Experimental three-photon quantum nonlocality under strict locality conditions

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Quantum correlations, often observed as violations of Bell inequalities1–5, are critical to our understanding of the quantum world, with far-reaching technological6–9 and fundamental impact. Many tests of Bell inequalities have studied pairs of correlated particles. However, interest in multi-particle quantum correlations is driving the experimental frontier to test larger systems. All violations to date require supplementary assumptions that open results to loopholes, the closing of which is one of the most important challenges in quantum science. Seminal experiments have closed some loopholes10–16, but no experiment has closed locality loopholes with three or more particles. Here, we close both the locality and freedom-of-choice loopholes by distributing three-photon Greenberger–Horne–Zeilinger entangled states17 to independent observers. We measured a violation of Mermin’s inequality18 with parameter 2.77 ± 0.08, violating its classical bound by nine standard deviations. These results are a milestone in multi-party quantum communication19 and a significant advancement of the foundations of quantum mechanics20.

In his breakthrough work, John Bell1 derived upper bounds on the strength of correlations exhibited by local hidden variable (LHV) theories, very general models of nature in which measurement outcomes in one region of space are independent of the events in other space-like separated regions. Quantum mechanical correlations can violate these bounds. Greenberger, Horne and Zeilinger (GHZ) considered a scenario involving three-particle entangled states. Surprisingly, and unlike the case of two-particle Bell inequalities, nonlocality could manifest in perfect correlations of measurement outcomes, enabling an all-or-nothing test of local realism that can be refuted with a single measurement event17,21. The GHZ argument was converted into the form of an inequality by Mermin18, which we experimentally tested.

An ideal Bell inequality experiment requires separating two or more particles by a large distance and making high-efficiency local measurements on those particles using randomly chosen settings, then comparing these results22,23. The first Bell inequality tests were carried out using two-photon cascades in atomic systems3–5. Mermin’s inequality was violated using three-photon entanglement from a parametric downconversion source24. However, these and the many other Bell experiments that followed are subject to one or more loopholes that could, in principle, be exploited to yield a violation even though nature is in fact describable by an LHV model. The ‘detection loophole’ concerns LHV models that take advantage of low detection efficiency25; it has been closed for two-particle Bell inequalities in ion trap and photon experiments10–12 as well as in a six-ion experiment16. Those experiments that do not close this loophole must appeal to the ‘fair-sampling assumption’: properties of the subset of particles that are measured are representative of all the particles. The ‘locality loophole’ (L) exploits configurations where the choice of measurement settings and measurement outcomes at distant locations are not causally disconnected, that is, are not outside each others’ forward and backward light cones. This loophole has been closed for two-particle Bell inequalities in photon experiments13–15. The ‘freedom-of-choice loophole’ (FoC) exploits arrangements where the choice of measurement settings is not causally disconnected from the source. This loophole was recently identified and closed using photons15. A loophole-free Bell inequality test has yet to be performed.

No attempts have been made to close locality loopholes in Bell experiments involving three or more particles24–26. The primary reason for this is source brightness. While entangled photon pairs have been generated and detected at rates in excess of 1 MHz (refs 27,28), entangled photon triplets have only been observed at rates on the order of hertz29,30, necessitating long measurement times. In addition, further experimental challenges include high sensitivity to loss, causality relations requiring a complex experimental set-up and demanding stability requirements.

Here, we have overcome these challenges and report the experimental violation of the three-particle Mermin’s inequality closing both the locality and FoC loopholes, having to make only the fair-sampling assumption. Our experiment utilized a high-fidelity, high-rate source of entangled multi-photon states, high-transmission telescopes, ultra-short latency quantum random number generators and fast-switching polarization analysers, independent and synchronized recording of detection events and the random numbers, a high-bandwidth wireless data transmission network and complex on-line analysis software. Each of these elements operated in concert with the multi-hour stability required to extract conclusive results. With this set-up, we created three-photon polarization-entangled states, separated each photon from a triplet to different, distant locations, and independently measured each photon. The correlations were extracted from measurements of each photon polarization using randomly chosen settings. Our
composite analysis of the space–time arrangement of the experiment establishes independence of the important events.

