Exact electronic properties and processes relies on solving the Schrödinger equation to find the many-electron wave function. However, this problem scales exponentially with the number of electrons and practical digital algorithms rely on approximations that fail for strong electronic correlation. Therefore, accurate simulations of technologically important processes, such as transition metal catalysis and high-temperature superconductivity, remain challenging. Gate-based quantum computation offers a revolutionary approach with the potential to overcome this exponential scaling and provide a general solution to quantum chemistry.

Current and near-term quantum devices are impaired by noise and are limited to shallow quantum circuits. The most promising near-term approaches optimise the parameters of an ansatz for the quantum state, and algorithms such as the variational quantum eigensolver (VQE) have the potential to outperform classical computational methods with modest quantum resources. However, the best choice of ansatz for strongly correlated wave functions is far from clear, raising the question of whether near-term quantum computing will be useful for addressing unsolved quantum chemical problems.

We present a method for designing highly compact ansätze for quantum computational chemistry by combining products of symmetry-preserving fermionic operators with a global optimisation algorithm, the Discretely Optimised Variational Quantum Eigensolver (DISCO-VQE), which optimises both the discrete sequence of operators and their variational parameters. Numerical simulations using this approach demonstrate that accurate wave functions for strongly correlated electronic systems can be parametrised with efficient quantum circuits. These advances provide a route towards practical simulations of strong electron correlation on quantum devices, bringing us closer to solving challenging problems in quantum chemistry.

Unitary product states

All ansatz-based approaches apply a unitary transformation \( U \) to an initial state \( |\Phi_0\rangle \) in the form of a quantum circuit, constructed through a series of parametrised building blocks as \( U = U_1(\theta_1) \cdots U_N(\theta_N) \). The parameters \( \theta_i \) are optimised on a classical computer by extremising an objective function evaluated using the quantum computer. This product of unitary transformations defines a family of wave functions that we term unitary product states (UPS). The UPS paradigm is completely general and encompasses every ansatz that has been proposed for quantum computational chemistry so far.

The predominant ansätze use either “hardware-efficient” unitary operators constructed from qubit operations that minimise the depth of a specific quantum circuit, or “physically-motivated” fermionic unitary operations based on the second-quantised representation of electronic systems. Second-quantised fermionic operators ensure that the quantum circuit satisfies particle number symmetry and Pauli antisymmetry, making them appropriate for electronic states. While physically-motivated operators typically reduce the number of ansatz parameters, each fermionic operator requires multiple gate operations that lead to deep quantum circuits. Therefore, there is an apparent trade-off between symmetry-preservation and gate efficiency. Conserving physical symmetries ensures that truncated wave functions predict accurate properties beyond the energy and can be used as initial states for fault-tolerant quantum phase estimation, where efficiency depends on the overlap of the initial and exact states.

Initial developments using fermionic operators were based on Unitary Coupled Cluster (UCC) schemes, inspired by the success of many-body Coupled Cluster theory for classical computing. The UCC unitary transformation is an exponential of a sum of anti-Hermitian fermionic operators, usually restricted to one- and two-body excitations. The first step to represent the UCC ansatz as a quantum circuit is converting this operator into the UPS form. Since fermionic operators generally do not commute, expanding the sum requires a Trotter approximation that is only exact in the infinite limit: \( e^{A+B} = \lim_{m \to \infty} (e^{A/m} e^{B/m})^m \).

Instead, focus has shifted to a particular UPS form of the UCC wave function called disentangled UCC. While it has been shown theoretically that exact electronic states can be represented using a suitably-ordered infinite product of one- and two-body fermionic operators, in practice, finite expansions are sensitive to the operator order and often fail for strong correlation. Moreover, spin symmetry corresponding to the \( S_z \) operator is not conserved because the non-commuting spin components of unpaired two-body operators appear independently in the unitary product. The ADAPT-VQE algorithm,
and its extensions,\textsuperscript{14–17} iteratively build a UPS from a pool of one- and two-body fermionic operators, selecting the operator with the largest energy improvement at each step. Although promising, these approaches are not guaranteed to converge to the exact state\textsuperscript{18,19} or identify the most accurate representation with the fewest number of operators, and can spontaneously break spin symmetry.

