PHYSICAL REVIEW LETTERS

Heralded Generation of Ultrafast Single Photons in Pure Quantum States

Peter J. Mosley,[1,*] Jeff S. Lundeen,[1] Brian J. Smith,[1] Piotr Wasylczyk,[1,2] Alfred B. U'Ren,[3]
Christine Silberhorn,[4] and Ian A. Walmsley[1]

[1]*Clarendon Laboratory, University of Oxford, Parks Road, Oxford, OX1 3PU, United Kingdom*
[2]*Institute of Experimental Physics, Warsaw University, ul. Hoza 69, 00-681 Warsaw, Poland*
[3]*Division de Fisica Aplicada, Centro de Investigacion Cientifica y de Educacion Superior de Ensenada,
Ensenada, Baja California, 22800 Mexico*
[4]*Max-Planck-Forschungsgruppe fur Optik, Gunther-Scharowsky Strasse 1/Bau 24, 91058 Erlangen, Germany*
(Received 30 October 2007; published 3 April 2008)

We present an experimental demonstration of heralded single photons prepared in pure quantum states from a parametric down-conversion source. It is shown that, through controlling the modal structure of the photon pair emission, one can generate pairs in factorable states and thence eliminate the need for spectral filters in multiple-source interference schemes. Indistinguishable heralded photons were generated in two independent spectrally engineered sources and Hong-Ou-Mandel interference observed between them without spectral filters. The measured visibility of 94.4% sets a minimum bound on the mean photon purity.

Optical quantum-information processing (QIP) either by linear gates or by the formation of cluster states relies on the coalescence of two independent photons incident on a beam splitter [1]. This Hong-Ou-Mandel interference (HOMI) [2] effect will only occur completely if the two photons are both pure and indistinguishable [3]. There are two common approaches to generating single photons: cavity quantum electrodynamics (CQED) in which a single emitter such as an atom, ion, quantum dot, or crystal vacancy is placed in a resonant optical cavity, and photon pair generation in a bulk medium, for example by four-wave mixing in a fiber [4] or by three-wave mixing in a nonlinear crystal [2]. However, it has not been proven that single photons generated by either technique can fulfill both requirements of purity and indistinguishability.

In this Letter we show that single photons derived from pair generation by parametric down-conversion (PDC) in a birefringent nonlinear crystal can be prepared directly in pure states. Usually, this purity is limited by correlations within each pair arising from the constraints of energy and momentum conservation between the pump and daughter fields [5]. Hence photon pairs are emitted into many highly correlated spatiotemporal modes and, due to these quantum correlations, the act of detecting one photon (known as heralding) projects the remaining single photon into a mixed state. This renders it useless for most quantum-information processing applications. A common strategy is to discard photons from the mixed state by spectral filtering to approximate a single mode [6–8]. This has the drawback that the purity of such a subensemble only approaches unity as the filter bandwidth tends to zero. Hence, to achieve high purity one must use narrow band filters, making the generation rate of photons in usable modes prohibitively low.

For any spectrally filtered source of photon pairs, the trade off between purity and count rate imposes a funda-mental limit on the production rate of heralded photons and, in the situation where *both* arms are filtered, also the heralding efficiency (see Fig. 1). However, in an ideal heralded single photon source all three parameters—purity, count rate, and heralding efficiency—must be high. If the purity is low then the fidelity of any operation will be poor, yet on the other hand, limited production rates result in any protocol taking a prohibitively long time. Simply reducing the filter bandwidth and increasing the pump power does not solve this problem. As one pumps harder, the background contribution from simultaneous generation of multiple pairs increases, reducing the visibility of any interference effects [4,9]. Furthermore, low heralding efficiencies combined with random fluctuations in the properties of consecutive photons from filtered sources have forced all previous implementations of optical QIP protocols to remain in the postselected regime—successful gate operations must be conditioned on the presence of the logical qubits. This problem of postselection is a well-known obstacle to scaling up QIP schemes [10]. Additionally, the problem of spectral filtering to eliminate correlations becomes more extreme as one moves to the production of higher photon numbers in each mode. Loss quickly degrades entanglement in photon number, required, for example, in continuous variable entanglement distillation. Here, the need for high-visibility interference between photons from independent sources as well as number entanglement within the emission from each source demands that the photon pairs are generated initially in states that are uncorrelated in all degrees of freedom other than number [11].

