Appl. Phys. B 77, 803–807 (2003)
DOI: 10.1007/s00340-003-1321-5

Applied Physics B
Lasers and Optics

S. GAERTNER[1,2]
M. BOURENNANE[1,2]
M. EIBL[1,2]
C. KURTSIEFER[2]
H. WEINFURTER[1,2,✉]

High-fidelity source of four-photon entanglement

[1] Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany
[2] Sektion Physik, Ludwig-Maximilians-Universität, 80799 München, Germany

Received: 7 August 2003
Published online: 4 November 2003 •

ABSTRACT A polarisation-entangled four-photon state can be generated directly by a second order parametric down-conversion process. We use this emission to characterise the properties of a four-qubit state and to analyse its entanglement based on the violation of a four-particle Bell inequality. The observed high count rates and the fidelity of the polarisation correlations are the basis for the realisation of several new multiparty quantum communications schemes, such as secure multiparty key distribution and quantum telecloning.

PACS 03.67.Mn; 03.65.Ud; 42.50.Ar; 42.65.Lm

1 Introduction

The process of spontaneous parametric down-conversion (SPDC) currently offers the best way to generate entangled photon pairs. In this process, photons from an intense light beam are converted into pairs of daughter photons in an optically nonlinear material. In the conversion, conservation laws cause strong correlations between various properties of the generated photons. Particularly, type-II SPDC offers a method of directly generating pairs of polarisation-entangled photons [1]. The entanglement of such systems has been extensively studied and has been used for several implementations of quantum communication like quantum teleportation [2], quantum dense coding [3], and entanglement-based quantum cryptography [4], and for many tests of local hidden variable theories [5].

Recently, pulsed down-conversion enabled the simultaneous observation of more than just two photons, which formed the basis for the first experiments with 3- and 4-photon GHZ states [6], in which interferometric setups were used to generate the desired multiphoton entanglement out of two pairs of photons. However, the fragility of interferometric setups obstructs detailed investigations and possible applications in new multiparty quantum communication schemes [7–9].

Four-photon entangled states can be directly obtained from parametric down-conversion without overlapping single photons [10, 11]. In an extension to [11], we analyse here the properties of the four-photon state that is invariant under a simultaneous change of the four analysis directions. Due to its high symmetry, this state can be used for decoherence-free communication of quantum information and is the basis for quantum telecloning. In addition, a possible pair entanglement of the state could be ruled out based on the violation of a generalised Bell inequality [12]. The stability of the source together with the significantly increased rate and fidelity of the four-photon entanglement now enables for the first time the realisation of multiparty quantum communication.

2 Four-photon entanglement from parametric down-conversion

Analysing the process of SPDC, one observes that not only pairs of entangled photons can be emitted. The emission of four photons becomes possible in a second order process when two photons of the pump light are simultaneously down-converted. The state of four photons emitted into two spatial modes a_0 and b_0 can be written as

$$\frac{c^2}{2}(a^\dagger_{0H}b^\dagger_{0V} - a^\dagger_{0V}b^\dagger_{0H})^2\,|0\rangle\ , \tag{1}$$

where, for example, $a^\dagger_{0H}$ ($b^\dagger_{0V}$) is the creation operator for a horizontally (vertically) polarised photon in mode a_0 (b_0). The coefficient c includes the pump intensity as well as the nonlinearity and length of the down-conversion crystal. Even for large pump intensities, usually obtained with frequency-doubled mode-locked Ti:Saphire lasers, the rate of four-photon emission is low due to the small optical nonlinearity. Thus higher order contributions that generate more than four photons can be neglected.

The remarkable feature of the four-photon state (1) is that it is not simply the product of two entangled pairs. Due to their bosonic nature, the emission of two photons with identical polarisation into the same direction is twice as probable as the emission of two photons with orthogonal polarisation. This very fundamental interference causes entanglement between the four photons emitted by type-II SPDC (which also holds for other degrees of freedom causing mode, frequency, or time-energy entanglement between the four photons). Of course, this effect also causes entanglement for a higher number of photons. Eventually, one can expect that

✉ Fax: +49-89/2180-5032,
E-mail: harald.weinfurter@physik.uni-muenchen.de

experiments using either interferometrically enhanced down-conversion [13] or amplified laser systems will allow easier access to states with more than four photons in the future.

