Broadband polarization-tunable photon pairs from an ultrathin nonlinear film

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Entanglement is a crucial feature of quantum systems, essential for various applications of quantum technologies. In photonics, the most demanded are polarization-entangled photons, typically generated via spontaneous parametric down-conversion (SPDC). However, the phase-matching condition limits the operation of bulk SPDC sources. This restriction can be overcome by utilizing nanoscale ‘flat-optics’ sources. Here we demonstrate, for the first time, broadband polarization-entangled photons from an ultrathin nonlinear layer. By changing the pump polarization, we achieve unprecedented tunability of the two-photon polarization state and its degree of entanglement, which is impossible in a single bulk source of photon pairs. Together with the continuous-variable entanglement, polarization entanglement makes nanoscale SPDC sources a promising platform for integrated quantum photonics.

Introduction

There is a pronounced tendency in photonics towards ‘flat’ optics \cite{1}, involving ultrathin films, 2D materials, and metasurfaces. Linear and nonlinear ‘flat’ optical elements are not only compact, integrable and efficient; they are also multifunctional and thus promise to outperform their bulk counterparts \cite{2}. Quantum optics is also on its way to ‘flat’ platforms \cite{3, 4}. The latter are so far mainly used as hosts for single-photon sources \cite{5} and linear converters of quantum light \cite{4}, both in space \cite{6, 7, 8} and in polarization \cite{6, 9}. Meanwhile, ‘flat-optics’ generation of polarization-entangled photons, which are ubiquitous in quantum technology, remains a challenge. Although photon pairs have been generated in ultrathin films through spontaneous four-wave mixing \cite{10} and spontaneous parametric down-conversion (SPDC) \cite{11, 12}, bulk crystals are still used as sources of polarization entanglement \cite{7, 9}. This shortcoming hinders the development of ‘flat’ quantum optics.

At the same time, ultrathin platforms provide unprecedented advantages in engineering photon pairs. Being free from the phase matching constraints, they can be fabricated of any materials, including those with especially large second-order susceptibility $\chi^{(2)}$ to boost the efficiency of pair generation through SPDC. To further enhance the process, one can take advantage of geometric and collective resonances in metasurfaces \cite{13}.

To produce polarization-entangled photons, we use another advantage of ultrathin SPDC sources: having relaxed phase matching, they enable several nonlinear interactions (type-0, type-I, type-II) simultaneously, the only restriction being the structure of the $\chi^{(2)}$ tensor. In lithium niobate, which has been the most efficient ‘flat’ source of SPDC so far, there is little freedom in the choice of nonlinear interactions because only one component of the second-order susceptibility tensor is substantial: $\chi^{(2)}_{zzz}$, leading to type-0 interaction. Accordingly, the pump and the daughter photons should be all polarized linearly along the $z$ direction \cite{11}.

Here we generate photon pairs in an ultrathin layer of gallium phosphide (GaP) whose $\chi^{(2)}$ tensor allows more versatility in SPDC. We find that these pairs feature nontrivial polarization properties, in particular polarization entanglement. Further, we show that by choosing the polarization of the pump we can easily tune the polarization state of the photon pairs, from maximally entangled to almost disentangled, and at the same time maintain the purity of the state. Such a performance is impossible with regular linear polarization elements without losses in the system \cite{9}. Moreover, in combination with the nearly unlimited frequency spectrum of photon pairs emitted from ultrathin materials, polarization entanglement results in a remarkably narrow Hong-Ou-Mandel dip or peak, depending on the experimental conditions, which we also demonstrate in experiment.
Results

Tunable polarization state of the photon pairs

We generate photon pairs via SPDC in a 400 nm-thick film of GaP, pumped by a 60-mW continuous-wave laser at 638 nm (Fig. 1a), whose linear polarization can be rotated by a half-wave plate (HWP). We focus the pump with lens L1, collect the SPDC radiation with lens L2 and filter it from the pump with a long-pass or band-pass filter (LP, BP). Further, non-polarizing beam splitter BS distributes photons between two arms A,B, where superconducting nanowire single-photon detectors (SNSPDs) detect them. A time tagger receives photo-detection pulses from both detectors and builds the histogram of their time differences (Fig. 1b), where the central peak clearly shows the simultaneity of photon arrivals in both arms. Events forming this peak, called ‘two-photon coincidences’, indicate the detections of photon pairs. The background of the histogram shows accidental two-photon coincidences, which are almost entirely caused by the photoluminescence of the GaP film. In all measurements, this background was subtracted.

