Quantum interference with femtosecond entangled two-photon fields

Jan Peřina, Jr.[a,b] *, Alexander V. Sergienko[b], Bahaa E. A. Saleh[b], and Malvin C. Teich[b]
[a]Joint Laboratory of Optics of Palacký University and the Academy of Sciences of the Czech Republic, 17. listopadu 50, 772 07 Olomouc, Czech Republic
[b]Quantum Imaging Laboratory, Department of Electrical and Computer Engineering Boston University, 8 Saint Mary's Street, Boston, MA 02215, USA

ABSTRACT

We investigate quantum interference effects of entangled two-photon states generated in a nonlinear crystal pumped by femtosecond pulses. Attention is devoted to the effects of the pump-pulse profile (pulse duration and chirp) as well as those originating in second-order dispersion, both in the nonlinear crystal and in the optical elements through which the down-converted photons propagate. The characteristics of the pump pulse, along with the dispersion, influence the visibility and the symmetry of the coincidence-count interference pattern. Nonlocal dispersion cancellation occurs in some cases.

Keywords: down-conversion, entangled two-photon interference, spontaneous processes, ultrafast nonlinear optics

1 INTRODUCTION

A great deal of attention has been recently devoted to the process of spontaneous parametric down-conversion in nonlinear crystals pumped by cw lasers.[1–4] The nonclassical properties of entangled two-photon light generated by this process have been used in many experimental schemes to elucidate distinctions between the predictions of classical and quantum physics.[2] A new frontier in these efforts is the generation of quantum states with three correlated particles (GHZ states),[5,6] which would be most useful for further tests of the predictions of quantum mechanics. One possibility is to construct such states from pairs of two-photon entangled states[7] which are synchronized in time, i.e. generated in a sharp time window. This can be achieved by using femtosecond pump beams. Successful quantum teleportation has been already observed using femtosecond pumping.[8]

For these reasons, a great deal of attention has recently been devoted both to theoretical and experimental investigations of the properties of pulsed spontaneous parametric down-conversion.[9–11] It has been shown that ultrashort pumping leads to a loss of visibility of the coincidence-count interference pattern at a beam splitter,[9,10] and narrowband frequency filters are required to restore the visibility.[7,9] In this contribution, we devote particular attention to the effects of pump-pulse chirp and second-order dispersion (in both the pump and down-converted beams)[12] on the visibility and shape of the photon-coincidence interference pattern produced at a beam splitter.[1] Dispersion cancellation, which has been extensively studied in the case of cw pumping,[13] is also predicted to occur under certain conditions for femtosecond down-converted pairs.

* e-mail: perina_j@risc.upol.cz

2 SPONTANEOUS PARAMETRIC DOWN-CONVERSION

Consider a nonlinear crystal pumped by a strong coherent-state field. Nonlinear interaction then leads to the spontaneous generation of two down-converted fields (the signal and the idler). The interaction Hamiltonian of the process can be written in the form[1]:

$$\hat{H}_{\rm int}(t) = \int_{-L}^{0} dz\, \chi^{(2)} E_p^{(+)}(z,t)\hat{E}_1^{(-)}(z,t)\hat{E}_2^{(-)}(z,t) + \text{h.c.}, \tag{1}$$

where $\chi^{(2)}$ is the second-order susceptibility, $E_p^{(+)}$ denotes the positive-frequency part of the electric-field amplitude of the pump field, and $\hat{E}_1^{(-)}$ ($\hat{E}_2^{(-)}$) is the negative-frequency part of the electric-field operator of down-converted field 1 (2). The nonlinear crystal extends from $z = -L$ to $z = 0$. The symbol h.c. means Hermitian conjugate.