It has been shown theoretically that by careful control of the modes available for PDC, each pair of daughter photons can be free from any correlations in their spatiotemporal degrees of freedom [5]. The PDC emission modes are restricted by adapting the idea of vacuum mode engineer-
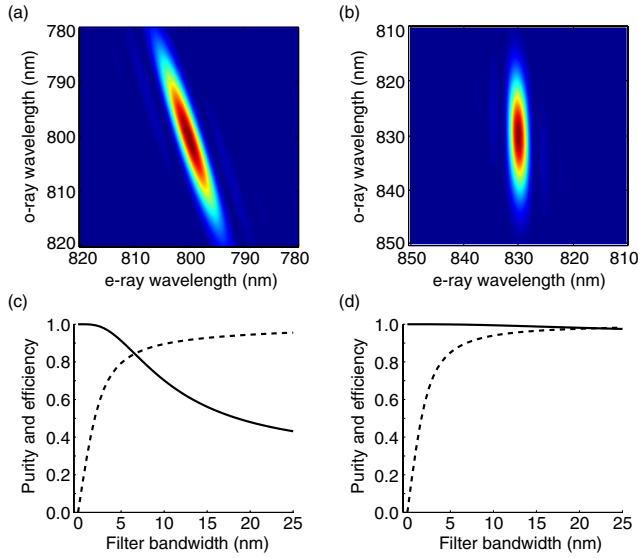
FIG. 1 (color online).   A numerically simulated comparison of collinear type-II pair generation in a 2 mm β-barium-borate (BBO) crystal pumped at 400 nm [(a),(c)] and a 5 mm potassium-dihydrogen-phosphate (KDP) crystal pumped at 415 nm [(b),(d)]. (a),(b) show the two-photon joint spectral probability distributions. Theoretical optimum heralding efficiency (dashed line) and purity of the heralded single photons (solid line) are plotted in (c) and (d) as a function of the bandwidth of filters in both PDC arms. We note that if we require a purity of 95%, for BBO the maximum possible heralding efficiency is necessarily reduced to 0.75, whereas for KDP this purity does not set an upper bound on the heralding efficiency as no spectral filtering is required. The pump was modeled as plane wave with a FWHM bandwidth of 4 nm.

ing, well known in CQED, to the case of bulk media and free space propagation. In CQED the cavity enclosing the single emitter ensures that it will radiate only into a single mode, however, a bulk medium contains many emitters. The coherent phasing of these determines the emission modes, which are generally numerous and highly correlated [12]. It is the multimode nature of the resulting fields that inevitably produces heralded photons in mixed states. In contrast, by choosing a nonlinear medium with the correct dispersion characteristics, we reduce the number of modes into which the down-converted fields can couple efficiently. Hence photon pairs can be created in uncorrelated states resulting in pure heralded single photons.

The two-photon component of the state emitted by a single down-converter can be written

$$|\Psi\rangle = \iint d\omega_e d\omega_o f(\omega_e, \omega_o)\hat{a}_e^\dagger(\omega_e)\hat{a}_o^\dagger(\omega_o)|0\rangle$$
$$= \sum_n \sqrt{\lambda_n}\, \hat{b}_n^\dagger \hat{c}_n^\dagger|0\rangle, \qquad (1)$$

where $\hat{a}_e^\dagger(\omega_e)$, $\hat{a}_o^\dagger(\omega_o)$ are the creation operators for photons in polarization modes e, o with frequencies ω_e, ω_o, respectively, $\hat{b}_n^\dagger$, $\hat{c}_n^\dagger$ are those for the Schmidt modes $\psi_n(\omega_e)$, $\phi_n(\omega_o)$, such that $\hat{b}_n^\dagger = \int d\omega_e \psi_n(\omega_e)\hat{a}_e^\dagger(\omega_e)$,

$\hat{c}_n^\dagger = \int d\omega_o \phi_n(\omega_o)\hat{a}_o^\dagger(\omega_o)$, and each mode has a weighting given by the Schmidt eigenvalue λ_n [5]. The Schmidt mode decomposition makes it clear that heralding with one photon from the state $\hat{\rho} = |\Psi\rangle\langle\Psi|$ projects the remaining photon into a state whose purity can be found from its reduced density operator $\hat{\rho}_e = \text{Tr}_o(\hat{\rho})$ to be $\text{Tr}(\hat{\rho}_e^2) = \sum_n\lambda_n^2$. This purity is determined by the factorability of the joint amplitude function $f(\omega_e, \omega_o)$ [13], but in general the Schmidt decomposition cannot be found analytically. Instead, one can calculate $f(\omega_e, \omega_o)$ numerically and compute its singular value decomposition, the matrix analogue of the Schmidt decomposition. The purity of the reduced state is then given by the sum of the squares of the singular values, which are equal to the Schmidt magnitudes [14]. Hence a necessary condition for pure heralded photons is a factorable joint amplitude, $f(\omega_e, \omega_o) = f_e(\omega_e)f_o(\omega_o)$, with only one spectral mode present.