To make the four-photon entanglement accessible, we split each of the two outputs of the type-II SPDC by non-polarising beam splitters. Furthermore, we select events such that one photon is detected in each of the resulting four outputs (a, b, c, and d) of the beam splitters (Fig. 1). The state of the four detected photons is then given by [10]

$$\left|\Psi^{(4)}\right\rangle_{abcd} = \sqrt{\frac{1}{3}}\Big[\,|HHVV\rangle + |VVHH\rangle - \frac{1}{2}\left(|HVHV\rangle + |HVVH\rangle + |VHHV\rangle + |VHVH\rangle\right)\Big]_{abcd}. \tag{2}$$

The four entries in the kets describe the polarisation (H, horizontal; V, vertical) of the photons in the arms a, b, c, and d. We want to emphasise that this state inherits from the state of (1) particular properties, due to the linearity of the beam splitters. For example, the detection of two H-polarised photons in the arms a and b has the same probability as all possible combinations of finding orthogonally polarised photons in these arms, and, most importantly, the state exhibits four-photon entanglement. In order to obtain the particular form of the above state, it is necessary to compensate for birefringence in the SPDC source and for the beam splitters with compensation crystals right behind the SPDC crystal and additional quartz plates in the reflected output arms of the beam splitters (not shown in Fig. 1) such that the two-photon state $\left|\Psi^{-}\right\rangle$ can be observed between arms a and c, a and d, etc.

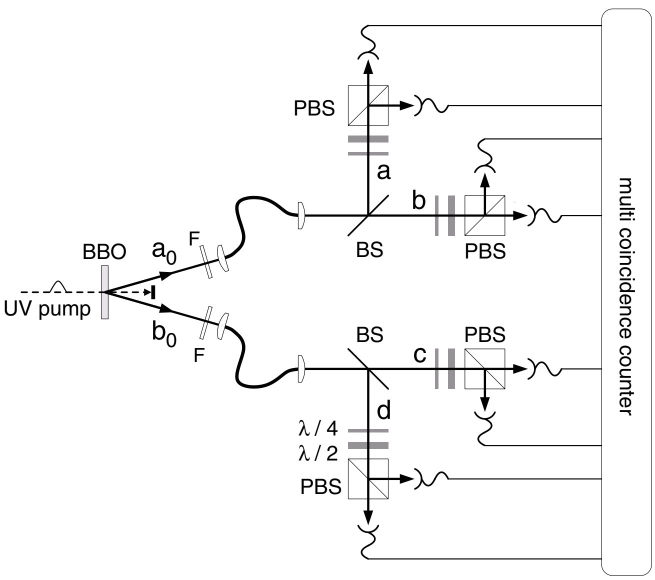

FIGURE 1 Experimental setup for observing four-photon entanglement obtained directly from type-II down-conversion. The four photons are emitted from the BBO crystal into two spatial modes a_0 and b_0, passed through 3-nm interference filters (F), and distributed into the four modes a, b, c, and d by 50%–50% beam splitters (BS). To characterise the polarisation-entangled four-photon state $\left|\Psi^{(4)}\right\rangle$ (2), a polarisation analysis in various bases is performed in each mode by using $\lambda/4$ and $\lambda/2$ plates in front of polarising beam splitters (PBS) and single photon avalanche detectors. Joint photodetection events in the four arms are recorded in a multi-coincidence unit

The state $\left|\Psi^{(4)}\right\rangle$ of (2) is a superposition of a four-photon GHZ state and a product of two Bell states

$$\left|\Psi^{(4)}\right\rangle_{abcd} = \sqrt{\frac{2}{3}}\,|GHZ\rangle_{abcd} - \sqrt{\frac{1}{3}}\left|\Psi^{+}\right\rangle_{ab}\left|\Psi^{+}\right\rangle_{cd}, \tag{3}$$

where the four-photon GHZ state is equal to

$$|GHZ\rangle_{abcd} = \frac{1}{\sqrt{2}}(|HHVV\rangle + |VVHH\rangle)_{abcd} \tag{4}$$

and the two Bell states are given by

$$|\Psi^{+}\rangle_{xy} = \frac{1}{\sqrt{2}}(|HV\rangle + |VH\rangle)_{xy}. \tag{5}$$

