. Therefore, any combinatorial optimization NP-hard problem may be encoded into the parameter assignments ($\{h_i, J_{ij}\}$) of H_P and may exploit adiabatic evolution as a method

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for reaching the ground state of H_P. More recently, an optically based coherent Ising machine was developed and is capable of finding the ground state of an Ising Hamiltonian describing a set of several hundred coupled spin 1/2 particles.[19−21] These challenging NP-hard problems are characterized by difficulty in devising a polynomial-time algorithm; therefore, solutions cannot be easily found using classical numerical algorithms in a reasonable time for large system sizes (N).[19−21] Such special purpose machines may help in finding the solutions to some of the most difficult computational problems faced today.

■ RESULTS AND DISCUSSION

The technical scheme for performing electronic structure calculations on such an Ising-type machine can be summarized in the following four steps: First, write down the electronic structure Hamiltonian in terms of creation and annihilation Fermionic operators, delivering the second quantization form. Second, use either the Jordan−Wigner or the Bravyi−Kitaev transformation to move from Fermionic operators to spin operators.[13,22] Third, reduce the spin Hamiltonian (which is generally a k-local Hamiltonian) to a 2-local Hamiltonian.[23] Finally, map this 2-local Hamiltonian to an Ising-type Hamiltonian.

This general procedure begins with a second quantization description of a Fermionic system in which N single-particle states can be either empty or occupied by a spineless Fermionic particle.[4,24] One may then use the tensor product of each individual spin orbital, written as $|f_0, ..., f_n\rangle$ to represent states in Fermionic systems where $f_j \in \{0, 1\}$ is the occupation number of orbital j. Any interaction within the Fermionic system can be expressed in terms of products of the creation and annihilation operators $a_j^\dagger$ and a_j, for $j \in \{0, ..., N\}$. Thus, the molecular electronic Hamiltonian can be written as

$$\hat{H} = \sum_{i,j} h_{ij} a_i^\dagger a_j + \frac{1}{2} \sum_{i,j,k,l} h_{ijkl} a_i^\dagger a_j^\dagger a_k a_l \tag{1}$$

The above coefficients h_{ij} and h_{ijkl} are the one- and two-electron integrals which can be precomputed in a classical way used as inputs to the quantum simulation. The next step is to develop and employ a Pauli matrix representation of the creation and annihilation operators. This can be done by employing the Bravyi−Kitaev transformation—or the Jordan−Wigner transformation—to map between the second quantization operators and the Pauli matrices $\{\sigma_x, \sigma_y, \sigma_z\}$.[22,25] The molecular Hamiltonian now takes the general form[13]

$$H = \sum_{i,\alpha} h_\alpha^i \sigma_\alpha^i + \sum_{ij\alpha\beta} h_{\alpha\beta}^{ij} \sigma_\alpha^i \sigma_\beta^j + \sum_{ijk\alpha\beta\gamma} h_{\alpha\beta\gamma}^{ijk} \sigma_\alpha^i \sigma_\beta^j \sigma_\gamma^k + ... \tag{2}$$

where the index $\alpha = x, y, z$ notes the Pauli matrix and the index i designates the spin orbital. Now, after having developed a k-local spin Hamiltonian (many-body interactions), one should use a general procedure to reduce the Hamiltonian to a 2-local (two-body interactions) spin Hamiltonian form;[26,27] this is a requirement, since the proposed computational devices are typically limited to restricted forms of two-body interactions. Therefore, universal adiabatic quantum computation requires a method for approximating a quantum many-body Hamiltonian up to an arbitrary spectral error using at most two-body interactions. Hamiltonian gadgets, for example, offer a systematic procedure to address this requirement. Recently, we have employed analytical techniques resulting in a reduction of the resource scaling as a function of spectral error for the most

commonly used subdivisions, three- to two-body and k-body gadgets.[28]

Here we present a universal method of mapping an n-qubit Hamiltonian, H, which depends on σ_x, σ_y, σ_z to an rn-qubit Hamiltonian, H', consisting of only products of σ_z. In this process, we increase the number of qubits from n to rn, where the integer r plays the role of a "variational parameter" to achieve the desired accuracy in the final step of energy calculations. With increasing r, which means the use of more qubits to calculate, we can achieve more accurate results; please see the Appendix for more details.

We use two steps to complete this mapping. The first step is to generate a mapping between the eigenstates. In an original n-qubit Hilbert, H, space, a general eigenstate is given by $\psi = \sum_i a_i \phi_i$; we can map this to the space of the rn-qubit Hamiltonian, H', which increases the space from n qubits to rn qubits. The mapping method is to repeat the basis (ϕ_i) b_i times, where b_i is determined by a_i defined through the relationship $a_i \approx \frac{b_i S(b_i)}{\sqrt{\sum_m b_m^2}}$, where $\sum_i b_i = r$. $S(b_i)$ is a sign function associated with b_i, as a_i can be positive and negative and repeat time can only be positive. If a_i is positive, we have $S(b_i) = 1$, and if a_i is negative, we have $S(b_i) = -1$. After this procedure, we get a new eigenstate $|\Phi\rangle \otimes_i \otimes_{j=1}^{b_i} |\phi_i\rangle$. This procedure is represented in Figure 1.
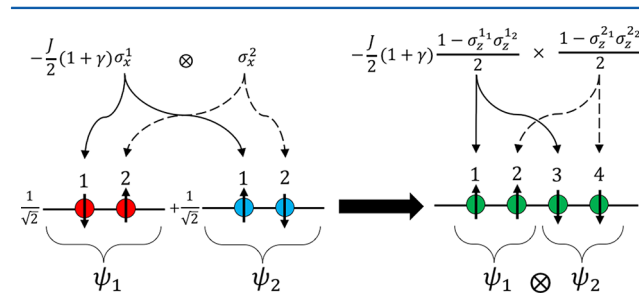

Figure 1. Example of mapping σ_x^2 between different bases from a state $|\Phi\rangle = \frac{1}{\sqrt{2}}|000\rangle + \frac{1}{\sqrt{2}}|010\rangle$ to state $|000010\rangle$, where 0 represents spin up and 1 represents spin down. The spin operators act on the second qubit in the original Hamiltonian basis and on the second and fifth qubits in the mapped Hamiltonian basis.

The second step is the mapping between the Hamiltonians, $H \to H'$. By mapping the eigenstate, we have a corresponding way to map the spin operator $(I_i, \sigma_x^i, \sigma_y^i, \sigma_z^i)$ on the ith qubit in n-qubit space to rn-qubit space by

$$\sigma_x^i \to \frac{1 - \sigma_z^{i_j} \sigma_z^{i_k}}{2} S'(j) S'(k) \quad \sigma_y^i \to \mathbf{i} \frac{\sigma_z^{i_k} - \sigma_z^{i_j}}{2} S'(j) S'(k)$$

$$\sigma_z^i \to \frac{\sigma_z^{i_j} + \sigma_z^{i_k}}{2} S'(j) S'(k) \quad I^i \to \frac{1 + \sigma_z^{i_j} \sigma_z^{i_k}}{2} S'(j) S'(k) \tag{3}$$

σ^{i_j} means acting on the ith qubit in jth n qubits in $|\Psi\rangle$ (or the spin operator σ acting on the $[(j-1)n + i]$ qubit). In addition, $S'(j)$, $S'(k)$ represents the sign of the jth, kth of the n qubits in the new state of the rn-qubit space. The introduction of $S'(j)$ is the same as the introduction of $S(b_i)$. Now, the newly transformed Hamiltonian, H', includes only products of σ_z and I, and is therefore diagonal (details of the transformation are presented with examples in the Appendix).

So far, we are able to transform our Hamiltonian to a k-local Hamiltonian including only products of σ_z terms. It is then
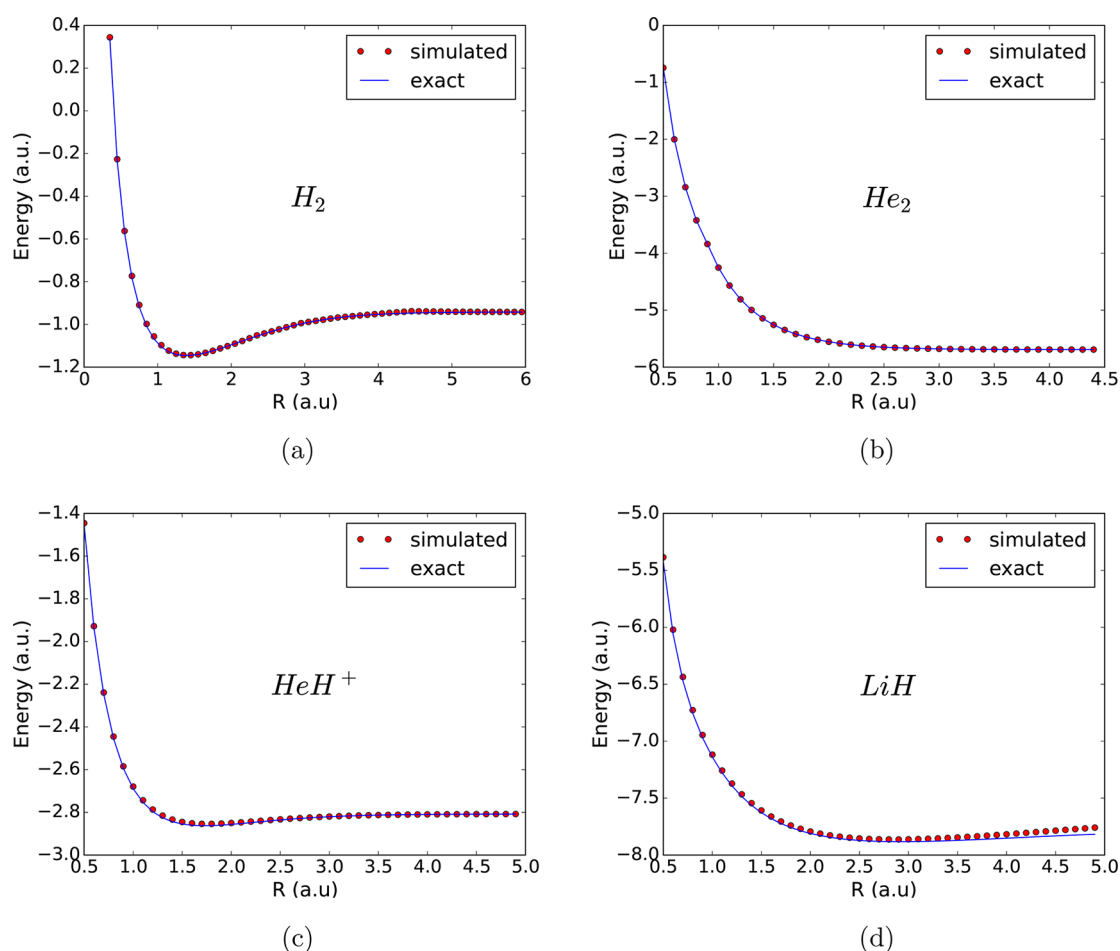
Figure 2. Comparing the numerical results of ground state energy of the Ising Hamiltonian with the exact STO-6G calculations of the ground state of H_2, He_2, HeH^+, and LiH molecules as one varies the internuclear distance R.

