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Frequency-bin entanglement of ultra-narrow band non-degenerate photon pairs

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Abstract
We demonstrate frequency-bin entanglement between ultra-narrowband photons generated by cavity enhanced spontaneous parametric down conversion. Our source generates photon pairs in widely non-degenerate discrete frequency modes, with one photon resonant with a quantum memory material based on praseodymium doped crystals and the other photon at telecom wavelengths. Correlations between the frequency modes are analyzed using phase modulators and narrowband filters before detection. We show high-visibility two photon interference between the frequency modes, allowing us to infer a coherent superposition of the modes. We develop a model describing the state that we create and use it to estimate optimal measurements to achieve a violation of the Clauser–Horne (CH) Bell inequality under realistic assumptions. With these settings we perform a Bell test and show a significant violation of the CH inequality, thus proving the entanglement of the photons. Finally we demonstrate the compatibility with a quantum memory material by using a spectral hole in the praseodymium (Pr) doped crystal as spectral filter for measuring high-visibility two-photon interference. This demonstrates the feasibility of combining frequency-bin entangled photon pairs with Pr-based solid state quantum memories.

1. Introduction

Photonic entanglement plays an important role in quantum information science, allowing the distribution of quantum correlations over long distances. It is therefore a crucial resource for several applications including quantum key distribution and quantum teleportation [1]. Photonic entanglement has been demonstrated with several processes, the most common being spontaneous parametric down conversion (SPDC). Various types of encoding have been investigated, including polarization, energy-time, time-bin and path encoding [2]. In recent years, several experiments have also investigated entanglement between frequency modes of the photons, so called frequency-bin entanglement. It is well known that the sum of the frequencies of twin photons produced in an SPDC process equals the pump photon frequency. This leads to a continuous variable entanglement between the frequency modes of the two photons. This effect and its consequences have been extensively studied (see, e.g. [3–5]). One can also define discrete frequency bins in the spectra of the SPDC photons where one bin of the signal is correlated with the corresponding bin from the idler photon. In this context the entanglement of the photons in the frequency-bin degree of freedom has been shown [6], and several related works have been reported [7–13].

For applications towards quantum networks [14] and long distance quantum communications using quantum repeaters [15], photons should interact strongly with material systems able to store their quantum states, i.e. photonic quantum memories [16–18]. In most cases, this requires the generation of narrowband photon pairs. Significant progress has been realized in recent years in the generation of narrow-band, memory compatible photon pairs using SPDC in nonlinear crystals [19–24] or Raman processes in cold atomic systems.
or rare-earth doped crystals [26–28]. Demonstrations of entanglement between narrow-band photon pairs have also been reported [19, 25, 29, 30]. Time-bin or energy-time encoding are well suited for long distance fiber transmission. The analysis of such type of entanglement is usually done with the help of interferometers in a configuration introduced by Franson [31]. A technical obstacle for narrowband photons with a long coherence time is that interferometers with very long path lengths are necessary.

In this paper, we investigate the entanglement of narrow-band quantum-memory-compatible photon pairs created by SPDC in the frequency domain. Our work is inspired by the previous works of Olislager and coworkers [6, 8]. In these works photon pairs with broad spectra were generated and the frequency-bins were defined by tunable filters. In contrast, we developed an SPDC source based on an optical parametric oscillator (OPO) operated below its oscillation threshold [22, 24]. Our source is highly non-degenerate, with one photon at 606 nm resonant with Pr doped crystals, while the other photon is at the telecommunication wavelength of 1436 nm. The double resonant feedback in the OPO implies that the photons can only be created resonant to a few modes of the cavity. Hence in our case the frequency-bins are generated intrinsically, which, to our knowledge, was not the case in previous demonstrations.

The entanglement between the optical modes is certified through a violation of the Clauser–Horne (CH) Bell inequality [32]. Our measurements are implemented by electro-optic modulators (EOMs) to mix the frequency bins and spectral filters to select particular frequency modes. We demonstrate high visibility two-photon interference showing coherent superposition of the frequency modes. We develop a theoretical model using the photon spectrum as input that reproduces very well our measured data. Contrary to previous demonstration of frequency-bin entanglement [6, 8], our photons exhibit a spectrum with a small number of frequency modes with different amplitude, leading to a non-maximally high-dimensional entangled state. This prevented us to use some of the assumptions done in [6]. We thus use our model to infer the best setting within our implementation to violate the inequality and show experimentally a significant violation.