The two-photon amplitude is given by the product of the pump distribution, $\alpha(\omega_e + \omega_o)$, with the phase-matching function of the nonlinear crystal, $\phi(\omega_e, \omega_o)$, so that $f(\omega_e, \omega_o) = \alpha(\omega_e + \omega_o)\phi(\omega_e, \omega_o)$. We select a pump wavelength and crystal for which the pump will propagate with the same group velocity as one of the daughter photons. Arranging the crystal dispersion this way sets $\phi(\omega_e, \omega_o)$ to be "vertical" in frequency space (see Fig. 2). Using an ultrafast, spectrally broad pump allows this phase-matching function to dominate the form of the two-photon amplitude, resulting in a factorable $f(\omega_e, \omega_o)$. An intuitive explanation for this is that traveling with equal group velocities forces one daughter photon to occupy the same narrow "time bin" as the pump pulse and therefore to be in a single broadband mode. Hence the timing jitter of each pair emission event is minimal. To fulfill this group velocity condition it is essential to have the flexibility provided by type-II phase matching where the pump photons decay into a pair in orthogonally polarized modes (e ray and o ray).

We have built a pair of sources exhibiting the characteristics required for pure-state generation by utilizing the dispersion characteristics of potassium-dihydrogen-phosphate (KDP) [15]. KDP has the property that the group velocity of e-polarized light at 415 nm is the same as that of o-polarized light at 830 nm, hence satisfying the group velocity matching condition for factorable states. This makes KDP the ideal crystal for this type of source as firstly it can be pumped by a frequency-doubled femtosecond Ti:sapphire laser, and secondly the daughter photons are easily detectable with silicon avalanche photodiodes. As spectral filters are not required, our sources deliver photon pairs at count rates comparable to those in other multisource down-conversion experiments (e.g., Refs. [6,8]) with an order of magnitude less pump power; per crystal, only 40 mW yields 6000 photon pairs per second.

In addition to being in pure states, we require single photons that are in well-defined spatial modes to imple-
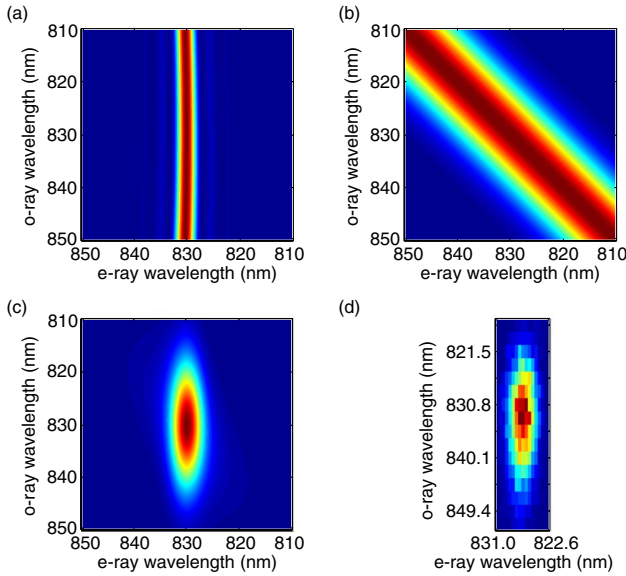
FIG. 2 (color online). Collinear phase-matching intensity (a) and pump intensity function (b) for a 5 mm KDP crystal and an ultrafast pump centered at 415 nm. The numerically calculated two-photon joint spectral probability distribution (c), with pump focusing and pair collection as per the experimental parameters. The measured two-photon joint spectral probability distribution is shown in (d); assuming no variation of the joint spectral phase, the singular value decomposition of the corresponding amplitude distribution gives a heralded photon purity of 98%.