Expanding the interacting fields into harmonic plane waves, the interaction Hamiltonian $\hat{H}_{\rm int}$ in Eq. (1) can be recast into the form:

$$\begin{aligned}\hat{H}_{\rm int}(t) &= C_{\rm int} \int_{-L}^{0} dz \sum_{k_p}\sum_{k_1}\sum_{k_2} \chi^{(2)} \mathcal{E}_p^{(+)}(0,\omega_{k_p} - \omega_p^0)\hat{a}_1^{\dagger}(k_1)\hat{a}_2^{\dagger}(k_2) \\ &\quad \times \exp\left[i(k_p - k_1 - k_2)z - i(\omega_{k_p} - \omega_{k_1} - \omega_{k_2})t\right] + \text{h.c.},\end{aligned} \tag{2}$$

where $C_{\rm int}$ is a constant. The symbol $\mathcal{E}_p^{(+)}(0,\omega_{k_p} - \omega_p^0)$ denotes the positive-frequency part of the envelope of the pump-beam electric-field amplitude at the output plane of the crystal; k_p stands for the wave vector of a mode in the pump beam, and ω_p^0 stands for the central frequency of the pump beam. The symbol $\hat{a}_1^{\dagger}(k_1)$ ($\hat{a}_2^{\dagger}(k_2)$) represents the creation operator of the mode with wave vector k_1 (k_2) and frequency ω_{k_1} (ω_{k_2}) in the down-converted field 1 (2).

The wave function $|\psi^{(2)}(t)\rangle$ describing an entangled two-photon state is given by:

$$|\psi^{(2)}(t)\rangle = \frac{-i}{\hbar}\int_{-\infty}^{t} dt' \hat{H}_{\rm int}(t')|\text{vac}\rangle, \tag{3}$$

where $|\text{vac}\rangle$ denotes a multimode vacuum state.

The description of a coincidence-count measurement with entangled states can be suitably formulated in terms of the two-photon amplitude $\mathcal{A}_{12}$ defined as a matrix element of the product of operators $\hat{E}_1^{(+)}(t_1)$ and $\hat{E}_2^{(+)}(t_2)$ sandwiched between the states $|\psi^{(2)}\rangle$ and $|\text{vac}\rangle$:

$$\mathcal{A}_{12}(t_1,t_2) = \langle \text{vac}|\hat{E}_1^{(+)}(t_1)\hat{E}_2^{(+)}(t_2)|\psi^{(2)}(t)\rangle. \tag{4}$$

The operator $\hat{E}_j^{(+)}$ of the positive-frequency part of the electric-field amplitude of the jth beam is defined as

$$\hat{E}_j^{(+)}(t_j) = \sum_{k_j} e_j(k_j) f_j(\omega_{k_j})\hat{a}_j(k_j)\exp(-i\omega_{k_j}t_j), \qquad j = 1,2, \tag{5}$$

where $e_j(k_j)$ denotes the normalization amplitude of the mode k_j, and $f_j(\omega_{k_j})$ characterizes an external frequency filter placed in the jth beam.

At the termination of the nonlinear interaction in the crystal, the down-converted fields evolve according to free-field evolution and the two-photon amplitude $\mathcal{A}_{12}$ depends only on the differences $\tau_1 = t_1 - t$ and $\tau_2 = t_2 - t$. When the down-converted beams propagate through a dispersive material of the length l, the two-photon amplitude $\mathcal{A}_{12}$ is given as follows:

$$\mathcal{A}_{12,l}(\tau_1,\tau_2) = C\int_{-L}^{0} dz \sum_{k_p}\sum_{k_1} f_1(\omega_{k_1}) \sum_{k_2} f_2(\omega_{k_2}) \mathcal{E}_p^{(+)}(0,\omega_{k_p}-\omega_p^0) \exp\left[i(k_p-k_1-k_2)z\right]$$
$$\times \exp\left[i(\tilde{k}_1+\tilde{k}_2)l\right] \delta(\omega_{k_p}-\omega_{k_1}-\omega_{k_2}) \exp[-i\omega_{k_1}\tau_1] \exp[-i\omega_{k_2}\tau_2]. \quad (6)$$

The amplitudes $e_1(k_1)$ and $e_2(k_2)$ from Eq. (5) are absorbed into the constant C and the wave vectors $\tilde{k}_j$ are appropriate for a dispersive material.