Due to the ultra-small thickness of the sample, the phase matching is relaxed, and there is no restriction on the types of nonlinear interactions leading to the pair generation. Therefore, possible polarization states of the pump, signal, and idler photons are only defined by the structure of second-order nonlinear tensor $\chi^{(2)}_{ijk}$, $i, j, k = x, y, z$. For GaP, the only nonzero elements of the tensor are $\chi^{(2)}_{xyz} = \chi^{(2)}_{yzx} = \cdots = \chi^{(2)}_{zxy}$ [14]. Because the last index determines the polarization of the pump, we observe photon pairs for both vertically (V) and horizontally (H) polarized pump (Fig. 1b, red and black points). The latter is apparently more efficient, due to the orientation of the film (Fig. 1a, inset).

To analyse the polarization state of the generated pairs, we apply two-photon polarization tomography [15], which is similar to the Stokes measurements on single photons [16] but involves coincidence registration. In this procedure, we independently select different polarization states in each beam splitter output port and measure the rate of coincidences. The polarization selection is performed by a quarter-wave plate (QWP) and a half-wave plate (HWP), followed by a polarizer (P). This combination, further called a polarization analyzer, can filter photons in an arbitrary polarization state. The combination of two such analyzers in two arms filters an arbitrary two-photon polarization state before the beam splitter. A set of nine such measurements enables the complete characterization of the two-photon polarization state (see “Methods”).

Figure 1: The experimental setup. (a) A continuous-wave pump is focused by lens L1 into a thin film of GaP, and photon pairs are collimated by lens L2 and filtered from the pump by long-pass filters LP and band-pass filter BP. Non-polarizing beam splitter BS sends the photons into arms A,B, each containing a quarter-wave plate (QWP) and a half-wave plate (HWP), a polarizer (P), and a superconducting nanowire single-photon detector (SNSPD). A time tagger builds a histogram of coincidences with respect to the arrival time difference (b). The histogram of coincidences is acquired for the horizontally (black) and vertically (red) polarized pump. (c) The delay line consists of 4 calcite plates, 5 mm thick each. The birefringence of calcite introduces a delay between orthogonally polarized photons, which is varied by tilting the plates.
A general two-photon polarization state in a single mode can be written as a superposition of vertically, horizontally, and orthogonally polarized pairs [17],

\[ |\Psi\rangle = C_1 |2\rangle_H |0\rangle_V + C_2 |1\rangle_H |1\rangle_V + C_3 |0\rangle_H |2\rangle_V, \quad \sum_{i=1}^{3} |C_i|^2 = 1, \]

where \(|N⟩_H,V\) are the Fock states with \(N\) photons polarized horizontally or vertically, respectively, and \(C_i\) are the normalized complex amplitudes. For such a state, the degree of polarization entanglement can be calculated through the Schmidt number [18]

\[ K = \frac{2}{2 - |C_1C_3 - C_2|^2}, \quad 1 \leq K \leq 2. \]

The maximal degree of entanglement is achieved for pairs with orthogonally polarized photons, while co-polarized photons have the minimal degree of entanglement.

Two-photon polarization tomography enables the reconstruction of state (1) or, for a mixed state, of the corresponding density matrix. Here we implement this procedure for the states produced with H- and V-polarized pump. In all these measurements, we use a 50 nm band-pass filter.

By varying the pump polarization, we activate different elements of the nonlinear susceptibility tensor and therefore tune the two-photon polarization state. The reconstructed density matrices of photon pairs generated by the H- and V-polarized pump are shown in Fig. 2a, 2b, and 2c, 2d, respectively.

Figure 2: **Two-photon reconstruction and polarization entanglement.** Panels a (c) and b (d) show the real and imaginary parts of the density matrix of the photon pair generated by the H- (V-) polarized pump. (e) The number of coincidences versus the linear polarization angle selected in arm A for the case of V-polarized pump (orthogonally polarized photons). In arm B, we select the basic linearly polarized states: horizontal (H), vertical (V), diagonal (D), anti-diagonal (A). The visibility reaches 96%, pointing towards a high degree of polarization entanglement. The absence of coincidences measured in the A-D basis results from the polarization Hong-Ou-Mandel effect.

