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Entanglement of biphoton states: qutrits and ququarts

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Abstract. We investigate, in a general form, entanglement of biphoton qutrits and ququarts, i.e. states formed in the processes of, correspondingly, degenerate and non-degenerate spontaneous parametric down-conversion. Indistinguishability of photons and, for ququarts, joint presence of the frequency and polarization entanglement are fully taken into account. In the case of qutrits, the most general three-parametric families of maximally entangled and non-entangled states are found, and anticorrelation of the degree of entanglement and polarization is shown to occur and to be characterized by a rather simple formula. Biphoton ququarts are shown to be two-qudits with the single-photon Hilbert space dimensionality \(d = 4\), which differentiates them significantly from the often used two-qubit model \((d = 2)\). New expressions for entanglement quantifiers of biphoton ququarts are derived and discussed. Rather simple procedures for a direct measurement of the degree of entanglement are described for both qutrits and ququarts.
1. Introduction

The problem of providing meaningful entanglement quantifiers for systems of indistinguishable particles has a long history. Significant work was done and numerous papers were published on the subject [1–10]. Nevertheless, as is clear from analysis of these and other works, there is still no single unified and widely accepted approach to the problem. On the other hand, there is a highly developed field of quantum information, dealing with abstract distinguishable qubits. Entanglement of qubits is well understood (at least for the case when there are only two of them), and existing approaches to entanglement measures in two-qubit systems are generally accepted by the community. There is a number of experimental implementations of bipartite qubit systems as well, using various physical resources to encode quantum information. Quantum optical experiments are of great importance among them. Usually they make use of biphotons generated in the process of spontaneous parametric down-conversion, and all theoretical tools developed for qubits are straightforwardly applied to entanglement in various photonic degrees of freedom. We may be surprised by this fact: indeed, photons are indistinguishable bosons, and it is far from evident how one can construct two distinguishable qubits with two photons. Even less evident is the fact that entanglement of biphotons may be quantified with measures, derived for distinguishable qubits. Still we have something that is hard to argue about: the results of numerous experiments clearly show that under some experimental conditions biphotons behave like a system of qubits. This question is omitted in the majority of experimental works, and is usually addressed only in a general context. We believe that analysis of the situation considering simple and well-known objects, such as biphoton states with photons having only one or two discrete degrees of freedom, will be of interest for the community.

The key insight to understanding the mentioned ambiguities is provided in Zanardi’s work [5]. The simple idea is that entanglement can only be defined for a particular
decomposition of a complex system into subsystems, and the amount of entanglement quantified with some measure may depend on the chosen decomposition. The Hilbert space of a quantum system is a unitized object with no \textit{a priori} division into subsystems. It is up to us to choose such a division and to induce a corresponding tensor product structure, giving rise to possible entanglement. Entangled systems differ from separable ones in their behavior under local operations, but the mere definition of which operations we should consider as local relies on a particular choice of subsystems. Of course, sometimes such a choice is obvious, that is, for example, the case of two distinguishable particles. But this is not the case when particles are indistinguishable. In such cases, as we show, separation for subsystems can be done as separation in particles’ variables. Although less obvious and somewhat more abstract, such separation provides conditions for describing intrinsic quantum correlations in systems of indistinguishable particles, which coexists with other types of entanglement and is accessible for experimental measurements.

2. Main ideas and outline

Specifically, we consider in this paper two kinds of states: biphoton qutrits and biphoton ququarts. In the first case, two one-photon modes are characterized only by polarization \(\sigma = H, V\) (horizontal or vertical). Biphonon purely polarization states (qutrits) can be formed in combinations of processes of degenerate spontaneous parametric down-conversion (SPDC), and their state vectors are given by arbitrary superpositions of three basis state state-vectors \(\{ |2_H, 0\rangle, |1_H, 1_V\rangle, |0, 2_V\rangle \}\). Features of biphoton qutrits are discussed in sections 3–6 of this paper.

In the second case, the underlying process is the non-degenerate SPDC, and photon modes are characterized by polarization \(\sigma\) and some other quantum number \(k\), which can take only one of two values \(k_a\) or \(k_b\). The quantum number \(k\) can mean either photon frequency, or angle, determining possible directions of photon propagation, or anything else. In this case, there are four one-photon modes (\(|1_{k_a,\sigma}\rangle\) and \(|1_{k_b,\sigma}\rangle\)) and four two-photon modes (biphoton configurations or biphoton basis state-vectors \(|1_{k_a,\sigma_1}, 1_{k_b,\sigma_2}\rangle, \sigma_1, \sigma_2 = \{H, V\}\). Superposition of four biphoton basis state-vectors forms a biphoton ququart, features of which are discussed in detail in section 7.

It may be reasonable to emphasize that we do not consider here entanglement or correlations either between qutrits and between ququarts [11, 12] or in any multiphoton multimode states with amounts of photons \(>2\) [13]. We assume here that the pump is not too strong and gives rise only to rare two-photon degenerate or non-degenerate SPDC pairs. We analyze only intrinsic entanglement of one-qutrit or one-ququart states. We assume that such investigation is important because qutrits and ququarts are key elements in many physical processes. Internal entanglement of these objects is one of their main fundamental characteristics, and knowing it is very important, especially because many widespread common opinions and evaluations in this field that need corrections are shown below.

For any pure bipartite state its basis state-vector can be presented as a sum of basis state-vectors, each of which is assumed to correspond to a given configuration of particles’ occupation numbers in \(i\)th modes \(\{n_i\}\), \(|\Psi\rangle = \sum_{\{n_i\}} C_{\{n_i\}} \{|\{n_i\}\rangle\}.\) This gives rise to a density matrix

\[
\rho = \sum_{\{n_i\}, \{n_j\}} C_{\{n_i\}} C_{\{n_j\}}^* \{|\{n_i\}\rangle \langle \{n_j\}|\].
\] (2.1)
By definition [5–8], the state determined by this state vector $|\Psi\rangle$ and density matrix $\rho$ is entangled only if it does not have and cannot be reduced to the single-configuration form

$$|\Psi\rangle \neq |\langle n_i|\rangle_0, \quad \rho \neq |\langle n_i|\rangle_0 \langle n_i|\rangle_0,$$  

(2.2)

where the subscript ‘0’ indicates some single given configuration. The entanglement defined by conditions (2.2) can be referred to as configurational entanglement. This definition is absolutely unambiguous in the case of bipartite states with distinguishable particles when all bipartite modes are non-degenerate, because two different distributions of two distinguishable particles in two different modes are physically different and distinguishable and represent two different configurations of the particles’ distribution numbers. In the case of indistinguishable particles, such as photons, definition (2.2) is assumed to hold. But the question about the number of modes and configurations is not so clear. Let us discuss the situation that arises by using the simplest example of the state of two degenerate photons with different polarizations and the state vector $a_H^\dagger a_V^\dagger|0\rangle = |1_H, 1_V\rangle$. The typical answer for the question about the number of biphoton modes and configurations in this state is ‘one mode’ $\{H, V\}$, ‘one configuration’ and, hence, no entanglement. But, first, in some works [2, 10] the state $|1_H, 1_V\rangle$ is treated as entangled (similar to the two-electron state of the same kind [11]). And, second, it is rather difficult to draw conclusions about the numbers of modes by looking at only the state vector or density matrix in the symbolical operator form, $\rho = |1_H, 1_V\rangle\langle 1_H, 1_V|$. To reduce $\rho$ to the form of a normal matrix with clearly written matrix elements, let us first write down the orthogonality condition for biphoton purely polarization state-vectors given by

$$\langle 1_{\sigma_1}, 1_{\sigma_2}|1_H, 1_V\rangle = \langle 0|a_{\sigma_1}a_{\sigma_2}a_H^\dagger a_V^\dagger|0\rangle = \delta_{\sigma_1,H}\delta_{\sigma_2,V} + \delta_{\sigma_1,V}\delta_{\sigma_2,H}. \quad (2.3)$$

Note that this expression follows directly from commutation rules for the photon creation and annihilation operators, and it would be wrong to leave only one term ($\delta_{\sigma_1,H}\delta_{\sigma_2,V}$) on its right-hand side (whereas the single-configuration description corresponds precisely to this approximation: only one term left in expressions like that of equation (2.3)). With the orthogonality relation (2.3) taken into account, we find that the matrix elements of the density matrix $\rho$ have the form

$$\langle 1_{\sigma_1}, 1_{\sigma_2}|\rho|1_{\sigma_1'}, 1_{\sigma_2'}\rangle = (\delta_{\sigma_1,H}\delta_{\sigma_2,V} + \delta_{\sigma_1,V}\delta_{\sigma_2,H})(\delta_{\sigma_1,H'}\delta_{\sigma_2,V'} + \delta_{\sigma_1,V}\delta_{\sigma_2,H'}). \quad (2.4)$$

Definitely, this matrix describes the entangled state. In terms of the number of modes and configurations in the state $|1_H, 1_V\rangle$, expressions (2.3) and (2.4) correspond to a two-mode and two-configuration state. To explain this result, we suggest an interpretation that is different from the traditional one. Instead of speaking about a single biphoton mode $\{H, V\}$, we say that the state we consider is characterized by two degenerate biphoton modes, $\{H, V\}$ and $\{V, H\}$. ‘Degenerate’ means in this case that the state vectors corresponding to these modes are identical, $|1_H, 1_V\rangle \equiv |1_V, 1_H\rangle$. Of course, the modes $\{H, V\}$ and $\{V, H\}$ are physically indistinguishable. But their existence is displayed clearly in the matrix elements (2.3) and (2.4). The suggested interpretation is valid not only for the state $|1_H, 1_V\rangle$ but also for any other states of two photons in different single-photon modes, as well as for the general-form biphoton qutrits and ququarts, and for any biphoton states of higher dimensionality. In all cases, the key point is in the correct calculation of matrix elements as shown in equation (2.3).

