Measurement of the spectral properties of the two-photon state generated via type II spontaneous parametric downconversion

Yoon-Ho Kim

Department of Physics, Pohang University of Science and Technology (POSTECH), Pohang 790-784, South Korea

Warren P. Grice

Center for Engineering Science Advanced Research, Computer Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

Received October 18, 2004

We report complete measurement of the spectral properties of photon pairs generated via spontaneous parametric downconversion. The measurements, which include not only single-photon spectra but also two-photon joint spectra, were performed for both cw and ultrafast-pumping configurations. In agreement with theoretical predictions, the spectra for the ultrafast-pumped case reveal asymmetries that are not present with cw pumping. © 2005 Optical Society of America

OCIS codes: 270.0270, 270.4180, 270.5290.

Entanglement between photons has played an important role in studies of the foundations of quantum physics and is relevant today in disciplines such as quantum information processing, quantum communication, and quantum-enhanced imaging and metrology. The most common method for generation of entangled photons is spontaneous parametric downconversion (SPDC).[1] Type II SPDC is of particular interest, not only because of the relatively narrow bandwidths of the photons but also because it is possible to generate polarization-entangled states directly.[2,3] Such states have been used in demonstrations of quantum cryptography and quantum teleportation, as well as in experimental tests of nonlocal quantum correlation, etc.[4–7] Although attention is often restricted to a particular type of entanglement, e.g., polarization, photons may be entangled in any of a number of degrees of freedom. As quantum technologies evolve, the need for more sophisticated two-photon states will grow, as will the need to characterize such states.

In this Letter we report the measurement of the spectral properties of the two-photon state. This measurement is of particular importance because it has been predicted that polarization-dependent spectral features can adversely affect the quality of polarization entanglement.[8,9] Although the theory on which this prediction is based has been found to be consistent with published experimental results,[10–12] to our knowledge no direct observation of these spectral differences has been reported previously. Before describing the experiment and results, we briefly review the theoretical description of type II SPDC.

The interaction Hamiltonian of the type II SPDC process may be written as

$$\mathcal{H} = \epsilon_0 \int \mathrm{d}^3\mathbf{r}\chi^{(2)} E_p(z,t) E_o^{(-)} E_e^{(-)} + \text{h.c.},$$

where the pump electric field $E_p(z,t)$ is considered classical. Operators $E_o^{(-)}$ and $E_e^{(-)}$ are the negative frequency parts of the quantized electric fields of the o-polarized (idler) and the e-polarized (signal) photons inside the crystal, respectively. The quantum state of type II SPDC can then be calculated by use of first-order perturbation theory $|\psi\rangle = -i/\hbar\int_{-\infty}^{\infty}\mathrm{d}t\mathcal{H}|0\rangle$, where $|0\rangle$ is the vacuum. Integrating over the length of the crystal, L, the quantum state is then calculated to be[8,9]

$$|\psi\rangle = C \int\int \mathrm{d}\omega_e \mathrm{d}\omega_o \mathcal{S}(\omega_e,\omega_o)\exp(-i\Delta L/2)a_e^\dagger a_o^\dagger|0\rangle,$$

where C is a constant, $\mathcal{S}(\omega_e,\omega_o) = \mathrm{sinc}(\Delta L/2)\mathcal{E}_p(\omega_e + \omega_o)$, $\Delta \equiv k_p(\omega_e+\omega_o) - k_o(\omega_o) - k_e(\omega_e)$, $a_e^\dagger = a^\dagger(\omega_e)$ is the creation operator for an e-polarized photon of frequency ω_e, and $a_o^\dagger$ is defined similarly. The pump pulse is described by $\mathcal{E}_p(\omega_e+\omega_o) = \exp[-(\omega_e+\omega_o - \Omega_p)^2/\sigma_p^2]$, where σ_p and Ω_p are the bandwidth and the central frequency of the pump pulse, respectively.