Mermin considered a scenario where three particles were measured at three stations, which we call Alice, Bob and Charlie. Each particle was measured by a device with two settings, for example, $a$ and $a'$ for the device at Alice, and two outcomes, $+1$ and $-1$. The correlation $E$ in the outcomes for settings $a$, $b$ and $c$ is defined as $E(a, b, c) = P^+ - P^-$, where $P^+$ ($P^-$) is the probability that the product of the outcomes is $+1$ ($-1$). LHV theories must obey Mermin’s inequality:

$$M = |E(a, b, c) - E(a, b', c') - E(a', b', c) - E(a', b', c')| \leq 2 \quad (1)$$

where $M$ is the Mermin parameter.

Mermin’s inequality can be maximally violated using three-particle GHZ states encoded, for example, in photon polarization:

$$|\text{GHZ}\rangle = \frac{1}{\sqrt{2}} (|\text{HHH}\rangle - i|\text{VVV}\rangle) \quad (2)$$

where $|\text{H}\rangle$ and $|\text{V}\rangle$ represent horizontal and vertical polarizations respectively. We define the diagonal/antidiagonal ($D/A$) states as $|D\rangle = \frac{1}{\sqrt{2}} (|\text{H}\rangle + |\text{V}\rangle)$ and $|A\rangle = \frac{1}{\sqrt{2}} (|\text{H}\rangle - |\text{V}\rangle)$ and right/left ($R/L$) states as $|R\rangle = \frac{1}{\sqrt{2}} (|\text{H}\rangle + i|\text{V}\rangle)$ and $|L\rangle = \frac{1}{\sqrt{2}} (|\text{H}\rangle - i|\text{V}\rangle)$. When we measure $|D\rangle$ or $|R\rangle$, we assign the outcome $+1$; when we measure $|A\rangle$ or $|L\rangle$, we assign $-1$. If we measure in the $R/L$ basis for settings

**Figure 1** | Experimental set-up. The downconversion (SPDC) source of triggered three-photon GHZ states and Alice were located in the RAC building. Two of the entangled photons were sent to the roof through optical fibres and transmitted to trailers Bob and Charlie through free-space optical links. Bob and Charlie were 801 m and 721 m away from the source, respectively, and the optical links used to send their photons were 772 m and 686 m long, respectively. Bob and Charlie measured their photon polarizations in one of two bases, determined by co-located QRNGs and fast Pockels cells (PCs), and recorded measurement values using time-tagging electronics. A third trailer, Randy, 446 m from the source, contained a QRNG that sent random bits to Alice over a 425 m RF link determining the setting of Alice’s PC. Randy contained time-tagging electronics to record the output of the QRNG for comparison with Alice’s record. Alice’s photon was delayed in a fibre spool before its polarization was measured and the result recorded using time-tagging electronics. HWP, half-wave plate; QWP, quarter-wave plate; PBS, polarizing beamsplitter; APD, avalanche photodiode; SMF, single-mode fibre.

**Figure 2** | Experimentally measured three-photon polarization correlations. The analysers were set to the $R/L$ basis for settings $a$, $b$ and $c$ and the $D/A$ basis for settings $a'$, $b'$ and $c'$. During a 1 h 19 min experiment, we measured 2,472 fourfold coincidence events, of which 1,232 were used to extract correlations for a test of Mermin’s inequality. The measured correlations were $E(a, b, c) = 0.689 \pm 0.040$, $E(a, b', c') = -0.710 \pm 0.042$, $E(a', b', c) = -0.718 \pm 0.038$ and $E(a', b', c') = -0.655 \pm 0.044$, yielding a Mermin parameter of $2.77 \pm 0.08$, which violates the local hidden-variable bound of 2 by over 9σ. Error bars represent one standard deviation based on Poisson statistics.
Figure 3 | Space-time analysis of the experiment. a, Simplified layout of the experiment showing the straight-line distances and angles between locations. b–e Six two-dimensional space–time diagrams fully describe the relationship between important events in the experiment in the laboratory frame, namely, the entangled photon creation event labelled ‘State creation’, choices of measurement bases and the measurements themselves. Insets: relevant locations for each panel. The earliest and latest times at which Bob’s measurement basis could have been selected are labelled ‘Bob basis early’ and ‘Bob basis late’, respectively. Labelling at Randy and Charlie follows this convention. The event corresponding to the measurement of a photon from the source at Bob is labelled ‘Bob measurement’, and similarly for Alice and Charlie. The light cones for the important events are shown as diagonal lines and shaded regions for the inequality.