For the experimental observation of this four-photon polarisation entanglement it is necessary to ensure the selection of single spatial modes as well as to erase the possible frequency correlations of the original two pairs of photons by appropriate filters [14].

3 Experimental setup

In our experiment (Fig. 1) we used the UV pulses of a frequency-doubled mode-locked Ti:Sapphire laser (pulse length 130 fs) to pump a 2-mm-thick BBO crystal at a wavelength of 390 nm. The pump beam was focused to a waist of 100 μm inside the crystal and the repetition rate was 82 MHz with an average power of ≈ 730 mW. The degenerate down-conversion emission into the two characteristic type-II crossing directions was passed through narrowband interference filters ($\Delta\lambda = 3$ nm) and coupled into single mode optical fibers (length 2 m) to exactly define the spatial emission modes. Behind the fibers the down-conversion light was split at dielectric 50%–50% beam splitters into four distinct spatial modes a, b, c, and d. To investigate the four-photon state $\left|\Psi^{(4)}\right\rangle$, polarisation analysis in each of the four outputs behind the beam splitters was performed by a combination of quarter- and half-wave plates together with polarising beam splitters. The four photons were detected by single photon Si-avalanche diodes and registered with an eight-channel multi-coincidence logic. This analysis system simultaneously registered every possible coincidence between the eight detectors and thus allowed efficient registration of the 16 relevant four-fold coincidences. The different detectors exhibit different efficiencies due to production tolerances. Therefore, rates presented here are corrected for the separately calibrated detector efficiencies, and the errors given are deduced from propagated Poissonian counting statistics of the raw detection events.

4 Analysis of the four-photon state

The conditional detection of one photon in each of the four polarisation analysers now allows the properties of the four-photon state $|\Psi^{(4)}\rangle$ to be studied. Figure 2a shows the 16 possible four-fold coincidences when all four polarisation analysers are oriented along H/V. In excellent agreement with (2), the rates of the $HHVV$ and $VVHH$ events are, within the errors, equal to the sum of all events in which the two photons detected in arms a and b, or in arms c and d, have

