

Experimental four-qubit bound entanglement

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Entanglement is one of the most puzzling features of quantum theory and of great importance for the new field of quantum information. Being a peculiar form of entanglement, bound entanglement emerges in certain mixed quantum states. This form of entanglement is not distillable by local operators and classical communication. Bound-entangled states are different from both the free entangled (distillable) and separable states. Here we report on the first experimental demonstration of a four-qubit polarization bound-entangled state, the so-called Smolin state. We have fully characterized its entanglement properties. Moreover, we have realized unlocking of the entanglement protocol for this state. The special properties of the Smolin state constitute a useful quantum resource for new multiparty communication schemes.

Quantum entanglement leads to the most counterintuitive effects in quantum mechanics^{1,2}. Entanglement plays a central role in the field of quantum information, leading to ongoing efforts for its quantitative and qualitative characterization. While entanglement of pure bipartite states is well understood^{3,4}, the entanglement of mixed and multipartite systems it is still under intense research. Studies of entanglement are particularly relevant for evaluating its use as a resource for multiparty quantum-communication protocols. Protocols such as super-dense coding, quantum teleportation and telecloning cannot be carried out without shared entanglement between the communicating parties. Entanglement, however, is a very fragile resource, easily destroyed by the decoherence processes owing to unwanted coupling with the environment⁵. Such uncontrollable influences cause, for example, noise, so that the entangled quantum states become mixed. Therefore it is important to know which mixed states can be distilled to maximally entangled states using local operations and classical communication, and then be useful for further information processing. It has been shown that any bipartite mixed entangled two-qubit and qubit-qutrit states can be distilled to singlet states^{6,7}, but this is not true for all mixed entangled states. An important theoretical discovery of a class of quantum entangled states where no entanglement can be distilled has been made. This entanglement class has been called bound entanglement^{8,9}. This new class of states is between separable and free-entangled states. The free entanglement can be distilled to the singlet form. Bound entanglement is not distillable and it is considered, in analogy with thermodynamics, as an entanglement that cannot be used to carry out useful information work, such as reliable transmission of quantum data through teleportation⁹. Importantly, it has been shown that bound entanglement naturally appears in quantum many-body systems^{10,11}. Beyond physics, in information theory, the concept of bound entanglement has led to a new type of information called bound information, which became itself a subject of a very active research area¹².

The Smolin state is a four-party unlockable bound-entangled state¹³, because no entanglement can be distilled with local operations and classical communication when the parties are separated. However, if any two parties come together and make joint measurements, entanglement can be distilled for the other two parties. The entanglement that is distilled will be pure and maximal but not available to the parties that have carried out the distillation. In this distillation protocol two parties have

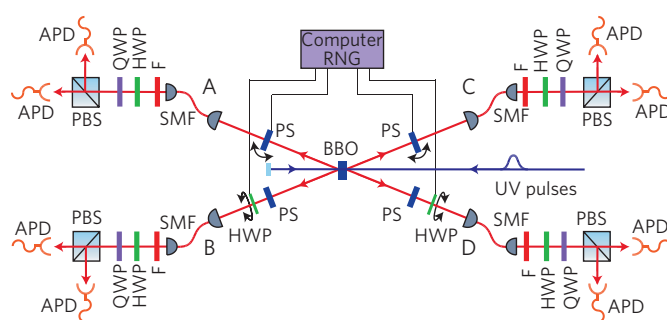


Figure 1 | Experimental set-up for the generation of a four-qubit polarization bound-entangled state. The experimental set-up consists of double-pass SPDC. A BBO crystal is pumped twice with ultraviolet pulses, creating four photons entangled pairwise. The four different Bell states in equation (2) are randomly prepared, using an RNG, prepared by single-qubit-flip (HWP) and phase-shift (PS) gates. The photons are coupled to single-mode fibres (SMFs) passing a narrowband filter (F) and brought to local polarization analysers. The photons are detected with avalanche photodiodes (APDs); see the Methods section for more details.