The same conclusions about entanglement of states like $|1_H, 1_V\rangle$ can be obtained from consideration in terms of ‘coordinate’- dependent basis wave functions and their superpositions.
For arbitrary bipartite states, the basis wave functions are defined as \( \Psi_{\{n_i\}}(x_1, x_2) = \langle x_1, x_2|\{n_i\} \rangle \) and their superposition is given by

\[
\Psi(x_1, x_2) = \sum_{\{n_i\}} C_{\{n_i\}} \langle x_1, x_2|\{n_i\} \rangle = \sum_{\{n_i\}} C_{\{n_i\}} \Psi_{\{n_i\}}(x_1, x_2). \tag{2.5}
\]

The coordinate-dependent density matrix following from equation (2.1) has the form

\[
\rho(x_1, x_2; x_1', x_2') = \sum_{\{n_i\},\{n_i'\}} C_{\{n_i\}} C_{\{n_i'\}}^* \langle x_1, x_2|\{n_i\} \rangle \langle \{n_i'\}|x_1', x_2' \rangle = \sum_{\{n_i\}} C_{\{n_i\}}^* C_{\{n_i\}} \Psi_{\{n_i\}}(x_1, x_2) \Psi_{\{n_i\}}^*(x_1', x_2'). \tag{2.6}
\]

For the simplest entangled biphoton state considered above, \(|1_H, 1_V\rangle\), \(x_{1,2} = \sigma_{1,2}\), the sums (2.5) and (2.6) have only one term and the coordinate-dependent wave function is given by

\[
\Psi(\sigma_1, \sigma_2) = \frac{1}{\sqrt{2}} (\delta_{\sigma_1, H}\delta_{\sigma_2, V} + \delta_{\sigma_1, V}\delta_{\sigma_2, H}). \tag{2.7}
\]

\(\Psi(\sigma_1, \sigma_2)\) coincides with the last expressions on the right-hand side of equation (2.3) divided by \(\sqrt{2}\), and the coordinate density matrix

\[
\rho(\sigma_1, \sigma_2; \sigma'_1, \sigma'_2) = \Psi(\sigma_1, \sigma_2) \Psi^*(\sigma'_1, \sigma'_2) \tag{2.8}
\]

coincides with that of equation (2.4), divided by 2 for proper normalization. (Note that in the two cases of equations (2.3), (2.4) and (2.7) the symbols \(\sigma_{1,2}\) play different roles: mode characterizers and ‘coordinates’, or photon variables.) These coincidences illustrate equivalence of the considerations in terms of state vectors and coordinate wave functions, if only in the case state-vectors matrix elements are calculated correctly, as in equation (2.3).

In terms of the coordinate-dependent wave functions, a bipartite state is entangled if its wave function cannot be factorized, i.e. cannot be presented in the form of a product of two single-particle functions,

\[
\Psi(x_1, x_2) \neq \varphi(x_1) \times \chi(x_2). \tag{2.9}
\]

A natural extension of this definition is given by the Schmidt decomposition, or Schmidt theorem [14, 15], according to which any entangled (unfactorable) bipartite wave function can be presented as a sum of factorized terms. These characteristics of entangled biphoton qutrits and ququarts are widely used in this work. In particular, in this way we find explicit conditions when the biphoton qutrits are entangled and, oppositely, factorized, in dependence on values of their parameters \(C_i\). For comparison, it can be mentioned that an extreme but widely spread opinion is that all biphoton qutrits are factorable and disentangled, which is not confirmed by our consideration. We also find the relationship between entanglement and polarization of biphoton qutrits, and a series of other results is derived.

Note that the orthogonality relation for state vectors (2.3), matrix elements of the density matrix (2.4), as well as the coordinate-dependent wave function (2.7) and the coordinate density matrix (2.8) are symmetric with respect to particle transpositions \(1 \leftrightarrow 2\). In this sense, it is possible to speak about symmetry entanglement as entanglement existing only in systems of indistinguishable particles and arising exclusively owing to the symmetry of wave functions or matrix elements dictated by the Bose–Einstein or Fermi–Dirac statistics. The state \(|1_H, 1_V\rangle\) has
only symmetry entanglement and nothing else. In more complicated states, such as the general- 
form biphoton qutrits and ququarts, the symmetry entanglement coexists with and is inseparable 
from the above-mentioned configurational entanglement, related to existence in such states of 
several, not necessarily degenerate, configurations (several terms in the sum of equation (2.1)). 
The Schmidt parameter $K$ and the Schmidt decomposition take into account both symmetry and 
configurational entanglement. On the other hand, as shown above, separation for symmetry and 
configurational entanglement is rather conventional. In a slightly modified interpretation, the 
symmetry entanglement can be included in the generalized configurational entanglement and 
then, mathematically, the main requirement consists simply in a correct calculation of matrix 
elements of the type (2.3).

3. State vectors and wave functions of biphoton qutrits

In the form of state vectors, purely polarization biphoton states (qutrits) are given by a 
superposition 

$$|\Psi\rangle = C_1 |2_H\rangle + C_2 |1_H, 1_V\rangle + C_3 |2_V\rangle,$$

(3.1)

where the basis state-vectors are given by 

$$|2_H\rangle = \frac{1}{\sqrt{2}} a_H^\dagger |0\rangle, \quad |1_H, 1_V\rangle = a_H^\dagger a_V^\dagger |0\rangle, \quad |2_V\rangle = \frac{1}{\sqrt{2}} a_V^\dagger |0\rangle.$$

(3.2)

$|0\rangle$ is the vacuum state-vector, and $a_H^\dagger$ and $a_V^\dagger$ are the creation operators of photons in modes 
with horizontal and vertical polarizations (with given equal frequencies and given identical 
propagation directions). $C_{1,2,3}$ are arbitrary complex constants $C_{1,2,3} = |C_{1,2,3}| e^{i\varphi_{1,2,3}}$, obeying 
the normalization condition 

$$|C_1|^2 + |C_2|^2 + |C_3|^2 = 1.$$

(3.3)

Actually, as the total phase of the state vector (3.1) or wave function (see below) does not affect 
any measurable characteristics of qutrits, one of the phases $\varphi_{1,2,3}$, or a linear combination of 
phases, can be taken equal to zero, and hence the general form of the qutrit’s state vector (3.1) 
is characterized by four independent constants (e.g. $|C_1|$, $|C_3|$, $\varphi_1$ and $\varphi_3$ with $\varphi_2 = 0$).

As qutrit (3.1) is a two-photon state, its polarization wave function depends on two 
variables. A general rule for obtaining multipartite wave functions from state vectors is known 
pretty well in quantum-field theory, and for bosons the corresponding formula has the form [16] 
in (slightly modified notation) 

$$\Psi(x_1, x_2, \ldots, x_n) = \langle x_1, x_2, \ldots, x_n | n_1, n_2, \ldots, n_k \rangle$$

$$= \frac{1}{\sqrt{n_1! n_2! \cdots n_k! n!}} \sum_{P} P(g_1(x_1)g_1(x_2)\ldots g_1(x_{n_1})g_2(x_{n_1+1})g_2(x_{n_1+2})\ldots$$

$$\times g_2(x_{n_1+n_2})\ldots g_k(x_{n_1+n_2+\cdots+n_{k-1}+1})\ldots g_k(x_n)),$$

(3.4)

where $x_1, x_2, \ldots, x_n$ are dynamical variables of identical boson particles, $g_j(x_i)$ are single-
particle wave functions (of $j$th modes and $i$th variables), $P$ indicates all possible transpositions 
of variables $x_i$ in wave functions $g_j(x_i)$, $n_1$, $n_2$, $\ldots$, $n_k$ are numbers of particles in modes, $n$ is 
the total number of particles in all modes and $k$ is the total number of modes; for empty modes, 
the corresponding single-particle wave functions have to be dropped.
In the case of qutrits, we have two modes \( j = H \) or \( V \) and two particles, \( n = k = 2 \). The polarization variables of two photons can be denoted as \( \sigma_1 \) and \( \sigma_2 \). In terms of wave functions, the single-photon wave functions \( g_j(x_i) \) are given by the Kronecker symbols. Thus, the qutrit basis wave functions corresponding to the basis state-vectors in equation (3.1) can be written as

\[
\Psi_{HH}(\sigma_1, \sigma_2) = \langle \sigma_1, \sigma_2 | 2_H \rangle = \delta_{\sigma_1, H} \delta_{\sigma_2, H},
\]

(3.5)

\[
\Psi_{HV}(\sigma_1, \sigma_2) = \langle \sigma_1, \sigma_2 | 1_H, 1_V \rangle = \frac{1}{\sqrt{2}} [\delta_{\sigma_1, H} \delta_{\sigma_2, V} + \delta_{\sigma_2, H} \delta_{\sigma_1, V}],
\]

(3.6)

\[
\Psi_{VV}(\sigma_1, \sigma_2) = \langle \sigma_1, \sigma_2 | 2_V \rangle = \delta_{\sigma_1, V} \delta_{\sigma_2, V}.
\]

(3.7)

The same basis wave functions can be written equivalently in the form of two-row columns, which is more convenient for calculation of matrices

\[
\Psi_{HH} = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix},
\]

(3.8)

\[
\Psi_{VV} = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix},
\]

(3.9)

\[
\Psi_{HV} = \frac{1}{\sqrt{2}} \left( \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} + \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \right) = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & 0 \end{pmatrix},
\]

(3.10)

where the upper and lower rows in two-row columns correspond to the horizontal and vertical polarizations and indices 1 and 2 numerate indistinguishable photons.

In a general form, the qutrit’s wave function corresponding to the state vector (3.1) is given by

\[
\Psi = C_1 \Psi_{HH} + C_2 \Psi_{HV} + C_3 \Psi_{VV},
\]

(3.11)

where \( \Psi_{HH}, \Psi_{HV} \) and \( \Psi_{VV} \) can be taken in the form of either (3.5)–(3.7) or (3.8)–(3.10).