straightforward to transform the Hamiltonian to 2-local by using ancilla qubits, as the following example illustrating the transformation from 3-local to 2-local Ising Hamiltonian.

$$\min(\pm x_1 x_2 x_3) = \min(\pm x_4 x_3 + x_1 x_2 - 2x_1 x_4 - 2x_2 x_4 + 3x_4)$$

$$x_1, x_2, x_3, x_4 \in \{0, 1\}$$

(4)

Here, we see how, by including x_4, one can show that minimizing 3-local is equivalent to minimizing the sum of 2-local terms. We can apply this technique in our reduction from k-local to 2-local terms, as shown in the Appendix.[29]

Finally, we succeed in transforming our initial complex electronic structure Hamiltonian from the second-quantization form to an Ising-type Hamiltonian which can be solved using existing quantum computing hardware.[14,19,30,31]

To illustrate the proposed method (details are in the Appendix), we present in Figure 2 the calculations for the hydrogen molecule (H_2), the helium dimer (He_2), the hydrogen–helium molecular cation (HeH^+), and lithium hydride (LiH). First, we used the Bravyi–Kitaev transformation and the Jordan–Wigner transformation to convert the diatomic molecular Hamiltonian in the minimal basis set (STO-6G) to the spin Hamiltonian of $(\sigma_x, \sigma_y, \sigma_z)$. We then used our transformed Hamiltonian in rn-qubit space to obtain a diagonal k-local Hamiltonian constructed of σ_z terms. Finally, we reduced the locality to obtain a 2-local Ising Hamiltonian of the general form

$$H' = \sum_i h_i' \sigma_z^i + \sum_{ij} J_{ij}' \sigma_z^i \sigma_z^j$$

(5)

To estimate the efficiency of this proposed method, we here derive the number of qubits required by constructing the final 2-local Hamiltonian in eq 5. Suppose we start with an original Hamiltonian in the second quantization representation containing n orbitals. The first step of the transformation into the Pauli operator representation requires $O(n)$ qubits and $O(n^4)$ terms. The second step of mapping the form containing all of the Pauli operators to the form containing only σ_z requires $O(rn)$ qubits and $O(2^n r^2 n^4)$ terms. The final step of reduction to 2-local requires $O(2^n r^2 n^7)$ qubits. Due to the computational costs of the current method, it is feasibly useful for small and medium size molecular systems. To push this method to a large size molecular system, we propose to combine our method with a recent approach by Chan and co-workers.[32] Instead of encoding the wave function using n Gaussian orbitals, leading to Hamiltonians with $O(n^4)$ second-quantized terms, Chan et al. utilize a dual form of the plane wave basis which diagonalizes the potential operator, leading to a Hamiltonian representation with $O(n^2)$ second-quantized terms. Moreover, one can repeat our initial calculations using the new procedure with $O(n^2)$ second-quantized terms. This will clearly save computational resources. There is of course much space for improvement, and one should continue to explore new ideas to further optimize the proposed method.
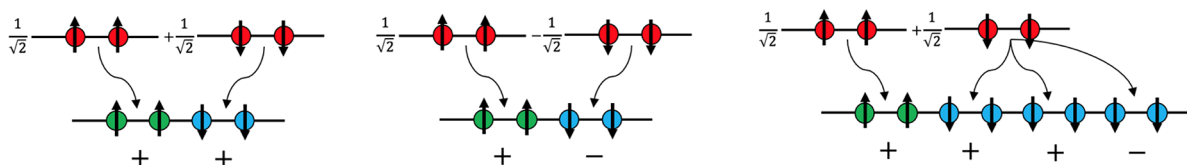
Figure 3. Left: If the 2-qubit state is $|\psi\rangle = \frac{1}{\sqrt{2}}|00\rangle + \frac{1}{\sqrt{2}}|11\rangle$, then the 4-qubit state is $|\Psi\rangle = |0011\rangle$ with $S'(1) = S'(2) = 1, r = 2, b_1 = b_2 = 1$, and $S(b_1) = S(b_2) = 1$. Middle: If the 2-qubit state is $|\psi\rangle = \frac{1}{\sqrt{2}}|00\rangle - \frac{1}{\sqrt{2}}|11\rangle$, then the 4-qubit state is $|\Psi\rangle = |0011\rangle$ with $S'(1) = -S'(2) = 1, r = 2, b_1 = b_2 = 1$, and $S(b_1) = -S(b_2) = 1$. Right: If the 2-qubit state is $|\psi\rangle = \frac{1}{\sqrt{2}}|00\rangle - \frac{1}{\sqrt{2}}|11\rangle$, then the 8-qubit state is $|\Psi\rangle = |00111111\rangle$ with $S'(1) = S'(2) = S'(3) = -S'(4) = 1, r = 4, b_1 = b_2 = 1$, and $S(b_1) = S(b_2) = 1$. ($S'(3)$ and $S'(4)$ cancel out, and thus, $S(b_2) = 1$).

■ CONCLUSION

We have developed a general procedure of mapping the original complex electronic structure Hamiltonian to a diagonal Hamiltonian in terms of a σ^z basis, which is experimentally implementable via today's Ising-type machines. Presently, implementation of σ^z, σ^x, and σ^y is not possible on current Ising machines nor while using quantum annealing as on the D-Wave machine. Our generalized procedure provides an opportunity to begin to utilize the current generation of quantum computing devices—of about 2000 connected spins—for important computational tasks in chemical physics. Incidentally, recent experimental results presented by the IBM group on few-electron diatomic molecules have shown that a hardware-efficient optimizer implemented on a 6-qubit super-conducting quantum processor is capable of reproducing the potential energy surface for such systems.[30] The development of efficient quantum hardware and the possibility of mapping the electronic structure problem into an Ising-type Hamiltonian may grant efficient ways to obtain exact solutions to the Schrödinger equation, thus providing a solution path to one of the most daunting computational problems in both chemistry and physics. Moreover, this study might open the possibility to connect with the rich field of statistical mechanics of he Ising model describing phase transitions.[33−37]

■ APPENDIX

Here, we present a procedure to construct a diagonal Hamiltonian with a minimum eigenvalue corresponding to the ground state of a given initial Hermitian Hamiltonian.

For a given initial Hermitian Hamiltonian, an eigenstate $|\psi\rangle$ can be expanded in a basis set $|\phi_i\rangle$ as $|\psi\rangle = \sum_i a_i|\phi_i\rangle$. This basis set consists of different combinations of spin-up and -down qubits. First, we will assume that all expansion coefficients, a_i, are nonnegative and will map the state to a new state $|\Psi\rangle$ according to the following rules:

- $|\Psi\rangle$ can be written as $|\Psi\rangle = \otimes_i \otimes_{j=1}^{b_i} |\phi_i\rangle$ and $r = \sum_i b_i$.
- If the original state $|\psi\rangle$ exists within an n-qubit subspace, the new state $|\Psi\rangle$ should be in an rn-qubit space, where r is the number of times we must replicate the n qubits to achieve an arbitrary designated accuracy.
- The number of times, b_i, we repeat the basis $|\psi\rangle$ in an rn-qubit state, $|\Psi\rangle$, approximates a_i by $\frac{b_i}{\sqrt{\sum_m b_m^2}}$. If r is large enough, b_i is proportional to a_i or we can just view $a_i \approx \frac{b_i}{\sqrt{\sum_m b_m^2}}$, where $\sqrt{\sum_m b_m^2}$ is the normalization factor. Here we introduce notation to be used throughout the remaining text:

Notation 1: We designate the ith qubit within the kth n-qubit subspace of the rn state space of $|\Psi\rangle$ as i_k.

Notation 2: We use $b(j)$ to represent the jth n-qubit in the space of $|\Psi\rangle$, which is in the basis $|\phi_{b(j)}\rangle$.

b_i may only be non-negative, yet a_i may be positive or negative. For a negative a_i, we will introduce a function $S(b_i)$ containing the sign information to account for b_i being non-negative. The mapping is described by the following rules:

- $S(b_i)$ is the sign associated with the b_i coefficient which is negative if a_i is negative and is positive if a_i is positive.
- For the rn-qubit state $|\Psi\rangle$, we can use a function $S'(i)$ to record the sign associated with each n qubits in the rn-qubit space. $S'(i)$ represents the sign of the ith n qubits in the rn-qubit space. Thus, $b_i = |\sum_{|\phi_{b(j)}\rangle=|\phi_i\rangle} S'(j)|$ and $S(b_i)$ is the sign of $\sum_{|\phi_{b(j)}\rangle=|\phi_i\rangle} S'(j)$.
- As before, b_i is the integer which approximates a_i by $\frac{b_i S(b_i)}{\sqrt{\sum_m b_m^2}}$. If r is large enough, we can just view $a_i \approx \frac{b_i S(b_i)}{\sqrt{\sum_m b_m^2}}$.
- $|\Psi\rangle$ can be written as $|\Psi\rangle = \otimes_i \otimes_{j=1}^{b_i} |\phi_i\rangle$ and $r = \sum_{i=1}^r |S'(i)|$.