The ideal unitary product \textit{ansatz} should meet the following requirements:

- Fidelity — symmetries of the electronic Hamiltonian should be preserved by each operator;
- Universality — any electronic eigenstate should be accessible;
- Practicality — the circuit depth and number of entangling gates should be small.

Here, we show that these requirements can be fulfilled by a symmetry-preserving unitary product state (s-UPS) that conserves Pauli antisymmetry, particle number symmetry, $\hat{S}_z$ and $\hat{S}^2$ spin symmetry using a pool of elementary fermionic operators that grows quadratically with the system size. We prove that this \textit{ansatz} is universal and demonstrate that it is gate-efficient. The appropriate choice and ordering of the operators is system-dependent and simultaneously optimising the sequence of operators and their variational parameters is challenging. We realise this goal by introducing the DISCO-VQE algorithm, which finds the best ordered set of operators for a given number of operators. Our approach yields gate-efficient quantum circuits that prepare an accurate s-UPS for strongly correlated systems using remarkably few variational parameters.

**Symmetry-preserving unitary product states**

Fermionic unitary operators are defined through an exponential of anti-Hermitian second-quantised operators $\hat{\kappa}_{pq...} := a_p^\dagger a_q^\dagger \cdots a_r^\dagger a_s \cdots$, where the indices $p,q \ldots$ denote arbitrary molecular spin orbitals and the absence (presence) of an overbar indicates a high-spin (low-spin) orbital. Individual operators may appear multiple times and each operator has its own continuous parameter, defined by the continuous coordinates $t = (t_1, \ldots, t_M)$. Because these anti-Hermitian operators in general do not commute, the \textit{ansatz} depends on the choice and ordering of the unitary operators, denoted by the ordered set of indices $\mu = (\mu_1, \ldots, \mu_M)$.

Using generalised fermionic operators, where $p,q,r,s$ correspond to arbitrary molecular spin orbitals, removes any connection between the operators in the s-UPS \textit{ansatz} and the separation of occupied or virtual molecular orbitals in some mean-field reference. In contrast to the many-body perturbation philosophy, the generalised one- and two-body operators are Lie algebra generators for unitary transformations in the $D$-dimensional Hilbert space and can rotate any initial state onto an arbitrary exact state. Universality of a UPS \textit{ansatz} built from all generalised one- and two-body fermionic operators has already been proved\textsuperscript{20} by realising that the full $1/2 D(D-1)$ dimensional Lie algebra\textsuperscript{21} can be generated using operators that can be decomposed as nested commutators of the generalised one- and two-body fermionic operators. The universality of the s-UPS \textit{ansatz} follows from the additional observation that generalised two-body operators can be obtained through nested commutators of one-body and paired two-body operators (supplementary materials, section S1).

Since our elementary operators are spin-adapted (i.e., the commutators $[\hat{S}_z^2, \hat{\kappa}]$ and $[\hat{S}_z, \hat{\kappa}]$ vanish) our \textit{ansatz} conserves the spin quantum numbers of the initial state, avoiding spin-constrained optimisation\textsuperscript{12} or spin projection.\textsuperscript{16} Conserving spin symmetry for each operator ensures that the s-UPS \textit{ansatz} is spin-pure for every truncation $M$. In contrast, spin adaptation of unpaired two-body operators is only obtained by exponentiating a sum of non-commuting operators, which requires a Trotter expansion to be applied on a quantum circuit. The spin adaptation is destroyed when this Trotter expansion is approximated.\textsuperscript{12} Furthermore, every Pauli-$Z$ operator in the qubit representation of the paired two-body operators $\hat{\kappa}_{pq}$ can be removed by cancellation, hence these operators can be implemented with a constant number of CNOT gates. In contrast, the CNOT count for unpaired two-body operators grows linearly with the number of orbitals (supplementary materials, section S2.2).

