Observation of nonlocal quantum interference between the origins of a four-photon state in a silicon chip

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Quantum mechanically, multiple particles can jointly be in a coherent superposition of two or more different states at the same time. This property is called quantum entanglement, and gives rise to characteristic nonlocal interference and stays at the heart of quantum information process. Here, rather than interference of different intrinsic properties of particles, we experimentally demonstrated coherent superposition of two different birthplaces of a four-photon state. The quantum state is created in four probabilistic photon-pair sources, two combinations of which can create photon quadruplets. Coherent elimination and revival of distributed 4-photons can be fully controlled by tuning a phase. The stringent coherence requirements are met by using a silicon-based integrated photonic chip that contains four spiral waveguides for producing photon pairs via spontaneous four-wave mixing. The experiment gives rise to peculiar nonlocal phenomena without any obvious involvement of entanglement. Besides several potential applications that exploit the new on-chip technology, it opens up the possibility for fundamental studies on nonlocality with spatially separated locations.

1. INTRODUCTION

In 1994, Herzog, Rarity, Weinfurter and Zeilinger demonstrated a remarkable quantum interference effect [1]. They overlapped the paths of emerging photon pairs from two probabilistic photon-pair sources in such a way that it cannot be distinguished – not even in principle – whether the pair has been created in the first or second source. As a result, the photons are in a coherent superposition of being created in either of the two sources. This generalization of superpositions of properties of photons to the mere origin of their creation has exciting consequences. They now tune a phase between the two crystals, and thereby manipulate the total amount of emerging photon pairs. Imagine for a moment, that we set the phase to \(\pi\), such that the two creation processes destructively interfere. In that case, even if two sources individually would create photon pairs, together they do not. Importantly, in contrast to a quantum eraser, here the photon pair has never been born in the first place. This effect has been called frustrated down-conversion [2,3].

Now, 27 years later, we experimentally demonstrate for the first time the multi-particle generalization of this mind-boggling quantum effect, which was theoretically proposed only 2 years ago via a detour to graph theory [4,5]. In our four-photon experiment, the paths of the photons are overlapped in such a way that there are exactly two different possibilities of how each of the four detectors sees a single photon. Taking advantage of high quality integrated optics at a silicon chip, we align the photon paths in such a way that it cannot be distinguished whether the pairs have been created in the first or second possibility. In that way, the origin of the four photons is in a coherent position and we can observe constructive and destructive interference of 4-photon systems that cannot be observed in the photon pairs themselves.

The multi-particle generalization of frustrated down-conversion has non-trivial consequences. In contrast to the two-photon case, this time the origin of the four-photon state consists of two locations that could be spatially separated. This opens a myriad of exciting foundational experiments, for example, investigating time delays of the interference effects [6]. Furthermore, by introducing the ability to change state properties in a distributed, remote way, it could also conceptually advance quantum technologies that are based on the idea of path identity [7,8]. Examples involve quantum metrology [9,10], quantum imaging [11], quantum microscopy [12,13], photon-pair shaping [14] and generation of complex multiphoton states [15,16]. Furthermore, the effect is the basis of new quantum computing schemes [5].

2. RESULTS

Physical interpretation of the four-photon interference

Let’s first look at the experiment by Herzog et al. [1], depicted in Fig.1a. Each of the two nonlinear crystals can probabilistically create a photon pair. If all properties of the photon pair, including their path, are identical, there is no way to dis-
tistinguish from which crystal a pair was created. In that case, the two-photon state is

\[
|\psi\rangle = g \left( |a, b\rangle + e^{i\varphi} |a, b\rangle \right) = g \left( 1 + e^{i\varphi} \right) |a, b\rangle ,
\]

where \(|a, b\rangle\) stands for a photon in path \(a\) and one in path \(b\), and \(g\) is related with the pair creation probability. For clarity, we ignore the vacuum and higher-order terms.

