Quantum simulation of the wavefunction to probe frustrated Heisenberg spin systems

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Quantum simulators are controllable quantum systems that can reproduce the dynamics of the system of interest in situations that are not amenable to classical computers. Recent developments in quantum technology enable the precise control of individual quantum particles as required for studying complex quantum systems. In particular, quantum simulators capable of simulating frustrated Heisenberg spin systems provide platforms for understanding exotic matter such as high-temperature superconductors. Here we report the analogue quantum simulation of the ground-state wavefunction to probe arbitrary Heisenberg-type interactions among four spin-1/2 particles. Depending on the interaction strength, frustration within the system emerges such that the ground state evolves from a localized to a resonating-valence-bond state. This spin-1/2 tetramer is created using the polarization states of four photons. The single-particle addressability and tunable measurement-induced interactions provide us with insights into entanglement dynamics among individual particles. We directly extract ground-state energies and pairwise quantum correlations to observe the monogamy of entanglement.

During the past years, there has been an explosion of interest in quantum-enhanced technologies. The applications are manyfold and reach from quantum metrology\textsuperscript{1} to quantum information processing\textsuperscript{2}. In particular, quantum computation has generated a lot of interest owing to the discovery of quantum algorithms\textsuperscript{3–5} which outperform classical ones. The first proposed application for which quantum computation can give an exponential enhancement over classical computation was suggested by Richard Feynman\textsuperscript{6,7}. He considered a universal quantum mechanical simulator, which is a controllable quantum system that can be used to imitate other quantum systems, and is therefore able to tackle problems that are intractable on classical computers. Since then the motivation to use a quantum simulator as a powerful tool to address the most important and difficult problems in multidisciplinary science has led to many theoretical proposals\textsuperscript{8–13}.

Vast technological developments allowed for recent realizations of such devices in atoms\textsuperscript{14–16}, trapped ions\textsuperscript{17–20}, single photons\textsuperscript{21–24} and NMR (refs 25,26). The quantum simulation of strongly correlated quantum systems (for example frustrated spin systems) is of special interest and would provide new results that cannot be otherwise classically simulated\textsuperscript{27}.

To manipulate and measure individual properties of microscopic quantum systems, complete control over all degrees of freedom for each particle is required. Typically, atoms in optical lattices\textsuperscript{14} are used for realizing physical systems that can simulate various models in condensed-matter physics. The fact that the experimental addressability of single atoms in optical lattices remains very challenging\textsuperscript{28–30} leads to studies of bulk properties of the atomic ensemble ($\approx 10^6$ atoms) instead of single particles. Therefore, we use single photons in separate spatial modes and measurement-induced interactions as a quantum simulator, thus the particles are individually accessible. The tunable interaction between two entangled photon pairs allows for the precise simulation of the ground state of a spin-1/2 tetramer. We obtain the ground-state energy and have direct access to the distribution of pairwise quantum correlations as a function of the competing spin–spin interactions.

We also observe the influence of monogamy\textsuperscript{31,32} in this strongly correlated quantum system.

**Analogue quantum simulator**

The main challenge in the understanding of strongly correlated quantum systems is to calculate the energies and ground-state properties of many-body systems, which becomes exponentially difficult with increasing number of particles when using a classical computer. In contrast, quantum simulators use quantum systems to store and process data, which enables them to polynomially mimic the evolution of the quantum system of interest. Typically, quantum simulations require methods to implement the Hamiltonian of the simulated system and probe its ground-state properties. In some cases, even if the ground-state wavefunction is known, obtaining all properties by using classical computers remains a challenging task. For example, for the one-dimensional antiferromagnetic Heisenberg model we can compute the spectrum and the wavefunction by using the Bethe ansatz\textsuperscript{33}, but it is not as easy to extract the correlation functions. This can be overcome by quantum simulators that are capable of generating and directly probing the quantum state of interest by controlling each quantum particle individually.

Usually, the system being simulated is defined by its Hamiltonian $H(t,J,B,...)$, which is dependent on parameters such as time, interaction strength, $J$, external field, $B$, and so on. In practice, one way of realizing a quantum simulator is based on discrete gate operations and the phase-estimation algorithm\textsuperscript{10,21}, referred to as a digital quantum simulator\textsuperscript{12}. An alternative approach uses the adiabatic theorem\textsuperscript{34}, where an initial Hamiltonian, whose ground state is easy to prepare, can be adiabatically evolved to a final Hamiltonian with a non-trivial ground state of interest\textsuperscript{8,9,11,13,26}.