ment linear optical gates and distribute across quantum networks. This is achieved by coupling the photon pairs into single-mode optical fiber. To obtain good coupling efficiency, one must properly focus the pump beam into the nonlinear crystals [16] and hence create preferential emission into a single spatial mode. The focusing conditions must be chosen carefully to maintain the factorability of the two-photon state; differences in the phase-matching conditions across the range of angles in the pump beam and emission pattern can introduce additional correlations not present in the case of plane-wave pumped KDP (illustrated in Fig. 1). Furthermore, the pump pulses from the second harmonic generation system are not transversely homogeneous. Efficient up-conversion of the Ti:sapphire laser requires tight focusing of the fundamental beam into the second harmonic generation crystal and the resulting range of phase-matching angles creates an output beam whose central frequency depends on this angle. The spread of frequencies in this spatially chirped pump beam is mapped to the down-conversion crystal so that each phase-matching angle sees a different pump wavelength. Careful matching of the sign and magnitude of the spatial chirp of the pump pulses allows the generation of states that have higher factorability than those that would result from homogeneous pump pulses without spatial chirp, while retaining favorable fiber coupling efficiencies. The required focusing and crystal parameters were determined using a numerical model based on Eq. (1), including the

spatial degree of freedom of the fields in addition to their frequency.

The form of the two-photon state from each crystal was verified directly by measuring the joint spectral probability distribution, $|f(\omega_e, \omega_o)|^2$. Photon pairs from one crystal were split and the photons sent to two grating monochromators. The coincidence rates were recorded for pairs of wavelengths to map the joint intensity distribution. Figure 2 shows that the measured distribution was indeed factorable and agreed well with the results of the numerical model.

Both the purity and indistinguishability of the heralded single photons were tested concurrently by interfering photons from two independent sources (Fig. 3). We observed the HOMI between the two heralded photons by monitoring the rate of fourfold coincidence events as a function of the relative optical delay in the two arms. The slow detectors integrated over the whole of each photon wave packet; therefore the detection was not time-resolved and no temporal filtering could take place. Although both sources were pumped by a single laser, one could use two lasers with no fixed phase relationship, the only requirement being that the pulses from both arrive simultaneously at the crystals [8,17].

Heralding with the o-ray photons, we obtained a HOMI dip with a visibility of $V_e = 94.4\% \pm 1.6\%$ between the e ray photons (Fig. 4). The visibility sets a lower bound on the average purity as it is equal to

$$V_e = \text{Tr}(\hat{\rho}_{e1}\hat{\rho}_{e2}) = \frac{\text{Tr}(\hat{\rho}_{e1}^2) + \text{Tr}(\hat{\rho}_{e2}^2) - O(\hat{\rho}_{e1}, \hat{\rho}_{e2})}{2}, \quad (2)$$
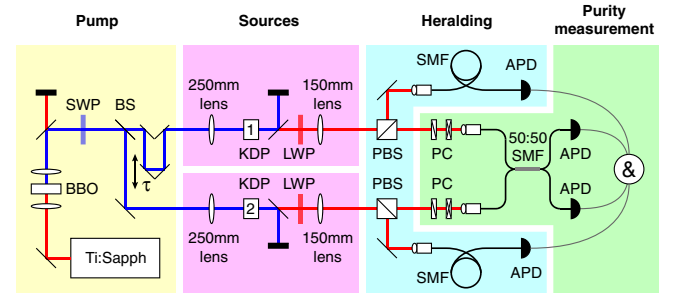