Universality of the s-UPS \textit{ansatz} is assured because the product of two unitary operators corresponds to an exponential of a sum of operators through the Baker–Campbell–Haussdorff expansion\textsuperscript{22} $e^{A+B} = e^A e^B + \frac{1}{2} [A,B] + \cdots$. Since these nested commutators generate the full Lie algebra, combining a sufficient number of them in a suitable order will eventually represent any unitary transformation in the Hilbert space. We postulate that $M \leq (D - 1)$ will allow any initial state to be rotated into an exact state, avoiding redundant parameters in the exact wave function, and provide numerical examples that support this. In practice, sufficient accuracy is expected with a much smaller value of $M$. This perspective contrasts with Trotterised UCC,\textsuperscript{9,11} where the unitary product form arises from expanding an exponential sum of operators as $e^A e^B = \lim_{m \to \infty} (e^{A/m} e^{B/m})^m$ and $n$-body operators are decomposed into an infinite unitary product through nested commutators (see Eq. (24) in Ref. 9).

**Discretely optimised variational quantum eigensolver**

While electronic states can be represented using a product of spin-adapted generalised one- and two-body exponential operators, this construction requires the correct ordered set of operators. Fixed operator orderings are typically a poor approximation to the optimal unitary product state, while simply increasing the number of operators ultimately leads to redundancies.\textsuperscript{13,24} In the absence of \textit{a priori} information, the
Basis-hopping uses random perturbations followed by minimisation to step between local minima on the continuous energy landscape and hence search for the global minimum (supplementary materials, section S3.1). After a series of basis-hopping steps, DISCO-VQE identifies the lowest-energy configuration that can be reached by performing a discrete cyclic permutation followed by reoptimisation of the continuous coordinates. Similarly, for operator mutations and pair swaps, we identify the lowest energy mutation or pair swap for each operator and accept this step if it lowers the energy (supplementary materials, section S3.2). By testing mutations and pair swaps for every operator individually, DISCO-VQE can completely change the discrete parameters in each macro-cycle and, in principle, the optimal ordered operator set could be discovered after one macro iteration. In practice, the combined space of continuous and discrete parameters also exhibits many local biminima and uphill moves are required to escape from energy traps (supplementary materials, section S3.3). The full DISCO-VQE algorithm is detailed in the supplementary materials, section S3.

**Exact wave functions using DISCO-VQE**

The universality of the s-UPS ansatz means that any exact wave function with an arbitrarily small energy error can be obtained using a finite number of unitary operators selected from the pool of spin-adapted one-body and paired two-body operators. DISCO-VQE calculations on the linear and tetrahedral structures of H₄ (supplementary materials, section S4) show that chemical accuracy (1.59 mEₕ) can be obtained with only nine and five operators, respectively (Fig. 2A–B). Furthermore, ground states with an energy error within the precision of our numerical simulations (10⁻⁹ Eₕ) can be identified using thirteen (linear) or eight (tetrahedron) operators. These results support our postulate that an exact s-UPS representation can be constructed with a finite number of operators less than the Hilbert space size and show that the spin-adapted one-body and paired two-body operators are sufficient for defining a universal ansatz.

Global optimisation of the continuous coordinates and discrete operator ordering is essential for realising the universality of the s-UPS ansatz. The algorithmic advance provided by DISCO-VQE is clear from comparison with ADAPT-VQE calculations using the same pool of operators. By adding one operator at a time, ADAPT-VQE performs a local optimisation that can converge onto a non-exact wave function, as demonstrated in Fig. 2A–B. This stagnation of the wave function is reminiscent of the symmetry roadblock described in Ref. 18 and indicates that ADAPT-VQE can struggle to create the cooperative operator interactions required to represent an exact state. In contrast, DISCO-VQE can escape these local minima in the discrete space and provides the first practical algorithm for globally optimising the s-UPS wave function.

Computational chemistry requires wave functions that provide consistent accuracy for every geometry of a molecule. DISCO-VQE achieves this task for linear H₄ by producing an exact wave function using thirteen operators at every bond length (Fig. 2C). Fixing the operators to the ordering identified for the exact solution at equilibrium gives near-exact energies across the full binding curve, with a maximum error of 0.4 μEₕ.
(Fig. S2), demonstrating that highly-accurate and transferable fixed ansätze can in principle be defined. In comparison, random fixed ansätze with thirteen operators are relatively accurate near equilibrium, but exhibit a large variation in the dissociation limit (Fig. 2C). Furthermore, ADAPT-VQE calculations provide an accurate description of equilibrium and dissociation, but have an appreciable error at intermediate geometries, where static and dynamic correlation compete (Fig. 2C). Only six operators are required to surpass the accuracy of ADAPT-VQE using DISCO-VQE, while chemically accurate binding curves can be obtained with ten operators (Fig. S3).

