

Supplementary Information - Demonstration of subspace-search variational quantum eigensolver using photonic orbital angular momentum qudits

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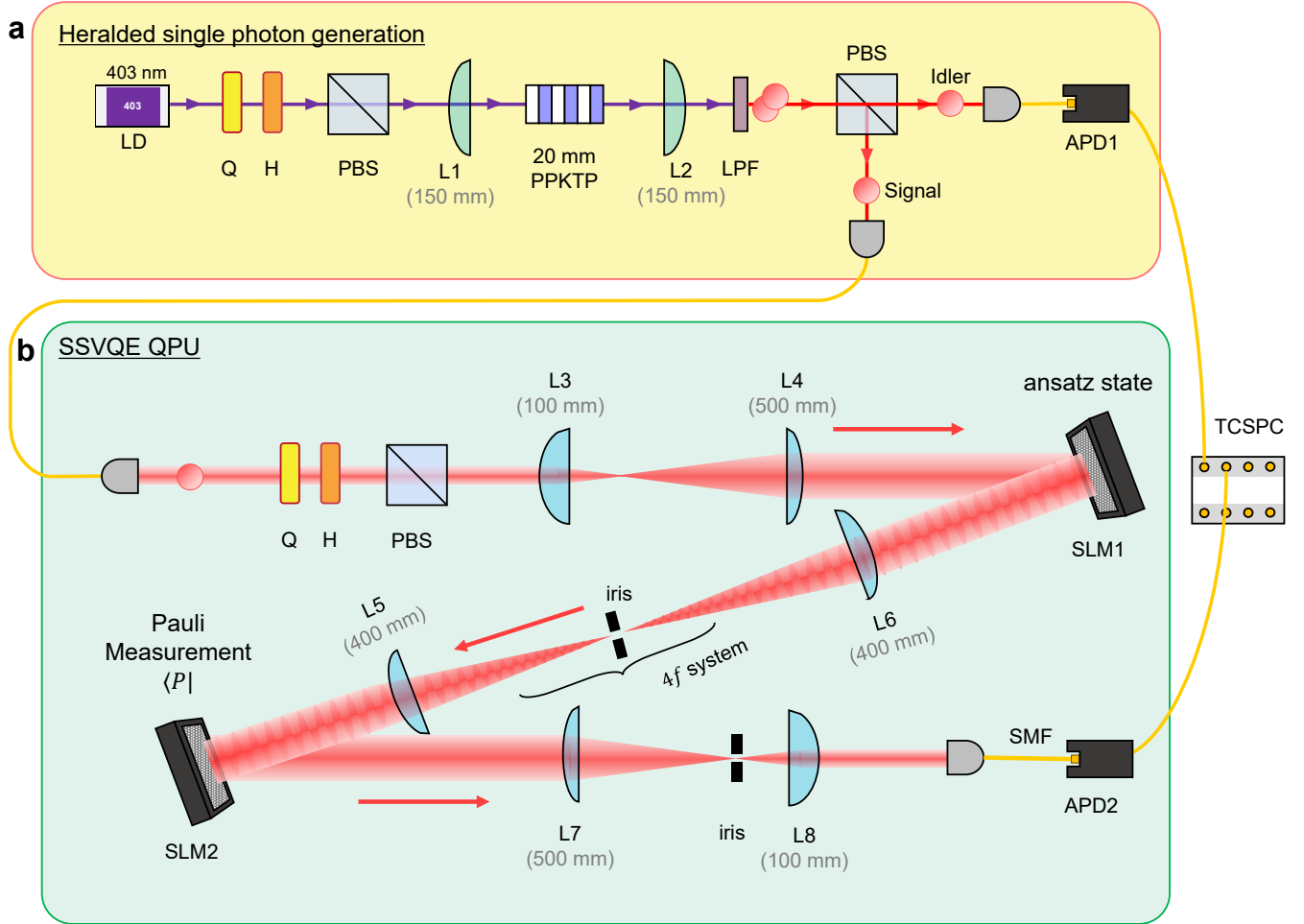
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SUPPLEMENTARY NOTE I. EXPERIMENTAL SETUP FOR SSVQE



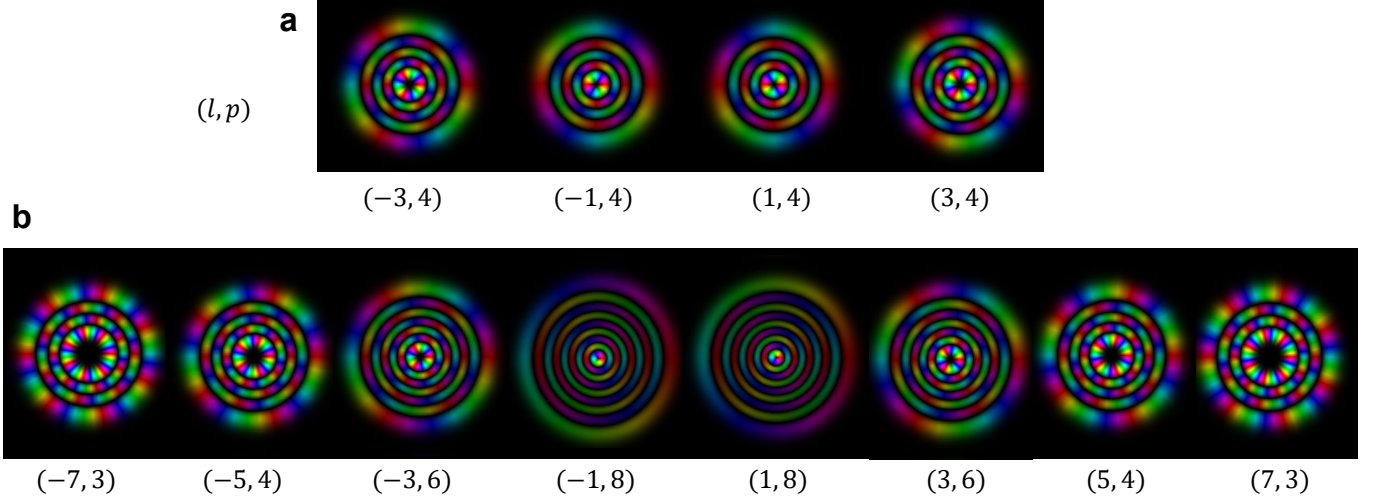
Supplementary Figure 1. **Experimental setup for SSVQE.** **a**, SPDC source for generating heralded single photons. **b**, Photonic SSVQE QPU based on OAM qudit encoding (LD, laser diode; HWP, half-wave plate; QWP, quarter-wave plate; PBS, polarizing beam splitter; LPF, long-pass filter; SLM, spatial light modulator; APD, avalanche photodiode; TCSPC, time-correlated single-photon counter).

A detailed experimental setup for our subspace-search variational quantum eigensolver (SSVQE) using the orbital angular momentum (OAM) of single photons is shown in Supplementary Fig. 1. A 20-mm-long periodically poled potassium titanyl phosphate (PPKTP) crystal is pumped by a 403 nm continuous-wave (CW) diode laser (OBIS 405 LX FP) with a free-space power of 40 mW. Photon pairs are generated via type-II spontaneous parametric down-conversion (SPDC) and separated by a polarizing beam splitter. The idler photon is detected by an avalanche photodiode (APD1; Excelitas SPCM-AQRH-14-FC) to herald the signal photon, which is sent to the SSVQE quantum processing unit (QPU).

The signal photon is prepared in horizontal polarization using a set of wave plates and expanded by a factor of five using lenses to approximate a plane-wave illumination. The beam is aligned to the center of the active area of a spatial light modulator (SLM; Holoeye Pluto-2.1 NIR 118), which prepares the desired ansatz qudit states. Holograms corresponding to the N ansatz states are displayed sequentially on SLM1, encoding the states in Laguerre–Gaussian (LG) modes. After SLM1, the prepared state propagates through a lens–iris–lens system (a $4f$ system) for spatial filtering [1]. The iris, placed at the Fourier plane, blocks all diffraction orders except the first-order beam.

For projective measurements, the beam is analyzed on SLM2 and converted into the fundamental Gaussian mode corresponding to the chosen measurement setting via a two-dimensional Fourier transform. The output is coupled into a single-mode fiber, which acts as a spatial filter for the Gaussian mode, and detected by a second avalanche photodiode (APD2). Two-photon coincidence events between APD1 and APD2 are recorded using a time-correlated single-photon counter (TCSPC; Swabian Time Tagger 20) with a 2 ns coincidence window.