to meet to unlock the entanglement. This is not necessary in general, because there exists a superactivation¹⁴ protocol where the unlocking is carried out by introducing one more party, and one more Smolin state distributed to four of the five parties. The usefulness of bound entanglement in quantum information tasks has been questioned, but it has been shown that bound-entangled states are a useful resource for multiparty secret sharing cryptographic protocol¹⁵, superactivation¹⁴ and remote information-concentration¹⁶ schemes. Recently, it has been proven that the Smolin state can maximally violate a simple two-setting Bell inequality similar to the standard Clauser–Horne–Shimony–Host inequality¹⁷, implying directly the possibility of a reduction of communication complexity protocol¹⁸. Next, a link between multipartite unlockable bound-entangled states and the useful stabilizer formalism was found¹⁹.

The four-qubit bound-entangled state ρ_S introduced by Smolin is defined as an equal mixture of all four Bell states and it is given by¹³

$$\rho_S = \sum_{i \in \{1,2,3,4\}} \frac{1}{4} |\psi_{AB}^{(i)}\rangle\langle\psi_{AB}^{(i)}| \otimes |\psi_{CD}^{(i)}\rangle\langle\psi_{CD}^{(i)}| \quad (1)$$

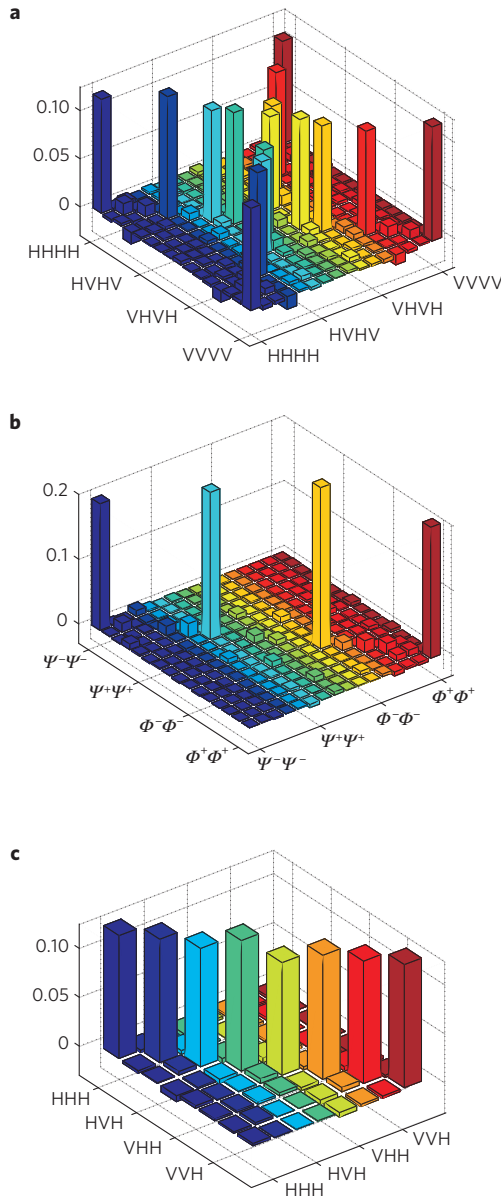


Figure 2 | Experimental results: the density matrix. Density matrix of the mixed four-photon bound entangled Smolin state ρ_S^{exp} . **a**, Matrix experimentally obtained from the quantum-state tomography in the computational basis H, V . Errors were obtained by a Monte Carlo simulation and have a maximum of 0.02 for both real and imaginary parts. **b**, ρ_S^{exp} in the Bell basis. **c**, Reduced density matrix of the three remaining qubits when one qubit is traced out.

where $\psi^i \in \{|\Psi^\pm\rangle, |\Phi^\pm\rangle\}$ are the Bell states given by

$$\begin{aligned} |\Psi^\pm\rangle &= \frac{1}{\sqrt{2}}(|01\rangle \pm |10\rangle) \\ |\Phi^\pm\rangle &= \frac{1}{\sqrt{2}}(|00\rangle \pm |11\rangle) \end{aligned} \quad (2)$$

where $|0\rangle$ and $|1\rangle$ are the qubit basis. Notice that the subscripts A, B, C and D denote the parties by whom the state is shared. The Smolin state can be expressed in a useful form as

$$\rho_S = \frac{1}{16} \left(\mathbb{1}^{\otimes 4} + \sum_{i \in \{1,2,3\}} \sigma_i^A \otimes \sigma_i^B \otimes \sigma_i^C \otimes \sigma_i^D \right) \quad (3)$$

Table 1 | Positive partial transpose for the bipartite cuts.