The function $|\mathcal{S}(\omega_e,\omega_o)|^2$, which is defined to be the joint spectrum of the two-photon state, is essentially a probability distribution for the photon frequencies.[8,9] Figure 1 shows plots of the calculated joint spectra for type II SPDC with two different

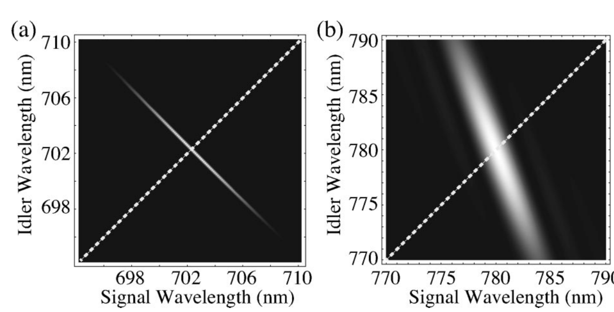

Fig. 1. Calculated joint spectrum functions for type II SPDC: (a) cw UV pumping and (b) ultrafast UV pumping. Dashed lines represent lines with the signal wavelengths equal to the idler wavelengths. Note the asymmetry in (b).

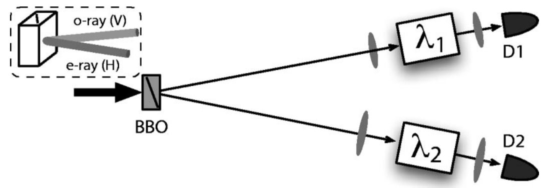

Fig. 2. Experimental setup: beamlike type II SPDC is used. λ_1 and λ_2 are monochromators.

pumping configurations. Figure 1(a) shows the calculated joint spectrum function for a 1-mm β-barium borate (BBO) crystal pumped by a 351.1-nm cw laser. For Fig. 1(b), the calculation is carried out for a 3-mm BBO crystal pumped by a frequency-doubled ultrafast laser. The pump is assumed to have a bandwidth of 2.5 nm and to be centered at 390 nm. (These pump wavelengths are typical in many SPDC experiments, and we use them in the subsequent experiment.) A comparison of the two plots reveals a couple of differences. Most notably, the plots differ both in the sharpness of the features and in the orientations of the joint spectra. The width of the joint spectrum is indicative of the degree of spectral entanglement between the two photons. Here the two-photon state from the cw-pumped crystal is more highly entangled, a consequence of the very narrow bandwidth of the pump field. For the ultrafast-pumped case, the spectral entanglement is not as strong. More importantly, the joint spectrum exhibits some asymmetry. This feature is a consequence of the asymmetric nature of the type II phase-matching conditions but is evident only when the pump is broadband. The asymmetry is believed to be the reason for the reduced quantum interference visibility observed in ultrafast type II SPDC.[8,9] The reduced visibility, which is consistent with the theoretical model described above, has been observed in experiments.[10–12] However, to our knowledge no direct measurements of $|\mathcal{S}(\omega_e,\omega_o)|^2$ have been reported previously.

Through a set of experiments we directly measured the joint spectra of type II SPDC for both cw UV and ultrafast UV laser pumping conditions. A schematic of the experiment is shown in Fig. 2. A BBO crystal is pumped either by an argon-ion laser (351.1 nm) or by the frequency-doubled output of a mode-locked Ti:sapphire laser (390 nm, $\delta\lambda \equiv 2.5$ nm). In each case the crystal is aligned for type II SPDC in a beamlike condition.[13,14] (See Fig. 1 in Ref. 14 for the emission pattern and the tuning curve of a beamlike type II SPDC.) In this configuration the frequency-degenerate photons propagate at a small angle with respect to the pump beam (see Fig. 2). The optical axis of the BBO crystal lies in the horizontal plane, and the pump laser beam is polarized horizontally. (The e ray of the crystal, therefore, is polarized horizontally.)

The BBO crystal was initially aligned to the beamlike configuration with two detectors (D_1 and D_2) with narrowband filters (10 nm) placed at the correct angles. Once the correct BBO setting was achieved, the narrowband filters were removed. For the spectral measurement, monochromators λ_1 and λ_2 were placed in front of each detector (see Fig. 2).[15] A lens focused the photons into the entrance slit, which is 200 μm wide. The exit slit of the monochromator is also 200 μm wide, and the output photons are collimated with a lens of the same focal length. Following a measurement of the single-photon spectrum of the idler photon (o ray), the same spectral measurement was performed on the conjugate photon, i.e., the signal photon (e ray), using monochromator λ_2 and D_2. The results of these measurements, which were carried out for both the cw UV and the ultrafast UV pulsed pump, are shown in Fig. 3. For both pumping schemes the SPDC photons were centered at the degenerate wavelengths: 702.2 nm in the cw-pumped case and 780 nm with the ultrafast pump. In addition, secondary nondegenerate peaks were also observed.[14] Comparison of the two pumping schemes shows features consistent with the theoretical descriptions discussed above. In particular, note that near the degenerate wavelengths the two photons from the cw-pumped configuration have identical spectra, whereas the ultrafast-pumped case yields a broader spectrum for one photon than for the other.