SUPPLEMENTARY NOTE II. CROSSTALK MATRIX OF LAGUERRE-GAUSSIAN MODES



Supplementary Figure 2. **LG modes used as basis states.** **a**, Modes used for the four-dimensional implementation. **b**, Modes used for the eight-dimensional implementation. (l, p) denote the azimuthal and radial indices of the LG modes.

To accurately prepare N orthogonal ansatz states, we experimentally verified the orthogonality of the employed basis states using a crosstalk matrix measurement. For the four-dimensional implementation ($d = 4$), we use a complete set of mutually unbiased bases (MUBs) summarized in Supplementary Table I [2]. In dimension d , a complete set of MUBs contains $(d + 1)$ bases, each comprising d orthonormal states, giving a total of $d(d + 1)$ states. We label the five bases for $d = 4$ as M0–M4. States within the same basis are orthogonal, yielding an ideal measurement probability of 1 (matched state) or 0 (orthogonal state), whereas measurements between different MUBs ideally give a uniform overlap of $1/d$.

To quantify the state-preparation and measurement performance, we measure the crosstalk matrix [3], a $d(d + 1) \times d(d + 1)$ matrix whose elements are

$$N_{ij} = |\langle \psi_j | \psi_i \rangle|^2, \quad (\text{S1})$$

where $|\psi_i\rangle$ denotes the prepared i th basis state and $\langle \psi_j |$ denotes the measured j th projection state. Here, N_{ij} is obtained from the two-photon coincidence counts for the corresponding preparation–measurement setting.

We further characterize each basis by the visibility V [3],

$$V = \frac{\sum_{i=1}^d N_{ii}}{\sum_{i,j=1}^d N_{ij}}. \quad (\text{S2})$$

The numerator $\sum_i N_{ii}$ corresponds to the diagonal (correct) projections, while the denominator $\sum_{i,j} N_{ij}$ sums over all projections within that basis.

To ensure robust orthogonality while equalizing coupling efficiencies, we construct the basis states using both the azimuthal index l and the radial index p of Laguerre–Gaussian (LG) modes. The phase maps for the employed basis states are shown in Supplementary Fig. 2. For $d = 4$, we obtain visibilities $V \in \{99.5, 99.5, 99.8, 99.3, 99.1\}\%$ for the five MUBs (M0–M4), as shown in Supplementary Fig. 3.

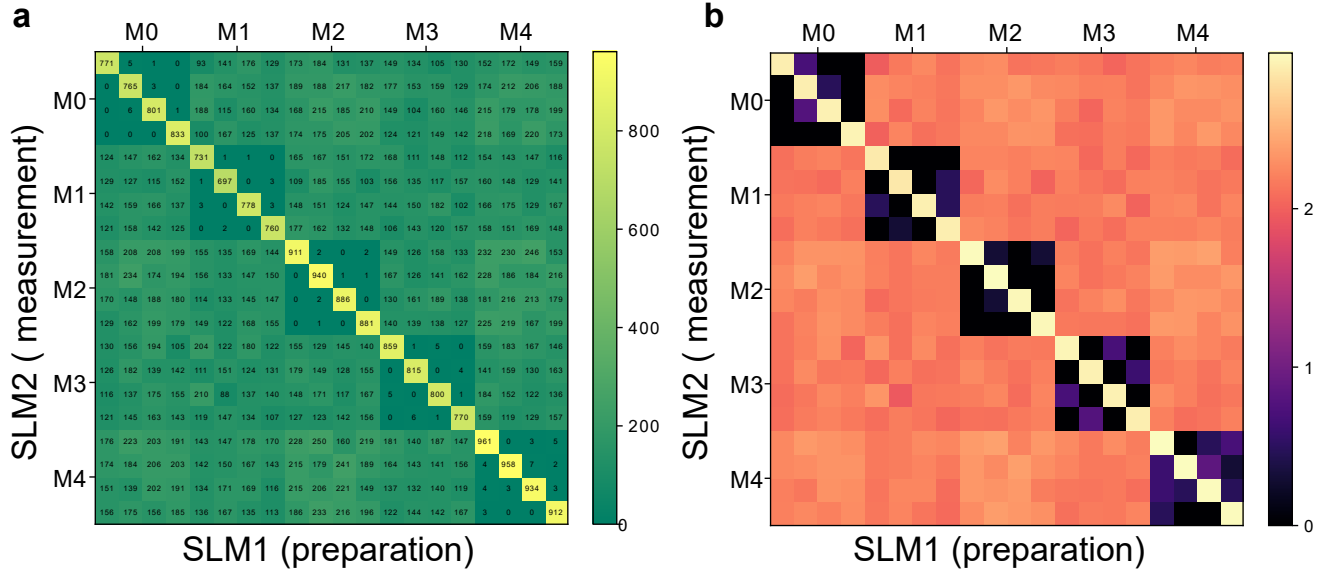
For the eight-dimensional implementation ($d = 8$), we employ nine equal-superposition bases that are mutually orthogonal within each basis, but do not form a complete set of MUBs (Supplementary Table II). The corresponding visibilities for the nine bases are shown in Supplementary Figs. 4 and 5.

Overall, we observe only small crosstalk between the intended orthogonal LG modes, with negligible counts between well-separated mode pairs. The residual nonzero crosstalk can contribute to experimental errors in state preparation and projective measurements, and we expect that improved alignment and mode matching will further reduce these errors, thereby improving the accuracy of the estimated energies.

