Source of polarization entanglement in a single periodically poled KTiOPO$_4$ crystal with overlapping emission cones

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Abstract: We present a simple polarization entanglement source based on type-II phase-matched parametric down-conversion in periodically poled KTiOPO$_4$. By use of noncritical phase matching in a noncollinear geometry the single-crystal source emits a cone of polarization-entangled photons. Two beams on opposite sides of the cone are selected for measurements to yield an observed flux of 820 pairs/s per mW of pump power with a two-photon fringe visibility of 96%. With this source we measure a violation of Bell’s inequality of 140 standard deviations in 160 s.

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References and links
Polarization entanglement is an essential quantum resource in many quantum optics experiments such as quantum teleportation [1] and quantum cryptography [2, 3]. Many different sources of polarization-entangled photons have appeared over the years, most of them based on spontaneous parametric down-conversion (SPDC) [4, 5, 6, 7, 8, 9, 10, 11, 12, 13]. Researchers have strived to obtain sources that generate a large number of pairs in a highly entangled state. A standard method uses a thin β-barium borate (BBO) crystal oriented for noncollinear type-II angle phase matching, similar to that described in Ref. [5]. In a type-II BBO source frequency-degenerate orthogonally-polarized signal and idler photons are emitted in two non-overlapping cones that intersect at two locations. The output beams at these two locations are polarization entangled if the timing delay due to BBO crystal birefringence is compensated. This convenient setup, however, is not efficient because the entangled photons from the intersecting locations is only a small fraction of the total emitted light.

Recently several groups have demonstrated more efficient sources of polarization-entangled photons. Kwiat et al. [6] have demonstrated a source that uses two BBO crystals in type-I phase matching in which the polarization-entangled photons are emitted in a cone. A similar cone output geometry for the entangled photons is obtained by De Martini’s group [11] by pumping a single type-I BBO crystal in both directions and cleverly combining the outputs. These two sources produce a very large number of entangled photons with a high degree of polarization entanglement. Another route to generating most of the SPDC output light as polarization-entangled photons and collecting them efficiently is the use of periodically poled nonlinear crystals, such as periodically poled KTiOPO4 (PPKTP), in a collinearly propagating geometry [12, 13]. Quasi-phase matching (QPM) in periodically poled crystals allows flexibility in the operating wavelengths, and in a collinearly propagating configuration the output cone collapses into an intense beam-like output. Kuklewicz et al. have utilized a single PPKTP crystal with a single-beam output that can be postselected by a 50–50 beam splitter to produce a high flux source of polarization-entangled photons [12]. However, the simple collinear output geometry suffers a 50% efficiency loss due to postselection, which in itself is often undesirable in some applications. Fiorentino et al. overcome this restriction by using a bidirectional pumping scheme with a single PPKTP crystal that has produced an ultrabright source of polarization entanglement at the expense of the need for phase locking an interferometric arrangement [13]. In this paper we present a new source of polarization-entangled photons based on a single PPKTP crystal. We take full advantage of type-II phase-matched PPKTP by propagating a single pump beam along one of the crystal’s principal axes that yields, under noncollinear angle phase matching, overlapping emission cones of signal and idler light. By collecting photons on the opposite sides of the emission cone we have detected a large number of highly polarization-entangled photons in a setup that is simpler and more robust than other sources with a comparable flux of photon pairs.

In the parametric down-conversion process a pump photon of frequency \(\omega_p\) splits into a signal and an idler photon in a nonlinear \(\chi^{(2)}\) crystal. For efficient SPDC generation energy and
Fig. 1. Definition of angles $\delta$ and $\phi$ to denote the direction of wave vectors. The axes of reference coincide with the crystal’s principal axes, $z$ is the direction along which the ferroelectric domains are oriented. The pump propagates along $x$ and is polarized along $y$. For collinear down-conversion signal and idler are polarized along $y$ and $z$, respectively.

momentum conservation must be satisfied

$$\omega_p = \omega_s + \omega_i, \quad (1)$$

$$k_p = k_s + k_i, \quad (2)$$

where $\omega$ is the frequency and $k$ is the wave vector with a modulus

$$|k_j| = 2\pi n_j (T, \lambda_j, \delta_j, \phi_j) / \lambda_j, \quad (3)$$

and the subscripts $j = p, s, i$ denote the pump, signal and idler, respectively. Equation (3) explicitly indicates the dependence of refractive index $n_j$ on the crystal temperature $T$, the direction of propagation determined by the angles $\delta_j$ and $\phi_j$ (as defined in Fig. 1), and the vacuum wavelength $\lambda_j$. In our experiments we use QPM in a periodically poled crystal, and the conventional phase matching condition (2) is modified to include the crystal’s grating wave vector

$$k_p = k_s + k_i + \frac{2\pi}{\Lambda} x. \quad (4)$$

The grating period $\Lambda$ serves as a free parameter to satisfy QPM condition (4) at user-determined wavelengths and propagation geometry such as the collinear configuration in which $k_{p,s,i} \parallel x$. The collinear geometry along a principal axis of the crystal is highly desirable in nonlinear optics because it optimizes the conversion efficiency by eliminating the walk-off of the interacting fields. In entanglement generation, collinear geometry can be utilized to yield a beam-like output. We note that in collinearly propagating configurations, wavelength tuning can be achieved by varying the crystal temperature.