Alternatively, the same general qutrit’s wave function (3.11) can be presented in the form of an expansion in a series of Bell states

\[
\Psi = C_+ \Phi^+ + C_- \Phi^-,
\]

(3.12)

where

\[
C_{\pm} = \frac{C_1 \pm C_3}{\sqrt{2}},
\]

(3.13)
The first step for finding the degree of entanglement is related to a transition from the wave function to the density matrix of the same pure biphoton state \( \rho = \Psi \Psi^\dagger \). The full density matrix of qudit (3.11) can be presented in the following two forms:

\[
\rho = |C_1|^2 \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}_2 + |C_3|^2 \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_2 + \frac{|C_2|^2}{2} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_2 \\
+ (0 0 1) \otimes (1 0 1) + (0 0 1) \otimes (1 0 1) + (0 0 1) \otimes (1 0 1) + (0 0 1) \otimes (1 0 1) \\
+ C_1 C_3^\ast \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_2 + C_1^\ast C_3 \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}_2 \\
+ \frac{C_1 C_2^\ast}{\sqrt{2}} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_2 + \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_2 \\
+ \frac{C_3 C_2^\ast}{\sqrt{2}} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}_2 + \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}_2 \\
+ \frac{C_3^\ast C_2}{\sqrt{2}} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_2 + \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_1 \otimes \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}_2
\]

(4.1)
The next step is the reduction of the density matrix with respect to one of the photon variables, e.g. of photon 2. Mathematically, this means taking traces of all matrices with subscript 2 in equation (4.1), which gives

$$
\rho_{r} = \text{Tr}_{2} \rho = \left(\begin{array}{cccc}
|C_1|^2 & C_1 C_2^* & C_1 C_3^* & C_1 C_3^* \\
\frac{1}{\sqrt{2}} C_1^* C_2 & \frac{1}{2} |C_2|^2 & \frac{1}{2} |C_2|^2 & \frac{1}{\sqrt{2}} C_1^* C_2 \\
\frac{1}{\sqrt{2}} C_1^* C_2 & \frac{1}{2} |C_2|^2 & \frac{1}{2} |C_2|^2 & \frac{1}{\sqrt{2}} C_1^* C_2 \\
C_3^* & \frac{1}{\sqrt{2}} C_3 C_2^* & \frac{1}{\sqrt{2}} C_3 C_2^* & |C_3|^2
\end{array}\right).
$$

(4.2)

It may be interesting to analyze a relation between the $4 \times 4$ density matrix (4.2) and the $3 \times 3$ coherence matrix introduced by Klyshko in 1997 [17]. The density matrix $\rho$ (4.2) is written in a natural two-photon basis,

$$
\left(\begin{array}{c}
1 \\
0 \\
0 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
1 \\
0 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
0 \\
1 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
0 \\
0 \\
1
\end{array}\right).
$$

(4.4)

The question is how it can be transformed to the basis of states $\Psi_{HH}$, $\Psi_{VV}$, $\Psi_{HV}$ plus the empty antisymmetric state $\Phi^{-}$ (3.15). Evidently, the transformation

$$
\left(\begin{array}{c}
1 \\
0 \\
0 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
1 \\
0 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
0 \\
1 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
0 \\
0 \\
1
\end{array}\right) \rightarrow \left(\begin{array}{c}
1 \\
0 \\
0 \\
0
\end{array}\right), \frac{1}{\sqrt{2}} \left(\begin{array}{c}
0 \\
1 \\
0 \\
1
\end{array}\right), \frac{1}{\sqrt{2}} \left(\begin{array}{c}
0 \\
1 \\
-1 \\
0
\end{array}\right), \left(\begin{array}{c}
0 \\
0 \\
0 \\
1
\end{array}\right)
$$

(4.5)

is provided by the matrix

$$
U = \left(\begin{array}{cccc}
1 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\
0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\
0 & 0 & 0 & 1
\end{array}\right).
$$

(4.6)
Now, transformed to the basis \{\Psi_{HH}, \Psi_{VV}, \Psi_{HV}, \Psi^{-}\}, the density matrix \(\rho\) (4.2) takes the form

\[
\rho_{\text{transf}} = U \rho U = 
\begin{pmatrix}
|C_1|^2 & C_1 C_2^* & 0 & C_1 C_3^* \\
C_1^* C_3 & |C_2|^2 & 0 & C_2 C_3^* \\
0 & 0 & 0 & 0 \\
C_1^* C_3 & C_3 C_2^* & 0 & |C_3|^2
\end{pmatrix}.
\] (4.7)

A part of this matrix with non-zero lines and columns coincides with the 3 \times 3 coherence matrix [17, 18]

\[
\rho_{\text{coh}} = 
\begin{pmatrix}
|C_1|^2 & C_1 C_2^* & C_1 C_3^* \\
C_1^* C_3 & |C_2|^2 & C_3 C_2^* \\
C_1^* C_3 & C_3 C_2^* & |C_3|^2
\end{pmatrix}.
\] (4.8)

Although the coherence matrix \(\rho_{\text{coh}}\) (4.8) is widely used and analyzed in the literature, both \(\rho_{\text{coh}}\) and \(\rho_{\text{transf}}\) are hardly appropriate for reduction over one of the photon variables (e.g. 2) and for finding correctly the reduced density matrix \(\rho_r\) (4.3) because variables 1 and 2 are mixed not only in the matrix \(\rho_{\text{transf}}\) itself but also in the transformed basis of equation (4.5).

5. Degree of entanglement

As is known [14, 15], the trace of the squared reduced density matrix \(\rho_r^2\) determines purity of the reduced state coinciding with the inverse value of the Schmidt entanglement parameter \(K^{-1}\). The result of its calculation for the reduced density matrix of equation (4.3) is given by

\[
K^{-1} = \text{Tr}(\rho_r^2) = \left( |C_1|^2 + \frac{|C_2|^2}{2} \right)^2 + \left( |C_3|^2 + \frac{|C_2|^2}{2} \right)^2 + |C_1^* C_2 + C_2^* C_3|^2.
\] (5.1)

With normalization condition (3.3) taken into account, equation (5.1) can be reduced to a much simpler form:

\[
K = \frac{2}{2 - |2C_1 C_3 - C_2|^2}.
\] (5.2)

It is also known [19] that in the case of bipartite states with the dimensionality of the one-particle Hilbert space \(d = 2\), there is a simple algebraic relation between Wootters’ concurrence \(C\) [20] and the Schmidt entanglement parameter \(K\), owing to which

\[
C = \sqrt{2 \left( 1 - \frac{1}{K} \right)} = |2C_1 C_3 - C_2^2|.
\] (5.3)

Finally, in terms of the constants \(C_{\pm}\) (3.13), equations (5.2) and (5.3) take the form

\[
K = \frac{2}{2 - |C_2^2 - C_2^2 - C_2^2|^2}, \quad C = |C_2^2 - C_2^2 - C_2^2|.
\] (5.4)

Note that expressions for the concurrence \(C\) (equation (5.3) and the last formula of equation (5.4)) can also be derived directly from the original Wootters’ definition [20].
Figure 1. The Schmidt entanglement parameter $K_{\text{real}}$ (5.2), concurrence $C_{\text{real}}$ (5.3) and the von Neumann subsystem entropy $S_{\text{real}}$ (6.7) of the qutrit (3.11), (3.12) with real constants $C_{1,2,3}, C_{\pm}$ versus $C_{+}$ (3.13).

Indeed, for a pure bipartite state with dimensionality of the one-particle Hilbert space $d = 2$, concurrence is defined as

$$C = |\langle \Psi | \tilde{\Psi} \rangle|,$$  \hspace{1cm} (5.5)

where $\tilde{\Psi}$ is the function or state vector arising from $\Psi$ after the ‘spin-flip’ operation,

$$\tilde{\Psi} = (\sigma_y)_1 (\sigma_y)_2 \Psi^*,$$  \hspace{1cm} (5.6)

and $\sigma_y$ is the Pauli matrix, $\sigma_y = \left( \begin{array}{cc} 0 & -i \\ i & 0 \end{array} \right)$. For qutrits, $\Psi$ is given by equation (3.11) or (3.12). The rules of the ‘spin-flip’ transformation for one-photon wave functions are $|1\rangle \rightarrow -i |0\rangle$ and $|0\rangle \rightarrow i |1\rangle$. From here we easily find the spin-flip transforms of the qutrit basis wave functions (3.8)–(3.10)

$$\tilde{\Psi}_{\text{HH}} = -\Psi_{\text{VV}}, \quad \tilde{\Psi}_{\text{VV}} = -\Psi_{\text{HH}}, \quad \tilde{\Psi}_{\text{HV}} = \Psi_{\text{HV}}, \quad \tilde{\Psi}_{+} = -\Psi_{+}, \quad \tilde{\Psi}_{-} = \Psi_{-}$$  \hspace{1cm} (5.7)

and of the general-form qutrit wave function (3.11)

$$\tilde{\Psi} = -C_1^* \Psi_{\text{VV}} + C_2^* \Psi_{\text{HV}} - C_3^* \Psi_{\text{HH}} = -C_+^* \Psi_{+} + C_{-}^* \Psi_{-} + C_2^* \Psi_{\text{HV}}.$$  \hspace{1cm} (5.8)

Substitution of these expressions into equation (5.5) gives

$$C = | -2C_1^* C_3^* + C_2^* | \leq |C_{-}^* - C_{+}^* |^2 + C_2^* |,$$  \hspace{1cm} (5.9)

in complete agreement with equations (5.3) and (5.4).