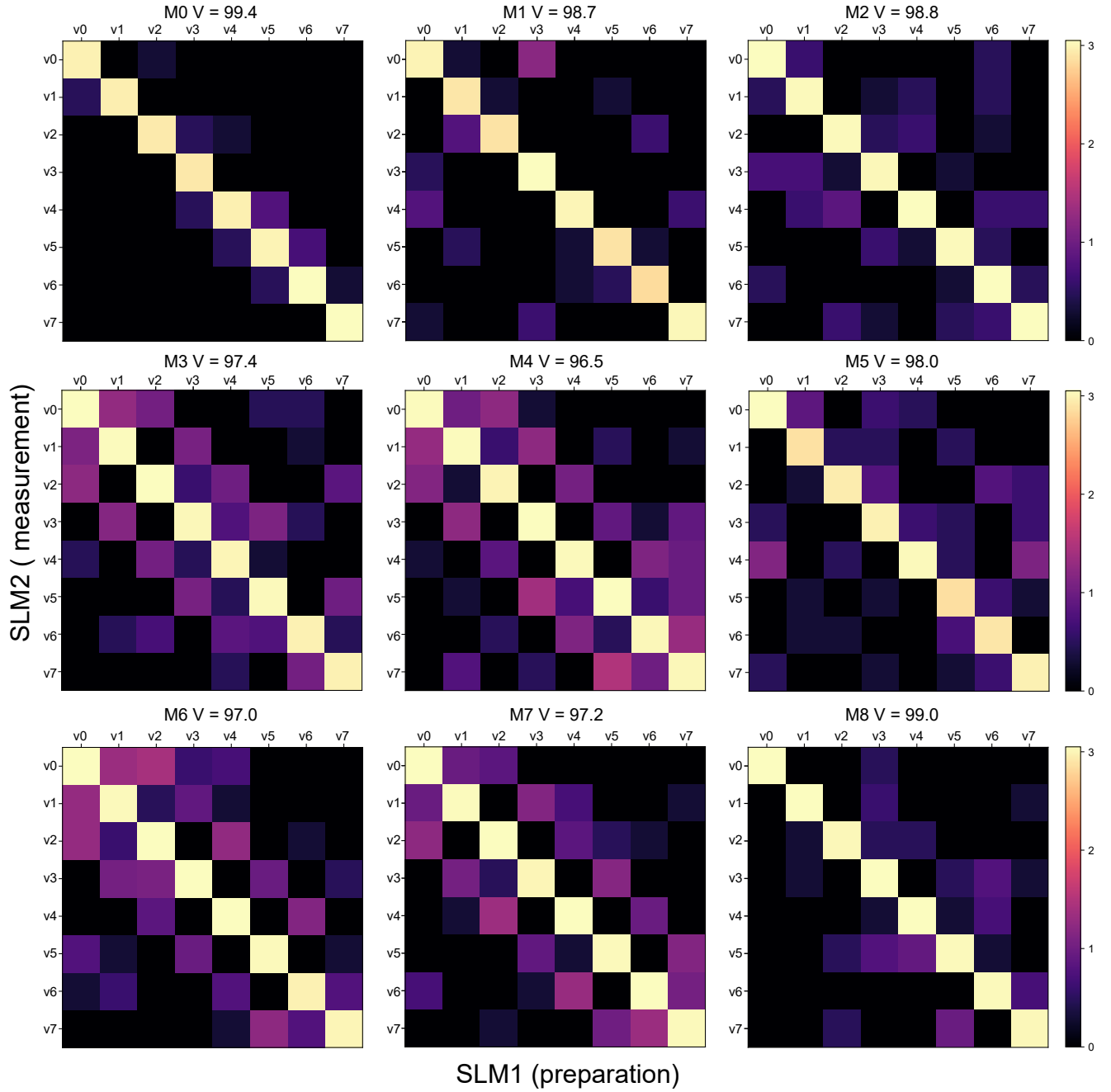
Group MUBs

M0	$\{(1, 0, 0, 0), (0, 1, 0, 0), (0, 0, 1, 0), (0, 0, 0, 1)\}$
M1	$\{\frac{1}{2}(1, 1, 1, 1), \frac{1}{2}(1, 1, -1, -1), \frac{1}{2}(1, -1, -1, 1), \frac{1}{2}(1, -1, 1, -1)\}$
M2	$\{\frac{1}{2}(1, -1, -i, -i), \frac{1}{2}(1, -1, i, i), \frac{1}{2}(1, 1, i, -i), \frac{1}{2}(1, 1, -i, i)\}$
M3	$\{\frac{1}{2}(1, -i, -i, -1), \frac{1}{2}(1, -i, i, 1), \frac{1}{2}(1, i, i, -1), \frac{1}{2}(1, i, -i, 1)\}$
M4	$\{\frac{1}{2}(1, -i, -1, -i), \frac{1}{2}(1, -i, 1, i), \frac{1}{2}(1, i, -1, i), \frac{1}{2}(1, i, 1, -i)\}$

Supplementary Table I. **Mutually unbiased bases (MUBs) for projective measurements in a four-dimensional Hilbert space.**



Supplementary Figure 3. **Experimental preparation-measurement crosstalk matrix for the four-dimensional implementation.** **a**, Crosstalk matrix displayed on a linear scale. **b**, Crosstalk matrix displayed on a logarithmic scale.

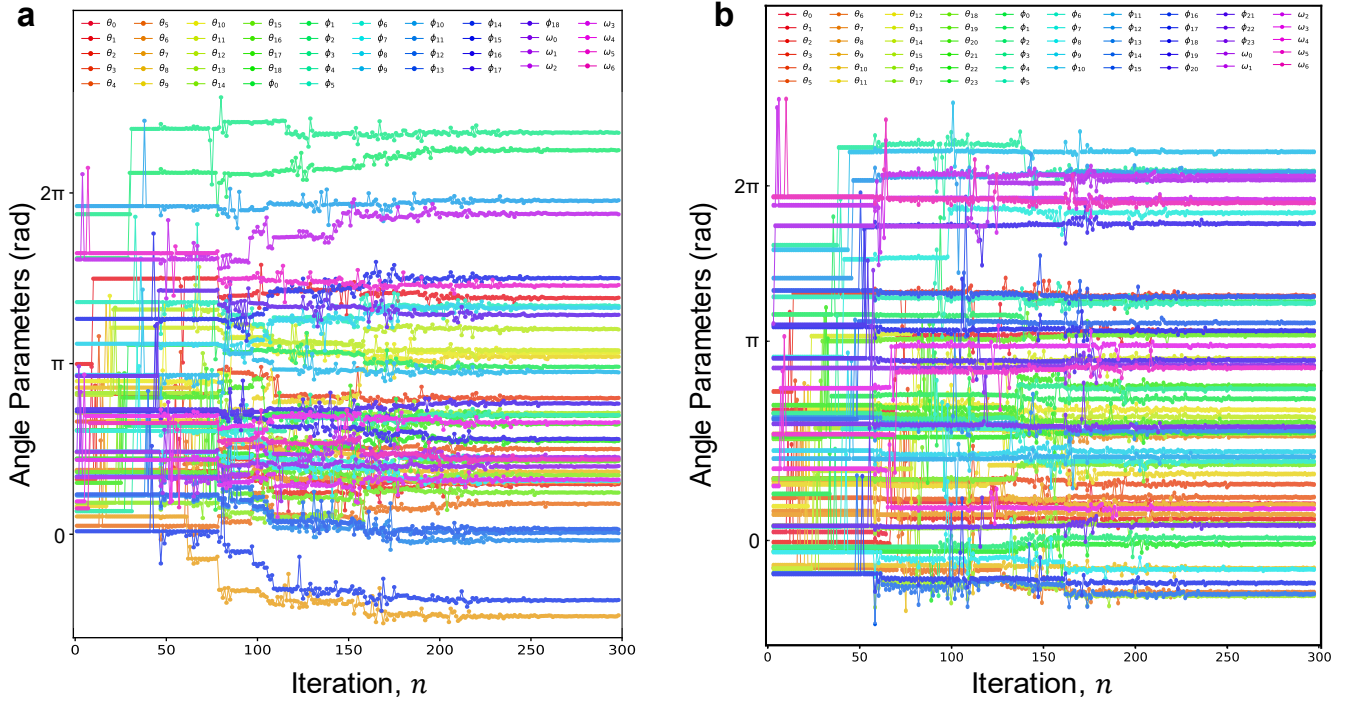


Supplementary Figure 5. Experimental preparation-measurement crosstalk matrix for the eight-dimensional implementation (log scale).