We assume the spectrum $\mathcal{E}_p^{(+)}(0,\Omega_p)$ of the envelope $\mathcal{E}_p^{(+)}(0,t)$ of the pump pulse at the output plane of the crystal in a Gaussian form:

$$\mathcal{E}_p^{(+)}(0,\Omega_p) = \xi_p \frac{\tau_D}{2\sqrt{\pi}\sqrt[4]{1+a^2}} \exp\left[-\frac{\tau_D^2}{4(1+a^2)}(1-ia)\Omega_p^2\right]. \quad (7)$$

The symbol ξ_p stands for the amplitude, τ_D is the pulse duration, and the parameter a describes the chirp of the pulse. The wave vectors k_p, k_1, k_2, $\tilde{k}_1$, and $\tilde{k}_2$ can be expressed in the following form including the effects of material dispersion up to the second order:

$$k_j(\omega_{k_j}) = k_j^0 + \frac{1}{v_j}(\omega_{k_j}-\omega_j^0) + \frac{D_j}{4\pi}(\omega_{k_j}-\omega_j^0)^2, \qquad j = p,1,2,$$
$$\tilde{k}_j(\omega_{k_j}) = \tilde{k}_j^0 + \frac{1}{g_j}(\omega_{k_j}-\omega_j^0) + \frac{d_j}{4\pi}(\omega_{k_j}-\omega_j^0)^2, \qquad j = 1,2, \quad (8)$$

where $1/v_j$ $(1/g_j)$ is the inverse of group velocity and D_j (d_j) stands for the second-order dispersion coefficient in the nonlinear crystal (dispersive material). The symbol ω_j^0 denotes the central frequency of beam j; $k_j^0 = k_j(\omega_j^0)$ and $\tilde{k}_j^0 = \tilde{k}_j(\omega_j^0)$. We further assume frequency filters with a Gaussian profile:

$$f_j(\omega_{k_j}) = \exp\left[-\frac{(\omega_{k_j}-\omega_j^0)^2}{\sigma_j^2}\right], \qquad j = 1,2, \quad (9)$$

where σ_j is the frequency width of the jth filter.

A typical experimental setup for coincidence-count measurement is shown in Fig. 1. We consider type-II parametric down-conversion. In this case two mutually perpendicularly polarized photons are provided at the output plane of the crystal. They propagate throught a birefringent material of a variable length l and then impinge on a 50/50 beamsplitter. Finally they are detected at the detectors D_A and D_B. The coincidence-count rate R_c is measured by a coincidence device C. The beams might be filtered by the frequency filters F_A and F_B which can be placed in front of the detectors. Analyzers rotated by 45 degrees with respect to the ordinary and extraordinary axes of the nonlinear crystal enable quantum interference between two paths to be observed; either a photon from beam 1 is detected by the detector D_A and a photon from beam 2 by the detector D_B, or vice versa.

In this experimental setup, the coincidence-count rate R_c is determined according to the relation:

$$R_c(l) = \frac{1}{4}\int_{-\infty}^{\infty} dt_A \int_{-\infty}^{\infty} dt_B \left|\mathcal{A}_{12,l}(t_A,t_B) - \mathcal{A}_{12,l}(t_B,t_A)\right|^2, \quad (10)$$

where the two-photon amplitude $\mathcal{A}_{12,l}$ is given in Eq. (6). The normalized coincidence-count rate R_n is then expressed as

$$R_n(l) = 1 - \rho(l), \quad (11)$$

where

$$\rho(l) = \frac{1}{2R_0}\int_{-\infty}^{\infty} dt_A \int_{-\infty}^{\infty} dt_B \,\mathrm{Re}\left[\mathcal{A}_{12,l}(t_A,t_B)\mathcal{A}_{12,l}^*(t_B,t_A)\right] \quad (12)$$