We stress that the CH inequality is used here as an entanglement witness, not as a proper nonlocality test, which would require more demanding experimental conditions such as space-like separation and high detection efficiency. However, these conditions are never needed when measuring entanglement witnesses. The advantage of using a Bell inequality for entanglement detection instead of a standard entanglement witness is that it invokes fewer assumptions to certify entanglement, namely a tensor product structure and the fair-sampling assumption, and it is dimension independent. Under these assumptions, entanglement can be detected without any further modeling of the state and measurements.

In the context of storing frequency-bin entanglement in quantum memories, rare-earth doped crystals are well suited because of their large inhomogeneous broadening of the optical transition that could be used to create frequency multiplexed quantum memories [33, 34]. Our source is compatible with praseodymium doped crystals, which have demonstrated excellent properties as quantum memories, including long storage times [35], high-efficiency [36], and spin-wave storage of single photons [37] and of time-bin qubits at the single photon level [38].

The paper is organized as follows. In section 2 we describe our theoretical model. In section 3, we describe the experimental setup and the experiments performed, including the demonstration of high-visibility two-photon interference and the violation of the Bell inequality. In section 4 we describe the two-photon interference experiments using a Pr doped crystal as spectral filter, with the goal of showing that our source is compatible with a quantum memory material. Finally, in section 5 we give a conclusion and an outlook for future experiments.

2. Model of frequency-bin entanglement

We report on the entanglement properties of pairs of photons generated in a cavity enhanced SPDC. In this process, a pump photon of frequency $\omega_p$ enters an optical cavity where the non-linear interaction with a $\chi^{(2)}$ crystal generates pairs of signal ($\omega_s$) and idler ($\omega_i$) photons satisfying energy conservation $\omega_p = \omega_s + \omega_i$. The bi-photon state can be described (to first order) as

$$\Psi = \int d\omega_s \int d\omega_i f_s(\omega_s, \omega_i) |\omega_s\rangle |\omega_i\rangle.$$  \hspace{1cm} (1)

We focus on the case where both, the signal and idler photons are resonant with the cavity but are not degenerate. The joint spectral amplitude of the cavity enhanced emission is $f_s(\omega_s, \omega_i)$, consisting of a sequence of equally high peaks of width $\delta \omega$, separated by $\Delta_{\text{FSR}}$, the cavity free spectral range (FSR). This peaked structure is modulated by the joint spectral amplitude of SPDC in the case without cavity. The explicit form of $f_s(\omega_s, \omega_i)$ is derived in [40].

As we will see in the next sections, our experimental parameters are chosen such that the widths of the peaks in the spectrum, $\delta \omega$, are two orders of magnitude smaller than the FSR, $\Delta_{\text{FSR}}$, which makes each peak perfectly
The action of the EOM can be described by the unitary \( U \). Consequently, we can rewrite equation (1) as

\[
|\Psi\rangle = \sum_n f_n |n\rangle |n\rangle.
\] (2)

The state of equation (2) is naturally frequency-bin entangled. In order to reveal this entanglement we need to manipulate and measure the frequency modes, which we do using EOMs and narrow-band frequency filters. EOMs are optical devices signal-controlled by a radio frequency (RF) \( \Omega \) of amplitude \( c \) and phase \( \gamma \). When applied to a monochromatic frequency state \( |\omega\rangle \) the action of the EOM can be described by the unitary transformation

\[
U(c, \gamma)|\omega\rangle = \sum_n U_n(c, \gamma)|\omega + n\Omega\rangle,
\] (3)

where \( U_n(c, \gamma) = J_n(c) e^{i\gamma - \frac{\pi}{2}} \) and \( J_n(c) \) are the \( n \)th order Bessel functions of the first kind. Then the effect of the EOM is to create a superposition between states which are displaced by integer multiples of \( \Omega \) in the frequency space. By making \( \Omega \) equal to the FSR one can use the EOM to implement a unitary transformation \( U \) on the discrete basis \( \{|n\rangle\} \).