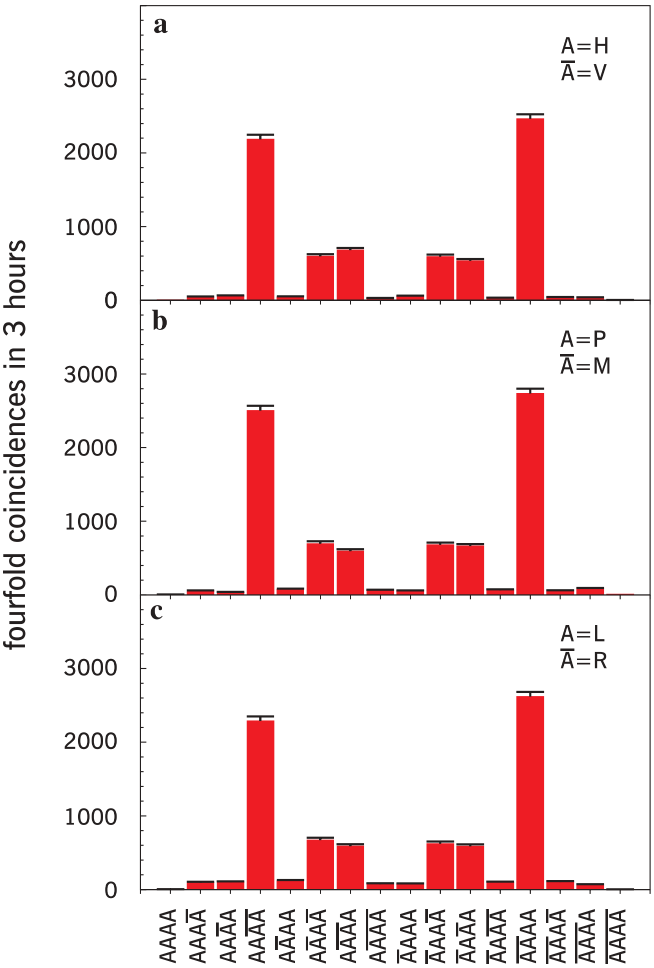

FIGURE 2 Four-fold coincidence counts corresponding to a detection of one photon in each of the four polarisation analysers in the H/V basis (**a**). The four-photon state $\left|\Psi^{(4)}\right\rangle$ exhibits two GHZ components with four times the counts than components corresponding to a product of EPR states. The state is invariant under a four-lateral unitary transformation and thus the characteristic coincidence pattern is the same when all four photons are analysed in the ±45° basis (**b**) or the left/right circularly polarised basis (**c**)

orthogonal polarisation. In these measurements we registered about 15000 twofold and 0.4–0.5 four-fold coincidences per second.

The four-photon state $\left|\Psi^{(4)}\right\rangle$ exhibits high symmetry. In particular, it is invariant under identical changes of detection bases for all four photons, expressed by the four-lateral unitary transformation

$$U^{\otimes 4}|\Psi^{(4)}\rangle_{abcd} = |\Psi^{(4)}\rangle_{abcd}\,, \tag{6}$$

where $U^{\otimes 4} = U_a \otimes U_b \otimes U_c \otimes U_d$ denotes the tensor product of four identical unitary transformations U. This property makes the state well suited for decoherence-free quantum information processing [18]. Experimentally the invariance can be demonstrated by joint identical basis transformation of each photon. Figure 2b and c show the four-photon coincidences for analysis along $+45°/-45°$ linear polarisation, and along left/right (L/R) circular polarisation. Due to the invariance, the four-fold coincidence pattern does not change under such transformations.

A characteristic property of entangled states is the possibility of obtaining perfect correlations between the measurement results of the four observers. For this purpose the observers in the four modes ($x = a, b, c, d$) perform measurements corresponding to a polarisation observable with eigenvectors $|l_x, \phi_x\rangle = \sqrt{1/2}(|R\rangle_x + l_x e^{i\phi_x}|L\rangle_x)$ and eigenvalues $l_x = +1, -1$. Quantum mechanics predicts for the correlation function (defined as the expectation value of the product of the four polarisation observables)

$$E(\phi_a, \phi_b, \phi_c, \phi_d) = \frac{2}{3}\cos(\phi_a + \phi_b - \phi_c - \phi_d) + \frac{1}{3}\cos(\phi_a - \phi_b)\cos(\phi_c - \phi_d)\,. \tag{7}$$

Figure 3 shows the dependence of the correlation function on the angle ϕ_b, for the other analysers fixed at angles $\phi_a = \phi_c = \phi_d = 0$, corresponding to H/V linear polarisation. The experimental value of the correlation function can be obtained from the 16 four-photon coincidence counts c_{l_a,l_b,l_c,l_d} via

$$E(\phi_a, \phi_b, \phi_c, \phi_d) = \frac{\sum_{l_a,l_b,l_c,l_d} l_a\, l_b l_c l_d \cdot c_{l_a,l_b,l_c,l_d}}{\sum_{l'_a,l'_b,l'_c,l'_d} c_{l'_a,l'_b,l'_c,l'_d}(\phi_a, \phi_b, \phi_c, \phi_d)}\,. \tag{8}$$

