Experimental proposal for the generation of entangled photon triplets by third-order spontaneous parametric downconversion in optical fibers

María Corona,[1,2,*] Karina Garay-Palmett,[1] and Alfred B. U'Ren[1]

[1]*Instituto de Ciencias Nucleares, Universidad Nacional Autónoma de México, Apartado Postal 70-543, 04510 Distrito Federal, Mexico*

[2]*Departamento de Óptica, Centro de Investigación Científica y de Educación Superior de Ensenada, Apartado Postal 2732, Ensenada, Baja California 22860, Mexico*

**Corresponding author: maria.corona@nucleares.unam.mx*

Received August 27, 2010; revised December 6, 2010; accepted December 9, 2010; posted December 14, 2010 (Doc. ID 134114); published January 11, 2011

We present an experimental proposal for the generation of photon triplets based on third-order spontaneous parametric downconversion in thin optical fibers. Our analysis includes expressions for the quantum state, which describes the photon triplets and for the generation rate in terms of all experimental parameters. We also present, for a specific source design, numerically calculated generation rates. © 2011 Optical Society of America

OCIS codes: 190.4370, 190.4410.

The generation of multiphoton entangled states represents an important goal in quantum optics, both for fundamental tests of quantum mechanics and for quantum-enhanced technologies. Spontaneous parametric downconversion (SPDC) in second-order nonlinear crystals is the physical process of choice for most entangled photon pair experiments. Within the past decade, the process of spontaneous four-wave mixing (SFWM) based on the third-order nonlinearity in optical fibers has emerged as a viable alternative to SPDC [1]. The same third-order nonlinearity that makes the SFWM process possible could also serve as the basis for a different process: third-order spontaneous parametric downconversion (TOSPDC) [2–4]. While in the SFWM process two pump photons are annihilated in order to generate a photon pair, in the TOSPDC process a single pump photon is annihilated in order to generate a photon triplet. In this Letter, we describe a proposal for the generation of photon triplets in thin optical fibers.

A number of approaches for the generation of photon triplets have been either proposed or demonstrated with extremely low collection efficiencies, including triexcitonic decay in quantum dots [5], combined second-order nonlinear processes [6], and approximate photon triplets formed by SPDC photon pairs together with an attenuated coherent state [7]. The clearest demonstration of photon triplet emission, albeit with low count rates, is a recent remarkable experiment involving two cascaded second-order SPDC processes [8]. In contrast, our proposed technique permits the *direct* generation of photon triplets, without postselection, and a straightforward source modification would lead to the direct generation of three-photon Greenberger–Horne–Zeilinger (GHZ) states [9]. Our proposal likewise permits experimental studies of the largely unexplored three-particle continuous variable (spectral) entanglement.

Borrowing from second-order SPDC terminology, we refer to the three photons in a given TOSPDC triplet as signal-1 (r), signal-2 (s), and idler (i). Here we consider the TOSPDC process in an optical fiber, with the three generated photons in the same transverse mode and where all four fields are copolarized. It can be shown [10] that, in the spontaneous limit, the state of the emitted radiation can be written in terms of the three-photon component $|\Psi_3\rangle$ as

$$|\Psi\rangle = |0\rangle_r|0\rangle_s|0\rangle_i + \zeta|\Psi_3\rangle, \tag{1}$$

$$|\Psi_3\rangle = \sum_{k_r}\sum_{k_s}\sum_{k_i} G_k(k_r,k_s,k_i) \times \hat{a}^\dagger(k_r)\hat{a}^\dagger(k_s)\hat{a}^\dagger(k_i)|0\rangle_r|0\rangle_s|0\rangle_i, \tag{2}$$

written in terms of the wavenumbers k_μ for the three generated modes ($\mu = r,s,i$), the creation operator $\hat{a}^\dagger(k)$, and the joint amplitude function $G_k(k_r,k_s,k_i)$. For a pulsed pump, centered at ω_p^o, with bandwidth σ, and with a Gaussian spectral envelope $\alpha(\omega) = \exp[-(\omega-\omega_p^o)^2/\sigma^2]$, the quantity ζ (related to the conversion efficiency) is given by

$$\zeta = \left[\frac{2(2\pi)^{3/2}\epsilon_0^3 c^3 n_p^3 P L^2\gamma^2}{\hbar^2(\omega_p^o)^2\sigma}\right]^{1/2}(\delta k)^{3/2}. \tag{3}$$

ζ has been written in terms of the fiber length L, the pump peak power P, the refractive index $n_p \equiv n(\omega_p^o)$, the mode spacing δk, the vacuum electrical susceptibility ϵ_0, and the nonlinear coefficient γ, defined as

$$\gamma = \frac{3\chi^{(3)}\omega_p^o}{4\epsilon_0 c^2 n_p^2 A_{\text{eff}}}, \tag{4}$$

where $\chi^{(3)}$ is the third-order susceptibility and $A_{\text{eff}} = [\int \mathrm{d}x \int \mathrm{d}y f_p(x,y) f_r^*(x,y) f_s^*(x,y) f_i^*(x,y)]^{-1}$ is the effective interaction area, written in terms of the transverse distribution $f_\mu(x,y)$ for mode μ.