An adiabatic quantum simulator can be built by engineering interactions among particles using tunable external parameters (for example an external magnetic field). The system will remain in its ground state if the system parameters change gradually enough.

Our experimental technique combines the advantages of both approaches by using a tunable quantum gate without

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Figure 1 | Mimicking an adiabatic quantum evolution with an analogue quantum simulator. a, Adiabatic quantum evolution. The system is prepared in an initial ground state |Ψ(t0, B0, J0,...)⟩. Then the gradual change of the system parameters (t, B, J and so on) causes an adiabatic evolution of the system to the final ground state of interest |ψ(t, B, J,...)⟩. b, Analogue quantum simulation. The adiabatic evolution of the system to be simulated is mapped onto a controllable evolution of a quantum system. A set of tunable gates gives access to the change of parameters. c, Model used to study the valence-bond states. The nearest-neighbour Heisenberg-type interactions of strengths J and weighted superposition of all different localized valence-bond states. If all the spins are covered by valence bonds, which are bond states might explain high-temperature superconductivity in cuprates. A quantum simulator capable of preparing such states is restricted to total spin-zero subspace for arbitrary superpositions of dimer-covering states is thus sufficient in cuprates because it was conjectured that transition from a localized valence-bond state to the superposition of different valence-bond states, corresponds to a quantum spin liquid, the so-called resonating valence-bond state.

The smallest configuration for studying and simulating these phenomena on a two-dimensional square lattice is four spin-1/2 particles forming a tetramer. In the case of such a spin-1/2 tetramer the Heisenberg-type interactions lead to the creation of three possible dimer-covering configurations for the localized valence-bond states, \(|Φ+⟩ = |ψ+⟩_1|ψ+⟩_2|ψ+⟩_3|ψ+⟩_4\) and \(|Φ−⟩ = |ψ−⟩_1|ψ−⟩_4|ψ−⟩_2|ψ−⟩_3\), where \(|ψ±⟩\) is the singlet of particle i and j (Fig. 1c). As the total spin-zero subspace for this system is two dimensional, these three dimer-covering states are not independent and \(|Φ+⟩\) can be written as \(|Φ+⟩ = |Φ⟩_2 − |Φ⟩_1\) in the \(|Φ⟩_2/|Φ⟩_1\) basis. This state, \(|Φ+⟩\), like any other equal superposition of these two dimer-covering states, represents a resonating valence-bond state. Particularly interesting states are resonating valence-bond states with s-wave pairing symmetry, \(|Φ⟩_1 + |Φ⟩_3\) (up to normalization), and with the exotic d-wave pairing symmetry, \(|Φ⟩_1 − |Φ⟩_3\). The studies of these states are of high interest, because it was conjectured that transition from a localized valence-bond configuration to the superposition of different valence-bond states might explain high-temperature superconductivity in cuprates. A quantum simulator capable of preparing such arbitrary superpositions of dimer-covering states is thus sufficient for simulating any Heisenberg-type interactions of four spin-1/2 particles on a two-dimensional lattice. It is the particular strength of our optical quantum simulator that the simulated ground states can be restricted to the spin-zero singlet subspace by using the quantum interference of photons at a tunable beam splitter.

For our quantum simulation we consider four spin-1/2 particles on a square lattice (tetramer) that interact through Heisenberg-type interactions. According to Marshall’s theorem, the ground state is restricted to total spin-zero subspace for arbitrary antiferromagnetic interactions, independent of their type and

the necessity of either discretizing the quantum evolution or engineering the physical interactions for an adiabatic quantum simulation. Thus, we consider our simulator as an analogue quantum computer, where the change of the quantum evolution can be obtained by a tunable quantum gate. The advantages of using analogue quantum gate operations seem to be far reaching as the number of gate operations can be less demanding than for digital quantum simulators. Figure 1 shows the concept of this analogue simulator. In our experiment, we use the analogue quantum simulator to directly probe the wavefunction instead of the Hamiltonian. We take advantage of single-particle addressability and the tunable interaction of our quantum simulator to prepare various ground states experimentally. Once the wavefunction is prepared, it is feasible to measure all the correlation functions and study the dynamics of entanglement in this strongly correlated system.
strength. Here we model our spin tetramer with nearest-neighbour interactions of strengths $J_1$ and $J_2$ (Fig. 1c) by the Hamiltonian