For the experimental data and numerical calculations we present here, we use a 10 mm-long PPKTP crystal with a poling period $\Lambda = 9 \mu m$. The pump has a wavelength of $\sim 400$ nm, is polarized along the $y$ axis (see Fig. 1), and propagates along the $x$ axis (with $\delta_p = 0$ and $\phi_p = 0$). For collinear propagation ($\delta_j = 0$ and $\phi_j = 0$), signal and idler are orthogonally polarized along $y$ and $z$ axes, respectively. Under noncollinear angle phase matching, the wavelengths and directions of propagation of the down-converted photons are obtained by solving the phase matching condition (4) with the energy constraint of Eq. (1). For a hydrothermally grown PPKTP crystal, we use the Sellmeier equations in Ref. [15] to obtain the wavelength dependence of the refractive indices. The temperature dependence of the KTP indices is obtained from Ref. [16]. We note that the data of Ref. [16] are for a wavelength range of 515–1064 nm.
While these temperature formulas work well at the ∼800-nm signal and idler wavelengths, they do not extrapolate well to our pump wavelength near 400 nm. Instead, we have used our second-harmonic generation results [17] to obtain a fit parameter for the temperature derivative at the pump wavelength

\[ \frac{\partial n_p}{\partial T} \bigg|_{T=20^\circ C} = 28 \times 10^{-6} / ^\circ C. \]  

(5)

With the help of the Sellmeier equations of Ref. [15] and the temperature dependence of Ref. [16] we set out to solve Eq. (4) numerically, and the angular phase matching results are shown in Figs. 2, 3, and 4. For Fig. 2, we calculate the output wavelengths as a function of the external elevation angle of propagation \( \delta (\phi_l = 0) \) for collinear frequency-degenerate down-conversion. In this configuration the frequency-degenerate signal and idler photons (\( \lambda_s = \lambda_i = 2\lambda_p \)) are emitted collinearly with the x-propagating pump beam. We have previously used this configuration to generate postselected polarization entanglement [12]. To obtain Fig. 2 we set the crystal temperature at \( T = 35.65^\circ C \), the pump wavelength at 398.6 nm, and \( \phi = 0 \) for both signal and idler, and external angles of propagation are used. We note that the angle of propagation outside the crystal is different from the internal angle due to refraction at the crystal-air interface. The signal curve (solid blue) and the idler curve (dashed red) are tangential to each other at the expected collinearly propagating angle of \( \delta = 0 \) at frequency degeneracy. For non-collinear propagation, \( \delta \neq 0 \), the signal wavelength is always shorter than the idler wavelength, and hence the two down-converted photons are spectrally distinguishable.

Figure 3 shows a similar angular phase matching plot for the case of noncollinear downconversion that is obtained by changing the crystal temperature to \( T = 34.7^\circ C \). In this configuration the signal and idler photons are not frequency degenerate at the collinearly propagating direction \( \delta = 0 \), and frequency degeneracy (\( \lambda_s = \lambda_i \)) occurs at a nonzero \( \delta \) angle of propagation. As clearly evident in Fig. 3 the interesting case of spectrally indistinguishable signal and idler emission (where the blue and red curves intersect) occur at two locations that are symmetric with

![Fig. 2. Plot of phase-matched wavelengths versus external propagation angle \( \delta \) for signal (solid blue) and idler (dashed red), calculated for \( \phi = 0 \). Operating condition: collinear frequency-degenerate signal and idler at \( \delta = 0 \). Green line represents the degenerate wavelength equal to \( 2\lambda_p = 797.2 \) nm.](image-url)
Fig. 3. Plot of phase-matched wavelengths versus external propagation angle $\delta$ for signal (solid blue) and idler (dashed red), calculated for $\phi = 0$. Operating condition: noncollinear frequency-degenerate signal and idler in two overlapping cones. Green line represents the degenerate wavelength equal to $2\lambda_p = 797.2$ nm.