The measurement of frequency entanglement with EOMs is analogous to the measurement of high-dimensional energy-time or time-bin entanglement using unbalanced interferometers in the Franson scheme \([31, 39]\). The EOMs may be seen as the equivalent of multipath interferometers, with different arm lengths and transmission. An important difference is that, in principle, the EOMs add an in

\[
[\{f(n)\}] \text{ is negligible. Hence we make the}
\]

In our experiment we are interested in measuring coincidences between signal and idler photon of a pair after they each passed an EOM and a spectral filter selecting a single frequency-bin. The state of the pair before filtering can thus be written as:

\[
|\Psi\rangle = U_0 \otimes |\Psi\rangle = \sum_n \sum_k \sum_m U_k(c_s, \alpha) U_m(c_i, \beta) f_n |n + k| - n + m\rangle,
\]

\[
U_k(c_s, \alpha) U_m(c_i, \beta) = J_k(c_s) J_m(c_i) \exp \left\{ i(k\alpha + m\beta - \pi/2(k + m)) \right\}.
\] (4)

If the spectral filters select the frequency bins \(|a\rangle\) and \(|b\rangle\), we can define the mode number separation \(d = a - b\). The coincidence probability can then be calculated by:

\[
P_d(c_s, \alpha, c_i, \beta; l) = \langle \langle -l + d | \Psi \rangle \rangle^2,
\]

\[
= \sum_n \sum_k \sum_m U_k(c_s, \alpha) U_m(c_i, \beta) f_n |n + k| - n + m\rangle
\]

which, due to orthogonality and normalization of the modes, results in

\[
P_d(c_s, \alpha, c_i, \beta; l) = \left| \sum_n \sum_k f_n |n + k| - n + m\rangle \langle -l + d| - n - k + d\rangle \right|^2.
\] (6)

In general, EOMs in conjunction with spectral filters can be used to implement a variety of measurement settings, in this case projectors

\[
\Pi_a(c_s, \gamma) = \Pi_a(c_s, \gamma)|n\rangle \langle n| \ U^*(c_s, \gamma),
\] (7)

from where one obtains the outcome probabilities

\[
P(ab|xy) = \text{Tr}[\Pi'_a(x)| \otimes \Pi'_b(y)| \rho],
\] (8)

where we abbreviate the notation by using \( x(y) \) for the amplitude and phase of EOM, corresponding to the measurement choices, and \( a(b) \) for the measurement outcomes on signal (idler) mode.

In an experiment with photons, the probability outcome is well approximated by photon coincidence counts \( C(ab|xy) \) normalized by the total number of events \( \sum_{ab} C(ab|xy) \), that is

\[
P(ab|xy) = \frac{C(ab|xy)}{\sum_{ab} C(ab|xy)}.
\] (9)

In practice we do not have access to all modes \(|a, b\rangle\). In our experiments we can select only the central mode in the signal arm and only three modes in the idler, i.e. the central and the next neighboring modes, as we have no reference laser available for actively stabilizing the filter frequency elsewhere. However, as described by the Bessel functions, for small modulation amplitude, the effect of the EOM for \( |d| > 1 \) is negligible. Hence we make the
Ca b x yab a b

\[ \sum_{a,b} C(ab|xy) \approx \sum_{|a-b| \leq 1} P(a, b|xy) \approx 1. \]

For stronger modulation amplitudes \( c_a, c_b \), this assumption still holds for a range of phases around \( |\alpha - \beta| \approx \pi \), as seen in figure 1(b). In the following section we use our model to provide an estimation about the error that this assumption for the experiment introduces.

Finally, by combining probabilities as we described above we can compute the CH inequality [32]

\[
\begin{align*}
\sum_{a,b} P(00|x_0y_0) + P(00|x_0y_1) + P(00|x_1y_0) - P(00|x_1y_1) - P(0|x_0) - P(0|y_0) \leq 0,
\end{align*}
\]

where \( x_i \) and \( y_j \) refer to the different measurement choices. We identify the marginal probabilities with \( P_B a x y = \sum_{b} P(a, b|x,y) \) and \( P_A b x y = \sum_{a} P(0, b|x,y) \). However, since we can only detect in the central mode in the idler arm, we make the assumption that \( P(0|x_0) = kP(0|y_0) \), where \( k \) is a constant. Assuming symmetric spectra for the signal and idler photons (which is justified by energy conservation) before the EOMs and equal modulation indices in the signal and idler EOM, we expect \( k = 1 \). However, in practice small variations of the modulation indices may arise. To take this effect into account, we use the model described above to estimate \( k \) with our experimental parameters (see section 3.2).