Theory	AB CD	AC BD	AD BC
0.25	0.25 ± 0.01	0.24 ± 0.01	0.23 ± 0.01
0.25	0.24 ± 0.01	0.23 ± 0.01	0.24 ± 0.01
0.25	0.21 ± 0.01	0.21 ± 0.01	0.21 ± 0.01
0.25	0.20 ± 0.01	0.22 ± 0.01	0.22 ± 0.01
0	0.04 ± 0.01	0.04 ± 0.01	0.04 ± 0.01
0	0.02 ± 0.01	0.03 ± 0.02	0.03 ± 0.01
0	0.02 ± 0.01	-0.02 ± 0.02	-0.01 ± 0.01
0	0.01 ± 0.01	-0.01 ± 0.02	-0.01 ± 0.01
0	0.01 ± 0.01	0.02 ± 0.02	0.02 ± 0.01
0	-0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01
0	0.00 ± 0.01	0.00 ± 0.01	0.00 ± 0.01
0	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01
0	0.00 ± 0.01	0.01 ± 0.01	0.00 ± 0.01
0	0.00 ± 0.01	0.01 ± 0.01	0.00 ± 0.01
0	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.01
0	0.00 ± 0.01	0.00 ± 0.01	0.01 ± 0.01

Eigenvalues of the partial transpose of the bipartite AB|CD, AC|BD and AD|BC cuts. All the eigenvalues are positive or zero up to the error bars.

where $\mathbb{1}^{\otimes 4} = \mathbb{1} \otimes \mathbb{1} \otimes \mathbb{1} \otimes \mathbb{1}$, $\mathbb{1}$ is the 2×2 unity matrix, the symbol \otimes is the tensorial product and σ_i for $i \in \{1, 2, 3\}$ are the Pauli spin matrices. In this form we can see that the state is invariant when interchanging particles. An important property of state ρ_S is that, if we take the partial trace on any of the qubits (equation (3)), the remaining three qubits are in a maximally mixed state $\mathbb{1}^{\otimes 3}/8$ and consequentially there are no correlations left between the remaining parties. This corresponds, for example, to situations where the information about a qubit is not accessible, or a party is not willing to cooperate.

If the four parties sharing the Smolin state are located in separated laboratories, the state, as it is written in the form (equation (1)), is separable across the bipartite cut AB|CD and owing to the invariance of the state it is also separable across the other bipartite AC|BD and AD|BC cuts. Therefore each party is separated from the other parties and this implies that no entanglement can be distilled between any two parties or any pair of parties.

If the parties C and D come together in the same laboratory and make a projective measurement in Bell basis $\{\Psi^+, \Psi^-, \Phi^+, \Phi^-\}$, and send the result of their measurement (classical information) to A and B, who will then know which of the Bell states they share, parties A and B can convert their state into the standard singlet state through local operations and classical communication. Furthermore, it has been shown that the Smolin state cannot be written in the separable form A|BCD, that is, $\rho_S = \sum_i \alpha_i |\psi_a^{(i)}\rangle \langle \psi_a^{(i)}| \otimes |\psi_{bcd}^{(i)}\rangle \langle \psi_{bcd}^{(i)}|$; the Smolin state ρ_S is entangled and not distillable and therefore by definition is called a bound-entangled state¹³.

