Two-color photon polarization entanglement using a single nonlinear crystal

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We demonstrate a scheme for producing polarization-entangled photons of different wavelengths. The scheme is a variation of the conventional single-crystal, frequency-degenerate type-II scheme and offers similar brightness, quality of entanglement, and ease of alignment.

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The development of sources of photons suitable for handling quantum information [1] in various contexts is an increasingly important task, as different technologies for the various components of quantum networks [2]—quantum memory, quantum logic, quantum channels, and quantum repeaters—continue to develop. A particularly important resource in quantum information is entanglement [1,3]. Photons produced by spontaneous parametric down-conversion (SPDC) can be entangled simultaneously in several degrees of freedom: frequency (or time), momentum (or position), and polarization. In this process, high energy photons (typically provided by an ultraviolet “pump” laser) interact with an optically nonlinear crystal and are induced to decay into a pair of photons of lesser energy. Conservation of energy, momentum, and angular momentum constrains the emission process and results in the quantum correlations between the daughter photons. Polarization entanglement is a natural choice for implementing entangled quantum bits, since polarization is described by two orthogonal states and is easily measured and manipulated using standard optical components. While there have been many studies involving polarization-entangled photons (PEPs) of the same wavelength [3,4], there have been few involving photons of different wavelengths [5–9]. Nondegenerate PEPs are called for when the photons in each pair must travel through different media with different transmission windows, e.g., through optical fiber and the atmosphere, to reach their intended recipients. They may also be expected to serve a critical role as interfaces between quantum network components operating in various contexts is an important task, as different technologies for the various components of quantum networks [2]—quantum memory, quantum logic, quantum channels, and quantum repeaters—continue to develop. A particularly important resource in quantum information is entanglement [1,3]. Photons produced by spontaneous parametric down-conversion (SPDC) can be entangled simultaneously in several degrees of freedom: frequency (or time), momentum (or position), and polarization. In this process, high energy photons (typically provided by an ultraviolet “pump” laser) interact with an optically nonlinear crystal and are induced to decay into a pair of photons of lesser energy. Conservation of energy, momentum, and angular momentum constrains the emission process and results in the quantum correlations between the daughter photons. Polarization entanglement is a natural choice for implementing entangled quantum bits, since polarization is described by two orthogonal states and is easily measured and manipulated using standard optical components. While there have been many studies involving polarization-entangled photons (PEPs) of the same wavelength [3,4], there have been few involving photons of different wavelengths [5–9]. Nondegenerate PEPs are called for when the photons in each pair must travel through different media with different transmission windows, e.g., through optical fiber and the atmosphere, to reach their intended recipients. They may also be expected to serve a critical role as interfaces between quantum network components operating at different wavelengths [10]. In addition, allowing the photons to have different wavelengths allows one to access various dispersive regimes and thereby tailor the spectral correlations to different applications [11].

The typical scheme for producing PEPs makes use of a single crystal oriented to achieve type II phase matching in the forward (or near-forward) direction. The crystal is oriented so that the emission cones of ordinary- and extraordinary-polarized photons of the same wavelength intersect [12]. An alternate scheme involves two similar crystals oriented for type I phase matching, in which photons of one polarization are produced in the first crystal and are superposed with photons of the orthogonal polarization produced in the second crystal [13]. Sibling photons from either crystal need not have the same wavelength, provided that both crystals can emit photon pairs at the same two wavelengths into the same two spatial modes. As noted by Kwiat, however, it should be possible to observe two-color polarization entanglement in the single-crystal scheme by looking at different portions of the emission cones [14]. In fact, as we will discuss in detail below, the output distribution generally contains a continuous range of polarization-entangled wavelengths, from degenerate to nondegenerate. In this paper we demonstrate polarization entanglement of photons at different wavelengths produced using a single crystal with type II phase matching. In terms of brightness, degree of entanglement, and ease of alignment, this two-color PEP scheme compares well with the conventional, degenerate PEP scheme.