Figure 3 shows the details about the mapping.

Theorem 1: With the mapping between $|\psi\rangle$ and $|\Psi\rangle$ as described above, we can find a mapping between the Hamiltonian in the space of $|\psi\rangle$ to the space of $|\Psi\rangle$.

Theorem 2: $\langle\phi_{b(j)}| \otimes_i I_i|\phi_{b(k)}\rangle$ is equal to $\langle\Psi| \prod_i \frac{1+\sigma_z^{i_j}\sigma_z^{i_k}}{2}|\Psi\rangle$, which means I_i in the space of $|\psi\rangle$ can be mapped to $\frac{1+\sigma_z^{i_j}\sigma_z^{i_k}}{2}$ in the space of $|\Psi\rangle$.

Proof: Clearly, I_i in the space of $|\psi\rangle$ is to observe if ith digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(k)}\rangle$ are the same or not. If they are the same, it yields 1; otherwise, 0. On the other hand, $\frac{1+\sigma_z^{i_j}\sigma_z^{i_k}}{2}$ in the space of $|\Psi\rangle$ is to check if the ith digits of the jth n-qubit subspace ($|\phi_{b(j)}\rangle$) and the kth n-qubit subspace ($|\phi_{b(k)}\rangle$) are the same or not. If they are the same, it yields 1; otherwise, 0. (For $\frac{1-\sigma_z^{i_j}\sigma_z^{i_k}}{2}$, we omit the operators for other digits which are the identity I.)

Thus, we get

$$\langle\phi_{b(j)}| \otimes I_i|\phi_{b(k)}\rangle = \langle\Psi| \prod_i \frac{1+\sigma_z^{i_j}\sigma_z^{i_k}}{2}|\Psi\rangle \tag{6}$$

If and only if all digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(k)}\rangle$ are the same, the left and right results are equal to 1; otherwise, they are equal to 0.

Theorem 3: $\langle\phi_{b(j)}| \otimes_{i<m} I_i \otimes \sigma_x^m \otimes_{i>m} I_i|\phi_{b(k)}\rangle$ is equal to $\langle\Psi| \prod_{i<m} \frac{1+\sigma_z^{i_j}\sigma_z^{i_k}}{2} \times \frac{1+\sigma_z^{m_j}\sigma_z^{m_k}}{2} \times \prod_{i>m} \frac{1-\sigma_z^{i_j}\sigma_z^{i_k}}{2}|\Psi\rangle$, which means σ_x^j in the space of $|\psi\rangle$ can be mapped as $\frac{1-\sigma_z^{i_j}\sigma_z^{i_k}}{2}$ in the space of $|\Psi\rangle$.

Chart 1

Algorithm 1
for i from 0 to $\lfloor \frac{r}{2} \rfloor$ **do**
Set the signs of the first i n qubits to be negative and the others to be positive. Set λ
to be a large number (at least larger than the ground state value and this will avoid the
case $\sum_m b_m^2 = 0$).
Construct H' and C.
while the ground eigenvalue of $(H' - \lambda C) < 0$ **do**
Calculate $H' - \lambda C$ and get the state $|\Psi'\rangle$ and the corresponding eigenvalue $\sum_m b_m^2 =$
$(\lambda' - \lambda)$.
Calculate C on $|\Psi'\rangle$ to get $\sum_m b_m^2$ then get λ'. Set λ to be λ'.
end while
λ' is the smallest eigenvalue with certain signs.
end for
Compare all λ' and the smallest one is the ground state energy of H.

Proof: Clearly, σ_x^i in the space of $|\psi\rangle$ is to check if the ith digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(k)}\rangle$ are the same or not. If they are the same, it yields 0; otherwise, 1. Similarly, $\frac{1 - \sigma_z^{i_j}\sigma_z^{i_k}}{2}$ in the space $|\Psi\rangle$ is to verify if the ith digits of the jth n-qubit subspace ($|\phi_{b(j)}\rangle$) and the kth n-qubit subspace ($|\phi_{b(k)}\rangle$) are identical. If they are the same, it gives 0; otherwise, 1. (For $\frac{1 - \sigma_z^{i_j}\sigma_z^{i_k}}{2}$, we omit operators for other digits which are the identity I.)

Thus, we get

$$\langle \phi_{b(j)}| \otimes_{i<m} I_i \otimes \sigma_x^m \otimes_{i>m} I_i |\phi_{b(k)}\rangle$$
$$= \langle \Psi| \prod_{i<m} \frac{1 + \sigma_z^{i_j}\sigma_z^{i_k}}{2} \times \frac{1 + \sigma_z^{m_j}\sigma_z^{m_k}}{2} \times$$
$$\prod_{i>m} \frac{1 - \sigma_z^{i_j}\sigma_z^{i_k}}{2} |\Psi\rangle \tag{7}$$

σ_x^i and $\frac{1 - \sigma_z^{i_j}\sigma_z^{i_k}}{2}$ have the same function in different space to check the digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(j)}\rangle$.

Also, σ_y^i and $\mathbf{i}\frac{\sigma_z^{i_k} - \sigma_z^{i_j}}{2}$ have the same function in different spaces. These operators are used to check the ith digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(j)}\rangle$. Also, σ_z^i and $\frac{\sigma_z^{i_j} + \sigma_z^{i_k}}{2}$ have the same function in different spaces to check the ith digits of $|\phi_{b(j)}\rangle$ and $|\phi_{b(j)}\rangle$. This can be easily verified by the above discussion.

Theorem 4: Any Hermitian Hamiltonian in the space of $|\psi\rangle$ can be written in the form of Pauli and identity matrices, which can be mapped to the space of $|\Psi\rangle$ as described above.

Notation 3: We denote the mapping between the jth n-qubit subspace and the kth n-qubit subspace in $|\Psi\rangle$, $\frac{1 - \sigma_z^{i_j}\sigma_z^{i_k}}{2}$ as $X_i^{(j,k)}$, $\frac{\sigma_z^{i_j} + \sigma_z^{i_k}}{2}$ as $Z_i^{(j,k)}$, $\mathbf{i}\frac{\sigma_z^{i_k} - \sigma_z^{i_j}}{2}$ as $Y_i^{(j,k)}$, and $\frac{1 + \sigma_z^{i_j}\sigma_z^{i_k}}{2}$ as $I_i^{(j,k)}$.

Proof: If H can be written as

$$H = \otimes_a \sigma_x^a \otimes_b \sigma_y^b \otimes_c \sigma_z^c \otimes_d I_d \tag{8}$$

We can write the mapped $H'_{(j,k)}$ as

$$H'_{(j,k)} = \prod_a X_a^{(j,k)} \prod_b Y_b^{(j,k)} \prod_c Z_c^{(j,k)} \prod_d I_d^{(j,k)} \tag{9}$$

It can be verified following the rules above that

$$\langle \Psi| H'_{(j,k)} |\Psi\rangle = \langle \phi_{b(j)}| H |\phi_{b(k)}\rangle \tag{10}$$

Thus, if we add the sign functions $S'(j)$ and $S'(k)$, we achieve

$$\langle \Psi| \sum_{\substack{j \neq k, j, k \leq r \\ j,k}} H_{(j,k)} S'(j) S'(k) |\Psi\rangle$$
$$= \sum_{\substack{j \neq k, j, k \leq r \\ j,k}} \langle \phi_{b(j)}| H |\phi_{b(k)}\rangle S'(j) S'(k)$$
$$= \sum_{\substack{j \neq k, j, k \leq 2^n \\ j,k}} b_j S(b_j) b_k S(b_k) \langle \phi_j| H |\phi_k\rangle$$
$$= \sum_m b_m^2 \sum_{\substack{j \neq k, j, k \leq 2^n \\ j,k}} a_j a_k \langle \phi_j| H |\phi_k\rangle \tag{11}$$

Also, in the same basis, we have

$$X_i^{(j,j)} = \frac{1 - \sigma_z^{i_j}\sigma_z^{i_j}}{2} = 0 \quad Y_i^{(j,j)} = \mathbf{i}\frac{\sigma_z^{i_j} - \sigma_z^{i_j}}{2} = 0$$
$$Z_i^{(j,j)} = \frac{\sigma_z^{i_j} + \sigma_z^{i_j}}{2} = \sigma_z^{i_j} \quad I_i^{(j,j)} = \frac{1 + \sigma_z^{i_j}\sigma_z^{i_j}}{2} = I \tag{12}$$

Thus, as before, we can also get

$$\langle \Psi| \sum_j^{j \leq r} H_{(j,j)} S'(j) S'(j) |\Psi\rangle$$
$$= \sum_j^{j \leq r} \langle \phi_{b(j)}| H |\phi_{b(j)}\rangle S'(j) S'(j)$$
$$= \sum_j^{j \leq 2^n} b_j^2 \langle \phi_j| H |\phi_j\rangle$$
$$= \sum_m b_m^2 \sum_j^{j \leq 2^n} a_j^2 \langle \phi_j| H |\phi_j\rangle \tag{13}$$

Combining the two together, we can get

$$\langle \Psi| \sum_{\substack{j,k \leq r \\ j,k}} H_{(j,k)} S'(j) S'(k) |\Psi\rangle = \sum_m b_m^2 \sum_{\substack{j,k \leq 2^n \\ j,k}} a_j a_k \langle \phi_j| H |\phi_k\rangle \tag{14}$$

We construct a matrix, C, in the space of $|\Psi\rangle$, which has elements $\sum_m b_m^2$ as

- $C = \sum_{\pm} \left(\sum_i \left(\prod_{k=1_i}^{n_i} \frac{1 \pm \sigma_z^k}{2} \right) S'(i) \right)^2$

- $\sum_{\pm}$ over all combinations of positive and negative signs of each digit in each ith of the n-qubit in space $|\Psi\rangle$.