Writing $G_k(k_r,k_s,k_i)$ in terms of frequencies leads to the joint spectral amplitude $G(\omega_r,\omega_s,\omega_i) = \ell(\omega_r)\ell(\omega_s)\ell(\omega_i)F(\omega_r,\omega_s,\omega_i)$, where $\ell(\omega) = \sqrt{(\hbar\omega)/(\pi\varepsilon_0 n^2(\omega))}$ [$n(\omega)$ is the index of refraction] and where the function $F(\omega_r,\omega_s,\omega_i)$ is given as the product of the pump spectral amplitude (PSA) function $\alpha(\omega)$ and the phase-matching (PM) function $\phi(\omega_r,\omega_s,\omega_i)$, in turn given by

$$\phi(\omega_r,\omega_s,\omega_i) = \mathrm{sinc}\left[\frac{L}{2}\Delta k(\omega_r,\omega_s,\omega_i)\right] e^{i\frac{L}{2}\Delta k(\omega_r,\omega_s,\omega_i)}, \quad (5)$$

written in terms of the phase mismatch $\Delta k(\omega_r,\omega_s,\omega_i) = k_p(\omega_r+\omega_s+\omega_i) - k(\omega_r) - k(\omega_s) - k(\omega_i) + \Phi_{\mathrm{NL}}$, which contains a nonlinear term $\Phi_{\mathrm{NL}} = [\gamma_p - 2(\gamma_{pr}+\gamma_{ps}+\gamma_{pi})]P$, expressed in terms of a self-phase-modulation coefficient γ_p and cross-phase-modulation coefficients $\gamma_{p\mu}$.

From Eqs. (2) and (3), it can be shown [10] that the number of triplets generated per second, N, is given by

$$N = \lim_{\delta k\to 0}\sum_{k_r}\langle\Psi|a^\dagger(k_r)a(k_r)|\Psi\rangle R$$
$$= \frac{2^3 3^2 \hbar c^3 n_p^3}{\pi^2(\omega_p^o)^2}\frac{L^2\gamma^2 PR}{\sigma^2} \times \int d\omega_r \int d\omega_s \int d\omega_i \frac{k_r'\omega_r}{n_r^2}\frac{k_s'\omega_s}{n_s^2}\frac{k_i'\omega_i}{n_i^2}|F(\omega_r,\omega_s,\omega_i)|^2, \quad (6)$$

in terms of the pump repetition rate R, $k_\mu' \equiv k'(\omega_\mu)$, and $n_\mu \equiv n(\omega_\mu)$ (the prime denotes a frequency derivative).

The generation of frequency-degenerate photon triplets requires the fulfilment of $k(3\omega) = 3k(\omega)$. In general, however, this is not trivial to attain; the large spectral separation between the pump and the generated photons implies that $k(3\omega)$ is typically considerably larger than $3k(\omega)$. Our proposed solution exploits the use of two different transverse modes in a thin fiber guided by air, i.e., with a fused silica core and where the cladding is the air surrounding this core. In particular, we will assume that, while the TOSPDC photons all propagate in the fundamental mode of the fiber (HE_{11}), the pump mode propagates in the first excited mode (HE_{12}) [11].

Figure 1(a) shows that, for a particular degenerate TOSPDC frequency ω (corresponding to $\lambda = 1.596$ μm), a PM radius exists ($r = 0.395$ μm) for which $3k(\omega) = k(3\omega)$ is fulfilled. Note that, for the pump powers to be considered here, Φ_{NL} can be neglected. We could choose a different ω, which would lead to a behavior similar to that shown in Fig. 1(a), with a different resulting r; the dependence of r versus ω (expressed as wavelength) is shown in Fig. 1(b). It turns out that, as illustrated in Fig. 1(c), a smaller r [or smaller TOSPDC wavelength, according to the relationship shown in Fig. 1(b)] leads to a larger value for the nonlinearity γ. Therefore, in order to obtain a large photon triplet flux (which scales as γ^2) it is advantageous to use small r and consequently a large pump frequency 3ω. While this could suggest the use of a UV pump, in this Letter we avoid the use of nonstandard fiber-transmission frequencies. We have chosen for our illustration in Fig. 1(a) a pump at 532 nm, resulting in photon triplets near the telecommunications band. Note that the small fiber radii required by our technique can be obtained, with limited interaction lengths, through fiber taper technology.