$$H = J_1 S_1 S_3 + J_2 S_2 S_4 + J_3 S_1 S_2 + J_4 S_3 S_4$$

where $S_i$ is the Pauli spin operator for spin $i$. All the properties of the system depend only on the coupling ratio $\kappa = J_2/J_1$; therefore, we re-normalize the Hamiltonian to

$$H(\kappa) = H_0 + \kappa H_1$$

where $H(\kappa) = H/|J_1|$ is the final Hamiltonian, $H_0 = S_1 S_3 + S_2 S_4$ the initial Hamiltonian and $H_1 = S_1 S_2 + S_3 S_4$ the competing Hamiltonian of $H_0$.

Due to the simplicity of the quantum system we study here, we can calculate the expected ground state analytically, which enables us to verify the experimental results. The ground state of the Hamiltonian given in equation (1) is

$$|\Psi(0)(\theta)\rangle = \frac{1}{\sqrt{n(\theta)}} (\cos 2\theta|\Phi_1\rangle + \cos^2 \theta|\Phi_2\rangle + \sin^2 \theta|\Phi_3\rangle)$$

where $n(\theta) = 1/2 + \cos^2 \theta + \sin^4 \theta$ is the normalization constant. The ground-state energy of $|\Psi(0)(\theta)\rangle$ is $E(0) = -2(1 + \kappa - 4\sqrt{1 - \kappa^2} + \kappa^2)$. The TDC’s angle, $\theta$, takes the values from the interval of $0 \leq \theta \leq \pi/2$. Using our photonic quantum simulator, we can mimic the adiabatic change of the Hamiltonian.
shown in equation (1), where the full range of the coupling ratio $-\infty \leq \kappa \leq +\infty$ is experimentally covered by tuning the angle of the TDC between $\arctan(1/\sqrt{2}) \leq \theta \leq \pi/2$. For $\kappa = 0$ ($\theta = \pi/4$) the ground state is $|\Phi_1\rangle$, whereas for $\kappa = +\infty$ ($\theta = \pi/2$) the ground state changes to $|\Phi_\infty\rangle$. These two cases are dimer-covering states. Tuning the coupling ratio to $\kappa = -\infty$ results in the equally weighted superposition $|\Phi_+\rangle + |\Phi_\infty\rangle$, whereas $\kappa = 1$ leads to the interesting resonating valence-bond state$^{37,38,40}$ $|\Phi_\pi\rangle + |\Phi_1\rangle$.

Here, we experimentally demonstrate an optical analogue of a quantum simulator by producing two polarization-entangled photon pairs (see Fig. 2a), $|\psi^-(\theta)\rangle_{12} = 1/\sqrt{2}(|HV\rangle_{12} - |VH\rangle_{12}$) and $|\psi^+(\theta)\rangle_{34} = 1/\sqrt{2}(|HV\rangle_{34} - |VH\rangle_{34})$, in the spatial modes 1 and 2 and 3 and 4. $|H\rangle$ and $|V\rangle$ denote horizontal and vertical polarization states, respectively. The tunable interaction among these singlet states is achieved by the TDC, followed by a projective measurement of one photon in each of the four output modes. This tunability enables us to continuously change the measurement-induced interaction between photons 1 and 3. The TDC is an optical fibre device that transfers optical signals between fibres as a beam splitter with controllable splitting ratio. The control of the splitting ratio is achieved by adjusting the relative positions of the fibres (Fig. 2b). The transmissivity and reflectivity of this TDC go as $\cos^2\theta$ and $\sin^2\theta$, respectively, where $\theta$ parameterizes the separation of the fibres. We calibrate the TDC’s transmissivity and reflectivity such that the modulating visibilities (Michaelson visibility) are above 95% for both inputs, as required for high-precision quantum control (Fig. 2c).