Although the single-photon spectra are instructive, they do not provide a complete picture. That is, they do not show how the two single-photon spectra are correlated. That information can be obtained only from a measurement of the joint spectrum. The joint spectrum was measured by scanning (0.4 nm each step) the two monochromators λ_1 and λ_2 simultaneously and recording the coincidence count rate be-

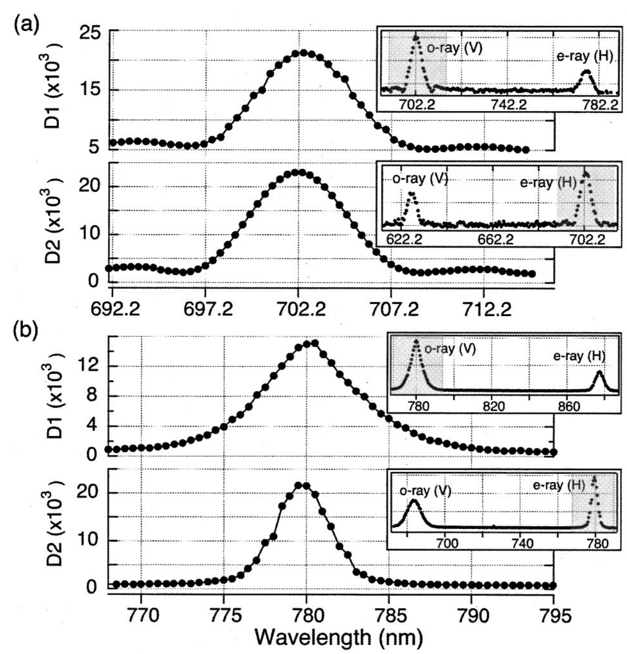

Fig. 3. Single-photon spectra of (a) cw UV pumped and (b) ultrafast UV pumped beamlike type II SPDC. Note that in (b) the single-photon bandwidths of the conjugate photons are noticeably different. Insets, full spectra of the SPDC. Entangled photons are found around the degenerate wavelengths, 702.2 or 780 nm. Secondary peaks at nondegenerate wavelengths, i.e., the e-ray peak at D_1 and the o-ray peak at D_2, are due to the tails of the tuning curves of the e ray and the o ray, respectively (see Fig. 1 in Ref. 14).

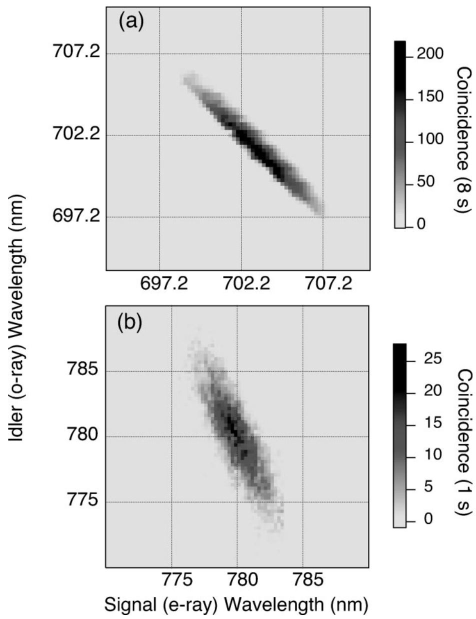

Fig. 4. Joint spectrum of type II SPDC for (a) cw UV pumping and (b) ultrafast UV pumping. The data are in good agreement with the theory plots shown in Fig. 1.

tween the two detectors. No bandwidth-limiting optics were placed in the paths of the photons. The results of these measurements are shown in Fig. 4. The experimental data agree well with the theoretical predictions shown in Fig. 1. It is clear from these plots that the single-photon spectra cannot adequately describe the spectral properties of the two-photon state. It is only when the spectra are measured in coincidence that the full picture begins to emerge. It can be seen, for example, that the spectral entanglement is weaker in the ultrafast-pumped case. That is, the cross section of the joint spectrum function at a given signal wavelength is much broader in ultrafast than in cw. This means that, for a given signal wavelength, a broader range of wavelengths is available for the idler. However, the reduced frequency entanglement in ultrafast type II SPDC is not necessarily unwelcome. The broadband pumping allows one to engineer the frequency entanglement of the two-photon state for specific quantum applications.[9]