In a special case of real constants $C_{1,2,3}$ and $C_{\pm}$, owing to normalization (3.3), the Schmidt entanglement parameter and concurrence (5.4) appear to be determined by the only real parameter $C_{+}$,

$$K_{\text{real}} = \frac{2}{1 + 4C_{+}^2 - 4C_{+}^2}, \quad C_{\text{real}} = |2C_{+}^2 - 1|.$$  \hspace{1cm} (5.10)

The functions $K_{\text{real}}(C_{+})$ and $C_{\text{real}}(C_{+})$ are shown in figure 1 together with the subsystem entropy found in the following section.
This picture demonstrates clearly that qutrits are non-entangled \((K_{\text{real}} = 1\) and \(C_{\text{real}} = 0\)) if \(C_+ = \pm 1/\sqrt{2}\) and, hence, \(C_+^2 + C_-^2 = \frac{1}{2}\). Consequently, the family of non-entangled qutrit wave functions with real coefficients is given by

\[
\Psi_{\text{NE, real}} = \frac{1}{\sqrt{2}}(\Psi_0 + \sin \phi \, \Psi_{HV} + \cos \phi \, \Psi_-)
\]

\[
= \cos^2\left(\frac{\phi}{2}\right) \Psi_{HH} + \frac{\sin \phi}{\sqrt{2}} \Psi_{HV} + \sin^2\left(\frac{\phi}{2}\right) \Psi_{VV}
\]

(5.11)

with arbitrary \(\phi\). If the constants \(C_{1,2,3}, C_\pm\) are complex, a general condition of no-entanglement is the same: \(C = 0\). As seen from the general expression for \(C\) (5.3), the concurrence depends on phases \(\varphi_{1,2,3}\) of the constants \(C_{1,2,3}\) only via the combination \(\varphi_1 + \varphi_3 - 2\varphi_2\). Hence, if \(\varphi_2 = \frac{1}{3}(\varphi_1 + \varphi_3)\), in the case of complex constants \(C_{1,2,3}\), equation (5.3) is reduced to \(C = |2|C_1||C_3| - |C_2|^2\), i.e. this case appears to be equivalent to the case of real constants \(C_{1,2,3}\). From here we find that the general three-parametric family of wave functions of non-entangled qutrits is given by

\[
\Psi_{\text{NE, general}}(\phi, \varphi_1, \varphi_3) = \frac{\sin \phi}{\sqrt{2}} e^{(i/2)(\varphi_1 + \varphi_3)} \Psi_{HV} + \cos^2\left(\frac{\phi}{2}\right) e^{i\varphi_1} \Psi_{HH} + \sin^2\left(\frac{\phi}{2}\right) e^{i\varphi_1} \Psi_{VV}
\]

(5.12)

with arbitrary \(\phi, \varphi_1\) and \(\varphi_3\). With these wave functions found explicitly, we can reconstruct the corresponding family of state vectors of non-entangled qutrits,

\[
|\Psi\rangle_{\text{NE, general}} = \frac{1}{\sqrt{2}} \left\{ \sin \phi \, e^{(i/2)(\varphi_1 + \varphi_3)} \hat{a}_H^+ \hat{a}_V^+ + \cos^2\left(\frac{\phi}{2}\right) e^{i\varphi_1} \hat{a}_H^{\dagger 2} + \sin^2\left(\frac{\phi}{2}\right) e^{i\varphi_1} \hat{a}_V^{\dagger 2} \right\} |0\rangle.
\]

(5.13)

Qutrits are maximally entangled when \(C = 1\) and \(K = 2\), and for wave functions with real constants \(C_{1,2,3}, C_\pm\) this occurs in two cases: \(C_+ = \pm 1\) and \(C_- = 0\). In the first of these cases, \(\Psi = \pm \Psi_+\). In the second case the maximally entangled wave function has the form of an arbitrary superposition of \(\Psi_{HV}\) and \(\Psi_-, \Psi = \sin \phi \, \Psi_{HV} + \cos \phi \, \Psi_-\). As before, this result can be generalized for the case of wave functions with complex coefficients such that \(\varphi_2 = \frac{1}{3}(\varphi_1 + \varphi_3)\). As a result, we obtain the following three-parametric family of wave functions of maximally entangled qutrits

\[
\Psi_{\text{max}}(\phi, \varphi_1, \varphi_3) = \sin \phi \, e^{(i/2)(\varphi_1 + \varphi_3)} \Psi_{HV} + \frac{\cos \phi}{\sqrt{2}} (e^{i\varphi_1} \Psi_{HH} - e^{i\varphi_3} \Psi_{VV})
\]

(5.14)

and the corresponding family of state vectors

\[
|\Psi\rangle_{\text{max}} = \left\{ \sin \phi \, e^{(i/2)(\varphi_1 + \varphi_3)} \hat{a}_H^+ \hat{a}_V^+ + \frac{\cos \phi}{2} (e^{i\varphi_1} \hat{a}_H^{\dagger 2} - e^{i\varphi_3} \hat{a}_V^{\dagger 2}) \right\} |0\rangle.
\]

(5.15)

Equations (5.14) and (5.15) are more general than the expression for the maximally entangled states of the work [21], which follows from (5.15) at \(\phi = 0\), \(|\Psi_{\text{max}}\rangle|_{\phi=0} = \frac{1}{\sqrt{2}} (e^{i\varphi_1} |2_H\rangle - e^{i\varphi_3} |2_V\rangle)\).

In the case where \(\phi = \pi/2\), equations (5.14) and (5.15) show that the state \(\Psi_{HV} (|1_H, 1_V\rangle)\) belongs to the family of maximally entangled states too (contrary to the opinion that the state \(|1_H, 1_V\rangle\) is factorable). A method of explicitly seeing whether qutrits are factorable or not consists in finding their Schmidt decompositions, which can contain either two products of Schmidt modes or only one product. This analysis is carried out in the following section.
But before switching to the Schmidt-mode analysis, let us discuss briefly the problem of qutrit polarization. If we define the biphoton polarization vector as \( \vec{\xi} = \text{Tr}(\rho \vec{\sigma}) \) [22], where \( \vec{\sigma} \) is the vector of Pauli matrices, from equation (4.3) we easily find
\[
\vec{\xi} = \{ \sqrt{2} \Re (C_1 C_2^* + C_2 C_3^*), - \sqrt{2} \Im (C_1 C_2^* + C_2 C_3^*), |C_1|^2 - |C_3|^2 \}.
\]
(5.16)
A direct comparison with the results obtained in 1999 by Burlakov and Klyshko [18] for polarization characteristics of qutrits shows that the polarization vector \( \vec{\xi} \) (5.16) coincides exactly with one half of the vector of Stokes parameters \( \vec{S} = \{ S_1, S_2, S_3 \} \) found by Burlakov and Klyshko [18], and the absolute value of \( |\vec{\xi}| \) coincides with their degree of polarization \( P \):
\[
\vec{\xi} = \frac{1}{2} \vec{S}, \quad |\vec{\xi}| = P = \frac{1}{2} \sqrt{S_1^2 + S_2^2 + S_3^2}.
\]
(5.17)
Note that the Stokes parameter and the degree of polarization were found in [18] in a way absolutely different from that used above for derivation of the polarization vector \( \vec{\xi} \). For this reason, coincidences (5.17) are rather non-trivial.

The derived expression for the polarization vector \( \vec{\xi} \) (5.16) can be compared with the general expression for the qutrit’s concurrence \( C \) (5.3) to give
\[
C^2 + P^2 = 1.
\]
(5.18)
Thus, the degrees of polarization and entanglement anticorrelate with each other: the maximally entangled qutrits are non-polarized, and maximally polarized states are non-entangled. In particular, all purely polarization Bell states are unpolarized and maximally entangled [23–25]. Although intuitively more or less expected and clear, as far as we know, anticorrelation of polarization and entanglement has never been presented in the rigorous mathematical form of equation (5.18).

6. Schmidt modes of qutrits and subsystem entropy

Entanglement means that the biphoton wave function cannot be factorized, whereas no-entanglement means that it is factorable. A transition from non-factorable to factorable wave functions can be reasonably explained in terms of Schmidt modes. Schmidt modes are eigenfunctions of the reduced density matrix, i.e. solutions of the equation \( \rho_r \psi = \lambda \psi \). As in the case of qutrits \( \rho_r \) (4.3) is the \( 2 \times 2 \) matrix, it has two eigenvalues \( \lambda_{\pm} \), its eigenfunctions are two-row columns \( \psi_{\pm} = (a_{\pm}, b_{\pm}) \), and the eigenvalue–eigenfunction equation has the form
\[
\rho_r \begin{pmatrix} a_{\pm} \\ b_{\pm} \end{pmatrix} = \lambda_{\pm} \begin{pmatrix} a_{\pm} \\ b_{\pm} \end{pmatrix}.
\]
(6.1)
The modes can be normalized, \( |a_{\pm}|^2 + |b_{\pm}|^2 = 1 \), and they are orthogonal to each other, \( a_+ a_- + b_+ b_- = 0 \). In terms of \( \lambda_{\pm} \), the Schmidt entanglement parameter equals \( K = (\lambda_+^2 + \lambda_-^2)^{-1} \).

In accordance with the Schmidt theorem, the biphoton wave function can be presented as a sum of products of Schmidt modes (Schmidt decomposition). In the case \( d = 2 \) (two-qubit states or qutrits), the Schmidt decomposition contains only two terms,
\[
\psi = \sum_{\pm} \sqrt{\lambda_{\pm}} \psi_{\pm}(1) \psi_{\pm}(2),
\]
(6.2)
where arguments of the Schmidt modes indicate variables of photons ‘1’ and ‘2’. This decomposition shows that in a general case the wave function \( \psi \) is non-separable. Exceptions
occur when one of the eigenvalues of the reduced density matrix, \( \lambda_+ \) or \( \lambda_- \), becomes equal to zero.