Since the normalization factor is common to all the involved probabilities, we can rewrite the inequality in terms only of number of coincidences.

\[
\begin{align*}
C(00|x_0y_0) + C(00|x_0y_1) + C(00|x_1y_0) - C(00|x_1y_1) - C(0|x_0) - C(0|y_0) \leq 0,
\end{align*}
\]

or

\[
\begin{align*}
S = 2 \frac{C(00|x_0y_0) + C(00|x_0y_1) + C(00|x_1y_0) - C(00|x_1y_1)}{C(0|A)(1 + k)} \leq 2.
\end{align*}
\]

Using a Matlab code we searched for the settings to achieve a high interference contrast or a significant violation of the Bell inequality (10). Figure 1(b) shows an example of such a simulation. As input we use the spectral distribution of the modes (figure 1(a)) that is specific for our system. The other input parameters are the modulation indices for signal and idler and a phase offset for idler \( (\alpha_i = 0) \). We keep one mode \( (|0_i) \) fixed as reference for heralding. The result is the phase dependent distribution of coincidences for each mode under consideration. The normalization is realized by building the sum over mode separations \( |d| \leq 4 \).

3. Experiment

We generate photon pairs via SPDC in an OPO. The details of the device can be found elsewhere [22, 24]. We use a narrowband pump laser at 426 nm to generate idler photons at 1436 nm (telecommunications E-band) and signal photons at 606 nm. The source design allows us to generate signal photons compatible with solid state quantum memories based on praseodymium doped crystals [37, 41]. The spectrum of the photons consists of several peaks, formed by longitudinal modes of the OPO cavity (see figure 1(a) and [24]). Hence, these modes are separated by the cavity FSR, which we measured as \( \Delta_{FSR} = 423.66(6) \text{ MHz} \). The measured biphoton bandwidth
is 2.8 \ MHz. Our source setup further contains tunable Fabry–Pérot cavities (FC) which we use to select a single mode of the spectrum. The lock of the SPDC cavity and the FP filter cavities, also explained in details in [22, 24], works as follows. We stabilize the length of the bow-tie cavity to a classical signal that we derive from the same 606 nm laser used to operate the memory employing the Pound Drever Hall technique. This reference signal, overlapping in the PPLN crystal with the pump laser light at 426 nm, gives rise to a difference frequency generated (DFG) beam at 1436nm. We send a feedback to the pump laser such that the transmission of this DFG signal through the SPDC cavity is maximized. Because of the wide non-degeneracy of the source and the double resonant feedback (see [22, 24]) only the photons resonant to a few cavity modes can exit the cavity. The exact number is determined by the dispersion of the PPLN crystal and the FSR of the cavity. In total we can identify up to eight modes contributing to the spectrum with different intensities. The FC filters in the signal and idler arms are actively stabilized by maximizing the transmission of the above-mentioned classical beams at 606 nm and 1436nm, respectively, using a microcontroller locking system similar to [42] which acts on piezo-electric actuators at the cavity mirrors. In the signal arm additional filtering is provided by an etalon whose role is to suppress the side clusters of modes lying outside of the operating range of the FC filter. Fulfilling this preconditions our source is well suited to demonstrate frequency-bin entanglement.

To analyze the frequency-bin entanglement we include EOMs in our setup as illustrated in figure 2. Each EOM is placed behind the source, before any spectral filter. We use a voltage controlled oscillator, tuned to 423.7 MHz (\(\approx\Delta_{\text{FSR}}\)) as master oscillator. We split its RF signal in two parts and feed it to the following components which are identical for the signal and idler branch. The further path for the RF signal contains a phase shifter, a variable attenuator and a high power amplifier to drive the EOM. Hence we can set the phase and power for each EOM individually, preserving coherence between the two EOMs. The effect of the EOMs on the optical spectrum is the creation of sidebands, as can be seen in the insets of figure 2, where we send a single mode, continuous wave laser beam through the EOMs and measure its spectrum by scanning the length of the filter cavities. It is worth mentioning that the widths of the peaks in this figure are limited by the resolution of the FCs, which is different for signal and idler. From the ratio of the peak height, we infer a modulation index \(c_i = 1.36\) for idler and \(c_i = 1.30\) for signal. The modulation index can be adjusted by the RF power sent to the EOMs. We found a linear relation between the modulation index and the RF amplitude. We use measurements of the RF amplitude at each data point to keep the modulation index constant.