- $\sum_i$ over all n-qubit collection in $|\Psi\rangle$ to check whether each n qubits is in a certain state.
- $\prod_{k=1,_i}^{n_i}$ over each qubits of ith n qubits in $|\Psi\rangle$.
- $\frac{1 \pm \sigma_z^k}{2}$ is to check whether kth qubits of ith n-qubit subspace is in a certain state. $\frac{1 + \sigma_z^k}{2}$ is 1 when the kth qubits are present in the basis $|0\rangle$ or 0 otherwise. $\pm$ is to go over combination by $\sum_{\pm}$.

So far, we have established a mapping between $|\psi\rangle$ and $|\Psi\rangle$. The Hamiltonian H in the space of $|\psi\rangle$ and $\sum_{(j,k)}^{j,k \le r} H'_{(j,k)}$ in the space of $|\Psi\rangle$. Also, we have constructed a matrix C to compute $\sum_m b_m^2$ corresponding to $|\Psi\rangle$. Thus, we have the final results:

$$\langle\Psi| \sum_{j,k}^{j,k \le r} H_{(j,k)} S'(j) S'(k) |\Psi\rangle = \sum_m b_m^2 \sum_{j,k}^{j,k \le 2^n} a_j a_k \langle\phi_j|H|\phi_k\rangle \tag{15}$$

Here, we present an algorithm combining $\sum_{(j,k)}^{j,k \le r} H'_{(j,k)}$ and C to calculate the ground state of the initial Hamiltonian H.

Notation 4: We mark the eigenvalue of $|\psi\rangle$ for H as λ'. According to the relationship above, the eigenvalue of $|\Psi\rangle$ for H' is $\sum_m b_m^2 \lambda'$. Thus, if we choose a λ and construct a Hamiltonian, $H' - \lambda C$. The eigenvalue of $|\Psi\rangle$ for $H' - \lambda C$ is $\sum_m b_m^2 (\lambda' - \lambda)$.

Theorem 5: The algorithm shown in Chart 1 converges to the minimum eigenvalue of H by finite iterations.

Proof:

Monotonic Decreasing If we can find an eigenstate $|\Psi\rangle$ of $H' - \lambda C$ with eigenvalue $\sum_m b_m^2 (\lambda' - \lambda) < 0$. Because $\sum_m b_m^2 \ge 0$, we get $\lambda' - \lambda < 0$. This means we can find an eigenstate $|\psi\rangle$ of H with an eigenvalue λ' and $\lambda' - \lambda < 0$. Thus, each time λ decreases monotonically.

The minimum eigenvalue: Here we prove that the minimum eigenvalue of H is achievable and the loop will converge when we obtain the minimum eigenvalue. According to **Monotonic Decreasing**, each time the eigenvalue we get will decrease. Because we have a finite number of eigenvalues, this means we will finally come to the minimum eigenvalue. Also, if we set λ to be the minimum eigenvalue of H, $H' - \lambda C = \sum_m b_m^2 (\lambda' - \lambda) \ge 0$ because $\sum_m b_m^2 \ge 0$ and $\lambda' - \lambda \ge 0$. Also, if λ' is just the minimum eigenvalue, we get $\lambda' - \lambda = 0$ and the loop stops.

Thus, we prove that the eigenvalue decreases and finally converges to the minimum eigenvalue of H.

Theorem 6: To account for the sign, we just need to set i from 0 to $\lfloor \frac{r}{2} \rfloor$ and set signs of the first ith n-qubit to be negative and the others to be positive in $|\Psi\rangle$.

Proof: If we have n qubits in the $|\Psi\rangle$ space with negative sign, where $|\Psi\rangle$ has total i n qubits with negative sign. If these n qubits are not in the first i n qubits, we can rearrange them to the first i n qubits by exchanging them with n qubits in the first i n qubits which have positive sign. Thus, all combinations can be reduced to the combination stated in **Theorem 6**.

Thus, we have established a transformation from an initial Hermitian Hamiltonian to a diagonal Hamiltonian and presented an algorithm to calculate the minimum eigenvalue of the initial Hamiltonian using the diagonal Hamiltonian.

Example

To illustrate the above procedure, we give details of the transformation for the simple model of two spin-$\frac{1}{2}$ electrons with an exchange coupling constant J in an effective transverse magnetic field of strength B. This simple model has been used to

discuss the entanglement for H_2 molecule.[38] The general Hamiltonian for such a system is given by

$$H = -\frac{J}{2}(1 + \gamma)\sigma_x^1 \sigma_x^2 - \frac{J}{2}(1 - \gamma)\sigma_y^1 \sigma_y^2 - B\sigma_z^1 - B\sigma_z^2 \tag{16}$$

where γ is the degree of anisotropy.

In the $\{|00\rangle, |10\rangle, |01\rangle, |11\rangle\}$ basis, the eigenvectors can be written as (here we just use the eigenvectors to show how we map the Hamiltonian, but in an actual calculation, we do not know the eigenvectors)

$$|\xi_1\rangle = \frac{1}{\sqrt{2}}(|10\rangle + |01\rangle)$$

$$|\xi_2\rangle = \frac{1}{\sqrt{2}}(|10\rangle - |01\rangle)$$

$$|\xi_3\rangle = \sqrt{\frac{\alpha - 2B}{2\alpha}}|11\rangle + \sqrt{\frac{\alpha + 2B}{2\alpha}}|00\rangle$$

$$|\xi_4\rangle = \sqrt{\frac{\alpha + 2B}{2\alpha}}|11\rangle - \sqrt{\frac{\alpha - 2B}{2\alpha}}|00\rangle \tag{17}$$

where $\alpha = \sqrt{4B^2 + J^2\gamma}$.

If we set $r = 2$, for example, if $|\psi\rangle = |\xi_1\rangle$, $|\Psi\rangle = |10\rangle \otimes |01\rangle$ with $S'(1) = 1$ and $S'(2) = 1$. If $|\psi\rangle = |\xi_2\rangle$, $|\Psi\rangle = |10\rangle \otimes |01\rangle$ with $S'(1) = -1$ and $S'(2) = 1$.

Abiding by the previous mapping, the mapped Hamiltonian H' and matrix C can be written as (this is illustrated in Figure 4)

$$H'_{(1,1)} = -B\sigma_z^1 - B\sigma_z^2$$

$$H'_{(2,2)} = -B\sigma_z^3 - B\sigma_z^4$$

$$\begin{aligned}
H'_{(1,2)} = &-\frac{J}{2}(1 + \gamma)\frac{1 - \sigma_z^1\sigma_z^3}{2}\frac{1 - \sigma_z^2\sigma_z^4}{2} \\
&-\frac{J}{2}(1 - \gamma)\frac{\sigma_z^1 - \sigma_z^3}{2}\frac{\sigma_z^4 - \sigma_z^2}{2} \\
&- B\frac{\sigma_z^1 + \sigma_z^3}{2}\frac{1 + \sigma_z^2\sigma_z^4}{2} \\
&- B\frac{\sigma_z^2 + \sigma_z^4}{2}\frac{1 + \sigma_z^1\sigma_z^3}{2}
\end{aligned}$$

$$H'_{(2,1)} = H'_{(1,2)} \tag{18}$$

$$\begin{aligned}
C = &\left(\frac{1 + \sigma_z^1}{2}\frac{1 + \sigma_z^2}{2}S'(1) + \frac{1 + \sigma_z^1}{2}\frac{1 + \sigma_z^2}{2}S'(2)\right)^2 \\
&+ \left(\frac{1 + \sigma_z^1}{2}\frac{1 - \sigma_z^2}{2}S'(1) + \frac{1 + \sigma_z^1}{2}\frac{1 - \sigma_z^2}{2}S'(2)\right)^2 \\
&+ \left(\frac{1 - \sigma_z^1}{2}\frac{1 + \sigma_z^2}{2}S'(1) + \frac{1 - \sigma_z^1}{2}\frac{1 + \sigma_z^2}{2}S'(2)\right)^2 \\
&+ \left(\frac{1 - \sigma_z^1}{2}\frac{1 - \sigma_z^2}{2}S'(1) + \frac{1 - \sigma_z^1}{2}\frac{1 - \sigma_z^2}{2}S'(2)\right)^2
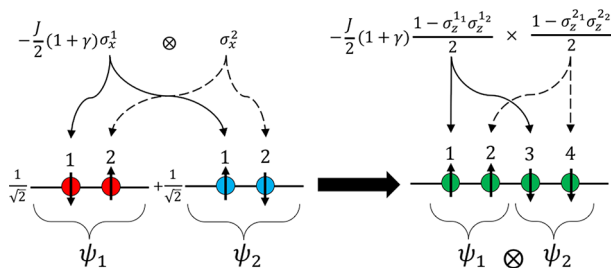\end{aligned} \tag{19}$$

If $S'(1) = S'(2) = 1$, we have

Figure 4. Mapped Hamiltonian $-\frac{J}{2}(1+\gamma)\sigma_x^1\sigma_x^2$ between different bases.

$$H' = \sum_{j,k}^{j,k\leq 2} H'_{(j,k)}S'(j)S'(k)$$

$$= H'_{(1,1)} + H'_{(2,2)} + H'_{(1,2)} + H'_{(2,1)} \tag{20}$$

We can write matrix C as

$$C = \begin{bmatrix} 4 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & 4 \end{bmatrix}$$

If $-S'(1) = S'(2) = 1$, we have

$$H' = \sum_{j,k}^{j,k\leq 2} H'_{(j,k)}S'(j)S'(k)$$

$$= H'_{(1,1)} + H'_{(2,2)} - H'_{(1,2)} - H'_{(2,1)} \tag{21}$$

We can write C in the matrix format:

$$C = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}$$

For $-S'(1) = S'(2) = 1$, we here display a procedural representation of our algorithm (we set $B = 0.001$, $J = -0.1$, and $\gamma = 0$):

1. First, when we choose $\lambda = 100$, we get that the minimum eigenvalue of $H' - 100C$ is -400 with $|\Psi\rangle = |0000\rangle$. Thus, we get $\sum_m b_m^2 = 4$ and $\lambda' = 0$.
2. When we set $\lambda = 0$, we get that the minimum eigenvalue of $H' + 0C$ is -0.2 with $|\Psi\rangle = |0110\rangle$. Thus, we get $\sum_m b_m^2 = 2$ and $\lambda' = -0.1$.
3. When we set $\lambda = -0.1$, we get that the minimum eigenvalue of $H' + 0.1C$ is 0 with $|\Psi\rangle = |0110\rangle$. We stop here and get that the minimum eigenvalue of H is -0.1.