The probability to detect a photon pair in \(a\) and \(b\) is \(P_{a,b} \approx 2g^2 (1 + \cos(\varphi))\). With \(\varphi = 0\), constructive interference increases the number of photon pairs by a factor of 4 compared to the number of pair generated by a single crystal. In the regime of destructive interference, for \(\varphi = \pi\), the two possibilities for pair generation cancel each other. That means, even though the crystals would produce photons independently when two crystals are set up in this way, no photon pair is created at all.

In our system, we use four photon-pair sources, I-IV and a phase \(\theta\) between the two layers, as shown in Fig.1b. Their paths are overlapped in such a way that detectors A-D only see photon quadruples if the source I & II or III & IV created a photon pair simultaneously. For small photon pair creation rate \(g\), we can write the state as,

\[
|\psi\rangle = g \left( |a, b\rangle + |c, d\rangle + |a, c\rangle e^{i\theta} + |b, d\rangle \right) + \ldots
\]

where \(|a, b\rangle\) stands for a photon in paths \(a\) and \(b\), and \(|a, b, c, d\rangle\) stands for a photon quadruple in the detectors A-D. No other combinations of two simultaneous photon-pair creations create a photon in each of the four detectors (see Methods). By careful design, the two possibilities for photon quadruplets can be created in a coherent superposition. In this way, we create a 4-photon quantum state that originates from either of two locations. Again, by introducing a phase \(\theta\) between the origins, we can observe constructively and destructively interfere of the four-photons creation process. For example, if the phase is \(\theta = \pi\), no four-photon state can be observed in the detectors A-D, while each of the individual crystals continue to create photon pairs.

We would like to argue how our experiment is related to nonlocality in quantum mechanics, in a stronger way than its two-photon analogy [11][17]. There is no nonlocality in the sense of Bell’s inequality or GHZ violation because the final four-photon term is not entangled. Yet, we suggest that our experiment shows nonlocality of a different form. This can be inferred by the fact that changing the phase in one path does not change any single-photon or two-photon count rates from the four crystals, but modulates only the four-fold count rates, theoretically with perfect visibility. This has a surprising consequence, as described in [13]: Imagine for a moment that the crystals III and IV in Fig[1b] are separated by a large distance. This configuration still satisfies all coherence requirements. Now if we modify the phase \(\theta\) (which we assume to be close to crystal IV), the single or two-photon counts in paths \(a\) and \(b\), which are separated by a large distance from the phase-shifter \(\theta\), do not change. If they would change, we could send messages faster than the speed of light. A computation that goes beyond the approximations in eq. (2) of the process confirms that (see SI). If we set the phase \(\theta = \pi\), no four-fold coincidences will emerge because of destructive four-photon interference. If now our detectors C and D both see a photon, we know that there cannot be one photon in each detector A and B simultaneously. That is because otherwise, we would have a four-fold event that does not occur in this setting. Consequently, we can infer that crystal III has not created a photon pair in this situation. Likewise, if \(\theta = 0\) and the detectors in C and D click, crystal III appears to produce four times as much photon pairs as in an incoherent situation. This conclusion is quite surprising because crystal III could be far away from the phase-setting \(\theta\), and it produces photons via an entirely spontaneous process. It appears that we can make conclusions about random processes that could be space-like separated, without entanglement being involved in any obvious way.

**Integrated quantum photonic chip**

We explore this multi-photon interference effect using an integrated silicon photonic chip. Integrated optics is consid-
FIG. 2: Experimental setup. (a) Microscopic photograph of the integrated silicon photonic chip. The total size of the chip is 3.8×0.8 mm². (b) Experimental setup to observe four-photon interference in a silicon photonic chip. An external pump laser is coupled to the chip. There, four spiral waveguide sources produce photon quadruples (Source I and Source II can produce a four-photon state as well as Source III and IV). The two possibilities for creating four photons are made indistinguishable by identifying all of their properties, including the photons’ paths. A relative phase between the two possibilities is introduced via the pump laser in the “Delay line” (at position marked $\theta$). A tunable MZI (MZI2) is used to change from two-photon interference (which is used for alignment and characterization) to four-photon interference. MZI, Mach-Zehnder interferometer; UMZI, unbalanced Mach-Zehnder interferometer; WDMs: wavelength-division multiplexers.