In our experiment the physical qubits are polarized photons, and the computational basis corresponds to horizontal H and vertical V linear polarization $|0\rangle \equiv |H\rangle$ and $|1\rangle \equiv |V\rangle$. The four-photon polarization bound-entangled state ρ_S can be obtained as follows: first we generate the product of two photon pairs in singlet state $|\psi^-\rangle$ by two spontaneous parametric down-conversion (SPDC) sources²⁰ (double pass through the nonlinear type-II β -barium borate (BBO) crystal; see Fig. 1). Next, we transform randomly the singlet $|\psi^-\rangle$ to any of the other Bell states. This is obtained by following simple local operations:

$$\begin{aligned} |\Psi^-\rangle &= \mathbb{1} \otimes \mathbb{1} |\Psi^-\rangle, \quad |\Psi^+\rangle = \sigma_z \otimes \mathbb{1} |\Psi^-\rangle \\ |\Phi^-\rangle &= \mathbb{1} \otimes \sigma_x |\Psi^-\rangle, \quad |\Phi^+\rangle = \sigma_z \otimes \sigma_x |\Psi^-\rangle \end{aligned} \quad (4)$$

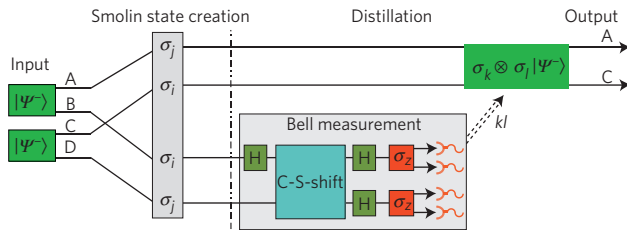


Figure 3 | Entanglement-distillation scheme. Distillation of entanglement: two singlet states $|\Psi^-\rangle$ are created and randomly manipulated by the operators in equation (4) (denoted by $\sigma_i \otimes \sigma_j$ in the figure), creating the Smolin state. By applying a Bell measurement on any two qubits, say B and D, the other two will be in the same Bell state as B and D were detected in. Here H are Hadamard operators and C-S-shift is a control sign-shift gate.

These operators were experimentally realized by motorized rotating half-wave plates (HWPs) and motorized tilting phase-shift crystals (PSs). To guarantee the equal mixture of the four Bell states, the drivers of all these motors are controlled by random-number generators (RNGs) connected to a computer, as shown in Fig. 1. All measurements in the four modes A, B, C and D are made with polarization analysis components, followed by single-photon avalanche photodiode (APD) detectors, and a multichannel coincidence unit. An average of three fourfold coincidences per 10 s was recorded.

To experimentally and fully investigate the properties of Smolin state, we have evaluated the four-photon 16×16 density matrix ρ_s by making 81 local polarization measurements in different bases of the linear and circular polarizations $|H/V\rangle$, $|+/-\rangle = (|H\rangle \pm |V\rangle)/\sqrt{2}$ and $|R/L\rangle = (|H\rangle \pm i|V\rangle)/\sqrt{2}$. The results of these measurements enable us to tomographically²¹ reconstruct the density matrix ρ_s^{exp} . Fourfold coincidences were recorded for the 81 projective measurements, each setting being measured for 3 h. To guarantee that the reconstruction algorithm does not allow unphysical results a maximum-likelihood technique was used. Figure 2a,b shows the real parts of the elements of the density matrix ρ_s^{exp} in the H/V and in the Bell bases, respectively. One can clearly observe the symmetric form of the state in both bases. As mentioned above, if one of the qubits of the Smolin state is lost or not accessible, then ρ_s will be reduced to a maximally mixed $\mathbb{1}^{\otimes 3}/8$ state. By tracing out one of the qubits in the ρ_s^{exp} matrix, we obtained the results presented in Fig. 2c, showing peaks on the diagonal, and with the same height. One can see in Fig. 2 that the noise is very low and uniform, enabling us to assume that ρ_s^{exp} can be written as $\rho_s^{\text{exp}}(p) = p\rho_s + (1-p)\mathbb{1}^{\otimes 4}/16$ where $0 \leq p \leq 1$. It has been shown that if $p \leq 1/3$ then $\rho_s(p)$ is separable. After calculating the state fidelity $F_s = \text{Tr}(\sqrt{\sqrt{\rho_s}\rho_s^{\text{exp}}\sqrt{\rho_s}})$ we obtained a very high value of $F_s = 0.933 \pm 0.002$. This value is well above the lower bound of the separability, which is 0.71.