A successful detection of a fourfold coincidence event from each spatial mode gives the four-photon state, $|\psi(\theta)\rangle_{1234} = (1/\sqrt{4})[|HHVV\rangle - \cos^2\theta(|HHVH\rangle + |VHHV\rangle) + \sin^2\theta(|HVHV\rangle + |VHVV\rangle) + \cos 2\theta(|HHVH\rangle + |VHVV\rangle)]$. The experimentally obtained density matrix, $\rho_{\text{exp}}$, is reconstructed from a set of 1,296 local measurements using the maximum-likelihood technique$^{41,42}$. For this, all combinations of mutually unbiased basis sets for individual qubits, that is $|H\rangle|V\rangle$, $|\pm\rangle = 1/\sqrt{2}(|H\rangle \pm |V\rangle)$, and $|R/L\rangle = 1/\sqrt{2}(|H\rangle \pm |V\rangle)$, are measured. The duration of each measurement for a given setting of the polarization analysers and the TDC was 200 s and the average detected fourfold coincidence rate was 3 Hz. In total eight density matrices for different settings of $\theta$ are reconstructed and are summarized in Supplementary Information. Uncertainties in quantities extracted from these density matrices are calculated using a Monte Carlo routine and assumed Poissonian errors.

The ground-state energy, $E^{(0)}$, is defined as the mean value of the Hamiltonian given in equation (1). As this Hamiltonian contains only pairwise interactions, we measured the expectation value of the corresponding pairwise correlations and obtained the data shown in Fig. 3, which show good agreement with theoretical prediction.

In Fig. 4, we show the experimentally obtained density matrices of the four valence-bond states, $|\Phi_+\rangle + |\Phi_\infty\rangle$, $|\Phi_\pi\rangle + |\Phi_1\rangle$ and $|\Phi_\infty\rangle$ and $|\Phi_\pi\rangle$, which correspond to the setting of $\theta = 0.197\pi$, $\theta = 0.25\pi$, $\theta = 0.304\pi$ and $\theta = 0.468\pi$. The state fidelity is defined as $F(\Psi, \rho) = (\Psi|\rho|\Psi)$, where $|\Psi\rangle$ is the target and $\rho$ is the experimentally obtained quantum state. Due to the high quality of our quantum simulator, we obtain four-photon state fidelities that range from $F = 0.712(4)$ to $F = 0.888(2)$ (see Supplementary Information).

**Quantum monogamy and complementarity**

Monogamy is one of the most fundamental properties of quantum entanglement$^{31,32}$. It restricts the shareability of quantum correlations among parties and is of essential importance in many quantum information processing protocols, including quantum cryptography and entanglement distillation. Recent work has shown that, in the context of condensed-matter physics, monogamy
is defined as $e_{ij} = (-1/3)\text{Tr}(\rho_i S_i S_j)$. Note that $\rho_{ij}$ is the density matrix of spins $i$ and $j$. The normalized Heisenberg energy per unit of interaction is also an entanglement witness$^{33,44}$ and reaches its maximum value of $e_{ij} = 1$ for the singlet state. The amount of entanglement can also be quantified by concurrence$^{45}$, which is directly related to $e_{ij}$ with $C(e_{ij}) = \max(0,(-1/2) + (3/2)e_{ij})$. For our four-spin system the dependencies of the pairwise energies with respect to the TDC’s angle $\theta$ are given by $e_{12} = -(\sin^2 \theta \cos 2\theta)/n$, $e_{13} = (\sin 2\theta \cos \theta)/n$ and $e_{14} = (\cos^2 \theta \cos 2\theta)/n$.

Remarkably, monogamy is manifested in the constraint of the energy distribution for the considered spin pair through a complementarity relation$^{46,47}$

$$e_{12}^2 + e_{13}^2 + e_{14}^2 = 1$$

This restricts the maximal amount of energy or entanglement associated with correlated spin systems (see Fig. 5a). For instance, in the experiment we obtain the normalized Heisenberg energy per unit interaction between photons 1 and 2 ($e_{12}$) with the correlation measurements in three mutually unbiased bases $(S_i^{(u)} \otimes S_j^{(u)})$ where $u = 1, 2, 3$.

As shown in Fig. 5a, the adiabatic change of the coupling between the four spins is simulated by tuning the angle of the TDC, $\theta$, from $-\pi/2$ to $\pi/2$. This corresponds to the full range of the coupling ratio $-\infty \leq \kappa = J_{ij}/J_{i'j'} \leq \infty$. In the ideal case, the maximum of $e_{ij}$ is unity, which corresponds to a singlet state shared by spins $i$ and $j$. However, imperfections in the generation of entangled photon pairs and the two-photon interference on the TDC reduce the measured value of $e_{ij}$ by a constant factor, independent of $\theta$. For the individual photon pairs we obtain the maximal Heisenberg energy of $e_{12}\text{max} = 0.920(7)$, $e_{13}\text{max} = 0.727(9)$ and $e_{14}\text{max} = 0.926(5)$.