Here we present the result of mapping the above Hamiltonian, eq 12, with $B = 0.001$, $J = -0.821R^{5/2}e^{-2R}$, and $\gamma = 0$[38] in Figure 5.

Reduce Locality of the Transformed Hamiltonian

Here we present the procedure to reduce the locality of H' from a k-local to a 2-local Ising-type Hamiltonian.

For $x, y, z \in \{0, 1\}$,[29]

$$xy = z \text{ if } xy - 2xz - 2yz + 3z = 0 \tag{22}$$

and

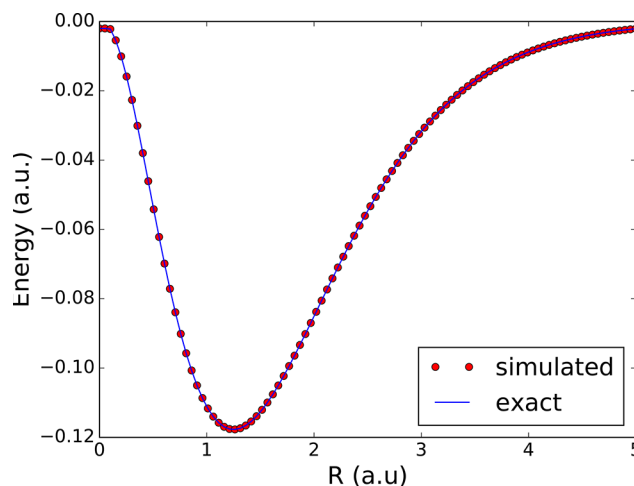$$xy \neq z \text{ if } xy - 2xz - 2yz + 3z > 0 \tag{23}$$

Figure 5. Comparing the ground state energy from exact (atomic units) of the original Hamiltonian H, eq 16, as a function of the internuclear distance, R (solid line), with the results of the transformed Hamiltonian H', eqs 20 and 21.

Thus, the 3-local $x_1x_2x_3$ can be transformed to 2-local by setting $x_4 = x_1x_2$:

$$\min(x_1x_2x_3) = \min(x_4x_3 + x_1x_2 - 2x_1x_4 - 2x_2x_4 + 3x_4)$$

$$x_1, x_2, x_3, x_4 \in \{0, 1\} \tag{24}$$

$$\min(-x_1x_2x_3) = \min(-x_4x_3 + x_1x_2 - 2x_1x_4 - 2x_2x_4 + 3x_4)$$

$$x_1, x_2, x_3, x_4 \in \{0, 1\} \tag{25}$$

We can prove that $\min(x_1x_2x_3 + f(x) = g_1(x)) = \min(x_4x_3 + x_1x_2 - 2x_1x_4 - 2x_2x_4 + 3x_4 + f(x) = g_2(x, x_4))$, where $f(x)$ is the polynomial of all variables (including x_1, x_2, x_3, and other variables, excluding x_4). If there exists x', this makes $g_1(x')$ be the minimum, and we can always make $g_2(x', x_4) = g_1(x')$ by choosing $x_4 = x_1x_2$. Then, if there exists x'', this makes $g_2(x'', x_4)$ be the minimum, and then $g_1(x'') \leq g_2(x'')$:

1. If $x_4 = x_1x_2$, $g_1(x'') = x_1x_2x_3 + f(x'') = x_4x_3 + x_1x_2 - 2x_1x_4 - 2x_2x_4 + 3x_4 + f(x'') = g_2(x'', x_4)$.
2. If $x_4 \neq x_1x_2$, $g_2(x'', x_4) = x_4x_3 + x_1x_2 - 2x_1x_4 - 2x_2x_4 + 3x_4 + f(x'') \geq x_4x_3 + 1 + f(x'') \geq x_1x_2x_3 + f(x'') = g_1(x'')$ because $x_4x_3 - x_1x_2x_3 \geq -1$.

Thus, we have $g_1(x') = g_2(x', x_4) \geq g_2(x'', x_4) \geq g_1(x'') \geq g_1(x')$. Thus, we have $g_1(x') = g_2(x', x_4) = g_2(x'', x_4) = g_1(x'')$, or all x makes $g_1(x)$ minimum would also makes $g_2(x, x_4)$ minimum and vice versa.

Thus, we obtain

$$\min\left(\sigma_z^1\sigma_z^2\prod_{i=3}^{n}\sigma_z^i\right) = \min\left(8\frac{1+\sigma_z^1}{2}\frac{1+\sigma_z^2}{2}\frac{1+\prod_{i=3}^n\sigma_z^i}{2}\right.$$

$$\left. - 8\frac{1+\sigma_z^1+\sigma_z^2+\prod_{i=3}^n\sigma_z^i+\sigma_z^1\sigma_z^2+\sigma_z^1\prod_{i=3}^n\sigma_z^i+\sigma_z^2\prod_{i=3}^n\sigma_z^i}{8}\right)$$

$$= \min\left(7 + \prod_{i=3}^n\sigma_z^i - 3\sigma_z^1 - 3\sigma_z^2 + 6\sigma_z^{n+1} + 2\prod_{i=3}^{n+1}\sigma_z^i - \sigma_z^1\prod_{i=3}^n\sigma_z^i\right.$$

$$\left. - \sigma_z^2\prod_{i=3}^n\sigma_z^i - 4\sigma_z^1\sigma_z^{n+1} - 4\sigma_z^2\sigma_z^{n+1} + \sigma_z^1\sigma_z^2\right) \tag{26}$$

Table 1. Comparing the Exact Ground Energy (au) and the Simulated Ground Energy (au) (Simulated by 4 × 2 qubits) as a Function of the Intermolecule Distance R (au)

R	g_0	g_1	g_2	g_3	g_4	exact	simulated
0.6	1.5943	0.5132	−1.1008	0.6598	0.0809	−0.5617	−0.5703
0.65	1.4193	0.5009	−1.0366	0.6548	0.0813	−0.6785	−0.6877
0.7	1.2668	0.4887	−0.9767	0.6496	0.0818	−0.7720	−0.7817
0.75	1.1329	0.4767	−0.9208	0.6444	0.0824	−0.8472	−0.8575
0.8	1.0144	0.465	−0.8685	0.639	0.0829	−0.9078	−0.9188
0.85	0.909	0.4535	−0.8197	0.6336	0.0835	−0.9569	−0.9685
0.9	0.8146	0.4422	−0.774	0.6282	0.084	−0.9974	−1.0088
0.95	0.7297	0.4313	−0.7312	0.6227	0.0846	−1.0317	−1.0415
1.0	0.6531	0.4207	−0.691	0.6172	0.0852	−1.0595	−1.0678
1.05	0.5836	0.4103	−0.6533	0.6117	0.0859	−1.0820	−1.0889
1.1	0.5204	0.4003	−0.6178	0.6061	0.0865	−1.0999	−1.1056
1.15	0.4626	0.3906	−0.5843	0.6006	0.0872	−1.1140	−1.1186
1.2	0.4098	0.3811	−0.5528	0.5951	0.0879	−1.1249	−1.1285
1.25	0.3613	0.37200	−0.523	0.5897	0.0886	−1.1330	−1.1358
1.3	0.3167	0.3631	−0.4949	0.5842	0.0893	−1.1389	−1.1409
1.35	0.2755	0.3546	−0.4683	0.5788	0.09	−1.1427	−1.1441
1.4	0.2376	0.3463	−0.4431	0.5734	0.0907	−1.1448	−1.1457
1.45	0.2024	0.3383	−0.4192	0.5681	0.0915	−1.1454	−1.1459
1.5	0.1699	0.3305	−0.3966	0.5628	0.0922	−1.1448	−1.145
1.55	0.1397	0.32299	−0.3751	0.5575	0.09300	−1.1431	−1.1432
1.6	0.1116	0.3157	−0.3548	0.5524	0.0938	−1.1404	−1.1405
1.65	0.0855	0.3087	−0.3354	0.5472	0.0946	−1.1370	−1.1371
1.7	0.0612	0.3018	−0.317	0.5422	0.0954	−1.1329	−1.1332
1.75	0.0385	0.2952	−0.2995	0.5371	0.0962	−1.1281	−1.1287
1.8	0.0173	0.2888	−0.2829	0.5322	0.09699	−1.1230	−1.1239
1.85	−0.0023	0.2826	−0.267	0.5273	0.0978	−1.1183	−1.1187
1.9	−0.0208	0.2766	−0.252	0.5225	0.0987	−1.1131	−1.1133
1.95	−0.0381	0.2707	−0.2376	0.5177	0.0995	−1.1076	−1.1077
2.0	−0.0543	0.2651	−0.2238	0.513	0.1004	−1.1018	−1.1019
2.05	−0.0694	0.2596	−0.2108	0.5084	0.1012	−1.0958	−1.0961
2.1	−0.0837	0.2542	−0.1983	0.5039	0.1021	−1.0895	−1.0901
2.15	−0.0969	0.249	−0.1863	0.4994	0.10300	−1.0831	−1.0842
2.2	−0.1095	0.244	−0.1749	0.495	0.1038	−1.0765	−1.0782
2.25	−0.1213	0.2391	−0.1639	0.4906	0.1047	−1.0699	−1.0723
2.3	−0.1323	0.2343	−0.1536	0.4864	0.1056	−1.0630	−1.0664
2.35	−0.1427	0.2297	−0.1436	0.4822	0.1064	−1.0581	−1.0605
2.4	−0.1524	0.2252	−0.1341	0.478	0.1073	−1.0533	−1.0548
2.45	−0.1616	0.2208	−0.125	0.474	0.1082	−1.0484	−1.0492
2.5	−0.1703	0.2165	−0.1162	0.47	0.109	−1.0433	−1.0437