The separability across the bipartite AB|CD, AC|BD and AD|BC cuts was tested using the Peres (positive partial transpose) criterion^{3,4}. The eigenvalues of the partial transpose of ρ_s^{exp} with respect to the bipartite AB|CD, AC|BD and AD|BC cuts are shown in Table 1. All the eigenvalues are positive or zero up to the error bars, indicating clearly the separability across these cuts. To rule out one–three-qubit separability and three-party entanglement a stabilizer witness²² was constructed (derivation of the witness is presented in the Methods section). The witness used is of the form

$$\mathcal{W}_s = \mathbb{1}^{\otimes 4} - \sigma_x^{\otimes 4} - \sigma_y^{\otimes 4} - \sigma_z^{\otimes 4}$$

We experimentally applied this witness method to the Smolin state and obtained the result $\text{Tr}(\mathcal{W}_s \rho_s^{\text{exp}}) = -1.576 \pm 0.016$, where the theoretical value is -2 . This shows that the experimental state ρ_s^{exp}

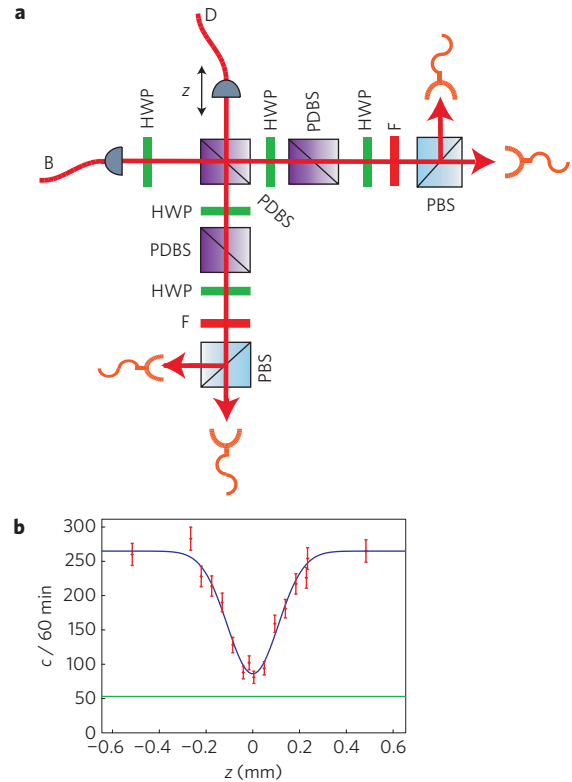


Figure 4 | Experimental set-up for Bell measurement. **a**, Bell measurement set-up for parties B and D. The set-up consists of a linear optical control sign-shift gate. **b**, The interference dip between two vertical-polarized photons from arms B and D needed for the Bell measurement. The visibility of the dip obtained was $V_{\text{exp}} = 67 \pm 2\%$ with a theoretical maximum of $V_t = 0.8$. Errors bars are calculated from Poissonian photon-counting statistics.

is entangled and does not have any cut of the form A|BCD, B|ACD, C|ABD, D|ABC, A|B|CD, C|A|BD and so on.

To further test the entanglement properties, a Clauser–Horne–Shimony–Holt-type Bell inequality of the form

$$|E_{1,1,1,1} + E_{1,1,1,2} + E_{2,2,2,1} - E_{2,2,2,2}| \leq 2 \quad (5)$$

has been used¹⁷. For the Smolin state ρ_s , this inequality is maximally violated when evaluated with the four observables $O_{1(2),1(2),1(2),1(2)} = \sigma_{x(z)} \otimes \sigma_{x(z)} \otimes \sigma_{x(z)} \otimes (1/\sqrt{2})(\sigma_x \pm \sigma_z)$, where $E_{i,j,k,l} = \langle O_{i,j,k,l} \rangle$. These are measured with only two settings for each of the four parties. Experimentally, all observables in the Clauser–Horne–Shimony–Holt inequality (equation (5)) were measured outside the quantum state tomography measurements. We achieved a value of 2.59 ± 0.02 , which corresponds to a violation of 29 standard deviations. We would like to point out that this result also demonstrates the first experimental Bell inequality violation aiming at the saturation of Csirel’son bound (maximal value) by mixed states²³.