Group Basis vectors $1/\sqrt{8}(\text{factor})$	
M0	$\{(1, 0, 0, 0, 0, 0, 0, 0), (0, 1, 0, 0, 0, 0, 0, 0), (0, 0, 1, 0, 0, 0, 0, 0), (0, 0, 0, 1, 0, 0, 0, 0),$ $(0, 0, 0, 0, 1, 0, 0, 0), (0, 0, 0, 0, 0, 1, 0, 0), (0, 0, 0, 0, 0, 0, 1, 0), (0, 0, 0, 0, 0, 0, 0, 1)\}$
M1	$\{(1, \xi^4, 1, \xi^4, 1, \xi^4, 1, \xi^4), (1, \xi^3, \xi^6, \xi, \xi^4, \xi^7, \xi^2, \xi^5), (1, \xi^2, \xi^4, \xi^6, 1, \xi^2, \xi^4, \xi^6), (1, \xi, \xi^2, \xi^3, \xi^4, \xi^5, \xi^6, \xi^7),$ $(1, 1, 1, 1, 1, 1, 1, 1), (1, \xi^7, \xi^6, \xi^5, \xi^4, \xi^3, \xi^2, \xi), (1, \xi^6, \xi^4, \xi^2, 1, \xi^6, \xi^4, \xi^2), (1, \xi^5, \xi^2, \xi^7, \xi^4, \xi, \xi^6, \xi^3)\}$
M2	$\{(1, \xi^4, 1, \xi^6, \xi^4, \xi^4, \xi^4, \xi^5), (1, \xi^3, \xi^6, \xi^3, 1, \xi^7, \xi^6, \xi^7), (1, \xi^2, \xi^4, 1, \xi^4, \xi^2, 1, 1), (1, 1, \xi^2, \xi^5, 1, \xi^5, \xi^2, 1),$ $(1, 1, 1, \xi^2, \xi^4, 1, \xi^4, \xi^2), (1, \xi^6, \xi^6, \xi^6, 1, \xi^2, \xi^6, \xi^3), (1, \xi^5, \xi^4, \xi^4, \xi^4, \xi^6, 1, \xi^3), (1, \xi^4, \xi^2, 1, 1, 1, \xi^2, \xi^5)\}$
M3	$\{(1, \xi^3, 1, \xi^7, 1, \xi^3, 1, \xi^7), (1, \xi^2, \xi^6, \xi^4, \xi^4, \xi^6, \xi^2, 1), (1, \xi, \xi^4, \xi, 1, \xi, \xi^4, \xi), (1, 1, \xi^2, \xi^6, \xi^4, \xi^4, \xi^6, \xi^2),$ $(1, \xi^7, 1, \xi^3, 1, \xi^7, 1, \xi^3), (1, \xi^6, \xi^6, 1, \xi^4, \xi^2, \xi^2, \xi^4), (1, \xi^5, \xi^4, \xi^5, 1, \xi^5, \xi^4, \xi^5), (1, \xi^4, \xi^2, \xi^2, \xi^4, 1, \xi^6, \xi^6)\}$
M4	$\{(1, \xi^4, \xi^2, \xi^4, 1, 1, \xi^2, 1), (1, \xi^2, 1, 1, \xi^4, \xi^3, \xi^4, 1), (1, \xi, \xi^6, \xi^5, 1, \xi^5, \xi^6, \xi), (1, \xi, \xi^4, \xi^3, \xi^4, \xi, 1, \xi^3),$ $(1, 1, \xi^2, \xi^7, 1, \xi^4, \xi^2, \xi^4), (1, \xi^7, 1, \xi^5, \xi^4, \xi^7, \xi^4, \xi^4), (1, \xi^6, \xi^6, \xi^2, 1, \xi^2, \xi^6, \xi^6), (1, \xi^4, \xi^4, \xi^6, \xi^4, \xi^4, 1, \xi^7)\}$
M5	$\{(1, \xi^3, \xi^2, \xi^5, \xi^4, \xi^7, \xi^6, \xi), (1, \xi^2, 1, \xi^2, 1, \xi^2, 1, \xi^2), (1, \xi, \xi^6, \xi^7, \xi^4, \xi^5, \xi^2, \xi^3), (1, 1, \xi^4, \xi^4, 1, 1, \xi^4, \xi^4),$ $(1, \xi^7, \xi^2, \xi, \xi^4, \xi^3, \xi^6, \xi^5), (1, \xi^6, 1, \xi^6, 1, \xi^6, 1, \xi^6), (1, \xi^5, \xi^6, \xi^3, \xi^4, \xi, \xi^2, \xi^7), (1, \xi^4, \xi^4, 1, \xi^4, \xi^4, 1)\}$
M6	$\{(1, \xi^4, \xi^4, \xi^2, \xi^4, \xi^4, 1, \xi^2), (1, \xi^3, \xi^2, \xi^7, 1, \xi^7, \xi^2, \xi^3), (1, \xi^2, 1, \xi^4, \xi^4, \xi^2, \xi^4, \xi^4), (1, \xi, \xi^6, \xi, 1, \xi^5, \xi^6, \xi^5),$ $(1, 1, \xi^4, \xi^6, \xi^4, 1, 1, \xi^5), (1, \xi^7, \xi^2, \xi^3, 1, \xi^3, \xi^2, \xi^6), (1, \xi^6, 1, \xi^7, \xi^4, \xi^6, \xi^4, \xi^7), (1, \xi^4, \xi^6, \xi^4, 1, 1, \xi^6, 1)\}$
M7	$\{(1, \xi^3, \xi^4, \xi^3, 1, \xi^3, \xi^4, \xi^3), (1, \xi^2, \xi^2, 1, \xi^4, \xi^6, \xi^6, \xi^4), (1, \xi, 1, \xi^5, 1, \xi, 1, \xi^5), (1, 1, \xi^6, \xi^2, \xi^4, \xi^4, \xi^2, \xi^6),$ $(1, \xi^7, \xi^4, \xi^7, 1, \xi^7, \xi^4, \xi^7), (1, \xi^6, \xi^2, \xi^4, \xi^4, \xi^2, \xi^6, 1), (1, \xi^5, 1, \xi, 1, \xi^5, 1, \xi), (1, \xi^4, \xi^6, \xi^6, \xi^4, 1, \xi^2, \xi^2)\}$
M8	$\{(1, \xi^4, \xi^6, 1, 1, 1, \xi^6, \xi^4), (1, \xi^3, \xi^4, \xi^5, \xi^4, \xi^3, 1, \xi^5), (1, \xi^2, \xi^2, \xi^2, 1, \xi^6, \xi^2, \xi^6), (1, \xi, 1, \xi^7, \xi^4, \xi, \xi^4, \xi^7),$ $(1, 1, \xi^6, \xi^4, 1, \xi^4, \xi^6, 1), (1, \xi^7, \xi^4, \xi, \xi^4, \xi^7, 1, \xi), (1, \xi^5, \xi^2, \xi^6, 1, \xi, \xi^2, \xi), (1, \xi^5, 1, \xi^3, \xi^4, \xi^5, \xi^4, \xi^3)\}$

Supplementary Table II. **Bases used for $d = 8$.** $M0$ denotes the computational basis and $M1 \dots M8$ denote the eigenbases of XZ^k ($k = 0, \dots, 7$), where $\xi = e^{2\pi i/8}$.

SUPPLEMENTARY NOTE III. THE DESIGN OF ANSATZ STATES AND ANGULAR PARAMETERS



Supplementary Figure 6. **Evolution of variational angle parameters for the Heisenberg chain ansatz.** **a**, Two-level implementation. **b**, Three-level implementation.

This section provides explicit expressions for the ansatz states used in the four-dimensional implementation, constructed according to the unitary-synthesis procedure described in Methods. In addition, for the eight-dimensional Heisenberg-chain model, a general $SU(8)$ unitary would require 63 parameters; however, when targeting a reduced subspace, the number of variational parameters can be lowered. In particular, the parameter count is reduced to 45 and 55 for estimating two and three energy levels, respectively, by excluding degrees of freedom associated with unused subspaces. The experimentally observed evolution of the angular parameters is shown in Supplementary Fig. 6.

$$|\psi_0\rangle_{4d} = \begin{bmatrix} e^{i\phi_2} e^{i\phi_5} \cos(\theta_2) \cos(\theta_5) + e^{i\phi_3} e^{i\phi_5} \sin(\theta_5) \cos(\theta_2) \cos(\theta_3) \\ (e^{i\omega_1} e^{i\phi_0} e^{i\phi_3} \cos(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_1} e^{i\phi_1} e^{i\phi_3} \sin(\theta_3) \cos(\theta_0) \cos(\theta_1)) e^{i\phi_5} \sin(\theta_5) \\ + e^{i\omega_1} e^{i\phi_0} e^{i\phi_2} e^{i\phi_5} \sin(\theta_2) \cos(\theta_0) \cos(\theta_5) \\ (e^{i\omega_2} e^{i\phi_0} e^{i\phi_3} \sin(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_2} e^{i\phi_1} e^{i\phi_3} \sin(\theta_3) \cos(\theta_0) \cos(\theta_1)) e^{i\phi_5} \sin(\theta_5) \\ + e^{i\omega_2} e^{i\phi_0} e^{i\phi_2} e^{i\phi_5} \sin(\theta_0) \sin(\theta_2) \cos(\theta_5) \\ e^{i\omega_3} e^{i\phi_1} e^{i\phi_3} e^{i\phi_5} \sin(\theta_1) \sin(\theta_3) \sin(\theta_5) \end{bmatrix}$$

$$|\psi_1\rangle_{4d} = \begin{bmatrix} e^{i\phi_2} \cos(\theta_2) \cos(\theta_5) + e^{i\phi_3} \cos(\theta_2) \cos(\theta_3) \cos(\theta_5) \\ (e^{i\omega_1} e^{i\phi_0} e^{i\phi_3} \cos(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_1} e^{i\phi_1} e^{i\phi_3} \sin(\theta_3) \cos(\theta_0) \cos(\theta_1)) \cos(\theta_5) \\ + e^{i\omega_1} e^{i\phi_0} e^{i\phi_2} \sin(\theta_2) \cos(\theta_0) \cos(\theta_5) \\ (e^{i\omega_2} e^{i\phi_0} e^{i\phi_3} \sin(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_2} e^{i\phi_1} e^{i\phi_3} \sin(\theta_3) \cos(\theta_0) \cos(\theta_1)) \cos(\theta_5) \\ + e^{i\omega_2} e^{i\phi_0} e^{i\phi_2} \sin(\theta_0) \sin(\theta_2) \cos(\theta_5) \\ e^{i\omega_3} e^{i\phi_1} e^{i\phi_3} \sin(\theta_1) \sin(\theta_3) \cos(\theta_5) \end{bmatrix}$$

$$|\psi_2\rangle_{4d} = \begin{bmatrix} e^{i\phi_4} \cos(\theta_2) \cos(\theta_3) \cos(\theta_4) \\ (e^{i\omega_1} e^{i\phi_0} \cos(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_1} e^{i\phi_1} \cos(\theta_0) \cos(\theta_1) \cos(\theta_3)) e^{i\phi_4} \cos(\theta_4) + e^{i\omega_1} e^{i\phi_4} \sin(\theta_4) \cos(\theta_0) \cos(\theta_1) \\ (e^{i\omega_2} e^{i\phi_0} \sin(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_2} e^{i\phi_1} \cos(\theta_0) \cos(\theta_1) \cos(\theta_3)) e^{i\phi_4} \cos(\theta_4) + e^{i\omega_2} e^{i\phi_4} \sin(\theta_4) \cos(\theta_0) \cos(\theta_1) \\ e^{i\omega_3} e^{i\phi_1} e^{i\phi_4} \sin(\theta_1) \cos(\theta_3) \cos(\theta_4) + e^{i\omega_3} e^{i\phi_4} \sin(\theta_4) \cos(\theta_1) \end{bmatrix}$$

$$|\psi_3\rangle_{4d} = \begin{bmatrix} \cos(\theta_2) \cos(\theta_3) \cos(\theta_4) \\ (e^{i\omega_1} e^{i\phi_0} \cos(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_1} e^{i\phi_1} \cos(\theta_0) \cos(\theta_1) \cos(\theta_3)) \cos(\theta_4) + e^{i\omega_1} \cos(\theta_0) \cos(\theta_1) \cos(\theta_4) \\ (e^{i\omega_2} e^{i\phi_0} \sin(\theta_0) \cos(\theta_2) \cos(\theta_3) + e^{i\omega_2} e^{i\phi_1} \cos(\theta_0) \cos(\theta_1) \cos(\theta_3)) \cos(\theta_4) + e^{i\omega_2} \cos(\theta_0) \cos(\theta_1) \cos(\theta_4) \\ e^{i\omega_3} e^{i\phi_1} \sin(\theta_1) \cos(\theta_3) \cos(\theta_4) + e^{i\omega_3} \cos(\theta_1) \cos(\theta_4) \end{bmatrix}$$