The stabilization procedure of the FC limits our access to the modes during the measurements. The cavity for idler can be stabilized to the modes \(|n_i\rangle = |1i\rangle, |0i\rangle, |1i\rangle\), while in the signal arm we can only select the central mode \(|n_s\rangle = |0s\rangle\). In contrast to our model or simulation, for the normalization of the coincidence rates we can only consider these modes. Hence we assume that higher mode separation can be neglected if the modulation is chosen low enough. This effect is illustrated in figure 3(a). The black solid line is the sum of all modes resulting from the simulation, while for the dashed black line the summation was restricted to the three central modes (as in the experiment). The gray dots in that figure are the sums of the experimental data points and overlap well with the dashed line. At phases around the maximum for \(d = 0\) the coincidence rate for \(|d| > 1\) is negligible, which can be seen in the good overlap between the black lines. We chose four data points in this

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4 One may wonder if the fact that the RF signal is distributed between the two parts could lead to a loophole in the CH test. However, this is not the case. First notice that the RF signal can just be seen as being part of the state prepared by the source. If the EOMs and spectral filters, as well as the RF phase shifters, are close to the detectors, a local choice of the settings is ensured. Second, as we know that the RF signal does not constitute any source of entanglement, any violation of the CH inequality must come from the entangled state produced by the SPDC source.
The solid lines show the results from our simulations. The cyan colored lines are simulations for higher order modes, not accessible in the experiment. The dashed blue (magenta) line corresponds to the solid blue (magenta) simulation for the case $|0, l\rangle$ (top) and $|+1, l\rangle$, normalized by the three central modes only, as in the experiment. (b) Shows the correlation histograms for minimum (bottom, $\beta = 178^\circ$, $\alpha = 51^\circ$) and maximum (top, $\beta = -29^\circ$, $\alpha = 51^\circ$). To determine the coincidence rate we integrate over an 400 ns interval, indicated by the vertical dashed lines. The $g^{(2)}_{\text{av}}$ values for these two measurements considering the same integration window are 1.8(4) and 62(2), respectively.

3.1. Frequency-bin two-photon interference

As the RF is matched to the cavity FSR, the side bands of one mode will overlap with the neighboring modes. This creates the superposition of frequency bins necessary to show interference. The phase dependence of the coincidence rate is described by Bessel functions and depends on the modulation indices (see equation (6)). With the help of our model we decided to use equal depths in the range $1 \leq c_s, c_i \leq 1.4$ as we expect a rather high interference contrast for the coincidence rate with these settings. Examples are shown in figure 3(b). We first set the phase of the idler EOM to $\alpha_i = 0^\circ$ and varied the signal EOM phase only (green curve in figure 3). Next we set the idler EOM phase to $\alpha_i = 51^\circ$ and repeated the measurement (blue curve in figure 3). In both cases the filter cavities were actively stabilized to the central mode (mode $|0, l\rangle$) of the spectra, corresponding to $d = 0$. For comparison, the magenta data points in the figure show the results when the idler cavity is stabilized to the side mode (mode $|+1, l\rangle$) while the signal filter is still centered (mode $|0, l\rangle$), corresponding to $d = 1$.

While we observe a dependence of the coincidence rate on the phase settings, no changes in the single rates are observed. Thus, this variation of the coincidence rate induced by the change of the relative RF phase shows two-photon interference which is a signature of the coherent superposition of the frequency-bins. From the raw data, shown in figure 3, we find an interference contrast of $V = 95(4)\%$. The solid lines shown in the figure are predictions of our simulation.

The very good overlap of the experimental data with the model is a first hint that we generate a frequency-entangled state.