A least-squares fit of a sinusoidal function to the data gave a visibility of the correlation of $V_{\mathrm{H/V}} = 92.29\% \pm 0.83\%$. For similar measurements with $\phi_a = \phi_c = \phi_d = \pi/2$ we obtained $V_{\pm 45°} = 88.18\% \pm 1.18\%$, and for an observable including right/left circular polarisation we obtained $V_{\mathrm{R/L}} = 84.49\% \pm 0.75\%$. The high visibility for the observed correlation functions is a measure of the quality of our state preparation, and largely depends on the ratio between the spectral bandwidth of the down-conversion pump light and the spectral width of the detected photons [14]. Note that the analysis angles giving perfect correlations of $\left|\Psi^{(4)}\right\rangle$ are different from those of a four-photon GHZ state. Due to the EPR contributions, this state cannot be used in a GHZ-type argument refuting local hidden variable models of quantum mechanics. However, the high symmetry enables perfect correlations for all possible

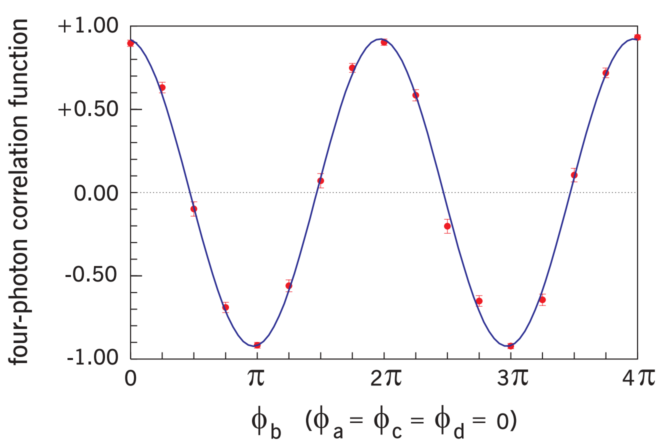

FIGURE 3 Four-photon polarisation correlation function for which the observer in modes a, c, and d analyzed their photons in the H/V-basis, while the observer in mode b varied the analysis angle. The *solid line* shows a fit to the experimental results, giving a visibility of $V = 92.29\% \pm 0.83\%$ compared with the theoretically expected value of 100% according to (7)

sets of common analysis directions, a feature which does not hold for GHZ states.

5 Violation of a four-party Bell inequality

The contribution of the product of the EPR states also changes the four-photon entanglement of $\left|\Psi^{(4)}\right\rangle$ relative to a GHZ state. However, the seemingly innocent question of how much entanglement is in this state cannot be answered for the moment, as clear measures of multiparticle entanglement are missing. Contrary to the case for two particles, multiparticle entanglement can be classified from numerous viewpoints still under discussion [15, 16]. Keeping in mind possible applications for multiparty quantum cryptography and secret sharing, we decided to analyse the state in terms of violation of a Bell inequality.

One can write down a Bell inequality which summarises all possible local realistic constraints on the correlation function for the case of each local observer measuring the polarisations along two alternative directions [10, 12]. Let us introduce the shorthand notation $E(\phi_a^k, \phi_b^l, \phi_c^m, \phi_d^n)$ for the correlation functions deduced from the observed count rates for the full set of 2^4 local directions, with $k, l, m, n = 1, 2$ denoting which of the two alternative phase settings was chosen by the local observer measuring in arm x $(x = a, b, c, d)$. The generalised Bell inequality gives an upper bound for the observed correlations in a local realistic description and reads [12]

$$S^{(4)} = \frac{1}{16} \sum_{s_a, s_b, s_c, s_d = \pm 1} \left| \sum_{k,l,m,n=1,2} s_a^k s_b^l s_c^m s_d^n \, E(\phi_a^k, \phi_b^l, \phi_c^m, \phi_d^n) \right| \leq 1 \,. \tag{9}$$

The maximal violation of this inequality for state (2) is obtained when three observers, $(x = a, c, d)$ perform polarisation analysis along $\pm\pi/4$ and the observer in mode b chooses between $\phi_b^1 = 0$ or $\phi_b^2 = \pi/2$. Then the quantum prediction is as high as $S_{QM}^{(4)} = 1.886$ [10] and results in a violation of the inequality (9) whenever the correlation function has visibility greater 53%. For a four-photon GHZ state one obtains $S_{QM}^{(4)} = 2\sqrt{2}$ and a critical visibility of $1/\sqrt{8} \approx 35\%$.