To pave the road for scalable quantum information processing, we fabricated an integrated silicon photonic chip. The device was manufactured in a standard silicon photonics foundry. All the processes required for state preparation, manipulation, and measurement are achieved through thermo-optical phase shifters and Mach-Zehnder interferometers. All phase shifters are controlled by one multichannel DC power supply and are calibrated with one continuous-wave laser.

In our experiment, a 200 GHz bandwidth pulsed laser centered at 1550.11 nm acts as the pump source. It is coupled into the chip using grating couplers and split coherently into two paths using a Mach-Zehnder interferometer configured to act as a 50:50 beam splitter (MZI1). The upper beam is split again at a 50:50 beam splitter (BS1) and acts as the pump for sources I and II, while the lower beam is delayed and subsequently pumps sources III and IV. Each photon-pair source (I-IV) is a 5 mm single-mode spiral silicon waveguide, with a high $\chi^{(3)}$ optical nonlinearity. In these silicon waveguides, two pump photons are annihilated, and signal-idler photon pairs can be generated with a spontaneously four-wave mixing process (SFWM). The signal and idler photons have a wavelength of 1545.32 nm and 1554.94 nm, respectively. After the sources I and II, an unbalanced Mach-Zehnder interferometer (UMZI1) is used to filter out the pump beam. Subsequently, the signal and idler photons in both paths are then separated using another unbalanced Mach-Zehnder interferometer (UMZI2). We estimated that the average error introduced by phase shifters in (U)MZIs was less than 1%. The two-arm length difference of UMZI1 is set to 80 $\mu$m, which shows a free spectral range of approximately 4.8 nm. The interferometer UMZI2 has two arms with a 40 $\mu$m length difference and a free spectral range of approximately 9.6 nm. The extinction ratio of both filters was estimated exceeding 20 dB (see SI for more details). In this way, we can achieve four photons in four paths as $|a⟩|b⟩$ and $|c⟩|d⟩$.

In paths $b$ and $c$, we place an MZI to act as a beam splitter with a tunable splitting ratio (MZI2). We use it for two functions (see inset). First, we can set the splitting ratio to zero, such that the photons remain in their paths. This allows us to observe two-photon frustrated pair-creation, as shown in Fig.1a. We use this to calibrate our chip and confirm the coherent creation of two-photon pairs, as demonstrated by Herzog et al. in bulk optics [1] and Ono et al. on-chip [19]. The second setting of the MZI2 swaps the paths of the photons in
h and c, resembling the configuration in Fig. 1b. After the tunable MZI, we use another pair of UMZI1 to combine signal and idler photons into one path.

The second, lower pump (in the Delay Line region, depicted in violet) is combined with the photons created in sources I and II using two UMZI1s (in the Combination region, depicted in red). The overlap is temporally controlled such that it enters into source III and IV at the same time as the photon pairs created in sources I and II. This is necessary to ensure the coherence criteria, such that it cannot be distinguished whether the photons are created in the sources I and II or in III and IV. Note that in contrast to Fig. 1, we introduce the phases $\varphi$ and $\theta$ into the laser which pumps source III and IV, rather than into the generated single photons. In this way, we can observe multiple oscillation cycles in one phase cycle ($0$ to $2\pi$). It can also be considered that we have changed the phase $\varphi$ or $\theta$ depicted in Fig. 1 with multiple times (details in SI).