For the H-polarized pump, the two-photon polarization state is a superposition of the horizontally and vertically polarized pairs with different weights: \(|C_1|^2 = 0.788 \pm 0.024\), \(|C_2|^2 = 0\), and \(|C_3|^2 = 0.212 \pm 0.017\). Since \(|C_1|^2 \gg |C_3|^2\), two photons within a pair are almost co-polarized horizontally. In contrast, photon pairs generated with the V-polarized pump contain orthogonally polarized photons: \(|C_1|^2 = 0.03 \pm 0.01\), \(|C_2|^2 = 0.97 \pm 0.06\), and \(|C_3|^2 = 0\). These results demonstrate a broad tunability of the two-photon polarization state, not achievable with a single bulk source of photon pairs and requiring a combination of nonlinear optical sources and additional linear elements [19].

Moreover, by changing the polarization of the pump we also tune the degree of polarization entanglement. In the case of an H-polarized pump, Eq. (2) yields for the degree of polarization entanglement \(K = 1.091 \pm 0.009\), meaning that photons within a pair are almost disentangled. For the HV photon pairs, obtained from a V-polarized
pump, the degree of entanglement is maximal, \( K = 1.92 \pm 0.11 \). Notably, this tunability is impossible with standard linear polarization elements, which maintain the degree of entanglement and can modify it only through lossy transformations [9].

**Bell inequality violation.**

To further demonstrate the polarization entanglement of photon pairs generated with the V-polarized pump, we measure the number of coincidences for different orientations of the analyzers. The analyzer in arm B is fixed at one of the basic polarization states, horizontal (H), vertical (V), diagonal (D), or anti-diagonal (A). The analyzer in arm A is then selecting linear polarization, rotated from 0° to 180°. The results are shown in Fig. 2e. A visibility of 96% witnesses strong polarization entanglement.

With these photon pairs, we can now test the Bell inequality in the Clauser-Horne-Shimony-Holt (CHSH) form [20],

\[
F \equiv \frac{1}{2} |\langle ab + a'b + ab' - a'b' \rangle| \leq 1,
\]

where the binary variables \( a, a', b, b' = \pm 1 \) for photons A, B can be chosen as their Stokes operators \( S_1^A, -S_2^A, S^B_1 + S^B_2 \) and \( S^B_1 - S^B_2 \), respectively [21]. By applying this equality to photons A, B in the two output ports of the beam splitter and performing the simultaneous measurement of their Stokes observables, we obtain \( F = 1.36 \pm 0.07 \), which violates the Bell inequality by five standard deviations.

Remarkably, generation of polarization-entangled pairs with high purity does not require any additional elements erasing the distinguishability between pairs of different polarization states, as is the case in bulk phase-matched sources [22, 23]. For orthogonally polarized photons, we obtain the value of the purity \( Tr(r^2) = 1.0 \pm 0.1 \). The indistinguishability is achieved due to the isotropic linear optical properties of GaP. A similar feature is expected in other nonlinear crystals with zinc-blende structures like GaAs or InAs [14].

**Two-photon spectrum**

Another remarkable property of photon pairs generated from ultrathin nonlinear layers is their extremely broad frequency spectrum, leading to a huge degree of time-frequency entanglement [12]. Here, we use this feature in combination with the polarization entanglement. We measure the spectrum of photon pairs using two-photon fiber spectroscopy, where the photons are transmitted through a group-velocity dispersion (GVD) material, such as optical fiber, and the wavelengths of the photons of the same pair are found from their temporal delay acquired in the fiber [24, 12]. To measure the two-photon spectrum, we remove the band-pass filter, leaving only long-pass filters with the cut-off wavelength at 1100 nm for the suppression of the pump and the photoluminescence, and place a dispersive fiber into the path of only one photon (Fig. 1a). Since the zero-dispersion wavelength of the fiber is red-shifted from the degenerate wavelength but is still within the SPDC spectral width, we select only blue-shifted photons in the fiber path using a short-pass filter, and mirror the second half of the spectrum, assuming the symmetry of the two-photon spectral distribution [25].

