Time uncertainty of a photon pair creation in a bulk periodically poled potassium titanyl phosphate pumped by a femtosecond laser

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Abstract. Periodically poled crystals, such as periodically poled potassium titanyl phosphate (PPKTP) or lithium niobate, recently became more attractive because of their high efficiency for producing a photon pair in both cw and pulse cases. The time uncertainty of photon pair creation in a crystal is crucial for many important applications. In this paper, we experimentally demonstrate that the time uncertainty of the photon pair creation in a PPKTP crystal is much larger than the coherence time of a femtosecond pump laser by using phase-sensitive two-photon interference using a Michelson interferometer, which means the femtosecond laser cannot be considered as a clock that announces the creation of the photon pairs. The experimental results can be well explained by an effective Franson-type interferometer, if the PPKTP is regarded as a narrow band filter at the same time. Our experiment also shows a way by which the Franson-type two-photon interference phenomenon can be observed even if the coherence length of the pump laser is less than the path-length difference between the two interfering beams.
Efficient generation of entangled photon pairs is essential for many applications in the quantum information field, such as quantum key distribution [1]–[4], teleportation [5]–[11], dense coding [12], etc. By far the most efficient way of obtaining this is spontaneous parametric down-conversion (SPDC) [13, 14]. In some applications, such as quantum teleportation, the generation of multi-photon entangled states [15], the interaction between entangled photon pairs generated from different sources is required. In the cw-laser pumped SPDC case, the photon pairs occur randomly within the coherence time of the pump laser. This huge uncertainty in time makes it difficult to use in the aforementioned applications. It has been proved that the uncertainty of the photon pair creation times in some materials, such as beta-barium borate (BBO) and lithium triborate (LBO), can be significantly reduced so that the photon pair can be a good candidate for such applications, if a femtosecond laser is used as a pump [6, 9]–[11]. Although a lot of important results have been obtained, many experiments, like teleportation or entanglement swapping, still suffer from low production rates leading to low signal-to-noise ratios and long measurement times. More recently, periodically poled bulk crystals or waveguides, such as periodically poled lithium niobate or potassium titanyl phosphate (PPKTP) were used for photon pair generation both in cw [16]–[21] and ultrashort pulsed pump [22]–[24] cases, and were experimentally shown to be very efficient compared to other bulk crystals. In this paper, we consider the generation of photon pairs by using a type-I phase matched PPKTP crystal. In [22] and [24], it was experimentally shown that a type-I phase matched PPKTP pumped by a femtosecond laser could be used to efficiently generate photon pairs. For example, about 3200 coincidence counts per second per milliwatt pump power were detected in a Hanbury–Brown–Twiss-type experiment by using a bulk PPKTP crystal in [24], compared to about $2.4 \times 10^4$ coincidence counts per second with 480 mW pump power in [25]. In [23], a femtosecond pumped type-II phase matched PPKTP was used for a conditional preparation of single photons. All these results showed that PPKTP is a very efficient pulsed photon pair source. Then the following question arises naturally: can the photon pairs generated from a PPKTP crystal pumped by a femtosecond laser also be good candidates for the just-mentioned applications? If the answer is affirmative, it is very promising for the realization of sources for quantum communication and metrology. Before we draw a conclusion, the uncertainty of the photon pair creation times should be checked to see whether it is really small enough to make them be a good candidate for these applications or not. This is the main motivation of this work. In order to check it, we perform a phase-sensitive two-photon interference experiment using an unbalanced Michelson interferometer, which may be considered as an effective Franson-type interferometer [26]. Experimentally, we find that even if the path-length difference between the two interfering beams is much larger than the coherence length of the pump laser, the two-photon coincidence rate still clearly shows oscillation with periodicity of the pump wavelength. At the same time, the single counts do not show any oscillation. Our experimental results clearly show that the uncertainty of the photon pair creation times is much larger than the coherence time of the pump laser, so that a femtosecond laser cannot be a trigger of the photon pair creation times. This also means that a multiphoton state cannot be generated directly in a PPKTP crystal [27]. This is mainly caused by the very narrow intrinsic bandwidth of the PPKTP crystal, therefore the PPKTP serves as a narrow band filter at the same time, prolonging the coherence time of the pump laser. This experiment shows that more attention should be given to the properties of the material in ultrashort applications. Besides, a very interesting point of this experiment is that the coherence length of the pump laser used is less than the path-length difference between the two interfering beams, in contrast to other experiments in which the coherence length of the pump
Figure 1. The measured spectrum of the SHG with a pump power of 148 mW. The FWHM is 0.093 nm. The temperature of PPKTP is 58 °C.