**Accuracy of truncated approximations**

Despite the promise of quantum computers, practical calculations on large molecular systems will still require approximate wave function truncations. Global optimisation of the s-UPS ansatz using DISCO-VQE provides the most accurate approximation for a given number of fermionic operators, while conserving spin symmetry at every truncation. For example, DISCO-VQE calculations on strongly-correlated linear H\textsubscript{6} (supplementary materials, section S5) achieve chemical accuracy using only 30 operators (Fig. 3A). In contrast, ADAPT-VQE using the symmetry-pure operator pool stagnates with between 14–40 operators and fails to identify the exact ground state (Fig. 3A). DISCO-VQE requires only 10 operators to surpass the accuracy of the ADAPT-VQE binding curve (Fig. 3B).

Furthermore, DISCO-VQE provides significantly greater accuracy using a smaller number of variational parameters than previous related methods with fermionic operators, including the un-Trotterised \(k\)-UpCCGSD\textsuperscript{24} and the Jastrow-based \(k\)-uCI\textsuperscript{32} approaches (Fig. 3C). Only 30 operators are required to surpass the accuracy of the un-Trotterised 2-UpCCGSD wave function with 90 operators\textsuperscript{11} or the 2-uCI approach with 150 operators,\textsuperscript{32} while the original ADAPT-VQE calculations using non-symmetry-preserving operators required at least 78 operators to obtain comparable results.\textsuperscript{13} Consequently, global optimisation using DISCO-VQE defines a new standard for the accuracy that can be obtained with a certain number of fermionic unitaries in linear H\textsubscript{6}.

Symmetrically stretched water and the dissociation of N\textsubscript{2} provide important testing grounds for the performance of wave function approximations. DISCO-VQE calculations for a truncated s-UPS ansatz produce highly accurate energies for the full H\textsubscript{2}O binding curve (supplementary materials, section S6), with a non-parallelity error\textsuperscript{33} (NPE) of 0.004 mE\textsubscript{h} (Fig. 4A). In contrast, ADAPT-VQE, using the same operator pool, fails to identify a chemically accurate s-UPS wave function at any bond length (Fig. 4A), while previous studies using the full pool of one- and two-body operators are much less accurate than DISCO-VQE, with an error of 1.5 to 3.0 mE\textsubscript{h}.\textsuperscript{34} The original un-Trotterised \(k\)-UpCCGSD ansatz required 126 variational parameters to reach an accuracy of 0.07 mE\textsubscript{h},\textsuperscript{24} compared to the 42 operators used for DISCO-VQE, while recent Trotterised \(k\)-UpCCGSD calculations using over 250 operators have a larger residual error ranging from 0.02 to 200 mE\textsubscript{h}.\textsuperscript{35} Therefore, using DISCO-VQE to globally optimise a s-UPS wave function significantly improves the accuracy using fewer fermionic operators than previous state-of-the-art methods.

For the strongly-correlated dissociation of N\textsubscript{2}, DISCO-VQE achieves chemical accuracy at nearly every geometry using 30 operators (supplementary materials, section S7), giving a very accurate binding curve (Fig. 4B). In comparison, previous UPS representations built from randomly ordered sets of
FIG. 3: (A) DISCO-VQE with 30 operators provides chemically accurate energies for the linear H$_6$ binding curve (STO-3G). (B) Comparison of DISCO-VQE errors and the previous state-of-the-art methods across the full binding curve. ADAPT-VQE calculations use the same operator pool. The un-Trotterised 1-UpCCGSD and 2-UpCCGSD results are taken from Ref. 11. The non-parallelity error (NPE) is the difference between the maximum and minimum error in the energy. (C) DISCO-VQE produces highly accurate energies at $R$(H−H) = 2.0 Å with fewer continuous parameters than previous algorithms. The $k$-uCJ results are taken from Ref. 32.