Our experimental configuration is shown in Fig. 1. The entangled photon source was located in a laboratory in the Research Advancement Centre (RAC) building. The source produced three-photon entangled states and a fourth trigger photon, which was sent to three measurement stations, Alice, Bob and Charlie. Photons from the entangled state were measured at the source. Photons to Bob and Charlie, located in trailers 801 m west and 721 m northwest of the source, via free-space links. Alice was co-located with the source and her photon was delayed in an optical fibre. The measurement basis choice for Alice was made by a fourth party, Randy, located in a trailer 446 m east of the source. Each receiver measured the polarization in one of two bases, chosen by a fast quantum random number generator (QRNG)31. All single-photon detection events and Randy’s QRNG results were recorded using time-tagging electronics. For more details see Supplementary Information.

To test Mermin’s inequality, we recorded time tags for 1 h 19 min, while the receivers measured either in the R/L or D/A bases. Over this time, the free-space link efficiencies averaged 33% and 32% to Bob and Charlie, respectively, and the long fibre efficiency at Alice was 14%. Once the experiment was completed, we extracted all fourfold events using a 3 ns coincidence window. This yielded 2,472 fourfold coincidence events, corresponding to an average rate of 0.5 Hz. The raw counts for each polarization setting are provided in the Supplementary Section ‘Additional experimental results – Main text Mermin inequality results’.
Figure 2 shows the measured correlations, which give a Mermin parameter of $2.77 \pm 0.08$, where the uncertainty is based on Poissonian count statistics. These results significantly violate Mermin’s inequality by over 9σ. Furthermore, for the measurement settings we used, our violation of Mermin’s inequality shows that our state was genuine tripartite entangled\(^2\).

We performed additional tests of Mermin’s inequality with different phase settings in the GHZ state, and in all cases we measured a significant violation and the correlations depended on the phase as expected. The results are described in the Supplementary Section ‘Additional experimental results-Additional phases’.

The timing and layout of the experiment are both critical for closing the locality and FoC loopholes. The space–time analysis of our set-up includes the locations of the components, measured delays, delays inferred from distance measurements, autocorrelation times of the QRNGs and the asynchronous QRNG sampling. This analysis is summarized in Fig. 3 and fully detailed in the Supplementary Section ‘Space-time analysis’. In Fig. 3a the distances and angles from the source/Alice to Bob, Charlie and Randy are shown. Note that Fig. 1 shows the free-space optical link distances, whereas Fig. 3 shows the straight-line distances between source and stations.

The space–time description of the experiment is inherently $3 + 1$-dimensional; however, we can extract all relevant information from two-dimensional slices, one for each pair of locations (Fig. 3b–e). All times and positions are given in the laboratory frame of reference. For compactness, Fig. 3b and d each depict a pair of two-dimensional slices, but, because the locations are non-collinear, the two halves of the diagram (positive and negative positions) should be interpreted independently.

For example, in Fig. 3b, the GHZ state is produced at the origin and labelled ‘State creation’. The red diagonal lines emanating from the origin define the past and future light cones of the source production event. Those events on or above the future light cone could be causally influenced by the source according to special relativity, while those events on or below the past light cone could causally influence the source. Those events outside the light cones of the source are independent and cannot influence nor be influenced by the source.

Now consider the right half of Fig. 3b, describing events relevant to the Source/Alice and Bob. Bob’s events take place at his position 801 m from the source. The photon arrives at Bob’s Pockels cell (PC) at time $t = 3.032$ ns. We consider the measurement to be complete when the photon detection event is stored in the memory of the time-tagging electronics; adding the optical and electronic delays, the measurement at Bob occurs at $t = 3.079 \pm 24$ ns and is labelled ‘Bob measurement’.