The laser used is much larger than the path-length difference [28]–[30]. Although this kind of source is not very useful for the aforementioned applications, it may be used to get a heralded high efficient single photon source [23].

The experimental proofs consist of two parts. Before giving them, we briefly give some parameters of the PPKTP crystal used in this experiment. It was bought from Raicol Crystal Company. The size of the crystal is: 1.05 mm (z) × 2.1 mm (y) × 5 mm (x), where z, y and x are mean height, width and length respectively. The grating period is of 3.25 µm. It is cut for type-I phase matching and is antireflection coated on both faces at 400 and 800 nm, and placed in a holder with a built-in Peltier device with a stability of 0.01 °C. The first proof given here is based on the measured spectrum of the second harmonic generation (SHG) of a strong 800 nm femtosecond mode-locked Ti: sapphire laser. The pulse width of the laser is less than 100 fs and the repetition rate is 80 MHz. We check the SHG power dependence on the temperature of the PPKTP crystal. The optimal temperature for our crystal and our laser is 58 °C. The spectrum of the SHG wave with 148 mW pump power is shown in figure 1. The measured full width at half maximum (FWHM) of the SHG wave by an optical spectrum analyser (ANDO AQ-6315A with a resolution of 0.05 nm) is about 0.093 nm. This intrinsic bandwidth determines the coherence time of the PPKTP crystal, which is about 2.5 ps, equals to about 750 µm coherence lengths assuming that the pulse shape is Gaussian. The main reason why PPKTP has a very narrow intrinsic bandwidth is that it has a large group velocity mismatch in the ultrashort pulsed regime case. 3 In [24], the FWHM of the SHG wave with a 2.12 mm long PPKTP crystal is approximately 0.18 nm, compared with the 3.2 nm wide SHG wave spectrum when pumping a 1 mm BBO crystal with the same pump laser.

3 Basically, the pulse width of the SHG wave can be approximately calculated by the formula \( \tau^2 \approx \frac{\tau_{p}^{2}}{1 + (L/L_{b})} \), where, \( \tau_{p} \) is the pulse width of the pump laser (SHG). L is the length of the crystal, \( L_{b} \) is the intrinsic length of the crystal determined by the group velocity mismatch, which can be calculated by the formula \( L_{b} = \tau_{p}/\Delta u \), where, \( \Delta u^{-1} = u_{p}^{-1} - u_{2}^{-1} \), and \( u_{p(2)} \) is the group velocity of pump laser (SHG) [31]. We calculated that \( \Delta u^{-1} \) of the BBO crystal is about 193 fs mm\(^{-1}\), when the SHG process from 800 to 400 nm is considered, compared with 1640 fs mm\(^{-1}\) of the PPKTP crystal. So, the PPKTP crystal shows the more larger pulse width compared to the BBO crystal.
Although we can infer indirectly from the spectrum of the SHG that the PPKTP acts effectively as a narrow band filter at the same time, prolonging the coherence time of the pump laser, this cannot be seen as a direct proof of the temporal behaviour of the photon pairs. In the following, we give a clearer proof. The basic idea can be explained by a two-photon interference experiment using a Franson-type interferometer. When we consider a Franson-type interference experiment done with a femtosecond laser, two different cases occur: when the path-length difference between the two interfering beams is larger than the coherence length of the single photons, but smaller than that of the pump laser, the coincidence rate, but not the single rate, depends on the fine path-length difference of the order of the pump laser wavelength. The maximal 100% visibility can be obtained if the two interfering terms ‘short–short’ and ‘long–long’ can be distinguished from the other two non-interfering terms ‘short–long’ and ‘long–short’ [26]. Otherwise, the maximal visibility is limited to 50%. This is a straightened condition since the coherence length of a femtosecond laser is very small demanding very small path-length difference. If the path-length difference is larger than the coherence length of the pump laser, the coincidences are also independent on the fine path-length difference. We hope to roughly estimate the actual uncertainty of the photon pair creation times by observing the two-photon interference and checking the relation between the path-length difference and the coherence length of the pump laser. Based on this idea, we perform a phase-sensitive two-photon interference experiment using a Michelson interferometer to directly check the temporal behaviour of the photon pairs generated in the PPKTP. An interesting solution given by Brendel et al [32] to solve the problem with a femtosecond laser should be mentioned. In their scheme, a new interferometer on the path of the pump laser is added in order to create a coherent superposition state of the pump photons. By this method, a ‘time-bin’ entangled state can be created. One advantage of this method is that the coherence of the pump laser is of no importance, the uncertainty of the pump photon’s arrival time at the crystal is replaced by two sharp values. So the path-length difference can be arbitrary chosen provided it is the same than that of the pump interferometer. The two-photon interference can be still observed even if the path-length difference is larger than the coherence length of the pump laser, and the maximal 100% visibility can also be expected using a post-selection on the two interfering terms.