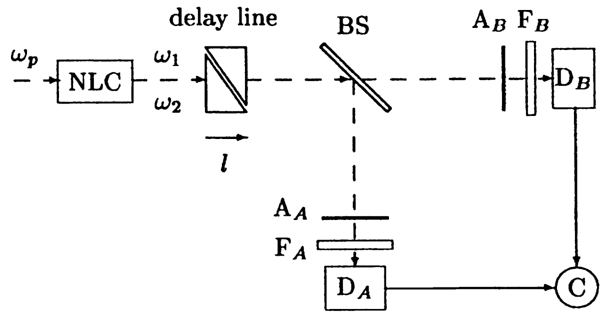

Figure 1: Sketch of the system under consideration: a pump pulse at the frequency ω_p generates down-converted photons at frequencies ω_1 and ω_2 in the nonlinear crystal NLC . These waves propagate through a delay line of length l and are detected at the detectors D_A and D_B; BS denotes a beamsplitter; A_A and A_B are analyzers; F_A and F_B are frequency filters; and C indicates a coincidence device.

and

$$R_0 = \frac{1}{2}\int_{-\infty}^{\infty} dt_A \int_{-\infty}^{\infty} dt_B \; |\mathcal{A}_{12,l}(t_A,t_B)|^2 . \tag{13}$$

The symbol Re denotes the real part of its argument.

3 DISCUSSION

We now proceed to examine the behavior of the normalized coincidence-count rate R_n on various parameters.

The profile of the interference dip in the coincidence-count rate[1] formed by the overlap of a pair of two-photon amplitudes can be understood as follows. The expression (12) for $\rho(l)$ can be rewritten in the form:

$$\rho(l) = \frac{1}{2R_0}\int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\tau \; \left[\mathcal{A}^{r}_{12,l}(t,\tau)\mathcal{A}^{r}_{12,l}(t,-\tau) + \mathcal{A}^{i}_{12,l}(t,\tau)\mathcal{A}^{i}_{12,l}(t,-\tau)\right] , \tag{14}$$

where $t = (t_A + t_B)/2$, $\tau = t_A - t_B$, $\mathcal{A}^{r}_{12,l} = \mathrm{Re}[\mathcal{A}_{12,l}]$, and $\mathcal{A}^{i}_{12,l} = \mathrm{Im}[\mathcal{A}_{12,l}]$. The symbol Im denotes the imaginary part of the argument. According to Eq. (14) the overlaps of the real and imaginary parts of the two-photon amplitudes $\mathcal{A}_{12,l}(t,\tau)$ and $\mathcal{A}_{12,l}(t,-\tau)$ determine the values of the interference term ρ. The amplitude $\mathcal{A}_{12,l}(t,-\tau)$ can be considered as a mirror image of the amplitude $\mathcal{A}_{12,l}(t,\tau)$ with respect to the plane $\tau = 0$. When only first-order dispersion in the optical material is taken into account, the shape of the two-photon amplitude $\mathcal{A}_{12,l}(t,\tau)$ does not depend on the length l; as l increases, the amplitude $\mathcal{A}_{12,l}(t,\tau)$ moves only in the t-τ plane. The shift in the τ-direction is important, because it changes the degree of overlap of the amplitudes and thus forms the shape of the dip.

The overlap of the two-photon amplitudes can be interpreted from the point-of-view of photon distinguishability.[9] When the overlap is complete, the detected photons cannot be distinguished and the interference pattern has maximum visibility. Incomplete overlap means that the photons can be "partially" distinguished and thus the visibility is reduced.

We further consider the role played by pump-pulse duration and chirp, second-order dispersion in the nonlinear down-converting medium, and second-order dispersion in the optical elements of the interferometer.