Figure 1(d) shows the pump transverse intensity, in mode HE_{12}, evaluated at $\lambda_p = 1.596/3 = 0.532$ μm. Figure 1(e) shows the transverse intensity for the generated photons, in mode HE_{11}, evaluated at $\lambda = 1.596$ μm. For this wavelength combination, we obtain from Eq. (4) that $\gamma = 19$ $(\mathrm{kmW})^{-1}$. In general, only a fraction of the free-space pump power will be coupled to the HE_{12} mode; assuming a transverse Gaussian light distribution at the air–fiber interface, the maximum attainable coupling efficiency is 29.8% for the fiber considered here.

Figure 2 shows a representation of the three-photon joint spectral intensity plotted as a function of the TOSPDC frequencies. Figure 2(a) shows the PM function, Fig. 2(b) shows the PSA function, while Fig. 2(c) shows the joint intensity. Note that the tilted orientation of the joint intensity, resulting from a narrow width along the direction $\omega_r + \omega_s + \omega_i$ and much larger widths in the two transverse directions, indicates the presence of spectral correlations amongst the three photons, which underlie the existence of three-partite entanglement. Note that these plots are clear generalizations of similar

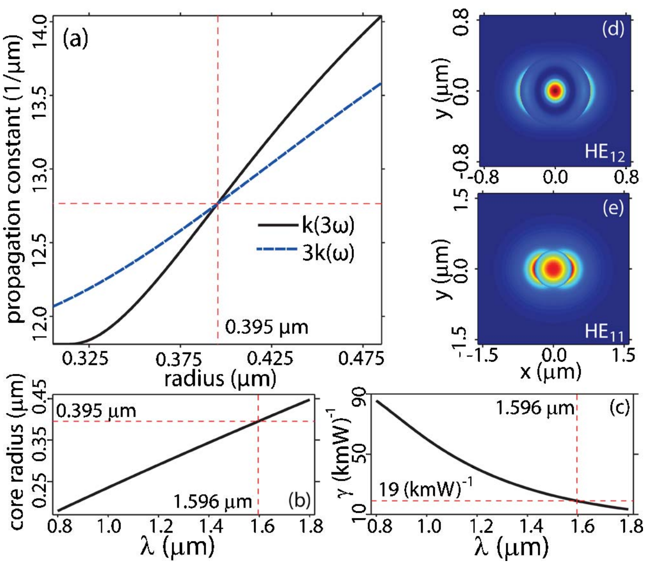

Fig. 1. (Color online) (a) $k(3\omega)$ and $3k(\omega)$ versus fiber radius. Plotted versus degenerate TOSPDC wavelength (b) PM radius and (c) nonlinearity γ. Transverse intensity for (d) the pump and (e) the TOSPDC photons.

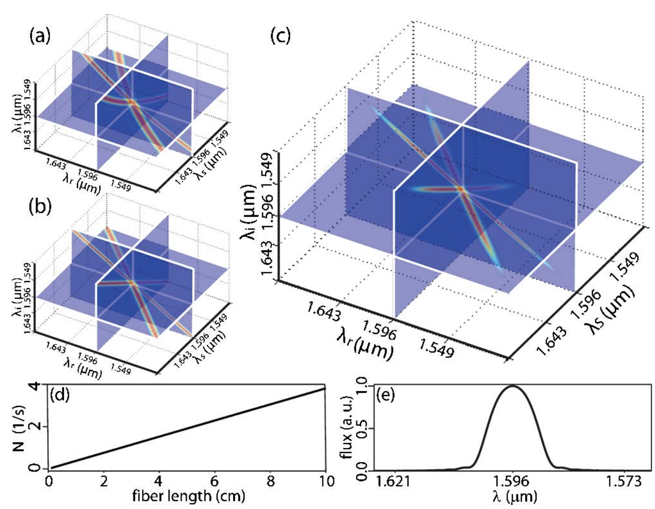

Fig. 2. (Color online) Representation, plotted versus the three generation frequencies, of the (a) PM, (b) PSA, and (c) joint intensity functions. (d) Generated flux versus fiber length. (e) TOSPDC single-photon spectrum.

plots for second-order SPDC [12]. We note that the values for L and σ (which control the widths of the PM and PSA functions, respectively) assumed in our source design, presented below, lead to acute spectral correlations which are difficult to plot clearly. Thus, in Figs. 2(a)–2(c) we have assumed $L = 0.6\ \mu$m and $\sigma = 5.1$ THz, which are considerably smaller and larger, respectively, compared to values assumed for our source design (see below) but that lead to the same qualitative behavior.