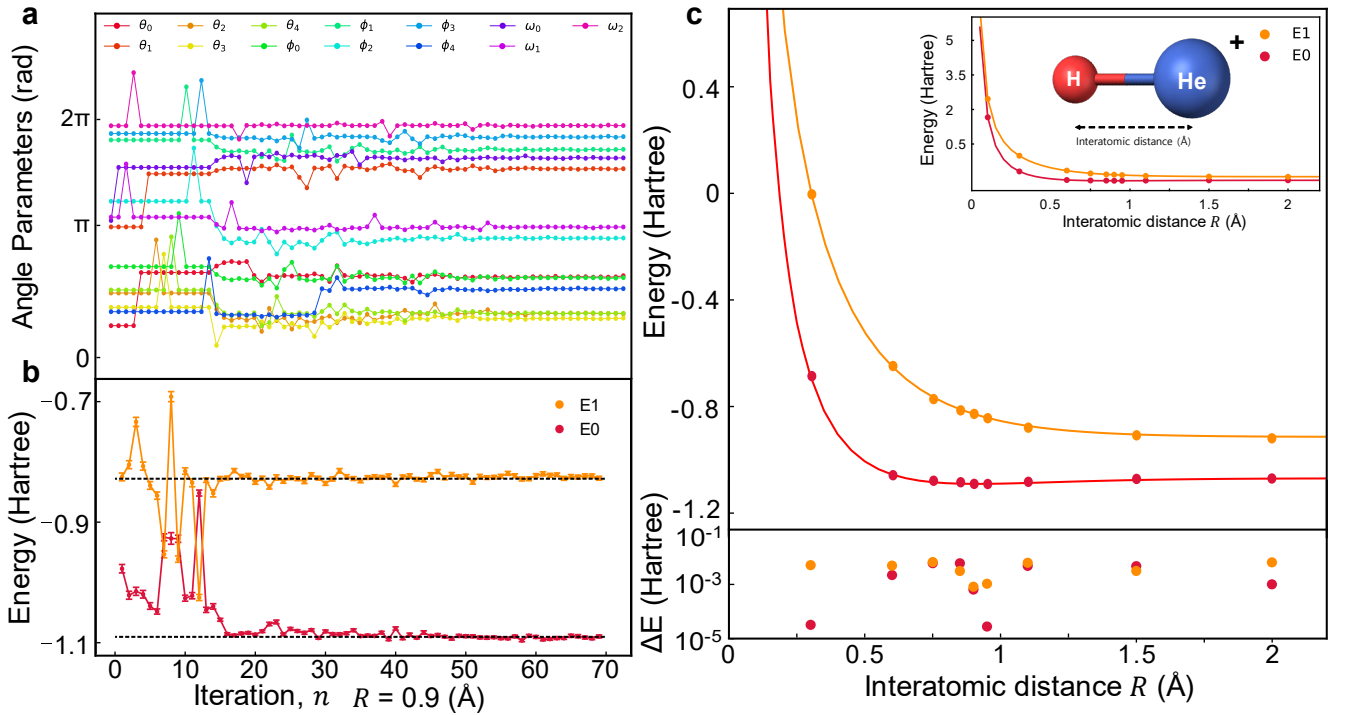
SUPPLEMENTARY NOTE IV. THE WEIGHT COEFFICIENTS OF ESTIMATED ENERGIES

Although SSVQE can, in principle, recover excited state energies independent of the choice of weight coefficients [4], in practice the optimization can become sensitive to $\{w_j\}$ due to finite-shot noise and small energy gaps between low-lying eigenstates. To improve convergence and reliably resolve closely spaced excited states (in particular, the first and second excited states), we therefore use different sets of weight coefficients depending on the number of targeted levels. The weight coefficients used for each case are listed in Supplementary Table III.

Target levels	Weight coefficients $\{w_j\}$
2-level	$\{0.6, 0.4\}$
3-level	$\{0.5, 0.38, 0.12\}$
4-level	$\{0.45, 0.38, 0.12, 0.05\}$

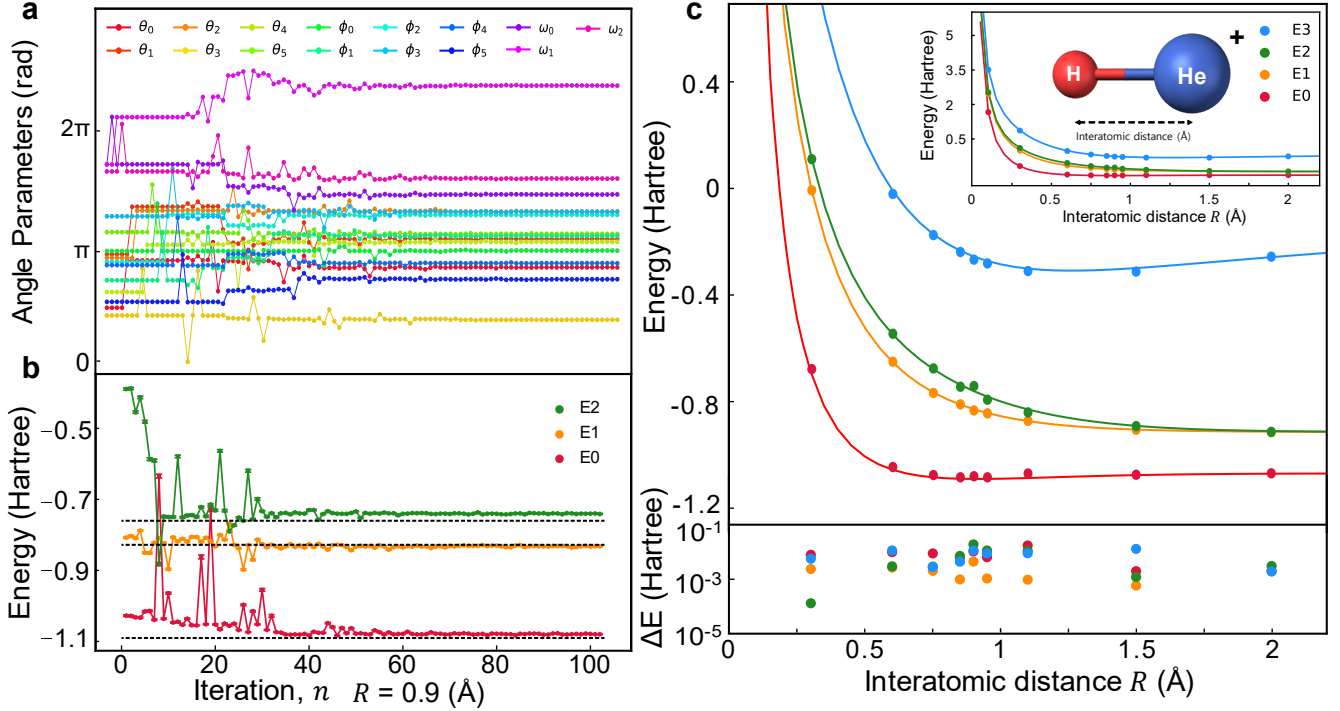
Supplementary Table III. **Weight coefficients used in SSVQE.** Weight coefficients $\{w_j\}$ used in the SSVQE cost function $L = \sum_j w_j \langle E_j \rangle$ for estimating 2-, 3-, and 4-level energy spectra. The weights were chosen to improve optimization stability under finite sampling noise, particularly in the presence of small energy gaps.