respect to the pump propagation direction. It is useful to examine the output emission characteristics for the special case of frequency degeneracy at different crystal temperatures. Figure 4 shows the calculated external emission angles of the degenerate signal photons ($\lambda_s = 2\lambda_p$) at temperatures of (a) 35.65°C, (b) 34.7°C, and (c) 32.8°C. Figure 4(a) has the same crystal temperature as Fig. 2 and shows that the degenerate signal output is collinear with the pump. The noncollinear case of Fig. 3, with an operating temperature that is lower than the collinear case, is shown in (b) with a full emission angle of $\sim 14$ mrad. As the temperature is further lowered, the emission cone becomes larger, showing a full divergence angle of $\sim 24$ mrad in (c). We note that the signal output cone has a slight ellipticity because the PPKTP crystal is biaxial. We have also calculated the emission angles for the idler, not shown here, to be exactly identical to those for the signal field, indicating that the signal and idler photons are emitted in overlapping cones at frequency degeneracy. This important feature is due to the fact that, thanks to periodic poling, one is able to phase match the degenerate down-conversion process in an angularly noncritical geometry in which the pump propagates along one of the crystallographic axes of the crystal. This feature of overlapping emission cones distinguishes our source from the angle phase-matched single-crystal BBO source [5] in which the signal and idler emission cones intersect but do not overlap, as shown in detail in Ref. [18]. We point out that our numerical calculations give a qualitative understanding of the angle phase-matched down-conversion process but the results are not necessarily quantitatively accurate, in particular the phase matching temperatures. We attribute these discrepancies to the differences among different types and manufacturers of KTP crystals and the empirical nature of Sellmeier equations.

Figure 5 shows snapshots of the orthogonally polarized signal and idler output light taken with a high-sensitivity charge-coupled device (CCD) camera (Princeton Instruments VersArray: 1300B) with an exposure time of 20 s. The pump wavelength for the measurements was 398.54 nm and the PPKTP crystal temperature was set at 22°C. To reduce background light, the pump was blocked with dichroic mirrors and a 0.11-nm-wide interference filter centered at 797.08 nm was inserted. We used a polarizer to select the signal or idler light for CCD imaging. We
Fig. 4. External propagation angles for degenerate down-converted signal \( (\lambda_s = 2\lambda_p = 797.2 \text{ nm}) \). Operating conditions: (a) collinear output \( T = 35.65^\circ\text{C} \), (b) noncollinear output cone with \( \sim 14 \text{ mrad} \) full divergence \( T = 34.7^\circ\text{C} \), and (c) noncollinear output cone with \( \sim 24 \text{ mrad} \) full divergence \( T = 32.8^\circ\text{C} \). Propagation cones are slightly flattened on the sides due to crystal birefringence.

note that the operating conditions for the snapshots in Fig. 5 were not the same as those used in our numerical calculations for Figs. 2–4. In particular, the temperature difference of \( \sim 10^\circ\text{C} \) in Fig. 5 and Fig. 4(c), which show the same divergence angles, can be attributed to the pump wavelength difference of 0.06 nm and the large temperature bandwidth of \( \sim 30^\circ\text{C} \). Both rings have a diameter of \( \sim 3.6 \text{ mm} \) corresponding to emission cones with a full aperture of 24 mrad (after taking into account the imaging optics in the measurements), which is the same as that of the calculated emission cone of Fig. 4(c). We observe that the horizontally-polarized signal and the vertically-polarized idler cones overlap, as predicted by our numerical results.

As indicated in Fig. 4 and verified in Fig. 5, two overlapping cones of frequency-degenerate, orthogonally polarized photons are emitted at a temperature lower than that for collinear frequency-degenerate down-conversion. These photons are spectrally and spatially indistinguishable but they experience a differential timing delay due to the crystal birefringence. This

Fig. 5. Snapshots (in false colors) of (a) signal and (b) idler down-converted photons taken with a CCD camera. Images were taken with a 10-mm-long PPKTP crystal (9 \( \mu \text{m} \) poling period) kept at a temperature of 22\(^\circ\text{C}\) and pumped with a 398.54-nm continuous-wave pump. Images were taken with a 0.11-nm interference filter centered around the degenerate wavelength of 797.08 nm; exposure time: 20 s. Ring diameters are \( \sim 3.6 \text{ mm} \) corresponding to a full angle aperture of \( \sim 24 \text{ mrad} \).
delay, which is longer than the photons’ coherence time, can be corrected by inserting a KTP compensating crystal with a length equal to half of the PPKTP down-conversion crystal [12]. With the compensating crystal in place, temporal distinguishability of the output photons is eliminated and we expect the photons on opposite sides of the cones to be polarization entangled [6].