Eigenvalues of matrix (4.3) can be found rather easily and can be reduced to a very simple form expressed via concurrence \( C \):

\[
\lambda_{\pm} = \frac{1}{2} (1 \pm \sqrt{1 - C^2}).
\]

(6.3)

In the case \( C = 1 \), when entanglement is maximal, \( \lambda_+ = \lambda_- = \frac{1}{2} \), i.e. two products in the Schmidt decomposition (6.2) are presented with equal weights and, clearly, the wave function is non-separable.

In the case \( C = 0 \) (no entanglement), equation (6.3) gives \( \lambda_+ = 1 \) and \( \lambda_- = 0 \). In this case, one of the products of Schmidt modes \( (\psi^- \psi_-) \) in the Schmidt decomposition disappears because the coefficient \( \sqrt{\lambda_-} \) in front of it vanishes. This is the reason for separability of the wave function in the case of no-entanglement. As for the remaining product in the Schmidt decomposition, the corresponding eigenfunction of the reduced density matrix \( \psi_+ \) at \( C = 0 \) can be found easily and has a reasonably simple form (5.12):

\[
\psi_+ = \begin{pmatrix} a_+ \\ b_+ \end{pmatrix} = \begin{pmatrix} \cos(\phi/2)e^{i\frac{1}{2}\varphi_1} \\ \sin(\phi/2)e^{i\frac{1}{2}\varphi_2} \end{pmatrix}.
\]

(6.4)

With this expression we find immediately that in the case \( C = 0 \), the Schmidt decomposition (6.2) gives the following representation for the factored biphoton wave function:

\[
\Psi_{\text{NE}} = \begin{pmatrix} \cos(\phi/2)e^{i\frac{1}{2}\varphi_1} \\ \sin(\phi/2)e^{i\frac{1}{2}\varphi_2} \end{pmatrix}_1 \begin{pmatrix} \cos(\phi/2)e^{i\frac{1}{2}\varphi_1} \\ \sin(\phi/2)e^{i\frac{1}{2}\varphi_2} \end{pmatrix}_2.
\]

(6.5)

Identity of equations (5.12) and (6.5) can be checked directly by means of substituting into equation (5.12), instead of \( \Psi_{\text{HH}}, \Psi_{\text{HV}}, \Psi_{\text{VV}} \), their column representations (3.8)–(3.10), calculation of direct products of columns in all terms of equation (5.12) and in equation (6.5), and presenting \( \Psi_{\text{NE}} \) in both cases in the form of the following four-row column:

\[
\Psi_{\text{NE general}}(\phi, \varphi_1, \varphi_3) = \begin{pmatrix} \cos^2(\phi/2)e^{i\varphi_1} \\ \frac{1}{2} \sin(\phi)e^{i(\varphi_1+\varphi_3)} \\ \frac{1}{2} \sin(\phi)e^{i(\varphi_1+\varphi_3)} \\ \sin^2(\phi/2)e^{i\varphi_3} \end{pmatrix}.
\]

(6.6)

Equation (6.3) can be used to find subsystem entropy [26] defined as

\[
S_t = -\text{Tr}(\rho_t \log_2 \rho_t) = -\sum_{\pm} \lambda_\pm \log_2 \lambda_\pm.
\]

(6.7)

In the case of qutrits with real coefficients, concurrence \( C_{\text{real}} \) itself is a function of \( C_+ \) (5.10). With \( C_{\text{real}}(C_+) \) substituted instead of \( C \) into equation (6.3) and then \( \lambda_{\pm}[C(C_+)] \) substituted into equation (6.7), we obtain a function \( S_{\text{real}}(C_+) \), which is plotted in figure 1. Although different from both \( C(C_+) \) and \( K(C_+) \), the subsystem entropy \( S_{\text{real}}(C_+) \) has the same main features as two other entanglement quantifiers: \( S_{\text{real}} \) is minimal and equals zero at \( C_+ = \pm 1/\sqrt{2} \) and \( S_{\text{real}} \) is maximal and equals 1 at \( C_+ = 0 \) and \( C_+ = \pm 1 \). Therefore, although Schmidt entanglement parameter \( K \), concurrence \( C \) and subsystem entropy \( S \), characterize the degree of entanglement in different metrics, their behavior is very similar, which confirms all conclusions made above about conditions of separability and non-separability of qutrits and their wave functions.
As mentioned above, one of the basis wave functions of qutrits, $\Psi_{HV}$ (3.6), is maximally entangled ($K = 2$, $C = 1$) and, hence, inseparable. Eigenvalues of the reduced density matrix of this state are degenerate, $\lambda_+ = \lambda_- = \frac{1}{2}$, and the Schmidt modes are easily found to be determined by the wave functions of photons polarized along the directions 45° and 135° with respect to the horizontal axis:

$$\psi_+^{HV} = \frac{1}{\sqrt{2}} \left( \begin{array}{c} 1 \\ 1 \end{array} \right), \quad \psi_-^{HV} = \frac{i}{\sqrt{2}} \left( \begin{array}{c} 1 \\ -1 \end{array} \right).$$  \hspace{1cm} (6.8)

With these expressions for the Schmidt modes, the Schmidt decomposition has form (6.2). However, it should be noted that in the case of degenerate eigenvalues of the reduced density matrices, $\lambda_+ = \lambda_-$, this is not the only possible form of the Schmidt decomposition. Indeed, if one can find two linear combinations $\varphi$ and $\chi$ of the functions $\psi_+$ and $\psi_-$ such that $\varphi$ and $\chi$ are orthogonal and normalized, then products of these functions can be used to rewrite the Schmidt decomposition (6.2) in the form

$$\psi_+^{HV} = \frac{1}{\sqrt{2}} (\varphi(\sigma_1)\chi(\sigma_2) + \varphi(\sigma_2)\chi(\sigma_1)).$$  \hspace{1cm} (6.9)

In this form, symmetry of the wave function is provided by the sum of two products of the Schmidt modes, each of which is not symmetric itself. For the state $\Psi_{HV}$ we obtain

$$\varphi = \frac{\psi_+ - i\psi_-}{\sqrt{2}} = \left( \begin{array}{c} 1 \\ 0 \end{array} \right), \quad \chi = \frac{\psi_+ + i\psi_-}{\sqrt{2}} = \left( \begin{array}{c} 0 \\ 1 \end{array} \right).$$  \hspace{1cm} (6.10)

and the Schmidt decomposition (6.9) takes the form of the original definition of the wave function $\Psi_{HV}$ (3.6).

In principle, it is easy to suggest methods of Schmidt mode separation in experiments. For example, in the case of state $\Psi_{HV}$ the SPDC biphoton beam has to be sent to a beam splitter (BS) non-selective with respect to polarizations. Then we obtain two channels with both split and unsplit pairs in each (see the following section for details). Unsplit pairs have to be ‘killed’, e.g. as was done in the experiment [27], and the remaining one-photon beams in both channels have to be sent to polarization beam BSs. One can use either the polarization BS separating horizontally and vertically or 45° and 135° polarized photons. Depending on this choice, one will get either the Schmidt modes of the form (6.10) or (6.8). One will obtain four channels with one Schmidt mode in each and with 100% correlations in each pair of channels related to a given product of the Schmidt modes in decomposition (6.2) or (6.9).

As for entanglement of the state $\Psi_{HV}$, here it is proved to occur in many ways and, of course, we always mean total entanglement, which includes both symmetry and configurational contributions. In the case of the $\Psi_{HV}$ state, total entanglement has a purely symmetry origin. This explains the difference between our result that the state $\Psi_{HV}$ is maximally entangled and the very often encountered opinion that this state is disentangled [3, 28]. Evidently, the last conclusion concerns only the configurational entanglement, which is missing, indeed, in the case of the single-configuration state $\Psi_{HV}$. Note, however, that separation of the symmetry and configuration entanglement in the case of purely polarization biphoton states is rather circumstantial. Indeed, in the basis turned 45° with respect to the horizontal–vertical one, the single-configuration state $\Psi_{HV}$ turns into the two-configuration one: $(\Psi_{45°-45°} - \Psi_{135°-135°})/\sqrt{2}$. This state has only configurational and no symmetry entanglement. But, fortunately, the total entanglement remains the same: the Schmidt parameter $K$ is basis-independent.

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7. Ququarts

7.1. Definitions and wave functions

As mentioned in the Introduction, in the generally accepted treatment, ququarts are considered as two-qubit states. In an abstract form, with unspecified pairs of distinguishable particles, the state vectors of ququarts are taken in the form

\[
|\Psi\rangle_{2\text{qb}} = C_1|00\rangle + C_2|01\rangle + C_3|10\rangle + C_4|11\rangle \tag{7.1}
\]

with one-qubit single-particle states $|0\rangle$ and $|1\rangle$. Written in the form of two-row columns, the wave function of state (7.1) takes the form

\[
|\Psi\rangle_{2\text{qb}} = C_1 \begin{pmatrix} 1 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix} + C_2 \begin{pmatrix} 1 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix} + C_3 \begin{pmatrix} 0 \\ 1 \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix} + C_4 \begin{pmatrix} 0 \\ 1 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix}. \tag{7.2}
\]

With this wave function one can easily obtain the reduced density matrix obtained by Bogdanov et al [31],

\[
\rho_{\text{r}}(2\text{qb}) = \begin{pmatrix} |C_1|^2 + |C_2|^2 & C_1 C_3^* + C_2 C_4^* \\ C_1^* C_3 + C_2^* C_4 & |C_3|^2 + |C_4|^2 \end{pmatrix}, \tag{7.3}
\]

the Schmidt entanglement parameter,

\[
K_{2\text{qb}} = \frac{1}{\text{Tr}(\rho_{\text{r}}^2)} = \frac{1}{1 - 2|C_1 C_4 - C_2 C_3|^2}, \tag{7.4}
\]

and the concurrence related to it [22, 31],

\[
C_{2\text{qb}} = \sqrt{2(1 - K_{2\text{qb}}^{-1})} = 2|C_1 C_4 - C_2 C_3|. \tag{7.5}
\]

Although this approach and equations (7.3)–(7.5) are rather widely used and accepted, we claim that they are inapplicable for biphoton ququarts formed by non-degenerate SPDC pairs. The first objection is the symmetry. Wave function (7.2) is asymmetric with respect to the permutation of particle variables $1 \leftrightarrow 2$, and this is strictly forbidden for any biphoton wave functions. A simple symmetrization of wave function (7.2) is insufficient and, actually, it does not help much, because it reduces wave function (7.2) to the qutrit’s wave function (3.11), with $C_2 + C_3$ playing the role of $C_2$ in equation (3.11) and with all peculiarities of ququarts completely lost.