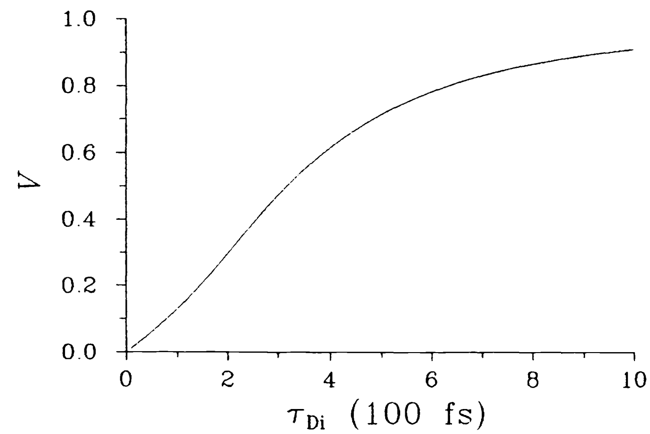

Figure 2: Visibility V ($V = \rho/(2-\rho)$) as a function of the pump-pulse duration τ_{Di}; $L = 3$ mm, $\sigma_1 = \sigma_2 = \infty$ nm, $a_i = 0$, and values of the other parameters are zero. In Figs. 2 and 3, the following parameters apply. Values of the inverse of group velocities appropriate for the BBO crystal with type-II interaction at the pump wavelength $\lambda_p = 397.5$ nm, and at down-conversion wavelengths $\lambda_1 = \lambda_2 = 795$ nm are: $1/v_p = 57.05 \times 10^{-13}$ s/mm, $1/v_1 = 56.2 \times 10^{-13}$ s/mm, and $1/v_2 = 54.26 \times 10^{-13}$ s/mm. We assume that the optical materials for the interferometer are quartz, for which $1/g_1 = 51.81 \times 10^{-13}$ s/mm and $1/g_2 = 52.08 \times 10^{-13}$ s/mm.

3.1 Role of pump-pulse parameters

It is well known that for a cw-pump field with no frequency filters included the coincidence-count rate $R_n(l)$ forms a triangular dip of width DL,[1] where $D = 1/v_1 - 1/v_2$. Visibility is 100%, reflecting maximum interference. An ultrashort pump pulse of duration τ_{Di} at the input plane of the crystal leads to a loss of visibility (see Fig. 2) but the width of the dip remains unchanged.[9] This can be understood from the shape[12] of the two-photon amplitude $A_{12,l=0}(t,\tau)$ which is confined in the τ-direction to the region $0 < \tau < DL$ for either cw or an ultrashort pump pulse; this confinement is responsible for the width of the dip. On the other hand the two-photon amplitude is confined by the ultrashort pump-pulse duration in the t-direction. The tilt of the amplitude in the t-τ plane leads to a loss of visibility since the overlap of the amplitudes $A_{12,l}(t,\tau)$ and $A_{12,l}(t,-\tau)$ for a given optimum value of l cannot be complete for a nonzero tilt. The shorter the pump-pulse duration, the smaller the overlap, and the lower values of visibility that result. Pump-pulse chirp (characterized by a_i at the input plane) introduces a phase modulation of the two-photon amplitude. This modulation decreases the overall overlap of the corresponding two-photon amplitudes, given as a sum of the overlaps of their real and imaginary parts. Increasing values of the chirp parameter a_i thus lead to a reduction of visibility. However, the width of the dip does not change. In fact, the visibility is determined by an effective pump-pulse duration τ_{eff} ($\tau_{\text{eff}} = \tau_{Di}/\sqrt{1+a_i^2}$) for a Gaussian pump-pulse profile. It can be shown that the dip remains symmetric for an arbitrary pump-pulse profile.

Frequency filters inserted into the down-converted beams broaden the two-photon amplitude both in the t- and τ-direction. Broadening in the τ-direction leads to wider dips, whereas that in the t-direction smooths out the effect of tilt discussed above and thereby results in a higher visibility. The narrower the spectrum of frequency filters, the wider the dip, and the higher the observed visibility.