FIG. 4: Wave functions built using DISCO-VQE accurately describe the potential energy surface for strongly correlated molecules. The number of operators in these DISCO-VQE calculations corresponds to the size of the operator pool in the STO-3G basis set, with (A) 42 operators for the symmetric stretch of H$_2$O and (B) 30 operators for the dissociation of N$_2$. The four lowest energy molecular orbitals are frozen in N$_2$. Un-Trotterised $k$-UpCCGSD results for N$_2$ are taken from Ref. 24.

all the generalised one- and two-body operators have residual errors ranging from 16 to 64 mEh, demonstrating the importance of globally optimising the operator ordering. The reduction in the number of parameters achieved by DISCO-VQE is demonstrated by comparing to the un-Trotterised $k$-UpCCGSD results, which require at least 180 variational parameters to obtain a better mean-average error$^{24}$ (Table S1). In contrast, ADAPT-VQE calculations with the same operator pool have a significant residual error and do not produce a smooth potential energy surface (Fig. 4B), while including non-symmetry-conserving one- and two-body operators breaks $S^2$ symmetry for truncated wave functions.$^{16}$ The s-UPS ansatz conserves the $S^2$ symmetry of the initial state at every truncation level and global optimisation using DISCO-VQE achieves a highly-accurate description of this challenging potential energy surface using a small number of operators.
Maximising the efficiency of quantum circuits

Applications on near-term quantum hardware must balance the accuracy of an algorithm against the number of two-qubit controlled-NOT (CNOT) gates in the quantum circuit, which contribute the largest source of hardware noise. This challenge has motivated the development of hardware-efficient ansätze where the pool of operators can be efficiently represented with a small number of CNOT gates, such as qubit-ADAPT-VQE\textsuperscript{14} and qubit-excitation-based (QEB) ADAPT-VQE.\textsuperscript{17} However, these methods do not satisfy fermionic antisymmetry and may also break particle number or $\hat{S}_z$ symmetry, giving an unphysical electronic state.

Despite employing a fermionic pool of operators, DISCO-VQE achieves chemical accuracy for linear $\text{H}_6$ using less than a third of the CNOT gates required for previous hardware-efficient ansätze (Fig. 5, see supplementary materials, section S2.3 for computational details). These extremely favourable CNOT counts are achieved by removing redundancies from the quantum circuit using discrete optimisation and through the CNOT efficiency of the the paired two-body operators (supplementary materials, section S2.2). For the first time, these data demonstrate that efficient quantum circuits can be achieved using symmetry-preserving fermionic operators without relying on a hardware-efficient ansatz design, creating a new paradigm for quantum-compatible electronic structure methods.

Balancing weak and strong electron correlation

Balanced descriptions of weak and strong electron correlation are essential for quantum chemistry but have proved remarkably difficult to achieve in practice. Many-body perturbation techniques can be quantitatively accurate for weak electron correlation, but fail to qualitatively describe strongly correlated systems where the molecular orbital picture breaks down. The accuracy of the DISCO-VQE results across the weakly correlated equilibrium geometry and strongly correlated dissociation limits of $\text{H}_6$, $\text{H}_2\text{O}$, and $\text{N}_2$ demonstrate that the s-UPS framework can quantitatively predict both types of electron correlation using a consistent wave function ansätze.

The accuracy of the s-UPS ansatz for strong correlation highlights the capability to progress beyond the many-body framework of coupled cluster theory, and should be considered as a parametrised unitary rotation in the Hilbert space.

The strongly correlated half-filled Hubbard lattice represents a challenging condensed matter physics problem for traditional many-body approximations.\textsuperscript{37} Here, the correlation strength is determined by the balance of the on-site electron repulsion $U$ and the one-electron hopping term between adjacent sites $t$. DISCO-VQE calculations with a truncated s-UPS wave function (supplementary materials, section S8) provide excellent accuracy in both the weak correlation ($U \ll t$) and strong correlation ($U \gg t$) limits (Fig. 6A). These data highlight the accuracy of the s-UPS ansatz for describing both weak and strong correlation using a consistent number of operators and variational parameters. In comparison, ADAPT-VQE provides a very accurate representation of the weakly correlated regime, but deteriorates as the correlation strength increases. Furthermore, the converged ADAPT-VQE wave function includes significantly more operators than DISCO-VQE for intermediate correlation ($U/t \approx 10$) but is less accurate (Fig. 6C). Consequent
quently, global optimisation using DISCO-VQE is essential for obtaining quantitatively accurate s-UPS wave functions for both weak and strong correlation with a consistent quantum resource cost.