SUPPLEMENTARY NOTE V. EXPERIMENTAL RESULTS FOR HeH^+



Supplementary Figure 7. **Experimental SSVQE results for two-level HeH^+ .** **a**, Iterative evolution of the 13 variational angle parameters used to prepare the two OAM-qudit ansatz states during SSVQE optimization at $R = 0.90$ Å (equilibrium bond length). **b**, Iterative optimization of the estimated ground and first excited state energies $\langle E_0 \rangle$ and $\langle E_1 \rangle$ at $R = 0.90$ Å. Solid colored curves (red and orange) denote experimental estimates as a function of iteration number n , and black dashed lines indicate theoretical values. Error bars represent one standard deviation obtained from 1,000 Monte Carlo resamplings. **c**, Estimated energies as a function of interatomic distance R (top). The energy deviations ΔE (bottom) are defined as the difference between the experimental estimates and the theoretical values.

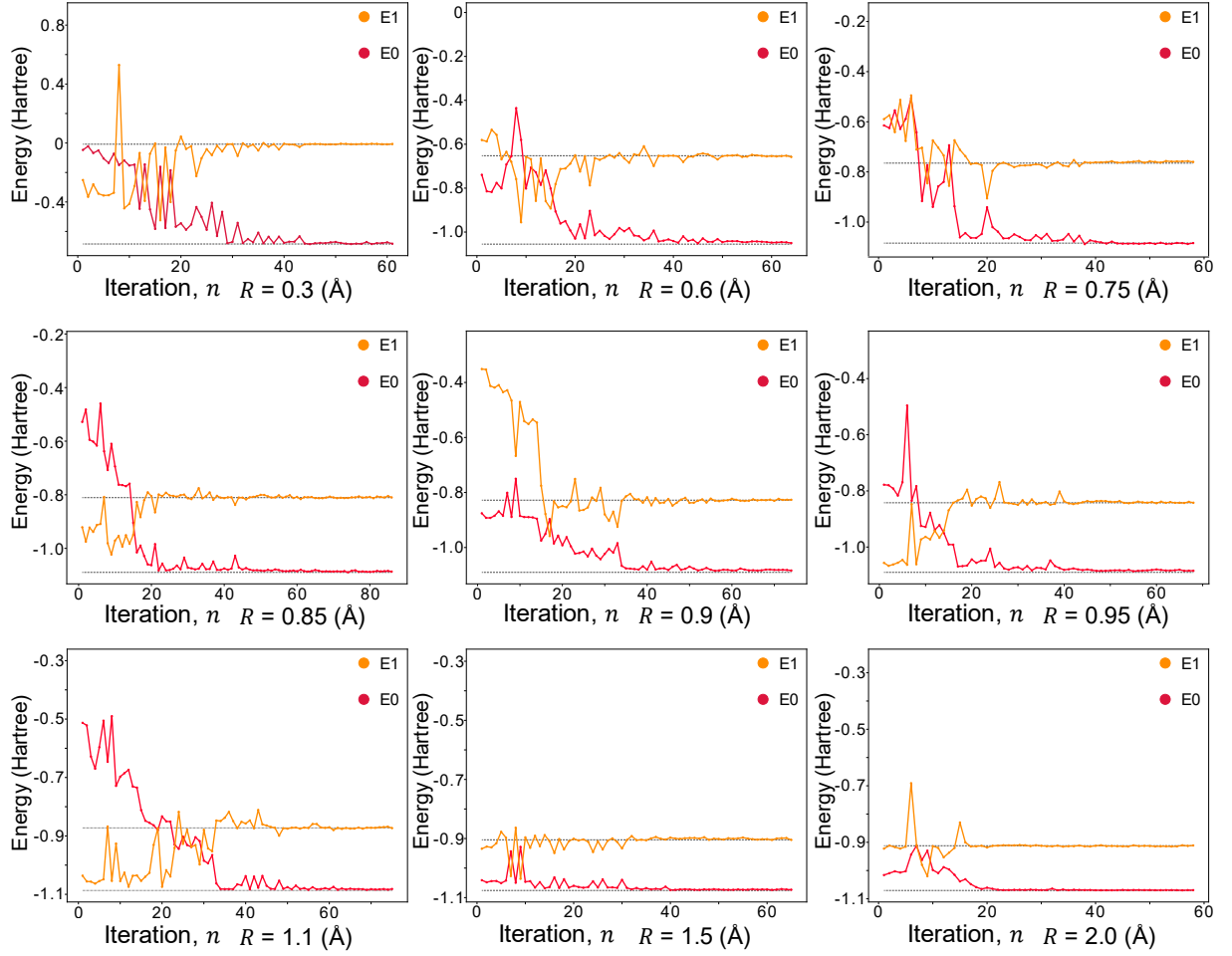
Our platform allows SSVQE to be performed in a subspace-selective manner by choosing the number of energy levels to estimate. In addition to the four-level results presented in the main text, we report SSVQE demonstrations targeting the lowest two and three energy levels of the HeH^+ Hamiltonian.



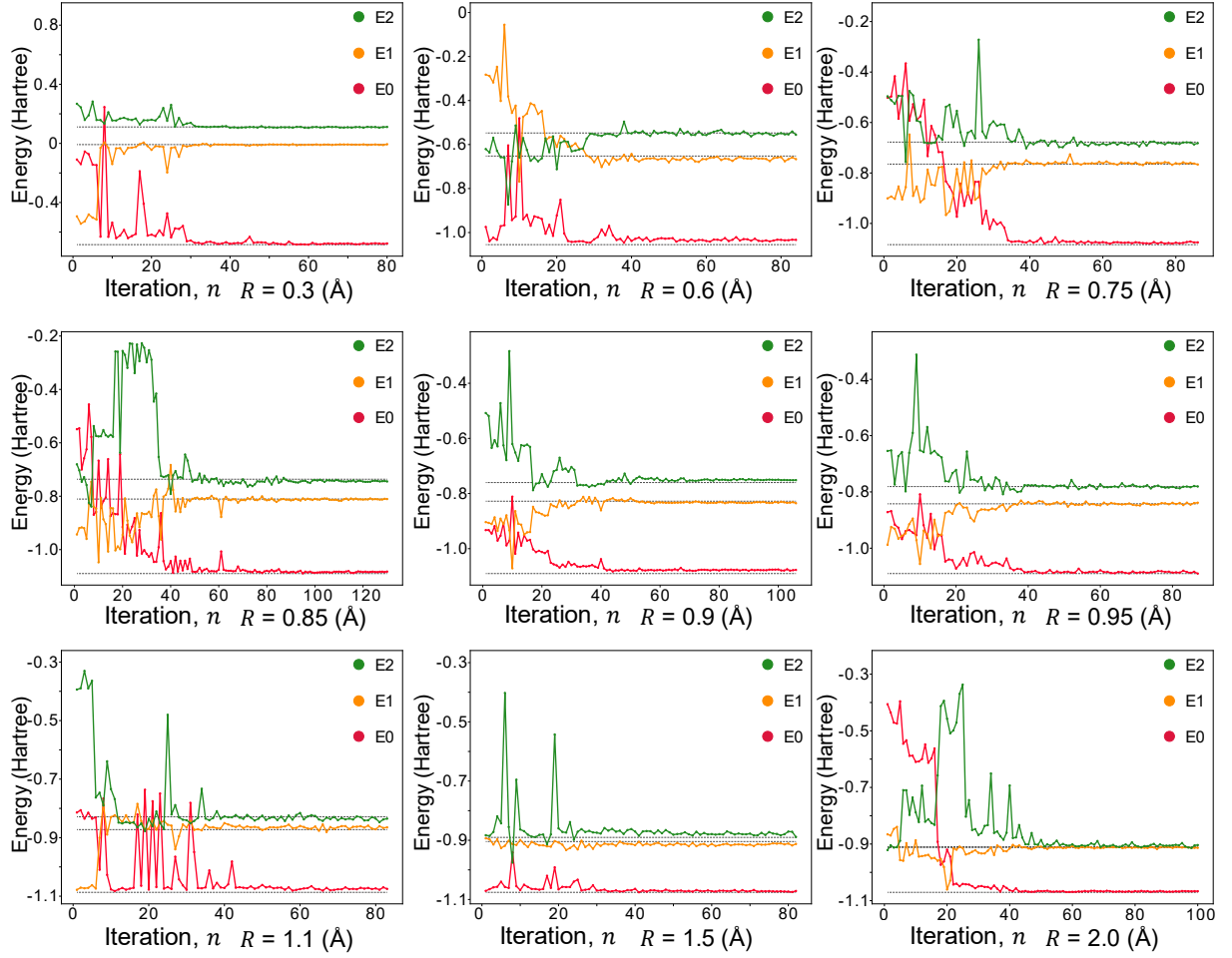
Supplementary Figure 8. **Experimental SSVQE results for three-level HeH^+ .** **a**, Iterative evolution of the 15 variational angle parameters used to prepare the three OAM-qudit ansatz states during SSVQE optimization at $R = 0.90 \text{ \AA}$ (equilibrium bond length). **b**, Iterative optimization of the estimated ground and excited state energies $\langle E_0 \rangle$, $\langle E_1 \rangle$, and $\langle E_2 \rangle$ at $R = 0.90 \text{ \AA}$. Solid colored curves (red, orange and green) denote experimental estimates as a function of iteration number n , and black dashed lines indicate theoretical values. Error bars represent one standard deviation obtained from 1,000 Monte Carlo resamplings. **c**, Estimated energies as a function of interatomic distance R (top). The energy deviations ΔE (bottom) are defined as the difference between the experimental estimates and the theoretical values.