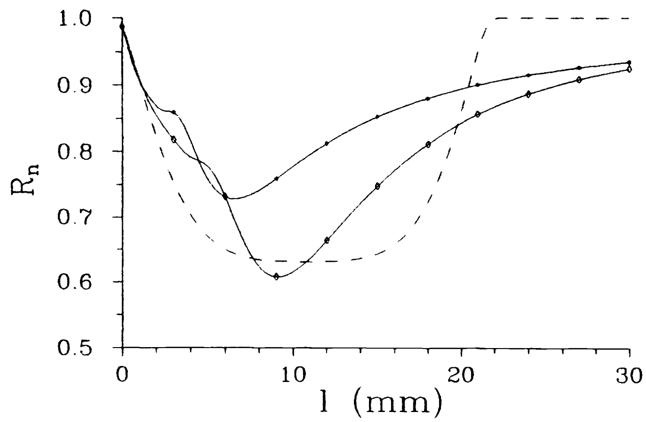

Figure 3: Coincidence-count rate $R_n(l)$ for various values of the second-order dispersion parameter $d = d_1 - d_2$ of an optical material; $d = 5 \times 10^{-26}$ s^2/mm ($\diamond$), $d = 1 \times 10^{-25}$ s^2/mm ($*$), and $d = 0$ s^2/mm (dashed curve, for comparison); $\tau_{Di} = 155$ fs, $L = 3$ mm, $\sigma_1 = \sigma_2 = 50$ nm, and values of the other parameters are zero.

3.2 Role of second-order dispersion in the nonlinear crystal

Second-order dispersion in the pump beam broadens the pump pulse as it propagates through the crystal and it also causes changes in its phase (chirp) as the pulse propagates. The effect of such pump-pulse broadening is transferred to the down-converted beams and modifies the interference dip as follows. An increase in the second-order dispersion parameter D_p leads to an increase of visibility, but no change in the width of the dip. For appropriately chosen values of D_p a small local peak emerges at the bottom of the dip. In the presence of narrow frequency filters the peak remains, but is suppressed.

Second-order dispersion in the down-converted beams (nonzero D_1, D_2) broadens the two-photon amplitude $A_{12,l}(t, \tau)$ both in the τ- and t-direction. This leads to a broadening of the dip, as well as an asymmetry and oscillations at its borders. Nonzero chirp results in a lower visibility, but tends to suppress oscillations at the borders of the dip. Frequency filters suppress asymmetry.

To observe the above mentioned effects caused by dispersion in a nonlinear crystal, rather high values of the dispersion parameters D_p, D_1, and D_2 are required. They are approximately an order of magnitude higher than those appropriate for the BBO crystals commonly used.

3.3 Role of second-order dispersion in optical elements comprising the interferometer

Second-order dispersion in an optical material (d_1, d_2) through which the down-converted photons propagate leads to asymmetry of the dip. The dip is particularly stretched to larger values of l (see Fig. 3) as a consequence of the deformation and lenghtening of the two-photon amplitude $A_{12,l}$ in a dispersive material. The higher the difference $d_1 - d_2$ of the dispersion parameters, the higher the asymmetry and the wider the dip; moreover its minimum is shifted further to smaller values of l (see Fig. 3). Asymmetry of the dip is also preserved when relatively narrow frequency filters are used though the narrowest filters remove it.

Asymmetry of the dip caused by second-order dispersion in an optical material can be suppressed in two cases. In the first case, for a pump pulse of arbitrary duration, dispersion cancellation occurs when the magnitude of second-order dispersion in the path of the first photon (given by $d_1 l$) equals that of the second photon (given by $d_2 l$). Dispersion cancellation is a result of completely destructive interference between the amplitudes $A_{12,l}(t, \tau)$ and $A_{12,l}(t, -\tau)$ for which there is nonzero overlap. When the pulse duration is sufficiently long (in the cw regime) dispersion cancellation occurs for arbitrary magnitudes of second-order dispersion present in the paths of the down-converted photons. Dispersion cancellation has its origin in the entanglement of photons.