Note that, by projecting the joint spectrum function either in the x or y axis, it is possible to obtain the single-photon spectrum for the signal or the idler photon. (This process is equivalent to integrating out one of the two frequency variables from the joint spectrum function $|S(\omega_e,\omega_i)|^2$.) By applying this to Fig. 4(b), one can see that the single-photon spectra for the signal and the idler photons would be different. This is consistent with the single-photon spectrum measurement shown in Fig. 3(b).

In summary, we have measured the joint spectra of the photon pairs produced via type II spontaneous parametric downconversion for both cw UV and ultrafast UV laser pumping conditions. The degree of spectral entanglement is explicitly revealed in these measurements, which confirm that the degree of frequency entanglement in ultrafast UV laser pumped type II SPDC is less than that of cw UV laser pumped type II SPDC, as predicted by theory. This observation, we believe, is an important first step toward engineered frequency entanglement for specific quantum applications such as quantum cryptography or multiphoton entanglement.[16]

This work was supported, in part, by the Advanced Research and Development Activity, the National Security Agency, the Korea Science and Engineering Foundation (grant R08-2004-000-10018-0), POSTECH Basic Science Research Institute Research Fund-2004, and a grant from the Ministry of Science and Technology of Korea. Oak Ridge National Laboratory is managed for the U.S. Department of Energy by UT-Battelle, LLC, under contract DE-AC05-00OR22725. Y.-H. Kim's e-mail address is yoonho@postech.ac.kr.

References

1. D. N. Klyshko, *Photons and Nonlinear Optics* (Gordon & Breach, New York, 1988).
2. P. G. Kwiat, K. Mattle, H. Weinfurter, A. Zeilinger, A. V. Sergienko, and Y. Shih, Phys. Rev. Lett. **75**, 4337 (1995).
3. Y.-H. Kim, S. P. Kulik, M. V. Chekhova, W. P. Grice, and Y. Shih, Phys. Rev. A **67**, 010301(R) (2003).
4. Y. H. Shih and C. O. Alley, Phys. Rev. Lett. **61**, 2921 (1988).
5. N. Gisin, G. Ribordy, W. Tittel, and H. Zbinden, Rev. Mod. Phys. **74**, 145 (2002).
6. C. H. Bennett, G. Brassard, C. Crepeau, R. Jozsa, A. Peres, and W. Wootters, Phys. Rev. Lett. **70**, 1895 (1993).
7. Y.-H. Kim, S. P. Kulik, and Y. Shih, Phys. Rev. Lett. **86**, 1370 (2001).
8. W. P. Grice and I. A. Walmsley, Phys. Rev. A **56**, 1627 (1997).
9. Y.-H. Kim and W. P. Grice, J. Mod. Opt. **49**, 2309 (2002).
10. W. P. Grice, R. Erdmann, I. A. Walmsley, and D. Branning, Phys. Rev. A **57**, R2289 (1998).
11. Y.-H. Kim, V. Berardi, M. V. Chekhova, and Y. Shih, Phys. Rev. A **64**, 011801(R) (2001).
12. Y.-H. Kim, S. P. Kulik, M. H. Rubin, and Y. Shih, Phys. Rev. Lett. **86**, 4710 (2001).
13. S. Takeuchi, Opt. Lett. **26**, 843 (2001).
14. Y.-H. Kim, Phys. Rev. A **68**, 013804 (2003).
15. In this experiment a 0.25-m monochromator (CS260 from Oriel) with a 600-line/mmgrating (blaze wavelength at 400 nm) and a 0.5-m monochromator (DK480 from CVI) with a 1200-line/mm grating blazed at 750 nm were used. For focusing and collimating the photons into and out of monochromators CS260 and DK480, a set of lenses with $f=25$ mm and $f=50$ mm were used, respectively.
16. W. P. Grice, A. B. U'Ren, and I. A. Walmsley, Phys. Rev. A **64**, 063815 (2001).