The obtained spectrum of the photon pairs is shown in Fig. 3a. Its width, here \( \approx 20 \text{ THz} \), is limited by the detection efficiency and the transmissivity of the optical fiber. It could be further increased by utilizing more broadband detectors and optical elements.

**Hong-Ou-Mandel effect**

As we see in Fig. 2e, ‘HV’ photon pairs generated by the V-polarized pump result in no coincidences if the polarization analyzers select diagonal (D) and anti-diagonal (A) polarization states. This is the polarization version of the Hong-Ou-Mandel (HOM) effect [27, 28] and another manifestation of polarization entanglement. Destructive quantum interference between the photons within a pair leads to the absence of the coincidences when the D-A polarization state is detected. In contrast, when the analyzers are both oriented diagonally or anti-diagonally, we observe the maximum of the coincidences due to the constructive interference.

By introducing a time delay between the photons of a pair, one observes a dip in the case of destructive interference and a peak in the case of constructive interference. The width of the dip or peak is determined by the spectrum of the photon pairs [26]. In our case of an ultrathin SPDC source, the broad spectrum leads to ultra-narrow correlations in time.

In the experiment, we introduce a variable time delay between orthogonally polarized photons before the non-polarizing beam splitter by means of a 5 mm birefringent crystal (calcite) plate. We vary the time delay by tilting the plate around its optic axis. For preventing the beam displacement, another calcite plate of the same thickness
Figure 3: Broadband polarization entanglement. (a) Spectrum of the photon pairs, with the width mainly limited by the detection efficiency and the fiber transmissivity. (b) The HOM dip (blue) and peak (red), measured (points) and calculated based on the Fourier transform of the spectrum in panel (a) (solid lines). The dashed lines show the Gaussian fit of the experimental data. The experimental width of the dip (peak) is $15 \pm 2$ fs ($12 \pm 2$ fs), which is much smaller than that predicted by the measured spectrum ($\approx 22$ fs).

is tilted symmetrically. Two additional calcite plates introduce a fixed time delay of the opposite sign, so that the total delay is scanned around zero (Fig. 1c).

The results of the HOM experiment are shown in Fig. 3b. The blue (red) triangles show the number of coincidences versus the delay between photons for the A-D (D-D) selected polarization state. The solid curves are theoretical dependences calculated using the spectrum in Fig. 3a [26]. As expected, and in accordance with Fig. 2e, we see a dip of coincidences at zero delay when the analyzers select orthogonal polarization states, A-D, and a peak when they select the same polarization states, D-D. At large time delays, quantum interference disappears because single-photon wavepackets do not overlap in time. The widths of the dip and the peak are $15 \pm 2$ fs and $12 \pm 2$ fs, respectively, which is much smaller than the width predicted from the spectrum in Fig. 3a ($\approx 22$ fs). This is because the measured two-photon spectrum is additionally narrowed by the spectral losses in the dispersive fiber used for two-photon spectroscopy.

Discussion

We see that the relaxed phase matching for SPDC in an ultrathin nonlinear film leads to the unprecedented polarization tuneability of the produced photon pairs. By changing the pump polarization, we utilize different components of the $\chi^{(2)}$ tensor, drastically tune the polarization state of the photon pairs, and change their degree of polarization entanglement. Such tuneability provides enormous freedom in photon pair polarization engineering, impossible in conventional systems without introducing losses. With such a system, one can change the degree of polarization entanglement on demand in a very easy way, which is useful for various applications of quantum technologies [29, 30].

We verify the high degree of polarization entanglement of one of the states through the experimental violation of the Bell inequality by 5 standard deviations. This is the first observation of polarization-entangled photons from ultrathin nonlinear films. The polarization-entangled state is of high purity and its preparation does not require linear optical polarization compensators, unavoidable with bulk SPDC sources.

Polarization entanglement comes in combination with an ultrabroad frequency spectrum, an extremely high degree of time/frequency entanglement [11] and ultra-narrow time correlations. Accordingly, we observe very narrow ($12 - 15$ fs) polarization HOM dip and peak for polarization-entangled photon pairs.