Supplementary Fig. 7 shows the two-level implementation, where only the ground and first excited states are estimated. At the equilibrium bond length $R = 0.90 \text{ \AA}$, we use 13 variational parameters with the initial values $\vec{\alpha}_0 = (\vec{\theta}_0, \vec{\phi}_0, \vec{\omega}_0)$ chosen arbitrarily. The optimization converges toward the theoretical energies with increasing iteration number.

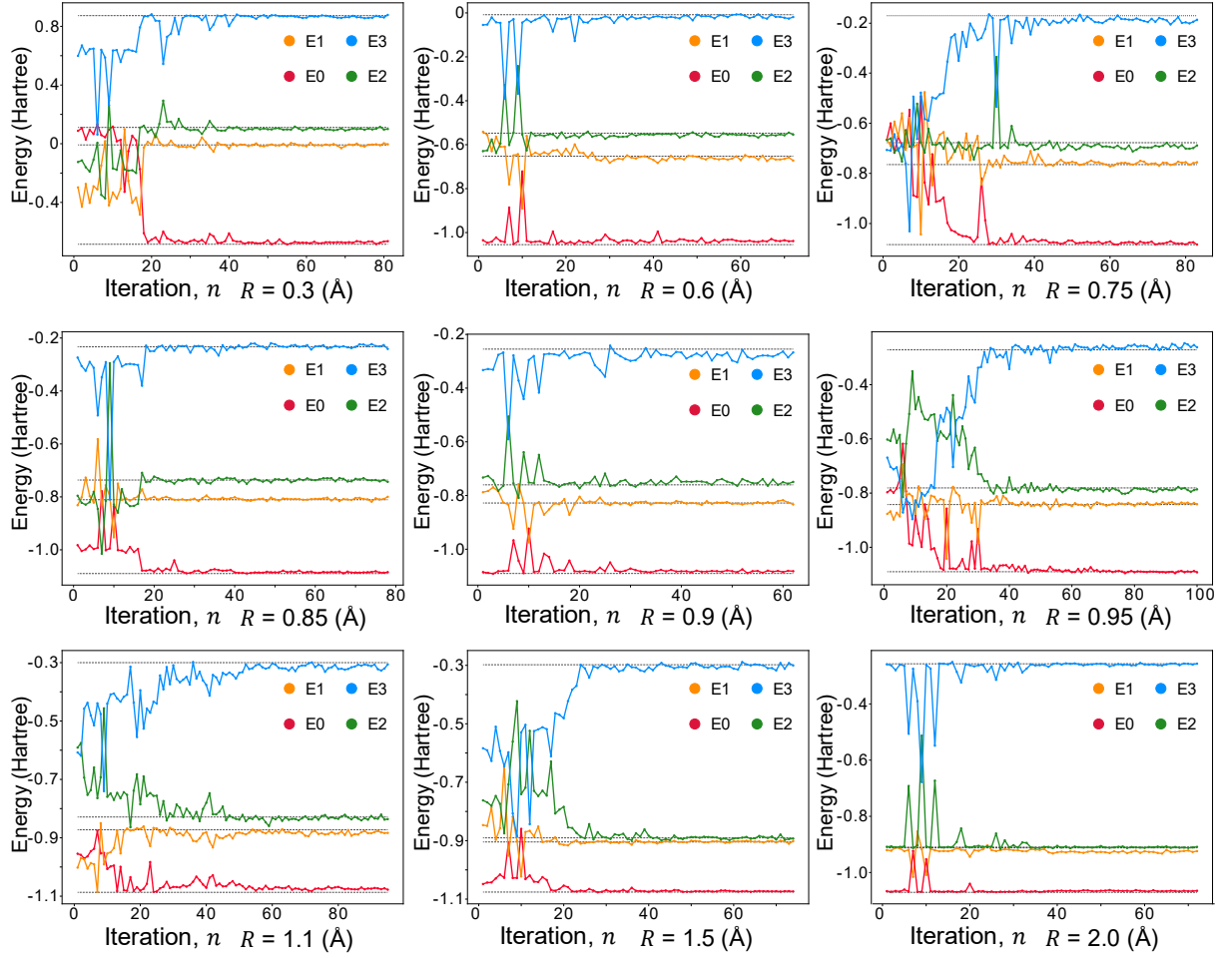
Supplementary Fig. 8 shows the three-level implementation. In this case, the energy of the remaining level can be inferred from the conserved trace of the Hamiltonian, $\text{Tr}(H)$, by subtracting the three experimentally estimated energies. By targeting fewer levels, we observe reduced experimental errors compared to the four-level case, leading to improved accuracy. Since only three ansatz states are prepared and measured, the total measurement time is correspondingly reduced, resulting in an approximately 25% decrease in experimental runtime. The average number of iterations to converge is $\sim 90.4 \pm 12.6$. For comparison, the full data sets for the two-, three-, and four-level configurations are provided in Supplementary Figs. 9–11.



Supplementary Figure 9. **Iterative SSVQE optimization for two-level HeH⁺ across interatomic distances.** Optimization trajectories of the estimated ground and first excited state energies, $\langle E_0 \rangle$ and $\langle E_1 \rangle$, shown for each interatomic distance R .

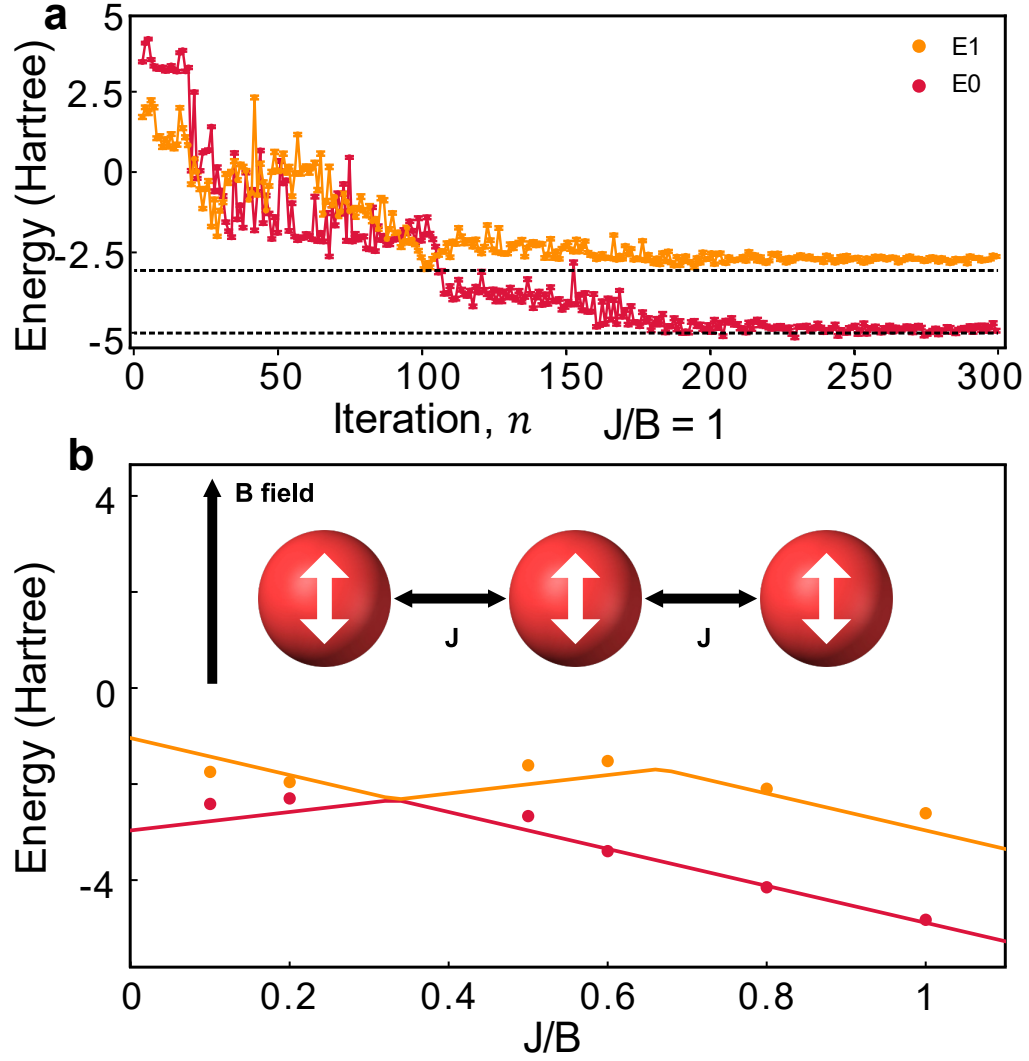


Supplementary Figure 10. **Iterative SSVQE optimization for three-level HeH⁺ across interatomic distances.** Optimization trajectories of the estimated ground, first, and second excited state energies, $\langle E_0 \rangle$, $\langle E_1 \rangle$, and $\langle E_2 \rangle$, shown for each interatomic distance R .