The experimental setup is shown in figure 2. The output of the mode-locked Ti:sapphire laser is doubled in a 1 mm type-I phase matched BBO crystal to generate violet pulses with a centre wavelength of 400 nm. The half waveplate and the polarizer are used to control the intensity of the 800 nm laser. Before the 400 nm laser pumps the PPKTP crystal, the violet pulses first pass through two 1.5 mm pinholes, and are attenuated by a continuous variable attenuator. The distance between the two pinholes is about 23 cm. The FWHM of the spectral bandwidth of the violet pulses measured by the optical spectrum analyser before the PPKTP is about 3.14 nm, and the centre wavelength is 400.51 nm. A lens with 100 cm focal length weakly focuses the violet pulses. The temperature of the PPKTP crystal is 40 °C. Note that in this case the optimal temperature is different in contrast to the SHG process, because their centre wavelengths are different. After passing through the PPKTP crystal, two red filters (RG715) are firstly used to cut the remaining violet pump pulses. Then the SPDC photon pairs are sent to a small Michelson interferometer. One of the reflective mirrors is mounted on a motorized micrometer stage (K101-20MS, Suruga). The

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4 A change in the temperature is equal to a change in the peak wavelength of the phase matching. The turning coefficient is quite small near the 390 nm spectral region, is about $1.2 \times 10^{-2}$ nm °C$^{-1}$ according to our calculation. This can explain the reason why there is quite large temperature difference even though the centre wavelength difference between SHG and parametric down conversion processes is quite small.
Figure 2. Experimental setup. HWP: half waveplate; POL: polarizer; BF: blue filter; AT: attenuator; DBS: dichroic mirror; P: pinhole; RF: red filter; M: mirror; OB: objective lens; SMF: single mode fibre; FC: fibre connector; FB: fibre beamsplitter; D(s): detector; C.C: coincidence circuit.

In our experiment, we first balance the two interfering beams by using the motorized micrometer stage, and measure the coincidence counts and one of single counts as a function of the voltage of the PZT. Then we change the path-length difference between the two interfering beams and repeat the same measurements. The experimental results are shown in figure 3. When the two beams are coarsely balanced, both coincidence and single counts show clear oscillations. It can be calculated from standard quantum mechanics that the variation of the coincidence rate corresponds to the mathematical formula $R_{cc} = 3 + 4 \cos \Theta + \cos(2\Theta)$ in the ideal case, where, $\Theta = 2\pi \Delta L/\lambda_{800}$, and $\Delta L$ is the path-length difference and $\lambda_{800}$ refers to the 800 nm wavelength.
Figure 3. Experimental results. (a) Coincidence counts and one of the single counts plotted as the function of the fine path-length difference when the two interfering path-length difference is coarsely zero. (b) Coincidence counts plotted as the function of the fine path-length difference when the path-length difference is coarsely nonzero. The coarse path-length difference is 80 µm in case (1), 240 µm in case (2) and 400 µm in case (3). Single counts in all cases are almost constant. (For example, the single counts of the detector D1 are 38 ks$^{-1}$ in all three cases, not shown.) All path-length differences are much larger than the coherence length of the pump laser.
The special formula comes from three possible processes of photon pair propagation inside the Michelson interferometer: both photons propagate along the same path, or along different paths. When the two photons propagate along the different paths, there is a photon bunching effect [33]. The solid line is the fitted line with the above formula. The fitted period of the oscillations is $1050 \pm 261$ nm. The small mismatch between the experimental data and the fitted curve is due to mechanical instabilities or misalignment of the interferometer, etc. The oscillations of the single counts correspond to the formula $R_s = 1 + \cos \Theta$ in the ideal case. This is also called white light interference [34]. The solid line is the fitted curve using the latter formula. The fitted period is $1000 \pm 246$ nm.