4 CONCLUSION

We have developed a model of spontaneous parametric down-conversion produced by an ultrashort pump pulse. The model includes frequency modulation of the pump pulse (chirp) and dispersion in both the nonlinear crystal and optical material through which the down-converted photons propagate. The influence of these features on the depth and asymmetry of a photon-coincidence dip at a beamsplitter has been established. The higher the chirp parameter, the lower the visibility. Second-order dispersion of the pump beam in the nonlinear crystal may result in the occurrence of a local peak at the bottom of the dip. Second-order dispersion of the down-converted beams in the crystal results in oscillations at the borders of the dip. Second-order dispersion of the down-coverted photons through optical materials that comprise the interferometer (e.g., the delay line) leads to asymmetry of the dip. These effects can be used to measure parameters of a pump beam (duration and chirp parameter) as well as dispersion parameters of both a nonlinear crystal and an arbitrary optical material. Dispersion cancellation has been revealed i) for long pump pulses and ii) when the amount of dispersion in the two down-converted beams is identical (for pump pulses of an arbitrary duration).

5 ACKNOWLEDGEMENTS

The authors thank J. Peřina and M. Atature for valuable discussions. This work was supported by the National Science Foundation under Grant No. ECS-9800300 and by Grant No. VS96028 of the Czech Ministry of Education.

6 REFERENCES

[1] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics*, Cambridge Univ. Press, Cambridge, 1995.

[2] J. Peřina, Z. Hradil, and B. Jurčo, *Quantum Optics and Fundamentals of Physics*, Kluwer, Dordrecht, 1994.

[3] D. N. Klyshko, *Photons and Nonlinear Optics*, Gordon and Breach Science Publishers, New York, 1988.

[4] B. E. A. Saleh, A. Joobeur, and M. C. Teich, "Spatial effects in two- and four-beam interference of partially entangled biphotons", *Phys. Rev. A* **57**, pp. 3991-4003, 1998.

[5] D. M. Greenberger, M. A. Horne, A. Shimony, and A. Zeilinger, "Bell's theorem without inequalities", *Am. J. Phys.* **58**, pp. 1131-1143, 1990.

[6] D.M. Greenberger, M. A. Horne, and A. Zeilinger, "Multiparticle interferometry and the superposition principle", *Phys. Today* **46** (8), pp. 22-29, 1993.

[7] A. Zeilinger, M. A. Horne, H. Weinfurter, and M. Zukowski, " Three-particle entanglements from two entangled pairs", *Phys. Rev. Lett.* **78**, pp. 3031-3034, 1997.

[8] D. Bouwmeester, J.-W. Pan, K. Mattle, M. Eibl, H. Weinfurter, and A. Zeilinger, "Experimental quantum teleportation", *Nature* **390**, pp. 575-579, 1997.

[9] G. Di Guiseppe, L. Haiberger, F. De Martini, and A. V. Sergienko, "Quantum interference and indistinguishability with femtosecond pulses", *Phys. Rev. A* **56**, pp. R21-R24, 1997.

[10] T. E. Keller and M. H. Rubin, "Theory of two-photon entanglement for spontaneous parametric down-conversion driven by a narrow pump pulse", *Phys. Rev. A* **56**, pp. 1534-1541, 1997.

[11] W. P. Grice and I. A. Walmsley, "Spectral information and distinguishability in type-II down-conversion with a broadband pump", *Phys. Rev. A* **56**, pp. 1627-1634, 1997.

[12] J. Peřina, Jr., A. V. Sergienko, B. E. A. Saleh, and M. C. Teich, "Dispersion in femtosecond entangled two-photon interference", to be published.

[13] T. S. Larchuk, M. C. Teich, and B. E. A. Saleh, "Nonlocal cancellation of dispersive broadening in Mach-Zehnder interferometers", *Phys. Rev. A* **52**, pp. 4145-4154, 1995.

[14] B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics*, Wiley, New York, 1991.

[15] J.-C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena*, Academic Press, San Diego, 1996.