To demonstrate the complementarity relation$^{48}$ we renormalize each energy $e_{ij}$ by its maximal value $e_{ij}\text{max}$ and obtain a good agreement with the theoretical prediction shown in Fig. 5b.

The advantage of the individual addressability for our particles in the ground state allows for the direct extraction of the pairwise quantum correlations. The pairwise quantum correlation is defined as

$$T(S_i^{(u)}, S_j^{(u)}) = \frac{C(S_i^{(u)} \otimes S_j^{(u)}, S_i^{(v)} \otimes S_j^{(v)})}{C(S_i^{(u)} \otimes S_j^{(u)}, S_i^{(v)}) + C(S_i^{(u)} \otimes S_j^{(u)}, S_j^{(v)}) - C(S_i^{(u)} \otimes S_j^{(u)}, S_i^{(v)}) - C(S_i^{(u)} \otimes S_j^{(u)}, S_j^{(v)}) - C(S_i^{(u)} \otimes S_j^{(u)}, S_i^{(v)}) - C(S_i^{(u)} \otimes S_j^{(u)}, S_j^{(v)})}$$

where $C(S_i^{(u)} \otimes S_j^{(u)}, S_i^{(v)} \otimes S_j^{(v)})$ are the coincidence counts between pair $i$ and $j$ along the direction of $S_i^{(v)}$ and that perpendicularly to $S_i^{(v)}$, respectively. In Fig. 6, the pairwise correlation functions for the ground states, $(|\Phi_+\rangle + |\Phi_\perp\rangle, |\Phi_\perp\rangle, |\Phi_\perp\rangle + |\Phi_\perp\rangle$ and $(|\Phi_\perp\rangle)$, are shown. As expected from the monogamy relation, in the cases of dimer-covered states, one pair of photons is maximally correlated, for example photons 1 and 3 in Fig. 6b, and photons 1 and 2 in Fig. 6d. In the cases of the equal superposition of two dimers (Fig. 6a) and the resonating valence-bond state (Fig. 6c), correlations are distributed among different pairs.

We demonstrate the feasibility of an all-optical analogue quantum simulator by enabling quantum control of the measurement-induced interaction among photonic quantum states. Various ground states, including the resonating valence-bond states for four interacting spin-1/2 particles, are generated and characterized by extracting the total energy and the pairwise quantum correlations. The simulation of a spin-1/2 tetramer also proves that the pairwise entanglement and energy distribution are restricted by the role of quantum monogamy. Our results provide promising insights for quantum simulations of small quantum systems, where individual addressability and control over all degrees of freedom on the
Figure 6 | Directly observed pairwise correlation functions of various valence-bond states. 

(a) The correlation tensors $T_{12}$ (photons 1 and 2), $T_{13}$ (photons 1 and 3) and $T_{14}$ (photons 1 and 4) are obtained from correlation measurements directly in the bases $X = \sigma_x$, $Y = \sigma_y$, and $Z = \sigma_z$. For a convenient graphical representation, the negative values of the correlation tensors are shown. The structures of the superposition state (a) and the resonating valence-bond state (c) show that the quantum correlations are equally distributed among two competing pairs. 

(b,d) Dimer-covering states, in which only one pair is maximally correlated in a singlet state.

single-particle level is required. This is of particular interest for quantum chemistry with small numbers of particles and might enable in the near future the simulation of aromatic systems and chemical reactions\(^ {49}\). Although it was shown that efficient scalable quantum computing with single photons, linear-optical elements and projective measurements is possible\(^ {30}\), the most important challenges will be (1) the realization of two- and more-qubit interactions with high fidelity, (2) generating systems with more qubits and (3) developing efficient methods of simulating other classes of complex Hamiltonians by using optical elements. Ideally, this and related work will open a new and promising avenue for the experimental simulation of various quantum systems.

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Author contributions
X.-M. and W.N. designed and carried out experiments, analysed data and wrote the manuscript. B.D. provided the theoretical analysis, analysed data and wrote the manuscript. A.Z. supervised the project and edited the manuscript. P.W. designed experiments, analysed data, wrote the manuscript and supervised the project.

Additional information
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