We consider the random number generated in the QRNG when a photon from a light-emitting diode (LED) hits the beamsplitter inside the device. The latest time that the QRNG output could determine the measurement setting for his received photon is 665 ns before the photon passes through the PC (that is, at $t = 2.367$ ns); this is labelled ‘Bob basis late’. This event occurs $304 \pm 25$ ns outside the source light cone, thus satisfying the FoC condition in this case. The earliest time at which the QRNG determines the setting is 1,200 ns earlier at $t = 1.167$ ns, which includes 1,000 ns for asynchronous sampling of the QRNGs, and 200 ns for the QRNG autocorrelation and is labelled ‘Bob basis early’. Alice is co-located with the source at position 0 m and her measurement, labelled ‘Alice measurement’, occurs at $t = 2.926$ ns. This event is $911 \pm 42$ ns outside the light cone (green line) of the earliest basis setting at Bob, satisfying the locality condition here.

We summarize results for the other five slices shown in Fig. 3b–e, with labels defined using the same convention. In all cases, the basis choice is made outside the light cone from the source creation event by at least $304 \pm 25$ ns, ensuring the FoC loophole is closed. The measurement at any location is completed outside the light cone of the earliest possible basis choice at each other location by at least $264 \pm 28$ ns, ensuring the locality loophole is closed.

To test if the observed correlations depend on fast random switching, we performed an additional experiment comparing the case of the measurement bases selection by QRNGs to the case where they were deterministically selected with 500 kHz periodic signals from function generators. Mermin’s inequality was violated in both cases, indicating no significant difference between deterministic and random switching. These results are provided in the Supplementary Section ‘Additional experimental results-Random to deterministic basis choices’.

Our results suggest several interesting avenues for future research, such as extending the set-up to close the locality loopholes for Mermin inequalities with larger numbers of particles, testing more general nonlocal hidden-variable models\(^3\), and studying the improvements in source technology and link efficiency required to perform a loophole-free Mermin inequality. Our set-up yielded a Mermin parameter only slightly reduced from that estimated from quantum state tomography measured directly at the source, demonstrating that multi-photon entanglement was distributed with high fidelity. Our experiment is thus an exciting new platform for multi-party quantum communications protocols, such as quantum secret sharing\(^19\) and third-man quantum cryptography\(^24\).