Correctly predicting the physical properties of strongly correlated systems is just as important as the electronic energy for understanding quantum phase behaviour. The strongly correlated limit of the half-filled Hubbard model is characterised by an antiferromagnetic phase where the electrons are unpaired and localised on individual sites. The double occupancy \( \langle n_p n_p \rangle \) measures the probability of simultaneously finding two electrons on the same site, which tends to zero for \( U/t \to \infty \) and 0.25 for \( U/t \to 0 \). Truncated s-UPS representations identified using DISCO-VQE accurately predict this change across different \( U/t \) values (Fig. 6B) and tend to the correct limits. Therefore, DISCO-VQE accurately predicts both the wave function and the energy, providing a physically complete description of strong electron correlation.

**Discussion and outlook**

We have shown that an arbitrary electronic state can be parametrised with symmetry-preserving unitary product states built from spin-adapted one-body and paired two-body fermionic operators. This ansatz conserves fermionic antisymmetry, particle number symmetry, and the quantum numbers corresponding to \( \hat{S}_z^2 \) and \( \hat{S}_y \) without any constrained optimisation or spin projection, and can parametrise any electronic state. The flexibility and accuracy of these wave functions comes with the additional challenge of identifying the optimal sequence of unitary operators. DISCO-VQE is the first algorithm that can address this challenge by performing a coupled discrete and continuous global optimisation of the wave function. Numerical simulations demonstrate that our approach delivers chemically accurate energies for weakly and strongly correlated molecules using significantly fewer operators and CNOT gates than previous state-of-the-art techniques.

While the discrete global optimisation in DISCO-VQE increases the computational cost relative to a fixed ansatz approach, there are several routes to mitigate this cost in practice. Firstly, the discrete search over operator mutations or pair swaps scales only linearly with respect to the size of the operator pool or the number of operators \( M \) in the s-UPS ansatz. Performing individual optimisations for each discrete step in parallel using multiple quantum computers would reduce the additional scaling to \( O(M) \). Secondly, since the continuous basin-hopping search represents a significant proportion of the computational cost, DISCO-VQE will benefit from the development of improved quantum numerical optimisation techniques.\(^{38,39}\) Finally, discrete combinatorial optimisation is a rich field and various algorithms have also been developed for optimising hardware-efficient ansätze.\(^{40–44}\) Identifying important heuristics to scan the discrete neighbourhood more selectively will further improve the efficiency of DISCO-VQE.\(^{26,27}\)

Symmetry-preserving unitary product states offer key advantages for practical quantum algorithms: they can parametrise an arbitrarily accurate wave function; they preserve the physical symmetries of the Hamiltonian at every truncation; and they have a low CNOT count suitable for noisy quantum hardware. Conserving physical symmetries is essential for creating suitable initial states for fault-tolerant quantum phase estimation, where efficiency depends on the overlap of the initial and exact states. Furthermore, formulating the generalised fermionic operators as Lie algebra generators for unitary rotations moves beyond the many-body unitary coupled cluster framework. This new perspective fully exploits the natural capabilities of qubit rotations in quantum computation, bridging the gap between fermionic operators and hardware-efficient ansätze. Ultimately, these advances set a new standard for quantitative, physically accurate, and gate efficient quantum circuits for simulating strongly-correlated chemical problems using near-term quantum computers.

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Author contributions:
H.G.A.B., D.P.T., and D.J.W. conceived the project. H.G.A.B., D.M.D., and D.J.W. designed the discrete optimisation algorithm. D.M.D., H.G.A.B., and D.P.T. designed and analysed the s-UPS ansatz. H.G.A.B. wrote the DISCO-VQE code and carried out the simulations. H.G.A.B. and D.M.D. performed the data analysis. All authors contributed to writing the manuscript. H.G.A.B. initiated and managed the collaboration.

Competing interests:
The authors declare that they have no competing interests.

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The data required to reproduce figures are hosted at https://dx.doi.org/10.5281/zenodo.6784056. All other data required to evaluate the conclusions are present in the paper or the supplemental materials.

Supplementary Materials
Supplementary Text
Figs. S1 to S3
Table S1
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