The quantum state of the electromagnetic field produced by spontaneous parametric down conversion can be written as

$$|\psi\rangle = |\text{vac}\rangle + \int A(k_{H\perp},\omega_H; k_{V\perp}, \omega_V) \times |1_{H,k_{H\perp},\omega_H} 1_{V,k_{V\perp},\omega_V} \rangle d^2k_{H\perp} d\omega_H d^2k_{V\perp} d\omega_V.$$  (1)

Here $1_p,k,\omega$ denotes a single photon with polarization $p$, wave vector component $k$ perpendicular to some chosen propagation axis, and angular frequency $\omega$. $H$ and $V$ denote horizontal and vertical polarization. $|\text{vac}\rangle$ denotes the vacuum (no-photon) state, which may be ignored in photodetection experiments. In the typical case that the nonlinear interaction is weak, terms involving more than two photons are negligibly small. Of primary interest is the joint probability amplitude $A$. In the case of a monochromatic, collimated pump, conservation of momentum and energy dictate that $k_{V\perp} = -k_{H\perp}$ and $\omega_V = \omega_{\text{pump}} - \omega_H$. In this case the joint amplitude can be expressed as function of the mode parameters of just one of the photons, say the horizontal photon

$$A(k_{H\perp}, \omega_H) \propto L \chi^{(2)} E_{\text{pump}} \text{sinc}(\Delta k L/2),$$  (2)

where $L$ is the length of the nonlinear crystal, $\chi^{(2)}$ is the nonlinear susceptibility, $E_{\text{pump}}$ is the pump field, and $\Delta k = (k_{\text{pump}} - k_{H\perp} - k_{V\perp})$ is the mismatch of the longitudinal components of the wave vectors, which is implicitly a function of $(k_{H\perp}, \omega_H)$. In writing Eq. (2), we suppress the fact that both $\chi^{(2)}$ and the constant of proportionality vary slightly with frequency. Figure 1 shows $|A(k_{\perp}, \omega)|^2$ and $|A(-k_{\perp}, \omega_{\text{pump}})$

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\( -\omega) \), which are the probability distributions of horizontally and vertically polarized photons, respectively, at several wavelengths. (Note that there is a direct mapping between frequency and wavelength, and between transverse momentum and angle of emission at a given wavelength.) One sees that there are several regions in which photons of given wavelength may be emitted with either polarization. These regions occur in pairs, such that if a photon is found to have a particular polarization in one region, a photon of the complementary polarization may be found in the other region. For example, an \( H \) photon may be emitted into region \( S \) and a \( V \) photon into region \( S' \), or a \( V \) photon into \( S \) and an \( H \) photon into \( S' \). The photons in regions \( S \) and \( S' \) exhibit polarization entanglement. The same is true of photons in regions \( R \) and \( R' \), but in this case the photons in each pair have different wavelengths.

In fact, all diametrically opposed regions along the dashed line in Fig. 1 show polarization entanglement. To understand this, we note that the distribution of \( H \) photons is characterized by the surface \( \Delta k (k_1 , \omega) = 0 \), which is the set of points \((k_1 , \omega)\) for which the phase matching envelope \( \text{sinc}(\Delta k L / 2) \) is maximized. The distribution of \( V \) photons is characterized by the surface \( \Delta k (\omega_p - \omega) = 0 \), which is the inversion of the first surface about the point of degeneracy \((\theta_p / 2)\). Wherever these surfaces intersect, photons of either polarization may be found, and polarization entanglement exists. Figure 2 shows a pair of surfaces typical of type II mixing. The spatial distribution of either photon at a fixed wavelength is a nearly circular ring. As wavelength increases, the ring radius increases for one of the polarizations and decreases for the other. (Recall that the surfaces are inverses of each other.) Provided that the rings at the degenerate wavelength cross each other, the surfaces will intersect at points along an ellipse such as that indicated by the dashed lines in Figs. 1 and 2. In this case there is a continuous set of modes exhibiting polarization entanglement.