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References

1. Bell, J. S. On the Einstein Podolsky and Rosen paradox. Physics 1, 195–200 (1964).
2. Clauser, J. F. et al. Proposed experiment to test local hidden-variable theories. Phys. Rev. Lett. 23, 880–884 (1969).
3. Friedman, S. J. & Clauser, J. F. Experimental test of local-hidden variable theories. Phys. Rev. Lett. 28, 938–941 (1972).
4. Fry, E. S. & Thompson, R. C. Experimental test of local hidden-variable theories. Phys. Rev. Lett. 37, 465–468 (1976).
5. Aspect, A., Grangier, P. & Roger, G. Experimental realization of Einstein–Podolsky–Rosen–Bohm gedankenexperiment: a new violation of Bell’s inequalities. Phys. Rev. Lett. 49, 91–94 (1982).
6. Bennett, C. H. & Brassard, G. in Proceedings of the IEEE International Conference on Computers, Systems, and Signal Processing 175–179 (IEEE, 1984).
7. Scarrani, V. et al. The security of practical quantum key distribution. Rev. Mod. Phys. 81, 1301–1350 (2009).
8. Ladd, T. D. et al. Quantum computers. Nature 464, 45–53 (2010).
9. Nielsen, M. A. & Chuang, I. L. Quantum Computation and Quantum Information (Cambridge Univ. Press, 2000).
10. Rowe, M. A. et al. Experimental violation of Bell’s inequality with efficient detection. Nature 409, 791–794 (2001).
11. Giustina, M. et al. Bell violation using entangled photons without the fair sampling assumption. Nature 497, 227–230 (2013).
12. Christensen, B. G. et al. Detection-loophole-free test of quantum nonlocality, and applications. Preprint at http://arxiv.org/abs/1306.5772 (2013).
13. Aspect, A., Dalibard, J. & Roger, G. Experimental test of Bell’s inequalities using time-varying analyzers. Phys. Rev. Lett. 49, 1804–1807 (1982).
14. Weihs, G. et al. Violation of Bell’s inequalities under strict Einstein locality conditions. Phys. Rev. Lett. 81, 5039–5043 (1998).
15. Scheidl, T. et al. Violation of local realism with freedom of choice. Proc. Natl Acad. Sci. USA 107, 19708–19713 (2010).
16. Barrett, J. et al. Demonstration of genuine multipartite entanglement with device-independent witnesses. Nature Phys. 9, 559–562 (2013).
17. Greenberger, D. M., Horne, M. A. & Zeilinger, A. in Bell’s Theorem, Quantum Theory, and Conceptions of the Universe (ed. Kafatos, M.) 73–76 (Kluwer Academic, 1989).
18. Mermin, N. D. Extreme quantum entanglement in a superposition of macroscopically distinct states. Phys. Rev. Lett. 65, 1838–1840 (1990).
19. Hillery, M., Bužek, V. & Berthiaume, A. Quantum secret sharing. Phys. Rev. A 59, 1829–1834 (1999).
20. Pan, J.-W. et al. Multiphoton entanglement and interferometry. Rev. Mod. Phys. 84, 777–838 (2012).
21. Greenberger, D. M. et al. Bell’s theorem without inequalities. Am. J. Phys. 58, 1131–1143 (1990).
22. Bell, J. B. Einstein’ssocks and the nature of reality. J. Phys. Colloq. 42(C2), 41–62 (1981).
23. Pearle, P. M. Hidden-variable example based upon data rejection. Phys. Rev. D 2, 1418–1425 (1970).
24. Pan, J.-W. et al. Experimental test of quantum nonlocality in three-photon Greenberger–Horne–Zeilinger entanglement. Nature 403, 515–519 (2000).
25. Zhao, Z. et al. Experimental violation of local realism by four-photon Greenberger–Horne–Zeilinger entanglement. Phys. Rev. Lett. 91, 180401 (2003).
26. Lavoie, J., Kaltenbaek, R. & Resch, K. J. Experimental violation of Svetlichny’s inequality. New J. Phys. 11, 073051 (2009).
27. Allepuz, J., Jeffrey, E. & Kwiat, P. Phase-compensated ultra-bright source of entangled photons. Opt. Express 13, 8951–8959 (2005).
28. Fedrizzi, A. et al. A wavelength-tunable fiber-coupled source of narrowband entangled photons. Opt. Express 15, 15377–15386 (2007).
29. Hübner, H. et al. Direct generation of photon triplets using cascaded photon-pair sources. Nature 466, 601–603 (2010).
30. Lamas-Linares, A., Howell, J. C. & Bouwmeester, D. Stimulated emission of polarization-entangled photons. Nature 412, 887–890 (2001).
31. Jennewein, T. et al. A fast and compact quantum random number generator. Rev. Sci. Instrum. 71, 1675–1680 (2000).
32. Gühne, O. & Toth, G. Entanglement detection. Phys. Rep. 474, 1–75 (2009).
33. Svetlichny, G. Distinguishing three-body from two-body nonseparability by a Bell-type inequality. Phys. Rev. D 35, 3066–3069 (1987).
34. Zukowski, M., Zeilinger, A., Horne, M. A. & Weinfurter, H. Quest for GHZ states. Acta Phys. Pol. B 93, 187–195 (1998).

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Author contributions
C.E., R.L., G.W., T.J. and K.J.R. conceived the experiment. C.E., E.M.S., K.F., J.L., B.L.H., Z.Y., C.P., J.P.B., R.P., L.R. and N.G. constructed the experiment. B.L.H. and L.K.S. performed the space–time analysis. C.E., E.M.S., K.F., C.P. and J.P.B. collected the data. C.E. analysed the data. All authors contributed to writing the manuscript.

Additional information
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Competing financial interests
The authors declare no competing financial interests.