$$\min\left(-\sigma_z^1\sigma_z^2\prod_{i=3}^n\sigma_z^i\right) = \min\left(-8\frac{1+\sigma_z^1}{2}\frac{1+\sigma_z^2}{2}\frac{1+\prod_{i=3}^n\sigma_z^i}{2}\right.$$

$$\left. + 8\frac{1+\sigma_z^1+\sigma_z^2+\prod_{i=3}^n\sigma_z^i+\sigma_z^1\sigma_z^2+\sigma_z^1\prod_{i=3}^n\sigma_z^i+\sigma_z^2\prod_{i=3}^n\sigma_z^i}{8}\right)$$

$$= \min\left(5 - \prod_{i=3}^n\sigma_z^i - \sigma_z^1 - \sigma_z^2 + 2\sigma_z^{n+1} - 2\prod_{i=3}^{n+1}\sigma_z^i + \sigma_z^1\prod_{i=3}^n\sigma_z^i\right.$$

$$\left. + \sigma_z^2\prod_{i=3}^n\sigma_z^i - 4\sigma_z^1\sigma_z^{n+1} - 4\sigma_z^2\sigma_z^{n+1} + 3\sigma_z^1\sigma_z^2\right) \qquad (27)$$

By repeating this, we can reduce the k-local in σ_z terms to a 2-local Hamiltonian.

Mapping the H$_2$ Hamiltonian to an Ising-Type Hamiltonian

Here, we treat the hydrogen molecule in a minimal basis STO-6G. By considering the spin functions, the four molecular spin orbitals in H$_2$ are

$$|\chi_1\rangle = |\Psi_g\rangle|\alpha\rangle = \frac{|\Psi_{1s}\rangle_1 + |\Psi_{1s}\rangle_2}{\sqrt{2(1+S)}}|\alpha\rangle \qquad (28)$$

$$|\chi_2\rangle = |\Psi_g\rangle|\beta\rangle = \frac{|\Psi_{1s}\rangle_1 + |\Psi_{1s}\rangle_2}{\sqrt{2(1+S)}}|\beta\rangle \qquad (29)$$

$$|\chi_3\rangle = |\Psi_u\rangle|\alpha\rangle = \frac{|\Psi_{1s}\rangle_1 - |\Psi_{1s}\rangle_2}{\sqrt{2(1-S)}}|\alpha\rangle \qquad (30)$$

$$|\chi_4\rangle = |\Psi_u\rangle|\beta\rangle = \frac{|\Psi_{1s}\rangle_1 - |\Psi_{1s}\rangle_2}{\sqrt{2(1-S)}}|\beta\rangle \qquad (31)$$

where $|\Psi_{1s}\rangle_1$ and $|\Psi_{1s}\rangle_2$ are the spatial functions for the two atoms, respectively, $|\alpha\rangle$ and $|\beta\rangle$ are spin up and spin down, and $S = {}_1\langle\Psi_{1s}|\Psi_{1s}\rangle_2$ is the overlap integral.[39] The one- and two-electron integrals are given by

$$h_{ij} = \int d\vec{r}\chi_i^*(\vec{r})\left(-\frac{1}{2}\nabla - \frac{Z}{r}\right)\chi_j(\vec{r}) \tag{32}$$

$$h_{ijkl} = \int d\vec{r_1}d\vec{r_2}\chi_i^*(\vec{r_1})\chi_j^*(\vec{r_2})\frac{1}{r_{12}}\chi_k(\vec{r_2})\chi_l(\vec{r_1}) \tag{33}$$

Thus, we can write the second-quantization Hamiltonian of H_2:

$$
\begin{aligned}
H_{H_2} =\ & h_{00}a_0^\dagger a_0 + h_{11}a_1^\dagger a_1 + h_{22}a_2^\dagger a_2 + h_{33}a_3^\dagger a_3 \\
& + h_{0110}a_0^\dagger a_1^\dagger a_1 a_0 + h_{2332}a_2^\dagger a_3^\dagger a_3 a_2 + h_{0330}a_0^\dagger a_3^\dagger a_3 a_0 \\
& + h_{1221}a_1^\dagger a_2^\dagger a_2 a_1 + (h_{0220} - h_{0202})a_0^\dagger a_2^\dagger a_2 a_0 \\
& + (h_{1331} - h_{1313})a_1^\dagger a_3^\dagger a_3 a_1 \\
& + h_{0132}(a_0^\dagger a_1^\dagger a_3 a_2 + a_2^\dagger a_3^\dagger a_1 a_0) \\
& + h_{0312}(a_0^\dagger a_3^\dagger a_1 a_2 + a_2^\dagger a_1^\dagger a_3 a_0)
\end{aligned} \tag{34}
$$

By using the Bravyi–Kitaev transformation,[25] we have

$$
\begin{aligned}
a_0^\dagger &= \frac{1}{2}\sigma_x^3\sigma_x^1(\sigma_x^0 - i\sigma_y^0) & a_0 &= \frac{1}{2}\sigma_x^3\sigma_x^1(\sigma_x^0 + i\sigma_y^0) \\
a_1^\dagger &= \frac{1}{2}(\sigma_x^3\sigma_x^1\sigma_z^0 - i\sigma_x^3\sigma_y^1) & a_1 &= \frac{1}{2}(\sigma_x^3\sigma_x^1\sigma_z^0 + i\sigma_x^3\sigma_y^1) \\
a_2^\dagger &= \frac{1}{2}\sigma_x^3(\sigma_x^2 - i\sigma_y^2)\sigma_z^1 & a_2 &= \frac{1}{2}\sigma_x^3(\sigma_x^2 + i\sigma_y^2)\sigma_z^1 \\
a_3^\dagger &= \frac{1}{2}(\sigma_x^3\sigma_z^2\sigma_z^1 - i\sigma_y^3) & a_3 &= \frac{1}{2}(\sigma_x^3\sigma_z^2\sigma_z^1 + i\sigma_y^3)
\end{aligned} \tag{35}
$$

Thus, the Hamiltonian of H_2 takes the following form:

$$
\begin{aligned}
H_{H_2} =\ & f_0\mathbf{1} + f_1\sigma_z^0 + f_2\sigma_z^1 + f_3\sigma_z^2 + f_1\sigma_z^0\sigma_z^1 + f_4\sigma_z^0\sigma_z^2 \\
& + f_5\sigma_z^1\sigma_z^3 + f_6\sigma_x^0\sigma_z^1\sigma_x^2 + f_6\sigma_y^0\sigma_z^1\sigma_y^2 + f_7\sigma_z^0\sigma_z^1\sigma_z^2 \\
& + f_4\sigma_z^0\sigma_z^2\sigma_z^3 + f_3\sigma_z^1\sigma_z^2\sigma_z^3 + f_6\sigma_x^0\sigma_z^1\sigma_x^2\sigma_z^3 \\
& + f_6\sigma_y^0\sigma_z^1\sigma_y^2\sigma_z^3 + f_7\sigma_z^0\sigma_z^1\sigma_z^2\sigma_z^3
\end{aligned} \tag{36}
$$

We can utilize the symmetry that qubits 1 and 3 never flip to reduce the Hamiltonian to the following form which just acts on only two qubits

$$
\begin{aligned}
H_{H_2} &= g_0\mathbf{1} + g_1\sigma_z^0 + g_2\sigma_z^1 + g_3\sigma_z^0\sigma_z^1 + g_4\sigma_x^0\sigma_x^1 + g_4\sigma_y^0\sigma_y^1 \\
&= g_0\mathbf{1} + H_0
\end{aligned} \tag{37}
$$

$$g_0 = f_0 \quad g_1 = 2f_1 \quad g_2 = 2f_3 \quad g_3 = 2(f_4 + f_7) \quad g_4 = 2f_6 \tag{38}$$

$$
\begin{aligned}
g_0 &= 1.0h_{00} + 0.5h_{0000} - 0.5h_{0022} + 1.0h_{0220} + 1.0h_{22} \\
&\quad + 0.5h_{2222} + 1.0/R \\
g_1 &= -1.0h_{00} - 0.5h_{0000} + 0.5h_{0022} - 1.0h_{0220} \\
g_2 &= 0.5h_{0022} - 1.0h_{0220} - 1.0h_{22} - 0.5h_{2222} \\
g_3 &= -1.0h_{00} - 0.5h_{0000} + 0.5h_{0022} - 1.0h_{0220} \\
g_4 &= 0.5h_{0022}
\end{aligned} \tag{39}
$$

where $\{g_i\}$ depends on the fixed bond length of the molecule. In Table 1, we present the numerical values of $\{g_i\}$ as a function of the internuclear distance in the minimal basis set STO-6G.