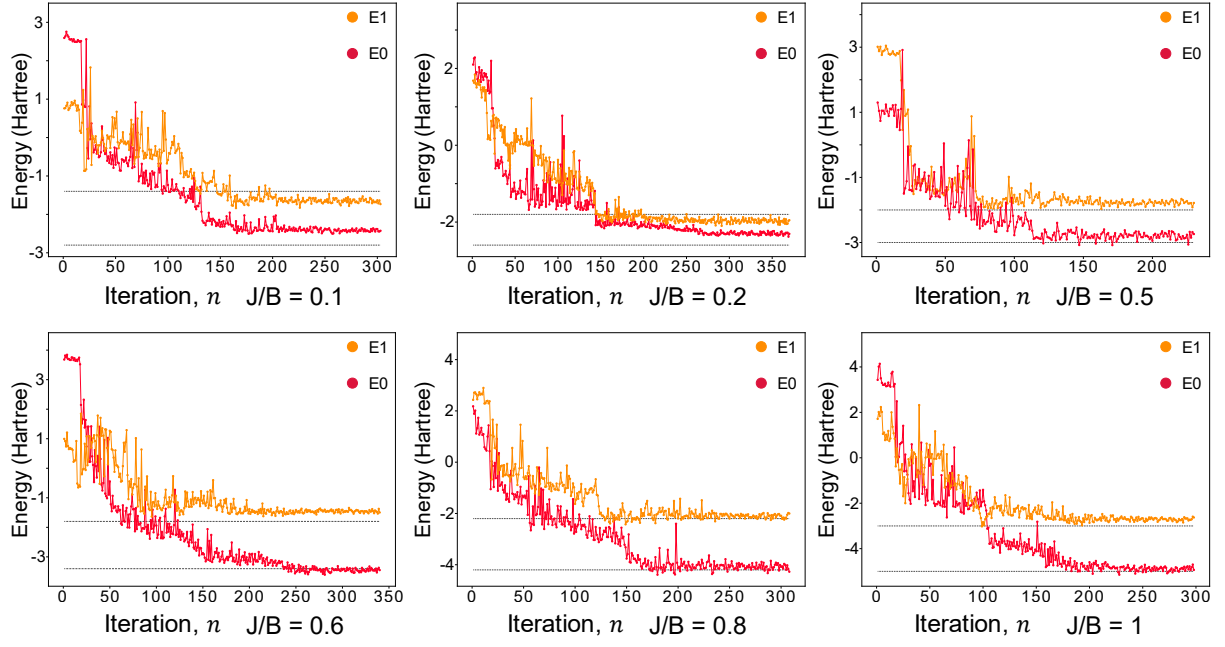
Supplementary Figure 11. **Iterative SSVQE optimization for four-level HeH^+ across interatomic distances.** Optimization trajectories of the estimated ground and excited state energies, $\langle E_0 \rangle$, $\langle E_1 \rangle$, $\langle E_2 \rangle$, and $\langle E_3 \rangle$, shown for each interatomic distance R .

SUPPLEMENTARY NOTE VI. EXPERIMENTAL RESULTS FOR HEISENBERG CHAIN MODEL

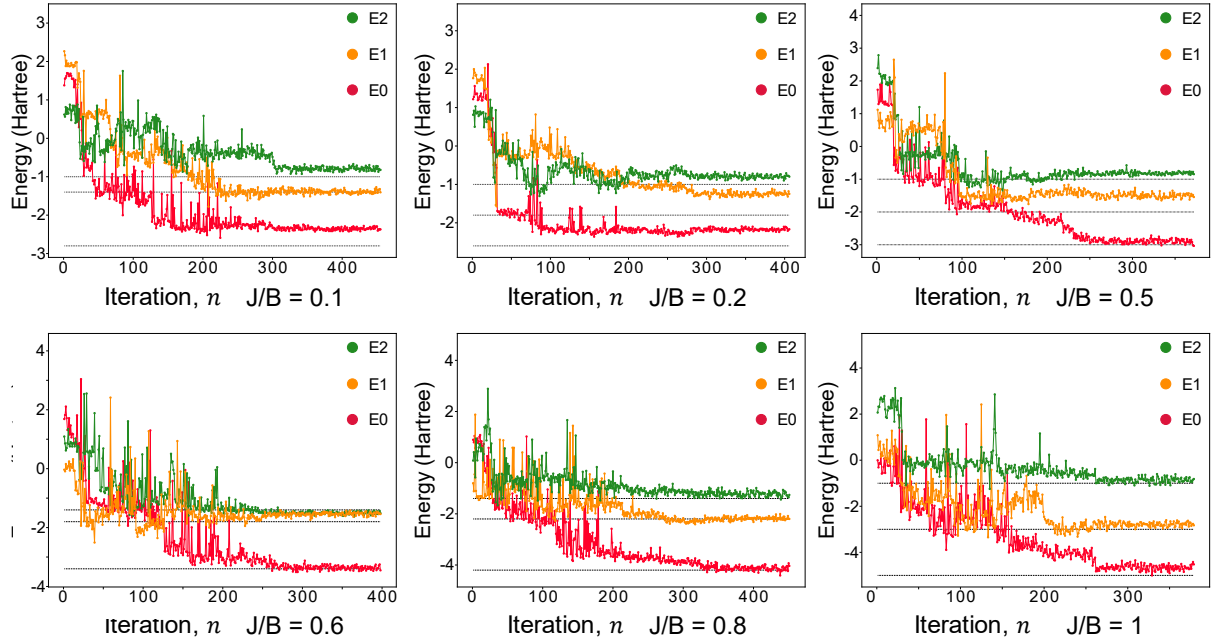


Supplementary Figure 12. **Experimental SSVQE results for two-level Heisenberg chain.** **a**, Iterative optimization of the estimated ground and first excited state energies $\langle E_0 \rangle$ and $\langle E_1 \rangle$ at $J/B = 1$, where the spin-spin interaction and the external field contribute comparably. Solid colored curves (red and orange) denote experimental estimates as a function of iteration number n , and black dashed lines indicate theoretical values. Error bars represent one standard deviation obtained from 1,000 Monte Carlo resamplings. **b**, Estimated energies as a function of the ratio J/B . The energy deviations ΔE are defined as the difference between the experimental estimates and the theoretical values.

We further demonstrate subspace-selective SSVQE on the Heisenberg-chain model by targeting only the lowest two energy levels. Supplementary Fig. 12 shows the two-level optimization results, where the estimated energies converge toward the theoretical values with increasing iteration number. At $J/B = 1$, we use 45 variational parameters and obtain an average convergence of $\sim 277.7 \pm 42.5$ iterations. Weight coefficients $w_j \in \{0.6, 0.4\}$ are used for the two targeted levels. For completeness, the full data sets for the two- and three-level configurations are provided in Supplementary Figs. 13 and 14, respectively.



Supplementary Figure 13. **Iterative SSVQE optimization for two-level Heisenberg chain across interaction ratios.** Optimization trajectories of the estimated ground and first excited state energies, $\langle E_0 \rangle$ and $\langle E_1 \rangle$, shown for each J/B .



Supplementary Figure 14. **Iterative SSVQE optimization for three-level Heisenberg chain across interaction ratios.** Optimization trajectories of the estimated ground, first, and second excited state energies, $\langle E_0 \rangle$, $\langle E_1 \rangle$, and $\langle E_2 \rangle$, shown for each J/B .

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