The second objection concerns photon degrees of freedom and dimensionality of the Hilbert space. In contrast to the traditional treatment, biphoton ququarts formed by non-degenerate SPDC pairs of photons have a higher dimensionality than qutrits. Single-photon states from which the biphoton ququarts are constructed are not qubits and they form the Hilbert space of dimensionality $d = 4$, rather than $d = 2$ occurring in the case of qutrits. Indeed, if for example SPDC photons in pairs have two different frequencies $\omega_{\text{h}}$ (high) and $\omega_{\text{l}}$ (low), $\omega_{\text{h}} > \omega_{\text{l}}$, one cannot say for sure which of the two photons gets a higher or lower frequency. Hence, the photon frequency $\omega$ becomes a second photon variable, additional to polarization and taking two values, $\omega_{\text{h}}$ (high) and $\omega_{\text{l}}$. In other words, each photon now has two degrees of freedom, polarization and frequency [32]. Together, they make four states that can be occupied by each photon, or four combined polarization–frequency modes, $H_{\text{h}}$, $H_{\text{l}}$, $V_{\text{h}}$ and $V_{\text{l}}$ (instead of two polarization modes $H$ and $V$ in the case of degenerate photons). Four modes correspond to the dimensionality of the single-photon Hilbert space $d = 4$, in contrast to $d = 2$ and two modes in...
the cases of qutrits and of the traditional two-qubit model of ququarts. The four single-photon polarization–frequency state vectors are now given by

\[ a_H^\dagger |0\rangle, \quad a_L^\dagger |0\rangle, \quad a_V^\dagger |0\rangle, \quad a_{VL}^\dagger |0\rangle. \]

(7.6)

The corresponding single-photon wave functions, describing states with two degrees of freedom, have two factors, depending on polarization (\(\sigma\)) and frequency (\(\omega\)) variables:

\[ \delta_{\sigma,H}\delta_{\omega,oh} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\text{pol}} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\omega} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, \]

\[ \delta_{\sigma,H}\delta_{\omega,oI} = \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\text{pol}} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\omega} = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}, \]

\[ \delta_{\sigma,V}\delta_{\omega,oh} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\text{pol}} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\omega} = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}, \]

\[ \delta_{\sigma,V}\delta_{\omega,oI} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\text{pol}} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\omega} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \]

(7.7)

The superscripts \(\text{pol}\) and \(\omega\) are used here for differentiating the polarization and frequency parts of the two-variable wave functions written in the form of direct products of two-row columns.

Because the dimensionality of the single-photon Hilbert space is \(d = 4\), these states are \(d = \) four-qudits, rather than qubits. Then, apart from qutrits being two-qubit states, the biphoton ququarts are also two-qudit states. Their basis state-vectors are given by all possible products of two different creation operators times the vacuum state. But if we assume that two photons in each non-degenerate SPDC pair definitely have different frequencies, the products of two creation operators corresponding to coinciding frequencies have to be excluded to give finally only four two-photon basis state-vectors:

\[ |\Psi_{HH}^{(qut)}\rangle = a_H^\dagger a_H^\dagger |0\rangle, \quad |\Psi_{HV}^{(qut)}\rangle = a_H^\dagger a_{VL}^\dagger |0\rangle, \quad |\Psi_{VH}^{(qut)}\rangle = a_{VL}^\dagger a_H^\dagger |0\rangle, \quad |\Psi_{VV}^{(qut)}\rangle = a_{VL}^\dagger a_{VL}^\dagger |0\rangle, \]

(7.8)

where, of course, all creation operators commute with each other. The basis wave functions corresponding to the state-vectors (7.8) are obtained with the help of general rules of quantum electrodynamics (3.4):

\[ \Psi_{HH}^{(qut)} = \left( \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\text{pol}} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\omega} \right) \otimes \frac{1}{\sqrt{2}} \left[ \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\omega} \otimes \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\omega} + \begin{pmatrix} 0 \\ 1 \end{pmatrix}^{\omega} \otimes \begin{pmatrix} 1 \\ 0 \end{pmatrix}^{\omega} \right] \]

\[ = \frac{1}{\sqrt{2}} \left\{ \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \right\}, \]

(7.9)
The six remaining and excluded wave functions are antisymmetric, and they cannot be properly taken into account. This excludes states \(|h, \sigma; h, \sigma'\rangle\) and \(|l, \sigma; l, \sigma'\rangle\), which give rise to six symmetric wave functions. They do not have fundamental reasons and are related rather to the most often met experimental conditions. The second, deeply fundamental restriction is symmetry. The six remaining and excluded wave functions are antisymmetric, and they cannot be realized with photons at all. Thus, in ququarts all 12 excluded basis wave functions either are

\[
\Psi_{HV}^{(qqrt)} = \frac{1}{\sqrt{2}} \left[ (1)^{pol} \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \otimes \left( \begin{array}{l} 0 \\ 1 \end{array} \right) + (1)^{pol} \left( \begin{array}{l} 0 \\ 1 \end{array} \right) \otimes \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \right]
\]

(7.10)

\[
\Psi_{VV}^{(qqrt)} = \frac{1}{\sqrt{2}} \left[ (0)^{pol} \left( \begin{array}{l} 0 \\ 1 \end{array} \right) \otimes \left( \begin{array}{l} 0 \\ 1 \end{array} \right) + (0)^{pol} \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \otimes \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \right]
\]

(7.11)

\[
\Psi_{VV}^{(qqrt)} = \frac{1}{\sqrt{2}} \left[ (0)^{pol} \left( \begin{array}{l} 0 \\ 1 \end{array} \right) \otimes \left( \begin{array}{l} 0 \\ 1 \end{array} \right) + (0)^{pol} \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \otimes \left( \begin{array}{l} 1 \\ 0 \end{array} \right) \right]
\]

(7.12)

In a general form, the state vector and wave function of the biphoton ququart are given by superpositions of four basis state-vectors (7.8) and four basis wave functions (7.9)–(7.12):

\[
|\Psi\rangle^{(qqrt)} = C_1|\Psi_{HH}^{(qqrt)}\rangle + C_2|\Psi_{HV}^{(qqrt)}\rangle + C_3|\Psi_{VH}^{(qqrt)}\rangle + C_4|\Psi_{VV}^{(qqrt)}\rangle
\]

(7.13)

and

\[
\Psi^{(qqrt)} = C_1\Psi_{HH}^{(qqrt)} + C_2\Psi_{HV}^{(qqrt)} + C_3\Psi_{VH}^{(qqrt)} + C_4\Psi_{VV}^{(qqrt)}.
\]

As they should be, all basis wave functions (7.9)–(7.12) and the general ququart wave function (7.14) are symmetric with respect to the particle permutations \(1 \leftrightarrow 2\). Also, these expressions properly take into account a higher dimensionality of the biphoton ququarts compared with qutrits and compared with the traditional simplified version of biphoton ququarts based on equations (7.1)–(7.2).

Once again, the dimensionality of the one-photon Hilbert space for non-degenerate photons is \(d = 4\). The dimensionality of the biphoton Hilbert space is \(D = d^2 = 16\). The question is why are ququarts characterized by only 4 rather than 16 basis wave functions. A general answer is because of some restrictions, owing to which 12 basis wave functions are excluded. One of these restrictions was mentioned above: we assume that the frequencies of photons are always different. This excludes states \(|h, \sigma; h, \sigma'\rangle\) and \(|l, \sigma; l, \sigma'\rangle\), which give rise to six symmetric wave functions. Their exclusion does not have fundamental reasons and is related rather to the most often met experimental conditions. The second, deeply fundamental restriction is symmetry. The six remaining and excluded wave functions are antisymmetric, and they cannot be realized with photons at all. Thus, in ququarts all 12 excluded basis wave functions either are

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missing in all possible superpositions or can be made present symbolically but with obligatory zero coefficients, like the only antisymmetric wave function in the qutrit wave function (3.16). As in the case of qutrits, the existence of ququarts with only four basis wave functions is crucially related to the requirement of symmetry of all biphoton wave functions.

Note, however, that as the ququart $16 \times 16$ density matrix contains many zeros, by a series of unitary transformation it can be reduced to the $4 \times 4$ coherence matrix plus 12 zero lines and columns (similar to what was done for qutrits in equations (4.6) and (4.7)). Moreover, the thus obtained $4 \times 4$ coherence matrix coincides with that of the two-qubit ququart’s model, although the reduced density matrices of the two-qubit and two-qudit do not coincide. But coincidence of the full coherence matrices in two-qubit and two-qudit systems indicates that there are some links between these two kinds of objects. Not going into any further details of this problem (to be described elsewhere), let us mention here that in experiments two-qubit ququarts can be obtained from two-qudit polarization–frequency biphoton ququarts with the help of a dichroic BS, separating photons by frequencies, $\omega_1$ in one channel and $\omega_2$ in the other. Then the states arising in each channel, considered independently of the other, are just the two-qubit ququarts.