The signal and idler photons are subsequently coupled out from the chip to a single-mode fibre array through two end couplers. For each output, we first use a broadband filter to filter out the pump light, and the signal and idler photons are divided into two optical fibres by using wavelength-division multiplexers (WDMs) with a bandwidth of 100 GHz. The signal and idler photons were delivered into four outputs, respectively. The visibility of the coincidence fringe was estimated as 100.0$\pm$5.1% with no background subtraction (the same below). Fig. 3 shows single counts of signal (solid blue line) and idler photons (solid red line). As expected, interference fringe of single-photon counts. Blue: Signal Photons, red: Idler Photons. Partial distinguishability, photon loss, unequal count rates and nonlinear noisy photons from the sources and dark counts in the detectors lead to a decrease in visibility.

**Experimental results**

The full controllability and high stability of the device make it possible for the practical observation of quantum interference between different creation processes, as shown in Fig. 1a and b. To confirm the quality of the device, we first demonstrate the two-photon frustrated pair generation process of Fig. 1a, 1b. We set MZI2 in the chip to the configuration where the photons stay in their path. At this time, our device can be regarded as two separated two-photon nonlinear interferometers, and we can modulate the phase in any of the interferometers. Considering to measure photons corresponding to the term $|c\rangle|d\rangle$, the resulting quantum state up to first order of SFWM is

$$|\psi\rangle_2 = 2|a\rangle|b\rangle + (1 + e^{i\varphi})|c\rangle|d\rangle. \quad (3)$$

Note that the amplitude before term $|a\rangle|b\rangle$ is meaningless. In Fig. 3, we show two-photon constructive and destructive interference results with each data point accumulated for 1 sec. To minimize multiphoton events, only 300 $\mu$W of the pulsed laser was coupled into the chip. Fig. 3 shows the coincidence counts of signal and idler photons by varying $\varphi$. The interference fringes are fitted with $1 + V \sin[\pi(\varphi - \varphi_c)/T]$, where $V$ is the fringe visibility, $\varphi_c$ is the initial phase, and $T$ is the oscillation period. The fringe visibility $V$ is defined as $V = (d_{\text{max}} - d_{\text{min}})/(d_{\text{max}} + d_{\text{min}})$, where $d_{\text{max}}$ and $d_{\text{min}}$ are the maximum and minimum of the fitted data, respectively. The visibility of the coincidence fringe was estimated as 100.0$\pm$5.1% with no background subtraction (the same below). Fig. 3 shows single counts of signal (solid blue line) and idler photons (solid red line). As expected, they also vary depending on $\varphi$ with the same period of the fringe obtained from coincidence counts. Visibilities of them are 43.0$\%$±4.0$\%$ (solid blue line) and 52.3$\%$±3.1$\%$ (solid red line), respectively.

In the final step, we aim to observe the interference depicted in Fig. 1b. To do so, we set the MZI2 in the setting where the incoming photons swap their paths. We get a two-photon state, which is expressed as

$$|\psi\rangle_4 = |a\rangle|b\rangle + e^{i\theta}|c\rangle|d\rangle + e^{i\theta/2}|c\rangle|d\rangle. \quad (4)$$

As we introduce the phase in the pump laser, it emerges in both terms created by crystals III and IV. Correspondingly, the four-photon state is expressed as

$$|\psi\rangle_4 = \left(1 + e^{i\theta}\right)|a\rangle|b\rangle|c\rangle|d\rangle. \quad (5)$$

From these equations, if we change the phase $\theta$, four-photon coincidence counts will increase or decrease, while two-photon coincidence counts will keep unchanged. This is a multiphotonic generalization of two-photon frustrated down-conversion.

We experimentally record four-photon coincidence counts when varying the phase $\theta$, as shown in Fig. 4. To reduce the impact of multiphoton noise, we set the pump power as 980
FIG. 4: Interference of four photons created in two separate locations. (a) The interference fringe has a visibility of 78.3±11.6% which clearly demonstrates that the four photons are indeed generated, with high quality, in two different locations. Here, each data collection took 30 minutes. (b) Fluctuation results of two-photon coincidence counts. As expected, the two-photon counts are nearly constant, and cannot explain the high visibility of the four-photon state, thus demonstrating genuine four-photon interference.