Figure 4 shows all 256 four-fold coincidence probabilities necessary for the analysis. They were recorded in blocks of 16 coincidence rates corresponding to the 16 phase settings (9), with an average measurement time of 1.5 h per frame. All together, the whole measurement including regular realignment and so on took 24 h. To evaluate the generalised Bell inequality we used the raw data without any correction for background, collection, or detection efficiency. The resulting value $S^{(4)} = 1.664 \pm 0.028$ clearly violates the boundary for local realistic theories and thus proves the entanglement of $\left|\Psi^{(4)}\right\rangle$. This value is also higher than the bound for bipartite entanglement ($S_{\text{bipartite}}^{(4)} \leq \sqrt{2}$) [16] and thus confirms that the observed state has at least tripartite entanglement. Yet, in order to unambiguously test four-particle entanglement, the Bell inequality is not suited, as there are tripartite entangled states giving values up to $S_{\text{tripartite}}^{(4)} \leq 2$. Although the possible tripartite entangled states do not exhibit the observed correlations and are thus ruled out by our measurements, the recently developed entanglement witness would be the proper tool [17].

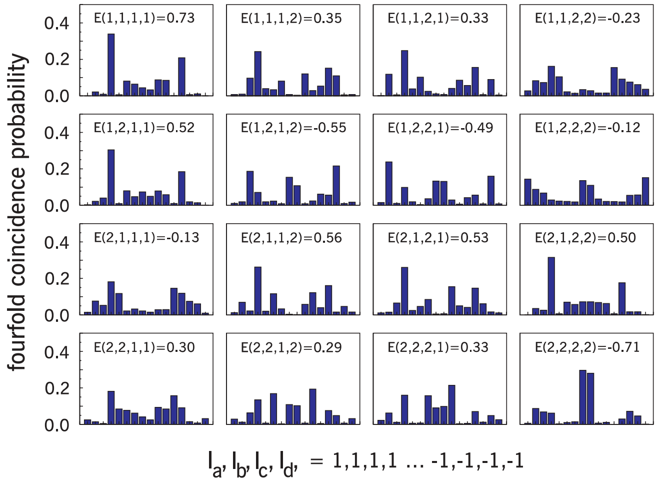

FIGURE 4 Four-fold coincidence probabilities for the evaluation of a four-particle Bell inequality. For the sixteen settings of the analyser phases ϕ_a, ϕ_b, ϕ_c, and ϕ_d, the normalised count rates $p_{k,l,m,n}$ obtained were used to evaluate a generalised Bell inequality (9), leading to $S^{(4)} = 1.664 \pm 0.028$. This value clearly exceeds the bound of 1 given for local realistic theories and proves the entanglement of the observed state. For this measurement the acquisition time for each individual frame was 1.5 h, with about 670 four-fold coincidence events per hour

6 Conclusion

In this contribution we characterised the properties of four-photon entanglement directly produced by parametric down-conversion. Bosonic interference occurring with the emission of two pairs is the origin for the particular properties of this four-photon state. The entanglement can be observed without additional interferometric setups. The high stability of the source enabled us to experimentally analyse the perfect correlations between measurement results of four observers and to perform a test of the entanglement based on the violation of a generalised four-photon Bell inequality. The high entanglement and visibility together with the ease of operation of this source shows its potential for multiparty quantum communication applications like secure three-party key distribution, two-party quantum telecloning [7], and multiparty quantum teleportation [8], and for decoherence-free quantum information processing [18] and solving the Byzantine agreement problem [19].

ACKNOWLEDGEMENTS This work was supported by the EU-Project RamboQ (IST-2002-6.2.1) and the Deutsche Forschungsgemeinschaft.