### 7.2. Degree of entanglement

As is known [19], in the case of pure two-particle states the dimensionality of the single-particle Hilbert space $d$ determines directly the maximal achievable value of the Schmidt entanglement parameter, $K_{\text{max}} = d$. Consequently, a two times higher dimensionality of the double-qubit Hilbert space (compared with qubits) doubles the maximal achievable degree of entanglement of ququarts if it is evaluated by the Schmidt entanglement parameter, $K_{\text{max}} = 4$ (to be compared with $K_{\text{max}} = 2$ in the case of qutrits). To find the entanglement quantifiers of ququarts in a general case, following the standard procedure, we have to start by finding the density matrix $\rho_{\text{qqrt}}$, corresponding to the wave function $\Psi_{\text{qqrt}}$ (7.14). The dimensionality of $\rho_{\text{qqrt}}$ is $16 \times 16$, which is too large to be shown explicitly. But the reduced density matrix $\rho_{\text{r qqrt}}$ is much more compact, its dimensionality is $4 \times 4$ and, explicitly, it is given by

$$
\rho_{\text{r qqrt}} = \frac{1}{2} \begin{pmatrix}
|C_1|^2 + |C_2|^2 & 0 & C_1 C_3^* + C_2 C_4^* & 0 \\
0 & |C_1|^2 + |C_3|^2 & 0 & C_1 C_4^* + C_3 C_4^* \\
C_1^* C_3 + C_2^* C_4 & 0 & |C_3|^2 + |C_4|^2 & 0 \\
0 & C_1^* C_2 + C_3^* C_4 & 0 & |C_2|^2 + |C_4|^2
\end{pmatrix}.
$$

(7.15)

The derived expression for the $4 \times 4$ reduced density matrix (7.15) differs significantly from the $2 \times 2$ density matrix of the paper [31] where ququarts were considered as two-qubit states. As explained above, key reasons for this and other differences between this paper [31] and others are in the symmetry of wave functions and in a higher dimensionality of ququarts compared with qutrits properly taken into account in equations (7.9)–(7.12) and (7.14).

By calculating the squared reduced density matrix $(\rho_{\text{r qqrt}})^2$ and its trace, we find that the Schmidt entanglement parameter of ququarts is simply twice that obtained in the two-qubit model (7.4):

$$
K = \frac{2}{1 - 2|C_1 C_4 - C_2 C_3|^2} = 2K_{2qb}.
$$

(7.16)

As for the concurrence, rigorously, its original Wootters’ definition [20] is invalid for two-qudit states with $d > 2$. But instead, one can use the so-called ‘I-concurrence’ [19], by definition,
determined via the Schmidt entanglement parameter \( K \) as \( C_I = \sqrt{2(1-K^{-1})} \). Defined in this way, I-concurrence does not provide any new information about the entanglement of ququarts compared with that provided by the Schmidt entanglement parameter \( K \). But it can be useful for comparison with other entanglement quantifiers, and for comparison with the results of the two-qubit model of ququarts. In a general case, equation (7.16) yields

\[
C_I = \sqrt{1 + 2|C_1 C_4 - C_2 C_3|^2}.
\]  

(7.17)

As follows from equations (7.16) and (7.17), the maximal achievable values of \( K \) and \( C_I \) for ququarts are \( K_{\text{max}} = 4 \) and \( C_{I\text{max}} = \sqrt{3/2} \), in agreement with the general expectations for qudits \( K_{\text{max}} = d \) and \( C_{I\text{max}} = \sqrt{2(1-d^{-1})} \) [19]. Minimal values of the Schmidt entanglement parameter and I-concurrence are achieved when \( C_1 C_4 = C_2 C_3 \), and in this case \( K = K_{\text{min}} = 2 \) and \( C_I = C_{I\text{min}} = 1 \). This means that all biphoton ququarts are entangled and non-factorable (in contrast to this, earlier [31], in a two-qubit model, it was assumed that at \( C_1 C_4 = C_2 C_3 \) ququarts are factorable). In particular, all basis states of ququarts (7.9)–(7.12) are minimally entangled. It may be interesting to note that the nature of entanglement of different basis states is different.

The wave functions \( \Psi_{\text{HH}}^{(\text{qqrt})} \) and \( \Psi_{\text{VV}}^{(\text{qqrt})} \) are seen to be factorized for polarization and frequency parts, and only their frequency parts are entangled, i.e. the states \( \Psi_{\text{HH}}^{(\text{qqrt})} \) and \( \Psi_{\text{VV}}^{(\text{qqrt})} \) have only a purely frequency entanglement. On the other hand, the wave functions \( \Psi_{\text{HV}}^{(\text{qqrt})} \) and \( \Psi_{\text{VH}}^{(\text{qqrt})} \) are not factorized for polarization and frequency parts, and in these cases we have a non-separable frequency–polarization entanglement.

Some examples of maximally entangled ququarts are

\[
\begin{align*}
\frac{1}{\sqrt{2}} (\Psi_{\text{HH}}^{(\text{qqrt})} & \pm \Psi_{\text{VV}}^{(\text{qqrt})}) \quad \left( C_1 = \pm C_4 = \frac{1}{\sqrt{2}}, \ C_2 = C_3 = 0 \right), \\
\frac{1}{\sqrt{2}} (\Psi_{\text{HV}}^{(\text{qqrt})} & \pm \Psi_{\text{VH}}^{(\text{qqrt})}) \quad \left( C_1 = C_4 = 0, \ C_2 = \pm C_3 = \frac{1}{\sqrt{2}} \right), \\
\frac{1}{2} (\Psi_{\text{HH}}^{(\text{qqrt})} & + \Psi_{\text{HV}}^{(\text{qqrt})} + \Psi_{\text{VH}}^{(\text{qqrt})} - \Psi_{\text{VV}}^{(\text{qqrt})}),
\end{align*}
\]  

(7.18)

and all functions similar to the last one but with the ‘minus’ sign located in front of the other terms, \( \Psi_{\text{HH}}^{(\text{qqrt})} \), \( \Psi_{\text{HV}}^{(\text{qqrt})} \) or \( \Psi_{\text{VH}}^{(\text{qqrt})} \).

To see how the degree of entanglement of ququarts changes from \( K_{\text{min}} = 2 \) to \( K_{\text{max}} = 4 \), let us consider, for example, the case \( C_1 = \cos \phi, \ C_4 = \sin \phi, \ C_3 = C_2 = 0 \), i.e. the wave function

\[
\Psi_\phi = \cos \phi \Psi_{\text{HH}}^{(\text{qqrt})} + \sin \phi \Psi_{\text{VV}}^{(\text{qqrt})}.
\]  

(7.19)

For this state, equations (7.16) and (7.17) give \( K(\phi) = 4/(1 + \cos^2 2\phi) \) and \( C_I(\phi) = \sqrt{1 + \frac{1}{2} \sin^2 2\phi} \). These functions are plotted in figure 2 together with subsystem entropy \( S_i(\phi) = -\sum_{i=1}^4 \lambda_i \log_2 \lambda_i \), where \( \lambda_i \) are eigenvalues of the reduced density matrix \( \rho_i^{(\text{qqrt})} \) (7.15). In the case of \( \Psi^{(\text{qqrt})} = \Psi_\phi \) (7.19), the matrix \( \rho_{i\phi}^{(\text{qqrt})} \) is very simple:

\[
\rho_{i\phi}^{(\text{qqrt})} = \begin{pmatrix}
\frac{1}{2} \cos^2 \phi & 0 & 0 & 0 \\
0 & \frac{1}{2} \cos^2 \phi & 0 & 0 \\
0 & 0 & \frac{1}{2} \sin^2 \phi & 0 \\
0 & 0 & 0 & \frac{1}{2} \sin^2 \phi
\end{pmatrix}.
\]  

(7.20)
Figure 2. The Schmidt entanglement parameter $K(\phi)$, $I$-concurrence $C_I(\phi)$ and subsystem entropy $S_r(\phi)$ for the ququart characterized by wave function $\Psi_\phi$ (7.19).

Evidently, its eigenvalues are $\lambda_1 = \lambda_2 = \frac{1}{2} \cos^2 \phi$ and $\lambda_3 = \lambda_4 = \frac{1}{2} \sin^2 \phi$, which yields

$$S_r(\phi) = 1 - 2(\cos^2 \phi \log_2 |\cos \phi| + \sin^2 \phi \log_2 |\sin \phi|).$$

Eigenvalues of the reduced density matrix $\rho_{q\phi}$ are twice degenerate, $\lambda_1 = \lambda_2$ and $\lambda_3 = \lambda_4$. If none of them equals zero, the Schmidt decomposition of the wave function $\Psi_\phi$ contains four terms of Schmidt-mode products. However, at $\phi = 0$, when the entropy $S_r(\phi)$ is minimal, two of these eigenvalues become zero, $\lambda_3 = \lambda_4 = 0$, and two Schmidt-mode products in the Schmidt decomposition disappear. Hence, in this case the degree of entanglement is minimal. However, the remaining two terms in the Schmidt decomposition indicate that in the case $\phi = 0$ the state $\Psi_\phi$ is entangled and non-factorable. The degree of entanglement of the state $\Psi_\phi$ is maximal when $\lambda_1 = \lambda_2 = \lambda_3 = \lambda_4 = 1/4$, i.e. when all four Schmidt-mode products enter the Schmidt decomposition with equal weights, and this occurs at $\phi = 45^\circ$ and $\phi = 135^\circ$.

In the general case, ququarts are multiparametric objects, and it is rather difficult to show all their features in a limited number of pictures. However, it may be interesting to show one more example, when the ququart wave function has the form

$$\Psi'_\phi = \frac{\cos \phi \Psi_{q\phi}^{(q\phi)} + \sin \phi \Psi_{q\phi}^{(q\phi)}}{\sqrt{2}} + \frac{\Psi_{q\phi}^{(q\phi)} + \Psi_{q\phi}^{(q\phi)}}{2},$$

for which $C_1 = \cos \phi/\sqrt{2}$, $C_4 = \sin \phi/\sqrt{2}$ and $C_2 = C_3 = 1/2$. The Schmidt entanglement parameter $K(\phi)$ and $I$-concurrence $C(\phi)$ of state (7.22) are shown in figure 3.