To verify that the single-crystal noncollinear configuration can indeed generate a cone of polarization-entangled photons, we used the experimental setup shown schematically in Fig. 6. A 398.6-nm loosely focused pump, derived from the second harmonic of a continuous-wave Ti:sapphire laser, was sent through a 10-mm-long hydrothermally grown PPKTP crystal with a grating period of 9 µm. The crystal was set on top of a thermoelectric heater and kept at a temperature of ∼32°C. Frequency-degenerate signal and idler photons were emitted in overlapping cones with a full aperture of ∼13 mrad. This divergence angle corresponds to the case of Fig. 4(b) which is calculated with a slightly different temperature due to the empirical nature of the Sellmeier equations. We collimated the cones with a lens (focal length, 150 mm) and spatially separated the two halves of the cone with a mirror. After going through irises that selected two opposite sections of the cones the down-converted photons passed through compensating crystals (5-mm-long KTP crystals with their y and z axes orthogonal to those of the PPKTP crystal) to eliminate the timing delays. The photons were analyzed by polarization analyzers, consisting of a half-wave plate and a polarizer, and filtered by 1-nm interference filters centered at 797 nm. Coincidence detection of the output photons was accomplished with single-photon counting modules (PerkinElmers SPCM-AQ14) and a fast electronic AND gate (coincidence window ≃ 2 ns). Accidental counts were negligible, given the low count rates (≤ 100,000 counts/s at each single photon counting module) and the narrow coincidence window.

Figure 7 shows the coincidence fringes we observed when the polarization analyzer angle θA in arm A (see Fig. 6) was varied while the other analyzer angle θB was fixed. For this data we used two irises with a diameter of 1 mm. The diameter of each iris was subtended by an angle of 6.7 mrad as seen from the crystal, which corresponds to a solid emission angle of 1.1 × 10⁻⁵ sr. The two collimated beams going through the irises had a 2-mm center-to-center separation that was subtended by an angle of 13.3 mrad as seen from the crystal. In Fig. 7 we show several sets of data taken for different values of θB: 0° (solid triangles), 45° (open diamonds), −45° (solid squares), and 90° (open circles). The curves in Fig. 7 are best fits to the data using the model coincidence function

\[
C (\theta_A) = A_1 \sin^2 (\theta_A - \theta_B) + A_2,
\]

where A1 and A2 are the fit parameters. The observed fringes indicate that, after the irises, the
generated state is the polarization-entangled biphoton triplet state

$$|\psi\rangle = \left( |H_A V_B\rangle + |V_A H_B\rangle \right) / \sqrt{2},$$

(7)

where the subscripts refer to the two detection arms (Fig. 6). A measure of the quality of the entangled state is given by the coincidence fringe visibility that can be obtained from the sinusoidal fits. We measure visibilities of $V_0 = 96 \pm 1\%$ for $\theta_B = 0^\circ$, $V_+ = 96 \pm 1\%$ for $\theta_B = 45^\circ$, $V_- = 96 \pm 1\%$ for $\theta_B = -45^\circ$, and $V_{90} = 94 \pm 1\%$ for $\theta_B = 90^\circ$; these visibility values indicate that the output state (7) was produced with high fidelity. We measure the $S$ parameter [19, 20] in the Clauser-Horne-Shimony-Holt (CHSH) version of Bell’s inequalities to be $S = 2.700 \pm 0.005$, thus violating the classical value of 2 by 140 standard deviations with a total measurement time of 160 s.

Figure 8 shows the dependence of the visibility $V_+$ on the iris aperture size. Opening the iris allowed more photons to reach the detector (as shown in the inset) but also decreased the visibility of the interference fringes as more photons that did not belong to the entangled cone were detected. For iris apertures larger than 2 mm the pair flux was limited by the size of the compensating crystal. For an aperture size of 1 mm and at $V_+ = 97 \pm 1\%$ we counted $\sim 820$ coincidences/s/mW of pump power. Observe that our collection system was not optimized to collect the entire entangled cone as was done in the system in Ref. [11]. We estimate that a more efficient collection system could increase the collected pairs by a factor of 3 without degrading the quality of the polarization entanglement.

The overlapping emission cone geometry we have presented is very similar to that used in Refs. [6] and [11]. Our setup is simpler than that presented in Ref. [6] in that we only need to use a single crystal. Compared with the setup in Ref. [11], we do not require a double passage of the pump beam through the crystal. The flux we have obtained is comparable to or larger than those obtained in Refs. [5, 6, 11, 12].
Ref. [5]) that allows us to use a long crystal without significantly degrading the quality of the generated polarization entanglement. Reference [13] reports a much larger flux than our current reported value but that experiment involves a more complicated interferometric setup that requires active stabilization.

In conclusion we have presented a source of polarization-entangled photons based on PPKTP. Using the unique properties of PPKTP we have been able to phase match the emission of frequency-degenerate, orthogonally polarized cones of signal and idler photons in an angularly noncritical geometry. The reported setup gives us the ability to generate a high-brightness cone of down-converted photons which show a high degree of polarization entanglement. This source is robust and simple and we believe it can be used in a variety of experiments that require the generation of highly entangled photon pairs.

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