µW. The integral time of each point is 30 min. The four-photon coincidence fringe shows a four-fold oscillation frequency, which is perfectly consistent with the theoretical prediction. The visibility of the four-photon coincidence fringe was estimated as 78.3±11.6%.

Two-photon coincidence counts with linear fitting (ab, ac, bd, cd) are given in Fig.4a, which hardly changed with the phase. The small fluctuation of two-photon coincidence counts is mainly due to the experimental measurement errors, and cannot explain the very large oscillation shown in Fig.1. Besides phases ϕ and θ, we also adjust other phases on the silicon chip and have observed similar frustrated interference phenomenon, which is given in Supplemental Information. These results prove that we observe a new type of nonlocal multi-photon quantum interference, which cannot be understood by the behaviour of local properties such as individual photon pair productions.

3. DISCUSSION AND CONCLUSION

We have shown experimentally that it is possible to create a four-photon quantum state coherently in two separate locations. As a consequence, we were able to observe genuine four-photon interference which does not occur in single-photon or two-photon states from the crystals. This was made possible by a highly stable, low-loss integrated photonics chip with four nonlinear photon-pair sources that are pumped coherently.

We would like to compare and contrast the multi-particle generalizations of the famous Hong-Ou-Mandel (HOM) interference-effect. There, multiple photons are mixed in multiports. If the output path combinations created by the multiport are indistinguishable, the different possibilities interfere\[22\]. This is even true if a subset of the photons is distinguishable \[23\][24\]. This interference effect is at the heart of BosonSampling experiments \[25\]. In contrast to that, rather than superposing intrinsic photonic properties such as the path of the photons, in our experiment, the two origins of the multi-photon state are indistinguishable and therefore in a coherent superposition. This introduces the notion of space in two different ways. First, the two locations of the four-photon creation process can be far apart. Second, the creation of a single four-photon event uses two crystals that can be at a large distance from each other. This opens entirely new experimental ways to study non-locality via multiphoton interference.

Beyond the immediate physical interest, our work also suggests advances in the context of quantum technology applications. First and foremost, for the photonic quantum system continuing to be scaled up, the quality of photonic sources and system losses need to be further improved\[26\][28\]. This involves the high-level control of on-chip photon-pair sources\[21\][26\][29\][31\]. Our experiment demonstrates a new level of control, by coherently creating and overlapping highly-distinguishable photon pairs from different on-chip sources. Thereby it opens the possibility of more complicated on-chip multi-photon interference, which is one requirement for a different type of special-purpose quantum computing scheme\[5\]. Also, destructive multi-photon interference in the exact way as demonstrated here is the basis of several new proposals for the generation of important photonic quantum states. This includes efficient new ways to generate heralded multi-photon and high-dimensional entangled states\[16\]. The new interference effect can readily be studied in connection to quantum metrology\[32\]. Furthermore, numerous proof-of-concepts applications have used the idea of path identity to enhance quantum imaging, microscopy, or spectroscopy\[3\]. The main advantage of these applications is the ability to probe objects with one wavelength for which detects are not available, and detecting the result in a wavelength that can easily be observed\[9\][11\][12\]. The very same advantage can be utilized in applications exploiting the new interference effect.

To conclude, we want to emphasize again that this effect might be best understood as interference between two possible ways to create photon quadruplets, rather than interference of the photons themselves. As the four locations of the photon creations can be spatially separated in a number of ways, variations of this experiment will allow for investigations of...
quantum nonlocality that does not rely in any obvious way on quantum entanglement.