For our experimental demonstration of nondegenerate polarization entanglement, we selected photons having the largest possible frequency difference at a given orientation of the crystal (photons from regions analogous to \( R \) and \( R' \) in Fig. 1). An argon ion laser, emitting 250 mW at 351 nm, pumped a 1-mm-long BBO crystal (Fig. 3). The pump beam, which was initially collimated with a \( w \) parameter of 0.8 mm, was weakly focused into the crystal with a lens of focal length 1000 mm. After the crystal, several dichroic mirrors reflected the pump beam while transmitting photons with wavelengths near 702 nm. A polarization rotator and birefringent optic (a second BBO crystal half the length of the first) were used to compensate for the spatial and temporal walkoff acquired by the photons in propagating through the first crystal. A beam-
splitter served to probabilistically send the photons in each pair in two different directions [19]. After passing through polarization rotators (achromatic half-wave plates) and polarizing beamsplitters, the photons were coupled into single-mode (SM600) optical fibers. The fiber tips were aligned to collect photons emitted from the first BBO crystal at angles 18 mrad above and below the pump. The divergence angle of the fiber modes was 7 mrad. The crystal was cut for degenerate emission in the forward direction. By tilting the crystal slightly (~0.1°), horizontally and vertically polarized photons near 690 nm were emitted into one fiber mode, while photons near 714 nm were emitted into the other. The photons collected by the fibers were detected by a pair of silicon detectors. The coincidence window was 4 ns. The count rates were approximately 3.5 × 10^4 s⁻¹ for photons of given polarization in the short- and long-wavelength arms, respectively. The count rates for the two arms were slightly different because the splitting ratio of the beam splitter was not exactly 50:50. In each arm, the count rate also depended slightly (~10%) on polarization, which is due to the fact that the H and V spatial distributions have different thicknesses at the collection regions (see Fig. 1) and couple to the fibers with different efficiencies.

To confirm that the collected photons were nongenerate, we temporarily inserted a monochromator (1 nm bandwidth) between one of the fibers and the corresponding detector, and measured the coincidence rate [20] as a function of wavelength (Fig. 4). The spectra of H and V photons in the one arm were, apart from overall scale, nearly identical and peaked at 690.4 nm. Because the frequencies of the photons in each pair must sum to the pump frequency, the photons in the other arm must have had complementary spectra peaked at 714.4 nm [21].

To demonstrate polarization entanglement, polarization correlations were measured at various angles. In experiments of this type, polarization-entangled photons will show strong correlations between horizontal and vertical polarizations and also between diagonal polarizations, whereas classically correlated (nontangled) photons would not. Typically, one measures the coincidence rate as a function of the angles of the two polarizers and reports the visibilities of the sinusoidal patterns that are observed. We found the coincidence rates for horizontal and vertical photons (not shown) to have a visibility of 99%, and coincidence rates for diagonally polarized photons (Fig. 5) to have a visibility of 87%. We note that the peak coincidence rates are not consistent across the entire range of the data. This is due to a wedge of 0.2 waves/mm in our waveplate, which caused the alignment between the photons and the fiber mode to vary as the plate was rotated. As confirmation of this hypothesis, we were able to restore any given peak to maximum value by a slight adjustment of the fiber positioner. The solid line, with a visibility of 97%, is the prediction of software developed by the author at ORNL. This software numerically evaluates the two-photon amplitude \( A(k_1, \omega_1; k_2, \omega_2) \approx \frac{\sin(\pi L/2)}{2} \exp[i(k_1 + k_2) \cdot L/2] \) and its projection onto a pair of Gaussian (fiber) modes. This calculation is performed for each of the polarization states \((H, V)\) and \((V, H)\), yielding two functions \( A_{HV}(\omega_1, \omega_2) \) and \( A_{VH}(\omega_1, \omega_2) \). The coincidence rate in the diagonal basis \((\theta = \pi/8)\) is then proportional to \( \int |A_{HV}|^2 \cos \alpha - |A_{VH}|^2 \sin \alpha \, d\omega_1 \, d\omega_2 \), where \( \alpha = 2(\theta - \theta') \) is the rotation of one photon relative to the other.