By applying the mapping method described above, we can get the Hamiltonian H' consisting of only σ_z (where i_1 and i_2 mean the 1 and 2 qubits of ith 2 qubits):

$$
\begin{aligned}
H' =\ & \sum_i g_1\sigma_z^{i_1} + g_2\sigma_z^{i_2} + g_3\sigma_z^{i_1}\sigma_z^{i_2} \\
& + \sum_{i\neq j}\left[g_1\frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(1 + \sigma_z^{i_2}\sigma_z^{j_2})}{4}S'(i)S'(j) \right. \\
& + g_2\frac{(\sigma_z^{i_2} + \sigma_z^{(j,2)})(1 + \sigma_z^{i_1}\sigma_z^{j_1})}{4}S'(i)S'(j) \\
& + g_3\frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2})}{4}S'(i)S'(j) \\
& + g_4\frac{(1 + \sigma_z^{i_1}\sigma_z^{i_1})(1 + \sigma_z^{i_2}\sigma_z^{j_2})}{4}S'(i)S'(j) \\
& \left. + g_4\frac{(\sigma_z^{i_1} - \sigma_z^{i_1})(\sigma_z^{j_2} - \sigma_z^{i_2})}{4}S'(i)S'(j) \right]
\end{aligned} \tag{40}
$$

According to the scheme for reducing locality, if we want to reduce $H' - \lambda C$, we can reduce H' and C separately. By applying the method for reducing locality, we can get a 2-local Ising-type Hamiltonian, H''. Here we show the example of all signs being positive, where a_{ij}^k, $k = 1, 2, 3, ..., 6$, are the indices of the new qubits we introduce to reduce locality.

$$
\begin{aligned}
H'' =\ & \sum_i g_1\sigma_z^{i_1} + g_2\sigma_z^{i_2} + g_3\sigma_z^{i_1}\sigma_z^{i_2} + \sum_{i\neq j}\left[g_1\frac{\sigma_z^{i_1} + \sigma_z^{j_1}}{4} + g_2\frac{\sigma_z^{i_2} + \sigma_z^{j_2}}{4} \right. \\
& + g_3\frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2})}{4} + g_4\frac{1 + \sigma_z^{i_1}\sigma_z^{j_1} + \sigma_z^{i_2}\sigma_z^{j_2}}{4} \\
& + g_4\frac{(\sigma_z^{i_1} - \sigma_z^{j,1})(\sigma_z^{i_2} - \sigma_z^{j_2})}{4} \\
& + g_4\frac{7 + \sigma_z^{i_2}\sigma_z^{j_2} - 3\sigma_z^{i_1} - 3\sigma_z^{j_1} + 6\sigma_z^{a_{ij}^1} - 4\sigma_z^{i_1}\sigma_z^{a_{ij}^1} - 4\sigma_z^{j_1}\sigma_z^{a_{ij}^1} + \sigma_z^{i_1}\sigma_z^{j_1}}{4} \\
& + (g_1 - g_4) \\
& \frac{14 + \sigma_z^{i_1} + \sigma_z^{j_1} - 6(\sigma_z^{i_2} + \sigma_z^{j_2}) + 6(\sigma_z^{a_{ij}^2} + \sigma_z^{a_{ij}^3}) + 2(\sigma_z^{i_1}\sigma_z^{a_{ij}^2} + \sigma_z^{j_1}\sigma_z^{a_{ij}^3})}{4} \\
& + (g_1 - g_4) \\
& \frac{-4(\sigma_z^{i_2} + \sigma_z^{j_2})(\sigma_z^{a_{ij}^2} + \sigma_z^{a_{ij}^3}) - (\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2}) + 2\sigma_z^{i_2}\sigma_z^{j_2}}{4} \\
& + g_2 \\
& \frac{10 - \sigma_z^{i_2} - \sigma_z^{j_2} - 2(\sigma_z^{i_1} + \sigma_z^{j_1}) + 2(\sigma_z^{a_{ij}^4} + \sigma_z^{a_{ij}^5}) - 2(\sigma_z^{i_2}\sigma_z^{a_{ij}^4} + \sigma_z^{j_2}\sigma_z^{a_{ij}^5})}{4} \\
& + g_2 \\
& \frac{-4(\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{a_{ij}^4} + \sigma_z^{a_{ij}^5}) + (\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2}) + 6\sigma_z^{i_1}\sigma_z^{j_1}}{4} \\
& + g_4\frac{7 + \sigma_z^{a_{ij}^1} - 3\sigma_z^{i_2} - 3\sigma_z^{j_2} + 6\sigma_z^{a_{ij}^6} + 2\sigma_z^{a_{ij}^1}\sigma_z^{a_{ij}^6}}{2} \\
& \left. + g_4\frac{-\sigma_z^{i_2}\sigma_z^{a_{ij}^1} - \sigma_z^{j_2}\sigma_z^{a_{ij}^1} - 4\sigma_z^{i_2}\sigma_z^{a_{ij}^6} - 4\sigma_z^{j_2}\sigma_z^{a_{ij}^6} + \sigma_z^{i_2}\sigma_z^{j_2}}{2} \right]
\end{aligned} \tag{41}
$$

We can write the corresponding count term C as

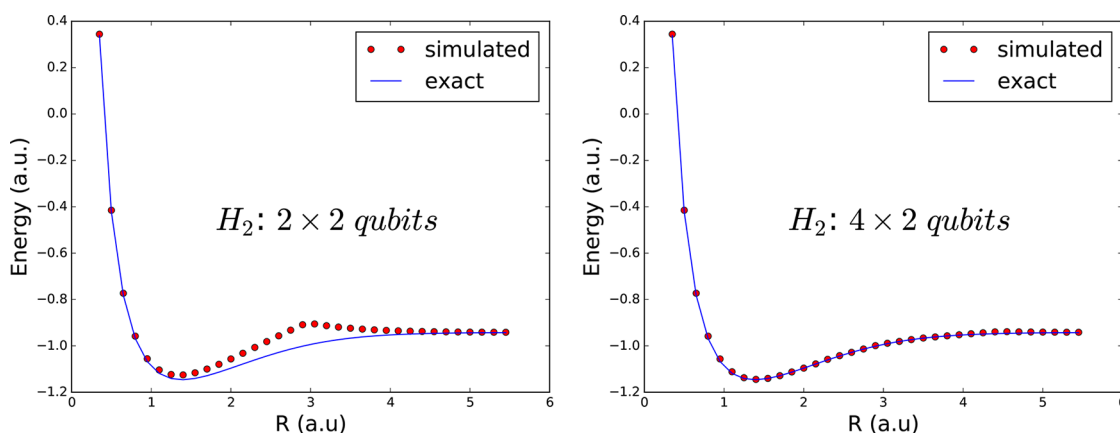$$C = \sum_{\pm}\left(\sum_i\frac{(1 \pm \sigma_z^{i_1})(1 \pm \sigma_z^{i_2})}{4}\right)^2 \tag{42}$$

Figure 6. Results of the simulated transformed Ising-type Hamiltonian with 2×2 qubits and 4×2 qubits compared with the exact numerical results for the ground state of the H_2 molecule.

By applying the method of reducing locality, the 2-local corresponding count term is

$$
\begin{aligned}
C' = \frac{1}{4} \Bigg\{ \sum_{i \neq j} \Big[& 7 + \sigma_z^{i_2}\sigma_z^{j_2} - 3\sigma_z^{i_1} - 3\sigma_z^{j_1} + 6\sigma_z^{a_{ij}^7} - 4\sigma_z^{i_1}\sigma_z^{a_{ij}^7} \\
& - 4\sigma_z^{j_1}\sigma_z^{a_{ij}^7} + \sigma_z^{i_1}\sigma_z^{j_1} + 2(7 + \sigma_z^{a_{ij}^7} - 3\sigma_z^{i_2} - 3\sigma_z^{j_2} \\
& + 6\sigma_z^{a_{ij}^8} + 2\sigma_z^{a_{ij}^7}\sigma_z^{a_{ij}^8}) + 2(-\sigma_z^{i_2}\sigma_z^{a_{ij}^7} - \sigma_z^{j_2}\sigma_z^{a_{ij}^7} \\
& - 4\sigma_z^{i_2}\sigma_z^{a_{ij}^8} - 4\sigma_z^{j_2}\sigma_z^{a_{ij}^8} + \sigma_z^{i_2}\sigma_z^{j_2}) + 14 + \sigma_z^{i_1} + \sigma_z^{j_1} \\
& - 6(\sigma_z^{i_2} + \sigma_z^{j_2}) + 6(\sigma_z^{a_{ij}^9} + \sigma_z^{a_{ij}^{10}}) \\
& + 2(\sigma_z^{i_1}\sigma_z^{a_{ij}^9} + \sigma_z^{j_1}\sigma_z^{a_{ij}^{10}}) - 4(\sigma_z^{i_2} + \sigma_z^{j_2})(\sigma_z^{a_{ij}^9} + \sigma_z^{a_{ij}^{10}}) \\
& - (\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2}) + 2\sigma_z^{i_2}\sigma_z^{j_2} + 1 + \sigma_z^{i_1}\sigma_z^{j_1} \\
& + \sigma_z^{i_2}\sigma_z^{j_2}] \Bigg\} + \sum_i \frac{(1 + \sigma_z^{i_1}) + (1 + \sigma_z^{i_2})}{4}
\end{aligned}
$$

$$(43)$$

As stated in the section "Reduce Locality of the Transformed Hamiltonian", a change in the locality would not change the state when calculating the ground state energy. Thus, we can still use C on certain qubits to calculate $\sum_i a_i^2$, and the algorithm we present above can still be used for the reduced Hamiltonian.

In Figure 6, we show our results from the transformed Ising-type Hamiltonian of 2×2 to 4×2 qubits compared with the exact numerical values. By increasing the number of qubits via $r = 2$, we increased the accuracy and the result matches very well with the exact results. Table 1 shows the numerical results.

Mapping the Hamiltonian for the He$_2$ Molecule to the Ising-Type Hamiltonian

As shown above for transforming the Hamiltonian associated with the H_2 molecule, we repeat the procedure for the helium molecule in a minimal basis STO-6G using the Jordan−Wigner transformation.