All experimental data are given with errors. A more interesting thing appears when the coarse path-length difference is much larger than the coherence length of the pump laser (which is in our experiment, the 400 nm laser with 3.14 nm bandwidth). The calculated coherence time is about 75 fs, which corresponds to about $23 \mu m$ coherence length assuming that the pulse shape is Gaussian), the coincidence counts still show clear oscillations. At the same time, we monitor the single counts and find the single counts remain constant (for example, the single counts of the detector D1 are $38 \pm 1$ with accidentals in all three cases, not shown in the related figure). The oscillations of the coincidence counts correspond to the formula $R_{c.c} = 1 + \frac{1}{2} \cos (2\Theta)$ in the ideal case. Obviously, there is no photon bunching effect in these cases with respect to the previously discussed balanced case. The experimental data for different path-length differences and the related fitted curves are shown in figure 3(b). The oscillation periods are $500 \pm 123, 532 \pm 131$ and $532 \pm 131$ nm respectively for the coarse path differences of 80, 240 and 400 $\mu m$. The visibilities are $42 \pm 4.1\%, 33.2 \pm 2.8\%$ and $23.9 \pm 3\%$ respectively calculated from the fitted curves including errors. Correcting for errors results in a few percentage changes. As we mentioned before, if the maximal 100% visibility is expected, the two interfering terms and two non-interfering terms should be distinguished. In our experiment, the path-length difference is so small that there is no way to differentiate them because of lower temporal resolution of the analyser. So, the maximal visibility is limited to 50%. The two-photon interference oscillation period is almost half of the single-photon interference. In all figures, the periods of all fits are larger than the theoretical values of 400 and 800 nm, we think mainly from the PZT used which has a large uncertainty, and maybe also from instability of the experimental system.

Now, we consider how to explain the results. Even if our experiment features collinear outputs and a single Michelson interferometer in contrast to other experiments [28, 30], this phenomenon can be effectively explained by a two-photon interference in a Franson interferometer [26]. The coherence length of the pump laser is much less than the path-length difference between the ‘long–short’ paths in our experiment compared to other Franson-type experiments [28]–[30], in which the coherence length of the pump laser is much larger than the path-length difference. The only reason that can explain our experimental phenomenon is that the PPKTP acts as a narrow band filter at the same time, prolonging the coherence length of the effective pump laser. Another difference between our experimental result and the results of the other experiments is that the visibility is gradually decreased with the increase of the path-length difference in our experiment. In contrast, the visibility remains constant in other experiments.

5 The error in the figure can be calculated by the following equation $E = \frac{\sigma}{\sigma_{cm}} = \sqrt{\frac{\sum (c_i - c_{i'}^2)^2}{n - 1}}$, where $c_{cm}$ is the mean of coincidence counts, $\sigma$ is standard deviation, $c_i$, is the coincidence counts of fitting, $c_i'$ is the actual coincidence counts and $n$ is the number of the data. The visibility uncertainty can be calculated by equation $\Delta V = \frac{2(c_{max} - c_{min})}{(c_{max} + c_{min})^2} E$, where $c_{max}$ and $c_{min}$ are maximum and minimum values of the coincidence counts.
The main reason is that the effective pump laser provides pulses with a finite bandwidth. The coherence becomes bad near the edge of the pulse. From another point of view, this is not surprising because the larger difference between the two interfering beams becomes comparable with the coherence length of the effective pump laser. This experiment clearly demonstrates that the femtosecond pump laser does not act as a clock as in other experiments by which the photon pair creation times can be known with a satisfying resolution. We also try to experimentally roughly evaluate the time uncertainty based on the fringe visibility as the function of path-length difference between two interfering beams \([35]\), and find that when the path-length difference is larger than about 1 mm, there is almost no coincidence oscillation, therefore the whole interference range is about 2 mm. If the Gaussian pump pulse shape is assumed, then the coherence length is about 0.88 mm. We also calculate the coherence length from the group velocity mismatch. It is about 1.08 mm assuming the Gaussian pulse shape of the pump laser with 100 fs FWHM pulse width. Both are almost comparable to the value calculated from the measured SHG bandwidth, and all are much larger than the coherence length of the pump laser.

Besides although our experiment could be explained by an effective Franson-type interferometer \([26]\), we observe the Franson-type two-photon interference even if the coherence length of the pump laser is less than the path-length difference between the two interfering beams, in contrast to many previously performed experiments in which the coherent time of the pump laser used is much larger than path difference \([28]–[30]\). Obviously, our experiment is different from the scheme presented in \([33]\) in which an artificial coherence of the pump laser is introduced by adding an interferometer on the pump path, in contrast to using the intrinsic bandwidth of the crystal to prolong the coherence time of the pump laser in our experiment.

In summary, although PPKTP can be used to highly efficiently generate photon pairs, the photon pairs occur within a larger time uncertainty compared to other crystal, such as BBO, LBO, even if an ultrashort pulsed laser is used as a pump. This large uncertainty in time is mainly determined by the intrinsic bandwidth of the PPKTP crystal, as this intrinsic narrow band filter prolongs the coherence time of the pump laser. This means a femtosecond laser cannot be considered as the clock that announces the creation of the photon pairs. We experimentally demonstrate this effect by using a phase-sensitive two-photon interference using a Michelson interferometer. The experimental phenomenon can effectively explained by a Franson-type interferometer. This experiment shows that the two-photon interference in a Franson-type interferometer can also be observed even if the coherence length of the pump laser is much less than the path-length difference between the two interfering paths.

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