REFERENCES

1 P.G. Kwiat, K. Mattle, H. Weinfurter, A. Zeilinger: Phys. Rev. Lett. **75**, 4337 (1995)
2 D. Bouwmeester, J.-W. Pan, K. Mattle, M. Eibl, H. Weinfurter, A. Zeilinger: Nature **390**, 575 (1997)
3 K. Mattle, H. Weinfurter, P. Kwiat, A. Zeilinger: Phys. Rev. Lett. **76**, 4656 (1996)
4 T. Jennewein, C. Simon, G. Weihs, H. Weinfurter, A. Zeilinger: Phys. Rev. Lett. **84**, 4729 (2000); D.S. Naik, C.G. Peterson, A.G. White,

A.J. Berglund, P.G. Kwiat: Phys. Rev. Lett. **84**, 4733 (2000); W. Tittel, J. Brendel, H. Zbinden, N. Gisin: Phys. Rev. Lett. **84**, 4737 (2000)

5 G. Weihs, T. Jennewein, C. Simon, H. Weinfurter, A. Zeilinger: Phys. Rev. Lett. **81**, 5039 (1998); for a recent review see W. Tittel, G. Weihs: Quantum Inf. Comput. **1**, 3 (2001)

6 D. Bouwmeester, J.-W. Pan, M. Daniell, H. Weinfurter, A. Zeilinger: Phys. Rev. Lett. **82**, 1345 (1999); J.-W. Pan, M. Daniell, S. Gasparoni, G. Weihs, A. Zeilinger: Phys. Rev. Lett. **86**, 4435 (2001)

7 M. Murao, D. Jonathan, M.B. Plenio, V. Verdral: Phys. Rev. A **59**, 156 (1998)

8 W. Duer, J.I. Cirac: J. Mod. Opt. **47**, 247 (2000)

9 M. Zukowski, A. Zeilinger, M.A. Horne, H. Weinfurter: Acta Phys. Polon. **93**, 187 (1998)

10 H. Weinfurter, M. Żukowski: Phys. Rev. A **64**, 010 102-1 (2001)

11 M. Eibl, S. Gaertner, M. Bourennane, C. Kurtsiefer, M. Żukowski, H. Weinfurter: Phys. Rev. Lett. **90**, 200 403 (2003)

12 R.F. Werner, M.M. Wolf: Phys. Rev. A **64**, 032 112-1 (2001); M. Żukowski, C. Brukner: Phys. Rev. Lett. **88**, 210 401-1 (2002)

13 A. Lamas-Linares, J.C. Howell, D. Bouwmeester: Nature **412**, 887 (2001)

14 M. Zukowski, A. Zeilinger, H. Weinfurter: Ann. N. Y. Acad. Sci. **755**, 91 (1995)

15 A. Wong, N. Christensen: Phys. Rev. A **63**, 044 301-1 (2001); W. Dür, J.I. Cirac, R. Tarrach: Phys. Rev. Lett **83**, 3562 (2001); F. Verstraete, J. Dehaene, B. De Moor, H. Verschelde: Phys. Rev A **65**, 052112-1 (2002)

16 D. Collins, N. Gisin, S. Popescu, D. Roberts, V. Scarani: Phys. Rev. Lett. **88**, 170 405-1 (2002)

17 M. Horodecki, P. Horodecki, R. Horodecki: Phys. Lett. A **223**, 1 (1996); B.M. Terhal: Phys. Lett. A **271**, 319 (2000); M. Lewenstein, B. Kraus, J.I. Cirac, P. Horodecki: Phys. Rev. A **62**, 052 310 (2000); O. Gühne, P. Hyllus, D. Bruß, A. Ekert, M. Lewenstein, C. Macchiavello, A. Sanpera: Phys. Rev. A **66**, 062 305 (2002); M. Bourennane, M. Eibl, S. Gaertner, C. Kurtsiefer, H. Weinfurter, O. Guehne, P. Hyllus, D. Bruss, M. Lewenstein, A. Sanpera: quant-ph/0309043 (2003)

18 M. Bourennane, M. Eibl, S. Gaertner, C. Kurtsiefer, H. Weinfurter: quant-ph/0309041 (2003)

19 M. Fitzi, N. Gisin, U. Maurer: Phys. Rev. Lett. **87**, 217 901-1 (2001); A. Cabello: Phys. Rev. A **68**, 012 304-1 (2003)