FIG. 3 (color online). A Ti:sapphire oscillator (830 nm, 50 fs, 76 MHz) was frequency doubled in a BBO crystal. After filtering out the fundamental frequency (SWP), the pulses were split at a 50:50 beam splitter (BS) to give a pair of 40 mW beams that were focused with two $f = 250$ mm SiO$_2$ lenses into two 5 mm long KDP crystals (labeled 1 and 2) cut for type-II phase matching at 830 nm. After the PDC crystals, the pump light was filtered out (LWP) and the photon pairs collimated with $f = 150$ mm lenses. The pairs were divided at two polarizing beam splitters (PBS). The two heralding photons were coupled into single-mode fibers (SMF) with $f = 15$ mm aspheric lenses and sent to silicon avalanche photodiodes (APD). The remaining single photons passed through half and quarter wave plates for polarization control (PC) and were directed to a 50:50 single-mode fiber coupler (50:50 SMF) with two APDs on the outputs. Fast electronics then monitored the coincidence rate between the four detectors. No narrow band spectral filters were used.
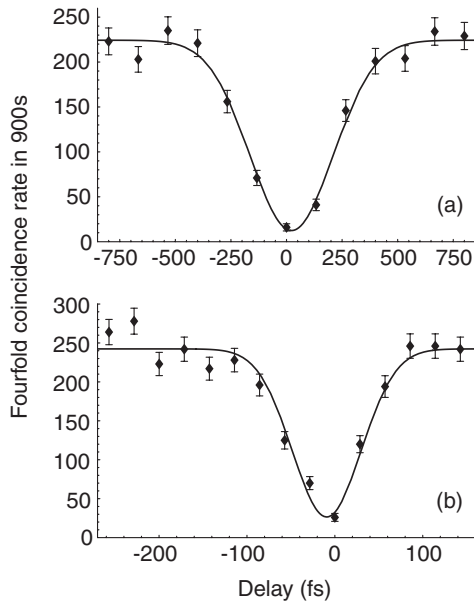
FIG. 4.   Interference of heralded photons from independent sources with no spectral filtering. Interference of e-ray photons (a) with $V_e = 94.4 \pm 1.6\%$, FWHM = 440 fs, and o-ray photons (b) with $V_o = 89.9 \pm 3.0\%$, FWHM = 92 fs. The o-ray visibility is reduced due to the insufficient bandwidth of the 50:50 coupler.

where $O(\hat{\rho}_{e1}, \hat{\rho}_{e2}) = \|\hat{\rho}_{e1} - \hat{\rho}_{e2}\|^2$ is the "operational distance" between the states of the two photons and ranges from 0 for perfect overlap to 2 for total distinguishability [18]. For two photons in pure states and single spatial modes, the operational distance is related to the overlap of their marginal spectral distributions by $O_{\text{pure}} = 2(1 - |\langle\psi_{e1}|\psi_{e2}\rangle|^2)$, where $|\psi_{ej}\rangle = \int d\omega_{ej} f_{ej}(\omega_{ej}) \hat{a}^{\dagger}_{ej}(\omega_{ej})|0\rangle$ with $j = 1, 2$ and the visibility is $V_{\text{pure}} = |\langle\psi_{e1}|\psi_{e2}\rangle|^2$. Measuring the e-ray photons' spectra, $F_{ej}(\omega_{ej})$, and calculating the overlap of the corresponding transform-limited pure states assuming $f_{ej}(\omega_{ej}) = \sqrt{F_{ej}(\omega_{ej})}$ yields $O_{\text{pure}} = 0.037$, a level of distinguishability that results in $V_{\text{pure}} = 98.1\%$. Because of the influence of this residual distinguishability, it is likely that our photons are of higher purity than the minimum bound set by the measured visibility, V_e. The FWHM of the e-ray dip was 440 fs, corresponding to a coherence time of 310 fs, commensurate with the spectra of the e-ray photons. In addition, we interfered the broadband o-ray photons, using the e rays as triggers. This gave a similar visibility ($89.9\% \pm 3.0\%$), but the interference dip was much narrower (92 fs FWHM) showing that the coherence time of the heralded o-ray photons was only 65 fs.

A benefit of using the e-ray photons to herald the o-ray photons is that a spectral filter in the e-ray arm can be used to increase the heralding efficiency by reducing the background without discarding any of the heralded photons themselves. The filter is chosen with a bandwidth similar to that of the e-ray photons (~4 nm) but narrower than the

background scatter. We have used this approach to measure detection efficiencies over 25%—very high for a pulsed PDC source using bulk optics. Taking into account the quantum efficiency of the signal detector (~60%), the heralding efficiency is almost 44%.

In conclusion, we have directly prepared fiber-coupled PDC photon pairs in factorable states. By successfully interfering almost identical photons produced simultaneously in two independent sources, we have shown that from these pairs we obtain single photons with a purity of approximately 95% at high heralding efficiencies (up to 44%). Because of its single-mode character, our source does not suffer from shot-to-shot fluctuations—the photons are delivered with precise timing and very low jitter. This source is of substantial importance for future photonic quantum-enhanced technologies, particularly for techniques requiring high photon number or multiple sources, where spectral filtering is ineffective.

*p.mosley1@physics.ox.ac.uk
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