From the written form of the Smolin state (equation (1)) it is obvious that if parties A and B come together they can perform a Bell measurement and determine which Bell state is shared by parties C and D. In this way A and B can distill entanglement to parties C and D. Surprisingly, this works for any two parties that join together. Here we experimentally realize the distillation protocol for the Smolin state: when parties B and D come together and make a Bell measurement, the states shared by parties A and C are the Bell states. The distillation protocol is illustrated in Fig. 3. To carry out a complete Bell analyser we used the two-qubit linear optics (with

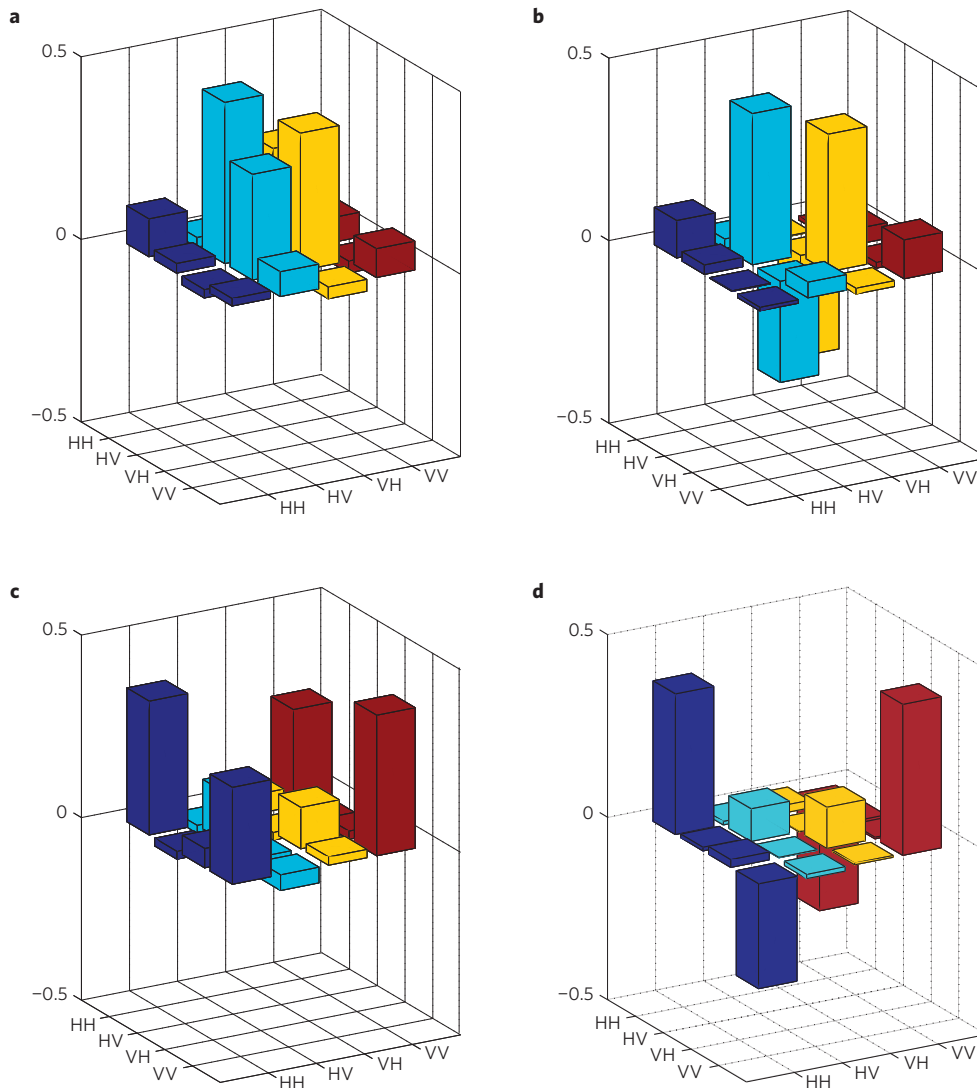


Figure 5 | Experimental results for the entanglement distillation. a–d, Experimental density matrices corresponding to the different Bell projections $|\Psi^+\rangle$ (a), $|\Psi^-\rangle$ (b), $|\Phi^+\rangle$ (c) and $|\Phi^-\rangle$ (d).