3.2. Bell inequality

The results in figure 3 show a very high two photon interference contrast, offering evidence of the entanglement of the state we generate. To demonstrate entanglement without assumptions on the dimension of the created state, we then show violations of the Bell inequality (equation (12)). However, although the results in figure 3 show a very high interference contrast, we cannot reach an S-value above the classical boundary with these settings. The reason for this surprising result is that it is not possible in these conditions to find phase settings for which the side modes are zero and all the coincidences are in the central mode. This is a consequence of the fact that the created state is non-maximally entangled. We confirm this intuition by our simulation which shows that if the modulation index remains constant for the four settings (although it may be different for signal and idler photons), only a negligible violation can be achieved (see figure 4 left). However, much more significant violations can be achieved if the modulation depth is alternated between the four settings (see figure 4 right).
From a technical point of view the differences in modulation during the alternation should not be too high, as we observed a reshaping of the optical beam by the EOMs depending on the thermal load (i.e. on the power of the RF), changing detection efficiency of the photons. In the experiment a switching of the modulation index by less than 0.5 has no observable influence on the transmission of the photons to the detectors. With this limitations, the best results are expected for an alternation between a high and a low modulation index and phase shifts around 180°. The final values we choose can be found in table 1. We accordingly shift the phase of the idler RF by 182°. To calibrate the global phase offset, in a first step we measured four fringes with the four settings and varied the phase of the signal EOM over the full accessible range. We used a random number generator to decide for each trial which setting (c_i,j, a_j, c_s,j for j = 0, 1) and phase (β_k) to use. In each trial we collected data for 10 s. The results were then added to the corresponding data point. The whole procedure was afterwards repeated with the idler filter set to the side modes 1_i ± 0, as necessary for the normalization. The normalized coincidences are shown in figure 5.

To compare our model with the data, we measured the modulation indices for the four settings with the procedure described above and put this information into the model. The only free parameter is a global phase shift between the model and the experimental data which we fit to the best overlap. The result is plotted in figure 5 for comparison. We additionally plotted the result in the case we normalize the simulation neglecting modes |d_i > 1 (dashed lines). The effect is smaller than the average error bar in the experiment proving the validity of our assumption. The effect of not-optimal normalization is smaller here compared to section 3.1 due to the lower modulation indices. Less modulation depth results in less mixing of frequency bins with neighboring modes. Quantitatively, the two methods introduce a deviation of 7% for the normalization of the x_0 y_1 setting, while the deviation is up to two orders of magnitude smaller for the other settings. As a matter of fact, the x_0 y_1 setting has higher modulation indices, thus the contribution of the modes with |d| > 1 is bigger. As explained in section 2, we assume that C(0|x_0) = kC(0|y_0). For the values of modulations used here (see table 1), we simulate the spectrum of the signal and idler field after their respective EOM and can then estimate the value of k. For the measurements shown on figure 5, we find k = 1.01.

The Bell inequality is based on the comparison of four coincidence probabilities, one for each combination of settings. Figure 5(a) shows the result for all allowed combinations of the experimental data. As we can see in figures 5(b) and (c), the violation is quite robust as there is a wide range of the measurement parameters resulting

**Table 1. Bell Settings.** The k parameter, which accounts for small variations of the signal and idler modulation indices (see section 2), is also reported.

| Experiment | Setting | x_0 | x_1 | y_0 | y_1 | k     |
|------------|---------|-----|-----|-----|-----|-------|
| Fringe     | Mod. idx.| 0.29| 0.85| 0.34| 0.81| 1.01  |
|            | Phase   | 0   | 182 | 314 | 181 |       |
| Bell points| Mod. idx.| 0.44| 0.56| 0.34| 0.81| 0.97  |
|            | Phase   | 0   | 182 | 361 | 171 |       |

**Figure 4.** Simulation of highest possible S-values for different settings. In the left graph the modulation index is held constant for all four Bell settings (although it can take different values for signal and idler photons). In the right graph we assume switching the modulation indices by 0.5 and phase shift by 180° between the four settings. In this combination higher limits for the S-value can be achieved.

Quantitatively, the two methods introduce a deviation of 7% for the normalization of the x_0 y_1 setting, while the deviation is up to two orders of magnitude smaller for the other settings. As a matter of fact, the x_0 y_1 setting has higher modulation indices, thus the contribution of the modes with |d| > 1 is bigger. As explained in section 2, we assume that C(0|x_0) = kC(0|y_0). For the values of modulations used here (see table 1), we simulate the spectrum of the signal and idler field after their respective EOM and can then estimate the value of k. For the measurements shown on figure 5, we find k = 1.01.
in $S > 2$. However, the highest violation of $S = 2.31(8)$ (calculated using $k = 1.01$) is achieved with the combinations summarized in table 1. For the error estimation we here take only into account the Poissonian statistics of the photon counts, no systematic errors are considered. The errors are then combined via error propagation calculations. This results in a violation of the classical bound by four standard deviations. Our model predicts a violation of $S = 2.30$ which is in perfect agreement with the experimental result. The deviation of the $S$-value due to the non-optimal normalization is below 1% and thus smaller than the statistical error of 3.5%.