We are currently investigating the cause of the discrepancy between the predicted and observed visibilities. Spectral mismatch is ruled out by the fact that measured spectra have the same shape. We also believe that the photons are not distinguishable by arrival times. Based on the measured lengths of the crystals, the delays experienced by photons of different polarization are equal to within much less than the photon coherence length. One possible explanation for the lower visibility is that the superposition state is actually of the form \( |\psi\rangle = |HV\rangle + e^{i\phi}|VH\rangle \) with \( \phi \neq 0 \). This situation occurs when the optical path lengths from the crystal to the two detectors are not the same for the different polarization states—due, for example, to errors in the Retardance of the wave plates or to misalignment of the birefringent compensating crystal. In this case, polarization correlations would still be perfect in the \( H-V \) (unrotated) basis, but upon rotation the correlated polarization states would be elliptical; correlations between diagonal linear polarizations would be imperfect. Nevertheless, efforts to improve the interference visibility by introducing additional wave plates and/or adjusting the crystal alignment have so far not been successful.
The entanglement that can be achieved in this scheme is ultimately limited by the fact that the spatial distributions of \((H, V)\) and \((V, H)\) pairs have different widths and therefore couple to the fibers with unequal efficiency. The coupling efficiency depends on the overlap between the fiber modes (assuming single-mode fibers) and the spatial distribution of the joint amplitude [15,16]. By adjusting the divergence angle of the fiber modes, one can make the coupling efficiencies for the two sets of polarizations nearly equal, restoring the entanglement [17]. Figure 6 shows the predicted collection rates and interference visibility (entanglement) as a function of fiber mode divergence angle (assuming a Gaussian mode) for the wavelengths of Fig. 1 and for the pump of the experiment. When the divergence angle is less than about 4 mrad, the nondegenerate \((H, V)\) and \((V, H)\) pairs are predicted to couple with nearly equal efficiency and exhibit nearly maximal entanglement. Furthermore, the collection rates will be comparable to those in the degenerate case.

It is known that collection efficiency in the degenerate case can be improved by focusing the pump [18]. Our simulations predict that focusing the pump more tightly can increase the collection rate by a factor of at least 10, but focusing the pump too much increases the difference in the widths of the emission rings and reduces the entanglement. While other PEP sources also exhibit asymmetric broadening of the emission modes when the pump beam is focused, mode matching for these sources can be less sensitive to such spatial asymmetries [17]. Sources involving collinear propagation along a principal axis of a periodically poled crystal are particularly bright and offer good mode-matching capability [7,8]. It may also be noted that the wavelengths of the source demonstrated here are not as disparate as those of some other sources. Nevertheless, the nondegenerate scheme described here offers performance comparable to the conventional degenerate scheme involving a single bulk crystal. Furthermore, it is conceivable that this nondegenerate scheme could have even better performance under conditions that have yet to be explored. The range of available wavelengths and the angular distribution are determined by the shape of the phase-matching surface (Fig. 2), which in turn is determined by the dispersion of the material. In other phase-matching regimes or other materials it may be possible, for example, to generate polarization entanglement between visible and infrared wavelengths with near-maximal entanglement and high collection efficiency.

In conclusion, we have demonstrated a variation of the conventional type-II SPDC source, producing polarization-entangled photons at different wavelengths. Such a source compares well with conventional sources and may be useful for bridging dissimilar quantum information-handling components.

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[19] The beam splitter was not strictly necessary, since in this experiment the signal and idler photons were emitted in slightly different directions and could have been spatially separated.
[20] We measured the coincidence rate instead of the rate of the spectrally filtered arm alone, since we are interested in only those photons whose partners can also be detected in the other (unfiltered) arm.
[21] The optics in our setup are relatively spectrally uniform over the range of observed wavelengths; however, it is reasonable to imagine that the chain of spectral filters introduces some minor differences between the spectra above and below the degenerate wavelength.