$1/9$ probability of success) control sign-shift (C-S-shift) gate²⁴. The experimental set-up for the Bell analyser is shown in Fig. 4. The fourfold coincidence dip in Fig. 4 indicates that the Bell analyser has a success rate of $Q = V_{\text{exp}}/V_i = 84\%$.

In the unlocking entanglement protocol, parties B and D make a joint Bell measurement and broadcast the result (kl) of their measurements through classical communication to parties A and C, who will know which Bell state they are sharing. In our experiment we instead let parties A and C gather all nine local measurements required for the two-qubit quantum-state tomography. By sorting the data that parties A and C have gathered from the four outputs of the Bell analyser (broadcast by parties B and D), four two-photon density matrices can be reconstructed, as shown in Fig. 5. The fidelities of the states sheared between A and C are $F = 0.84 \pm 0.01$, $F = 0.82 \pm 0.01$, $F = 0.80 \pm 0.01$ and $F = 0.83 \pm 0.01$ for the Bell states $|\Psi^+\rangle$, $|\Psi^-\rangle$, $|\Phi^+\rangle$ and $|\Phi^-\rangle$, respectively. To detect whether there is entanglement in the sheared states we used the standard two-qubit witness method and obtained $\text{Tr}(\mathcal{W}_s \rho_{\Psi^+}^{\text{exp}}) = -0.198 \pm 0.017$, $\text{Tr}(\mathcal{W}_s \rho_{\Psi^-}^{\text{exp}}) = -0.172 \pm 0.019$, $\text{Tr}(\mathcal{W}_s \rho_{\Phi^+}^{\text{exp}}) = -0.141 \pm 0.015$ and $\text{Tr}(\mathcal{W}_s \rho_{\Phi^-}^{\text{exp}}) = -0.186 \pm 0.016$, the theoretical value for all these witnesses being -0.5 . These results show clearly that all four states are entangled. In the entanglement distillation protocol, if parties A and C carry out a suitable local

operation, then they can distill a singlet $|\Psi^-\rangle$ with an average fidelity $F = 0.82 \pm 0.01$ and an average expectation value of the witness operator $\text{Tr}(\mathcal{W}_s \rho_{\Psi^-}^{\text{exp}}) = -0.173 \pm 0.010$.

We have for the first time demonstrated a high-fidelity mixed four-qubit polarization bound-entangled state. Using quantum-state tomography we have fully reconstructed its density matrix. We have demonstrated all-Smolin-state-entanglement properties through the violation of the Bell inequality, and the witness method. We have also shown a distillation scheme where any two parties can distill a singlet state for the other two parties. These properties make the Smolin bound-entangled state useful for novel multiparty quantum-communication schemes, for example, secret sharing, communication complexity reduction and remote information concentration. We believe that the results reported here will contribute to deeper understanding of the foundations of quantum mechanics.

Methods

Smolin four-qubit polarization bound-entangled state experimental set-up.

For the experimental observation of the Smolin state, the four-qubit polarization bound-entangled state that is an equal mixture of the product of two of all Bell states (equation (1)), a two-photon-pair emission in four spatial modes (A, B) and (C, D) during a double-pass pump pulse of type-II SPDC is used (Fig. 1). We use ultraviolet femtosecond pulses of a frequency-doubled mode-locked Ti:sapphire laser to pump