In addition, we repetitively measured the coincidence rate for the four settings (see table 1) to show a violation of the Bell inequality. We did these measurements with a reduced RF power in the idler EOM. Such a setting increases the stability of our FC lock system which in turn allows longer measurement times. On the other hand, we expect a lower violation of the Bell inequality ($S = 2.24$). In that case, we estimate the proportionality constant between the marginals to be $k = 0.97$. With the focus on these points only, a large number of coincidences could be recorded reducing the statistical error. This finally results in $S = 2.21(2)$ (calculated with $k = 0.97$), corresponding to a violation of 9.7 standard deviations.

4. Compatibility with a quantum memory material

In order to show the compatibility of our source with a quantum memory material, we then perform experiments using a narrow spectral hole in the absorption profile of a praseodymium doped $Y_2SiO_5$ crystal as frequency filter to select one frequency mode in the signal arm. The inhomogeneous broadening of the transition at 606 nm results in total absorption of all signal modes created by our OPO. However, we can create a transmission window via spectral hole burning. With the help of a narrow band tunable laser we empty two of the three hyperfine ground states. With this technique we tailor a window of 18 MHz featuring high transmission for the mode $|0\rangle$ only. We measure coincidences between the single mode signal photons filtered in that way and the idler photons filtered to a single mode with the help of the cavity, as explained before. The
coincidence rate is reduced due to increased losses in the signal arm and an additional duty cycle, necessary to periodically refresh the spectral filter hole. As shown in figure 6(b) we still observe interference fringes with high visibility.

We repeated this measurement for different pump powers of our source. Increasing the pump power increases the probability to create multiple pairs in the SPDC process. This reduces the fidelity of the heralded single photon. This effect can be observed in a decrease of the signal-idler cross correlation value $g^{(2)}_{si}$ (see also [24]). The result is summarized in figure 6(c). For all available pump powers we operate in a non-classical regime. Accordingly, we observe a decrease of frequency-bin interference visibility with pump power. Nevertheless, the lowest visibility for all accessible pump powers was still quite high with $V \geq 89.2(3)\%$. These results demonstrate the compatibility of our source with a QM material.

However, further steps will be needed before storing frequency-bin entanglement. We demonstrated previously that we can store heralded single photons created by this source in a solid state quantum memory [37, 41], using the atomic frequency comb (AFC) technique for storage. This involves creating a periodic absorption feature within the spectral hole we used for filtering. Due to the bandwidth limit, if a few MHz, imposed by the energy level scheme of the Pr ions, the majority of the ensemble, whose inhomogeneously broadened absorption is several GHz wide, is not involved in the storage. However, this can be exploited as a resource by creating, with the use of combinations of EOMs and AOMs, several AFC structures. This possibility has already been demonstrated in other materials [33, 34]. If the multiple AFCs are separated by the SPDC FSR, this would allow us to store the photon in a superposition of frequency bins which is essential to preserve the qudit state. The EOM for analysis of the entanglement will be placed behind the memory.

5. Conclusion

Frequency-bins are naturally defined for photons generated by an OPO-SPDC source. We here demonstrated for the first time the entanglement of such OPO-based photon pairs in the frequency-bin basis. The results can be well simulated by our generalized model based on the spectral distribution of the photons. With this model we analyzed the violation of the CH inequality that is accessible by our experiment. With our generated state, we observed that the settings used to obtain a high visibility in the two-photon interference were not suited for a strong violation of the Bell inequality, due to the shape of the photons’ spectrum. Nevertheless, with optimized settings we could significantly violate the inequality and thus show the entanglement of the photons. In a next step we replaced the signal filter cavity with a spectral hole in a quantum memory crystal and still observed interference effects, supporting prospects for storing frequency entangled photons. With our technique the photons are prepared in a high dimensional state. Future work will also focus in a better understanding of the dimensionality and the applications offered by this capability.
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