Evidently, a crucial consideration is the attainable source brightness, which we derive from numerical integration of Eq. (6); we assume $\sigma = 23.5$ GHz (which corresponds to a pulse duration of 100 ps), an average pump power of 200 mW, and $R = 100$ MHz. Figure 2(d) shows the number of generated photon triplets per second plotted as a function of the fiber length, exhibiting the expected linear dependence. We have limited the fiber length to $L = 10$ cm, for which we obtain a generation rate of 3.8 triplets per second; note that, based on recent publications, it is possible to obtain uniform-radius tapers with $\sim$445 nm radius over a 9 cm length [13]. Figure 2(e) shows, for a fiber length of $L = 10$ cm, the resulting single-photon spectrum for any of the three emission modes. Note that, because the three photons in a given triplet are in the same spatial and spectral mode, triplet splitting can only be attained probabilistically; in [10], we discuss spectrally nondegenerate TOSPDC.

There are some important differences between TOSPDC and SFWM. In common with second-order SPDC, the emitted flux in TOSPDC scales linearly with pump power and is constant (within the PM bandwidth) with respect to the pump bandwidth. In contrast, for SFWM the emitted flux scales quadratically with power and linearly with bandwidth [14]. Thus, SFWM sources tend to be significantly brighter than TOSPDC sources. Nevertheless, the source studied here leads to 3 orders of magnitude greater flux than that reported in [8]. Note also that while for many SFWM designs contamination due to spontaneous Raman scattering is a concern (if the signal and idler modes are within the Raman gain spectral window), the inherent large spectral separation between the pump and frequency-degenerate TOSPDC photons eliminates this concern.

Pump spectral broadening due to self-phase modulation can contribute to noise if pump and TOSPDC frequencies overlap. This may be controlled by exploiting the large pump-TOSPDC spectral separation and restricting the pump bandwidth and power. Further studies of noise mechanisms that could affect the performance of our proposed source may be necessary.

Note that a modified version of our source proposed here, with the TOSPDC fiber pumped in both directions, would permit the generation of GHZ states of the type $2^{-1/2}(|HHH\rangle + |VVV\rangle)$ (H/V represent horizontal/vertical polarization). This is similar to a scheme used for generating polarization-entangled photon pairs through SFWM [15].

We have presented an experimental proposal for the generation of photon triplets based on TOSPDC in thin optical fibers. We have shown that phase matching can be attained if the pump propagates in the mode HE_{12} while the generated photons propagate in the mode HE_{11}. We have presented an expression for the joint amplitude describing the photon triplets as well for the expected source brightness. We have presented a particular source design together with the expected generation rates. We expect that the technique presented will be useful for the exploration of multiphoton entangled states.

This work was supported by Consejo Nacional de Ciencia y Tecnología, Mexico; Dirección General de Asuntos del Personal Académico, Universidad Nacional Autónoma de México; and Fondo de Cooperación Internacional en Ciencia y Tecnología project 94142.

References

1. M. Fiorentino, P. L. Voss, J. E. Sharping, and P. Kumar, IEEE Photonics Technol. Lett. **14**, 983 (2002).
2. T. Felbinger, S. Schiller, and J. Mlynek, Phys. Rev. Lett. **80**, 492 (1998).
3. M. V. Chekhova, O. A. Ivanova, V. Berardi, and A. Garuccio, Phys. Rev. A **72**, 023818 (2005).
4. J. Douady and B. Boulanger, Opt. Lett. **29**, 2794 (2004).
5. J. Persson, T. Aichele, V. Zwiller, L. Samuelson, and O. Benson, Phys. Rev. B **69**, 233314 (2004).
6. T. E. Keller, M. H. Rubin, and Y. Shih, Phys. Rev. A **57**, 2076 (1998).
7. J. G. Rarity and P. R. Tapster, Phys. Rev. A **59**, R35 (1999).
8. H. Hübel, D. R. Hamel, A. Fedrizzi, S. Ramelow, K. J. Resch, and T. Jennewein, Nature **466**, 601 (2010).
9. D. M. Greenberger, M. A. Horne, A. Shimony, and A. Zeilinger, Am. J. Phys. **58**, 1131 (1990).
10. M. Corona, K. Garay-Palmett, and A. B. U'Ren are preparing a manuscript to be called "Theory for the process of third-order spontaneous parametric downconversion in optical fibers."
11. V. Grubsky and J. Feinberg, Opt. Commun. **274**, 447 (2007).
12. W. P. Grice and I. A. Walmsley, Phys. Rev. A **56**, 1627 (1997).
13. S. G. Leon-Saval, T. Birks, W. J. Wadsworth, P. St. J. Russell, and M. W. Mason, Opt. Express **12**, 2864 (2004).
14. K. Garay-Palmett, A. B. U'Ren, and R. Rangel-Rojo, Phys. Rev. A **82**, 043809 (2010).
15. For example: K. F. Lee, J. Chen, C. Liang, X. Li, P. L. Voss, and P. Kumar, Opt. Lett. **31**, 1905 (2006).