a 2 mm type-II BBO crystal at a wavelength of 390 nm with an average pump power of 650 mW. These pulses are reflected back by a small 3-mm-diameter mirror very close to the crystal, pumping the same BBO crystal again for a second SPDC process. The two emitted photon pairs in arms A and B (see Fig. 1), respectively C and D, are, after proper filtering, maximally polarization entangled and of the form $|HV\rangle_{AB(CD)} + e^{i\theta}|VH\rangle_{AB(CD)}$, where θ is the phase difference between horizontal and vertical polarizations due to birefringence in the crystal. This is corrected by placing in each photon path an HWP and 1-mm compensation BBO crystals. Two of the compensation crystals, placed in the path of modes B and D, are mounted on motorized tilting mounts creating single-qubit-phase (PS) gates. By switching between the phases 0 and π we can prepare the Bell $|\Psi^+\rangle$ or $|\Psi^-\rangle$ states. Motorized rotating HWPs are placed in the path of modes A and C, and by switching settings between 0° and 45° a single-polarization-qubit flip gate is realized. All these settings are controlled by random-number generators to guarantee equal probability of preparation of each of the four Bell states. To exactly define the spatial and spectral properties of the emitted four photons, they are coupled into single-mode fibres (SMFs) and passed through 3-nm-width narrowband interference filters (Fs). The polarization measurements are made in each mode by combining a quarter-wave plate (QWP) and HWP with a polarizing beam splitter (PBS). The photons are detected by Si avalanche photodiodes (APDs), and the coincidences are registered with an eight-channel multicoincidence logic.

Distillation experimental set-up. In Fig. 4, the distillation set-up is illustrated for parties B and D. The set-up is based on a linear optical control sign-shift gate (C-S-shift). This two-qubit gate consists of coherent interference of modes B and D at a polarization-dependent polarized beam splitter (PDBS). This PDBS has one and one-third transmission horizontal and vertical polarization photons, respectively. To obtain indistinguishability of photons B and D owing to their arrival times we adjusted the path length of the photon in mode D. In Fig. 4 the fourfold coincidence between the detectors versus the delay path between photons B and D is shown. The input state in both modes was a V polarized photon. The zero delay corresponds to the maximal overlap with a visibility of $V_{\text{exp}} = 67 \pm 2\%$ compared with the theoretical maximal value of $V_t = 80\%$.

Entanglement detection by the witness method. To detect the entanglement we used the so-called entanglement witness. According to this method a quantum state with the density matrix ρ is entangled if and only if there exists a Hermitian operator \mathcal{W} such that $\text{Tr}[\mathcal{W}\rho] < 0$, whereas $\text{Tr}[\mathcal{W}\sigma] \geq 0$ for all separable states σ holds⁴. It has been shown that a witness operator can be optimized²⁵ and decomposed in local projective measurements²⁶. Therefore, the experimental implementation of entanglement witnesses is easy, and we applied this method to detect the entanglement in the state $\rho_{\text{exp}}^{\text{exp}}$. The operators $\sigma_x^{\otimes 4}$, $\sigma_y^{\otimes 4}$, and $\sigma_z^{\otimes 4}$ are common stabilizers for all four products of Bell states $|\Psi^i\rangle \otimes |\Psi^j\rangle$. This implies that they stabilize the Smolin state²². For the derivation we can take any of the four pure states because they all behave in the same way when the stabilizers are used. A criterion for constructing a witness with stabilizers is that the stabilizers have to be non-commuting over all the sets we want to exclude. Since we are excluding separable and tri-separable states it does not matter that $\sigma_x^{\otimes 2}$, $\sigma_y^{\otimes 2}$ and $\sigma_z^{\otimes 2}$ are commuting. The witness is given by $\mathcal{W}_s = \alpha \mathbb{1}^{\otimes 4} - (\sigma_x^{\otimes 4} + \sigma_y^{\otimes 4} + \sigma_z^{\otimes 4})$, where $\alpha = \max_{|\psi\rangle \in P} (\langle \psi | \sigma_x^{\otimes 4} + \sigma_y^{\otimes 4} + \sigma_z^{\otimes 4} | \psi \rangle)$. Here P is the set of all separable and tri-separable states. For the set of separable states it is easy to show with the help of the Cauchy–Schwarz inequality in three dimensions that $\alpha = 1$. For the set of tri-separable states a computer program based on a simple annealing-like search for the maximum²⁷, over the set of states P , was used to obtain $\alpha = 1$. Any product of Bell states $|\Psi^i\rangle \otimes |\Psi^j\rangle$ will give $\alpha = 3$.

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Author contributions

E.A. carried out the experiment. E.A. and M.B. discussed the results and wrote the manuscript. M.B. supervised the project.

Additional information

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