The combination of polarization with other degrees of freedom enables achieving hyperentanglement. So far, SPDC in ultrathin films provided photon pairs with huge entanglement in frequency [11] and momentum [31]. Now, we complement this set with the polarization degree of freedom. Further extension can be done by considering entanglement in orbital-angular momentum (OAM) of photons [32, 33, 34].
Despite the success with subwavelength SPDC sources, reducing their thickness further is still a challenge. Monoatomic layers and surfaces are now in the focus of attention of nonlinear optics [35, 36, 37], due to their potentially giant nonlinearities, modified density of states, and the possibility to relatively simply combine them with bulk sources or metasurfaces. Surface SPDC [38] and SPDC in a monoatomic layer [39] have been indeed considered theoretically but no convincing evidence of pair generation from such sources exists so far.

Since the interaction volume in flat sources of photon pairs is significantly reduced, the price for the fascinating qualities of generated photon pairs is the relatively low conversion efficiency. The solution might be to use nano-structured materials to either increase the generation rate using geometric resonances [13] or to create giant nonlinearity [40]. Tailoring nonlinearity is also attractive for achieving a rich structure of $\chi^{(2)}$ and even a wider freedom in two-photon polarization engineering.

Methods

Sample. As a source of photon pairs, we use a 400 nm layer of GaP, which is a semiconductor with a zinc-blende symmetry. Its non-zero $\chi^{(2)}$ components are $\chi_{xyz} = \chi_{xzy} = \chi_{yzx} = \chi_{zxy} = \chi_{zyx}$ [14]. To get access to these components, the sample was grown with the crystal axis orientation as shown in the inset of Fig. 1a.

Photon pair generation and detection. The pump (60 mW continuous-wave diode laser centered at 638 nm) is focused into the sample with lens L1 (Fig. 1a), covering NA=0.05. Another lens (L2, NA= 0.16) collects the generated photon pairs. We cut off the pump with a set of long-pass filters (LP) and, for polarization measurements, select frequency-degenerate photon pairs with a band-pass filter (BP) centered at 1275 nm with 50 nm full width at half maximum. A non-polarizing beam splitter (BS) splits photon pairs into arms A and B, each of them containing a quarter-wave plate (QWP), a half-wave plate (HWP), a polarizer (P), and a superconducting nanowire single-photon detector (SNSPD). A time tagger builds a histogram of arrival time differences for the two detectors, examples being shown in Fig. 1b for the pump polarized horizontally (black) and vertically (red).

Two-photon polarization reconstruction. To reconstruct the single-mode two-photon polarization state, we use polarization tomography of single-mode photon pairs [41]. Since the density matrix of the two-photon polarization state has a dimension of $n = 3$, $n^2 = 9$ measurements (including one extra measurement for normalization) of coincidence count rate are required to fully reconstruct the state and verify its purity. The full protocol of the measurements for nine combinations of the analyzer settings in arms A,B can be found in Supplementary. In the more general case of photon pairs generated in two spatial modes, the dimension of the reduced density matrix is 4, and, as a consequence, 16 measurements are required for quantum tomography [19].

Two-photon fiber spectroscopy. The spectrum shown in Fig. 3a is measured using two-photon fiber spectroscopy [12, 24], where a 1 km long SMF-28 fiber in one channel (the inset in Fig. 1a) introduces a delay between the arrivals of the two photons, depending on their frequency separation according to the group-velocity dispersion. The correlation peak is broadened and, after the calibration with different spectral filters, its shape can be mapped to the two-photon spectrum.

Since the zero-dispersion wavelength of the fiber is around 1330 nm, there is no one-to-one correspondence between the photon wavelength and the delay over the whole spectral width of photon pairs. Thus, we perform the measurement of the blue-shifted part of the spectrum and mirror the red-shifted part, assuming that the two-photon spectrum is symmetric in frequency.

Time delays in Hong-Ou-Mandel effect. To introduce the time delay between orthogonally polarized photons, we use four 5 mm thick calcite crystal plates (Fig. 1c), with the optic axes vertical for two and horizontal for the other two plates. Tilting the plates around their optic axes changes their effective thickness. Due to the birefringence of calcite, the optical path for the horizontally and vertically polarized photons is different, which leads to the delay between them depending on the tilt angle of a plate. The zero delay is achieved when two orthogonally oriented crystal plates are tilted by the same angle. To compensate for the transverse displacement of the beam, we put a mirrored scheme with another two plates. By rotating the inner or the outer pair of plates we are able to achieve either negative or positive delay between two orthogonally polarized photons.

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