MATERIALS AND METHODS

Theory for photon pair generation. The interaction Hamiltonian for the photon pair generation by SFWM in the silicon waveguide is described as

$$\hat{H}_2 = i\hbar \chi (a^\dagger b^\dagger - ab),$$

where $\hbar$ is the reduced Planck constant, $\chi$ is proportional to the third-order nonlinear susceptibility $\chi^{(3)}$ and the amplitude of the pump, and $a^\dagger (a)\text{ and } b^\dagger (b)$ are the creation (annihilation) operators in paths $a$ and $b$. Therefore, the time evolution of the quantum state is given by

$$|\Psi\rangle = e^{-i\hat{H}/\hbar} |\text{vac}\rangle,$$

where $|\text{vac}\rangle$ is the vacuum state. In Fig. 1(b) of the main text, the interaction Hamiltonian is described as

$$\hat{H}_4 = \frac{1}{2}i\hbar \chi (a^\dagger b^\dagger + c^\dagger d^\dagger + a^\dagger c^\dagger + e^{i\theta} b^\dagger d^\dagger - ab - cd - ac - e^{i\theta} bd),$$

We assume that the pump light is strong and can be treated as a classical oscillator, and leading to the full state in eq.2. The state of four fold term is

$$|\phi\rangle = g^2(|2a, 2b, 0, 0\rangle + |2a, 0, 2c, 0\rangle + e^{i\theta} |0, 2b, 0, 2d\rangle + |0, 0, 2c, 2d\rangle + \sqrt{2}g^2(|2a, b, c, 0\rangle + e^{i\theta} |a, 2b, 0, d\rangle + |a, 0, 2c, d\rangle + e^{i\theta} |b, c, 2d\rangle + g^2(1 + e^{i\theta})|a, b, c, d\rangle).$$

We can see that exactly only one term exists when post-selecting one photon in each path (depicted in red), which stands for four-photon interferences (i.e. its amplitude changes when the phase $\theta$ changes).

Loss. Based on measurements on test structures on the same chip, we estimate losses as: 5.5 dB per grating coupler, 4 dB per end coupler, 2 dB cm$^{-1}$ of spiral waveguide propagation, 0.1 dB per cross and 0.2 dB per 2x2 multimode interferometer BS. All measurements are done at wavelength of 1550.11 nm. The associated channel efficiencies, $\eta_a$ and $\eta_i$ are estimated via $\eta_{\text{eff}} = \frac{C}{N_{\text{co}}}$, where $C$, $N_a$, and $N_i$ are, respectively, the coincidence, signal and idler channel counts. Because of the interference between photonic sources, we set the device as two separate two-photon nonlinear interferometers first and record two-photon interference counts. Then we select the maximum values to estimate channel efficiencies, and they are $\eta_s = -13.2$ dB, $\eta_a = -13.2$ dB, $\eta_c = -14.7$ dB and $\eta_i = -17.7$ dB, respectively.

Acknowledgements. The authors thank Alba Cervera-Lierta for useful comments on the manuscript. M.K. thanks Armin Hochrainer, Mayukh Lahiri, Manuel Erhard and Anton Zeilinger for valuable discussions on the concept of path identity. This work was supported by the National Natural Science Foundation of China (NSFC) (Nos. 61590932, 11774333, 62061160487, 12004373), the Anhui Initiative in Quantum Information Technologies (No. AHY130300), the Strategic Priority Research Program of the Chinese Academy of Sciences (No. XDB24030601), the National Key R & D program (No. 2016YFA0301700), the Postdoctoral Science Foundation of China (No. 2020M671860) and the Fundamental Research Funds for the Central Universities. This work was partially carried out at the USTC Center for Micro and Nanoscale Research and Fabrication. M.K. acknowledges support from the Austrian Science Fund (FWF) through the Erwin Schrödinger fellowship No. J4309.

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**Author contributions:** All authors contributed extensively to the work presented in this paper. L.T.F., M.Z. Y.J.C and D.X.D. prepared the samples. L.T.F., Y.J.C, M.K. and X.F.R. performed the measurements, data analyses and discussions. D.L., G.P.G., G.C.G. and M.K. provided support in experiment and conducted theoretical analysis. L.T.F., M.K. and X.F.R. wrote the manuscript, and M.K. and X.F.R. supervised the project. **Competing interests:** The authors declare no competing financial interests. **Date and materials availability:** Supplementary information is available in the online version of the paper. Correspondence and requests for materials should be addressed to M.K. or X.F.R.