The molecular spin Hamiltonian has the form

$$
\begin{aligned}
H_{\mathrm{He}_2} = & f_0 \mathbf{1} + f_1 \sigma_z^1 + f_2 \sigma_z^2 + f_3 \sigma_z^3 + f_4 \sigma_z^4 + f_5 \sigma_z^1 \sigma_z^2 \\
& + f_6 \sigma_z^1 \sigma_z^3 + f_7 \sigma_z^1 \sigma_z^4 + f_8 \sigma_z^2 \sigma_z^3 + f_8 \sigma_z^2 \sigma_z^4 \\
& + f_9 \sigma_z^3 \sigma_z^4 + f_{10} \sigma_x^1 \sigma_x^2 \sigma_y^3 \sigma_y^4 + f_{11} \sigma_x^1 \sigma_y^2 \sigma_y^3 \sigma_x^4 \\
& + f_{12} \sigma_y^1 \sigma_x^2 \sigma_x^3 \sigma_y^4 + f_{13} \sigma_y^1 \sigma_y^2 \sigma_x^3 \sigma_x^4
\end{aligned}
$$

$$(44)$$

The set of parameters f_i is related to the one- and two-electron integrals:

$$
\begin{aligned}
f_0 = & 1.0h_{00} + 0.25h_{0000} - 0.5h_{0022} + 1.0h_{0220} + 1.0h_{22} \\
& + 0.25h_{2222} + 4.0/R \\
f_1 = & -0.5h_{00} - 0.25h_{0000} + 0.25h_{0022} - 0.5h_{0220} \\
f_2 = & -0.25h_{0000} + 0.25h_{0022} - 0.5h_{0220} - 0.5h_{00} \\
f_3 = & 0.25h_{0022} - 0.5h_{0220} - 0.5h_{22} - 0.25h_{2222} \\
f_4 = & 0.25h_{0022} - 0.5h_{0220} - 0.25h_{2222} - 0.5h_{22} \\
f_5 = & 0.25h_{0000} \\
f_6 = & -0.25h_{0022} + 0.25h_{0220} \\
f_7 = & 0.25h_{0220} \\
f_8 = & 0.25h_{0220} \\
f_9 = & 0.25h_{2222} \\
f_{10} = & -0.25h_{0022} \\
f_{11} = & 0.25h_{0022} \\
f_{12} = & 0.25h_{0022} \\
f_{13} = & -0.25h_{0022}
\end{aligned}
$$

$$(45)$$

We can also use the mapping and reduction of locality as before to get the final Ising Hamiltonian. Here we just present the mapping result of some terms for illustration. For σ_z^1, the Hamiltonian between different bases can be mapped as

$$
\sum_{i \neq j} \frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(1 + \sigma_z^{i_2}\sigma_z^{j_2})(1 + \sigma_z^{i_3}\sigma_z^{j_3})(1 + \sigma_z^{j_4}\sigma_z^{i_4})}{16} S'(i)S'(j)
$$

$$(46)$$

For $\sigma_z^1 \sigma_z^2$, the Hamiltonian between different bases can be mapped as

$$
\sum_{i \neq j} \frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2})(1 + \sigma_z^{i_3}\sigma_z^{j_3})(1 + \sigma_z^{j_4}\sigma_z^{i_4})}{16} S'(i)S'(j)
$$

$$(47)$$

For $\sigma_x^1 \sigma_x^2 \sigma_y^3 \sigma_y^4$, the Hamiltonian between different bases can be mapped as

$$\sum_{i\neq j} \frac{(1 - \sigma_z^{i_1}\sigma_z^{j_1})(1 - \sigma_z^{i_2}\sigma_z^{j_2})(\sigma_z^{i_3} - \sigma_z^{j_3})(\sigma_z^{j_4} - \sigma_z^{i_4})}{16} S'(i)S'(j) \tag{48}$$

By reducing locality, we can get the 2-local Ising-type Hamiltonian. However, even just for $\sigma_x^1 \sigma_x^2 \sigma_y^3 \sigma_y^4$, the final 2-local Ising Hamiltonian would have about 1000 terms.

Mapping the Hamiltonian of HeH⁺ to an Ising Hamiltonian

Similar to H_2 and He_2 molecules, next we treat the HeH⁺ molecule in the minimal basis STO-6G using the Jordan–Wigner transformation. Using the technique defined above,[40] we can reduce the locality to

$$H_{HeH^+} = f_0 \mathbf{1} + f_1 \sigma_z^1 + f_2 \sigma_z^2 + f_3 \sigma_x^1 + f_4 \sigma_z^1 \sigma_z^2 + f_5 \sigma_x^1 \sigma_z^2$$
$$+ f_6 \sigma_z^1 \sigma_x^2 + f_7 \sigma_x^1 \sigma_x^2 + f_8 \sigma_y^1 \sigma_y^2 \tag{49}$$

The set of parameters f_i is related to the one- and two-electron integrals:

$$f_0 = 1.0 h_{00} + 0.25 h_{0000} + 0.5 h_{0220} + 1.0 h_{22} + 0.25 h_{2222}$$
$$\quad + 2.0/R$$
$$f_1 = -0.25 h_{0000} + 0.5 h_{0220} - 0.25 h_{2222}$$
$$f_2 = 0.25 h_{0000} + 0.5 h_{00} - 0.25 h_{2222} - 0.5 h_{22}$$
$$f_3 = -0.25 h_{0002} - 0.25 h_{0020} + 0.5 h_{0222}$$
$$f_4 = -0.5 h_{00} - 0.25 h_{0000} + 0.5 h_{22} + 0.25 h_{2222}$$
$$f_5 = -0.25 h_{0002} - 0.25 h_{0020} - 0.5 h_{0222}$$
$$f_6 = -1.0 h_{0022}$$
$$f_7 = 0.25 h_{0002} + 0.25 h_{0020} - 0.5 h_{0222}$$
$$f_8 = 0.25 h_{0002} + 0.25 h_{0020} + 0.5 h_{0222} \tag{50}$$

We can also use the mapping and reducing locality as before to get the final Ising Hamiltonian. Again, here we just present the mapping result of some terms for illustration:

For $\sigma_z^1 \sigma_z^2$, the Hamiltonian between different bases can be mapped as

$$\sum_{i\neq j} \frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2})}{4} S'(i)S'(j) \tag{51}$$

For $\sigma_z^1 \sigma_x^2$, the Hamiltonian between different bases can be mapped as

$$\sum_{i\neq j} \frac{(\sigma_z^{i_1} + \sigma_z^{j_1})(1 - \sigma_z^{i_2}\sigma_z^{j_2})}{4} S'(i)S'(j) \tag{52}$$

And if the coefficient of mapping term is positive, we can get the 2-local term as

$$\sum_{i\neq j} \left(\frac{\sigma_z^{i_1} + \sigma_z^{j_1}}{4} \right.$$
$$+ \frac{14 + \sigma_z^{i_1} + \sigma_z^{j_1} - 6(\sigma_z^{i_2} + \sigma_z^{j_2}) + 6(\sigma_z^{a_{ij}^1} + \sigma_z^{a_{ij}^2}) + 2(\sigma_z^{i_1}\sigma_z^{a_{ij}^1} + \sigma_z^{j_1}\sigma_z^{a_{ij}^2})}{4}$$
$$\left. + \frac{-4(\sigma_z^{i_2} + \sigma_z^{j_2})(\sigma_z^{a_{ij}^1} + \sigma_z^{a_{ij}^2}) - (\sigma_z^{i_1} + \sigma_z^{j_1})(\sigma_z^{i_2} + \sigma_z^{j_2}) + 2\sigma_z^{i_2}\sigma_z^{j_2}}{4} \right) \tag{53}$$

Mapping the Molecular Hamiltonian of LiH to an Ising Hamiltonian

Similar to H_2 and other molecules, next we treat the LiH molecule with four electrons in a minimal basis STO-6G and use the Jordan–Wigner transformation. Using the technique defined above,[41] we can reduce the locality to a Hamiltonian with 558 terms on 8 qubits. We just use 16 qubits for the simulations.

Computation Complexity and Error Analysis

Specifying the required amount of qubit resources can be derived throughout the multistep process of constructing the final Hamiltonian. As an example, let us consider a Hamiltonian with n total orbitals; transforming this Hamiltonian from second quantization to Pauli operators requires $O(n)$ qubits and a series of $O(n^4)$ expansion terms. This is due to the need to use two-body operator and four-body operator terms in formulating the second quantization Hamiltonian. Thus, the number of terms in the second quantization form of the Hamiltonian should be $O(n^4)$, yielding $O(n^4)$ terms to be described in the Pauli operator representation of the Hamiltonian.

Mapping the Hamiltonian from a Pauli operator representation to a diagonal matrix requires an additional $O(rn)$ factor of qubits, and thereby $O(2^n r^2 n^4)$ qubits. This is due to the fact that, for each term in the Pauli operator representation of the Hamiltonian, we now have $O(2^n r^2)$ terms in the diagonal Hamiltonian. This process leads us to the $O(r^2 n^4)$ mapping, which does—in fact—have $O(2^n r^2 n^4)$ terms.

Reducing the representation down to 2-local interactions requires $O(rn + 2^n r^2 n^7)$ qubits. Within this form, we do not know r. Consider that the qubit requirement during the reduction from a k-local to 2-local is given by $f(k)$. We have found that $f(k) < 10 f\left(\frac{2}{3}k\right)$ which gives us $f(k) = O(n^{\log_3^{10}})$ or $O(n^3)$ by the master theorem. Thus, for all terms, we finally have $O(rn + 2^n r^2 n^7)$ qubits.

However, we cannot clearly give a relationship between r and the desired accuracy in the solution. Similarly to the variation method trying to optimize results under particular conditions, this mapping tries to approach the optimal value of the desired ground eigenstate via a new state a by repetition of terms r times. Thus, we cannot calculate the errors associated with our optimal result compared to the exact result, because exact results are unknown within the varational method. Through repeated scaled calculation, we can see that in increasing r we obtain greater accuracy. This is likely due to the fact that by increasing r we increase the number of repetitious terms providing the necessary corrections allowing b_i to approach a_i.

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Notes

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