Asymmetry of these curves with respect to the substitution $\phi \rightarrow \pi - \phi$ indicates sensitivity of the degree of entanglement with respect to the sign of the product $C_1 C_3$, i.e. to phases of these real constants.

To conclude this subsection, let us describe here one more example of a rather peculiar polarization–angular ququart obtainable from qutrits with the help of a BS and post-selection. As mentioned earlier, polarization–frequency and polarization–angular ququarts are equivalent. Non-degeneracy of photons can be provided by the different direction of photons with equal
Figure 3. The Schmidt entanglement parameter $K(\phi)$ and $I$-concurrence $C_I(\phi)$ for the ququart characterized by the wave function $\Psi_\phi (7.22)$.

frequencies in the non-collinear SPDC process. Another way of providing non-collinear propagation of originally collinear and frequency-degenerate photons is in using a non-selective BS. After the BS, photons acquire new angular variables, $\theta_1$ and $\theta_2$, which can take one of two values each, $0^\circ$ and $90^\circ$. The combined polarization–angular wave function after the BS is given by the product

$$\Psi_{\text{after BS}} = \Psi^{\text{qtr}}(\sigma_1, \sigma_2) \times \frac{1}{2}(\delta_{\theta_1, 0} - \delta_{\theta_1, 90^\circ}) (\delta_{\theta_2, 0} - \delta_{\theta_2, 90^\circ}),$$

(7.23)

where $\Psi^{\text{qtr}}(\sigma_1, \sigma_2)$ is a purely polarization wave function of a biphoton qutrit before the BS. The wave function of equation (7.23) is not yet a complete analogue of ququarts considered above because of unsplit pairs in each channel after the BS (the terms proportional to $\delta_{\theta_1, 0} \delta_{\theta_2, 0}$ and $\delta_{\theta_1, 90^\circ} \delta_{\theta_2, 90^\circ}$ in equation (A.1)) . In the case of frequency non-degenerate biphoton states, similar terms would describe both photons with high or both with low frequencies, which we have excluded (see the discussion in the previous subsection). Because of these terms with coinciding angles $\theta_1$ and $\theta_2$, as mentioned above, the angular part of the combined wave function in equation (A.1) does not provide any additional entanglement to the polarization part $\Psi$ (qutrit). However, if we make a post-selection by killing (eliminating) in some way unsplit pairs in both channels after BS, we obtain a true polarization–angular ququart with the wave function of the form

$$\Psi^{\text{qqrt}} = \Psi^{\text{qtr}}(\sigma_1, \sigma_2) \frac{1}{\sqrt{2}}[\delta_{\theta_1, 0} \delta_{\theta_2, 90^\circ} + \delta_{\theta_1, 90^\circ} \delta_{\theta_2, 0}].$$

(7.24)

The peculiarity of the ququart, characterized by wave function (7.24), consists in factorization of parts, depending on polarization and angular variables. Hence the total Schmidt entanglement parameter of state (7.24) appears to be factorable too:

$$K^{\text{qqrt}} = K^{\text{qtr}} \times K^{\text{angle}},$$

(7.25)

where $K^{\text{qtr}}$ and $K^{\text{angle}}$ are the Schmidt entanglement parameters of the qutrit $\Psi^{\text{qtr}}$ and of the angular part of the wave function $\Psi^{\text{qqrt}}$. Actually, the angular part of the Schmidt entanglement
Parameter is $K^{\text{angle}} = 2$. Hence, equation (7.25) takes the simplest form: $K^{\text{qutrit}} = 2K^{\text{qtr}},$ i.e. in terms of the Schmidt entanglement parameter, post-selection after the BS doubles the degree of entanglement of the original qutrit. Although interesting enough, factorization of the Schmidt entanglement parameter is a specific feature of the ququart of the form (7.24). In a general case, the degree of entanglement of ququarts, characterized by any entanglement quantifier, is unseparable for angular/frequency and polarization parts.

8. Conclusions

To summarize, the consideration given above provided a systematic description of biphoton qutrits and ququarts and their features such as symmetry, dimensionality and entanglement. As both qutrits and ququarts are two-boson formations, their wave functions are obliged to be symmetric with respect to permutations of photon variables, and this condition is important for correct evaluation of the degree of entanglement. In our analysis, the degree of entanglement is evaluated by entanglement quantifiers such as the Schmidt entanglement parameter, concurrence and subsystem von Neumann entropy. All of them are good for pure bipartite states; they characterize the degree of entanglement in different metrics but, of course, their predictions must be compatible with each other. The Schmidt-mode analysis is also applied and shown to be very fruitful for finding families of non-entangled, factorable qutrits. For qutrits we found explicitly the most general three-parametric families of non-entangled and maximally entangled states. In particular, we showed that one of the basis states of qutrits, in which photons have different polarization (horizontal and vertical), belongs to the family of maximally entangled qutrits. Some other interesting features of qutrits are analyzed, such as e.g. anticorrelation of entanglement and polarization and so on. In the case of biphoton ququarts, the traditional two-qubit model is shown to be invalid. As ququarts are produced by biphoton beams with non-degenerate photons (either in frequencies or in directions of propagation), they have more degrees of freedom than qutrits, e.g. polarization and frequency. In such cases the frequencies of photons are variables rather than simply some given numbers, and frequency entanglement has to be taken into account together with polarization entanglement. Thus, biphoton ququarts are shown to be two-qudit rather than two-qubit states, with the dimensionality of the one-photon Hilbert space $d = 4$ and the dimensionality of the two-photon Hilbert space $D = d^2 = 16$ (in contrast with $d = 2$ and $D = d^2 = 4$ in the case of qutrits). This new understanding of the physics of biphoton ququarts gave new formulae for their entanglement quantifiers (7.16) and (7.17). One of the qualitative consequences of these results is that all ququarts are entangled and unseparable, in contrast to earlier predictions of the two-qubit model. For both qutrits and ququarts, schemes of their complete reconstruction from experimental data are suggested and described in detail in the two appendices below. The schemes are based on using a non-selective BS and carrying out full sets of coincidence photon-counting measurements in the usual horizontal–vertical basis and in the basis turned $45^\circ$.

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Appendix A. Scheme for direct experimental measurement of the qutrit’s degree of entanglement

We will show here that there is a rather simple method of measuring experimentally the degree of entanglement of qutrits (supposedly not known in advance). The key idea consists in splitting the original biphoton beam for two identical parts by a non-selective BS and performing a series of coincidence photon-counting measurements in channels I and II (see figure A.1(a)).

Measurements have to be done with polarizers installed in the horizontal (x) and vertical (y) directions, as well as along axes x’ and y’ turned 45° with respect to, correspondingly, the x- and y-axes (figure A.1(b)). As shown below, this set of measurements is sufficient for determining the degree of entanglement as well as for a full reconstruction of all parameters of arbitrary biphoton qutrits.

A.1. Beam splitter (BS)

In terms of biphoton wave functions, the BS adds an additional degree of freedom to each photon: the propagation angles $\theta_1$ and $\theta_2$, which can take only two values, 0 and 90°. In a nonselective BS each photon has equal probabilities of transmitting or being reflected. Thus, if the biphoton wave function in front of the BS is $\Psi_1$, after the BS it takes the form

$$
\Psi_{\text{after BS}} = \Psi \times \frac{1}{2}(\delta_{\theta_1,0} - \delta_{\theta_1,90}) (\delta_{\theta_2,0} - \delta_{\theta_2,90})
$$

$$
\equiv \Psi \otimes \frac{1}{2} \begin{pmatrix} 1 \\ -1 \\ -1 \\ -1 \end{pmatrix}_{\theta_1} \otimes \begin{pmatrix} 1 \\ -1 \end{pmatrix}_{\theta_2}.
$$

(A.1)

As the angular variables in $\Psi_{\text{after BS}}$ are separated from the polarization variables of $\Psi$ and, besides, parts depending on the variables $\theta_1$ and $\theta_2$ are also factorized, the angular factor in $\Psi_{\text{after BS}}$ (A.1) does not add any additional entanglement to this wave function compared with $\Psi$. That is the non-selective BS itself does not increase or diminish the degree of entanglement of any biphoton states. Coincidence measurements in the state arising after the BS are suggested here as a tool for determining qutrit parameters in front of the BS and the degree of entanglement of the original biphoton state.

As follows from equation (A.1) and from the described features of the BS, the latter splits between channels I and II only photons of half of all incoming biphoton pairs, whereas the other half remain unsplit, and these unsplit pairs are equally distributed between channels I and II. Let $N_{\text{HH}}^{\text{tot}}$, $N_{\text{VV}}^{\text{tot}}$ and $N_{\text{HV}}^{\text{tot}}$ be large ($\gg 1$) total numbers of biphoton pairs with coinciding (HH, VV) and differing (HV) polarizations of photons generated in a crystal for some given...
time. Then the total number of generated pairs is \( N_{\text{tot}}^{\text{pairs}} = N_{\text{HH}}^{\text{tot}} + N_{\text{VV}}^{\text{tot}} + N_{\text{HV}}^{\text{tot}} \) and the total number of generated photons is \( N_{\text{phot}}^{\text{tot}} = 2 N_{\text{tot}}^{\text{pairs}} \). Among all these generated photons there are \( N_{\text{H}}^{\text{tot}} = 2 N_{\text{HH}}^{\text{tot}} + N_{\text{HV}}^{\text{tot}} \) horizontally and \( N_{\text{V}}^{\text{tot}} = N_{\text{HV}}^{\text{tot}} + 2 N_{\text{VV}}^{\text{tot}} \) vertically polarized photons. Relative numbers of horizontally and vertically polarized photons can be interpreted as single-particle (absolute) probabilities for photons to have horizontal and vertical polarizations:

\[
\begin{align*}
  w^{(s)}_{\text{H}} &= \frac{N_{\text{H}}^{\text{tot}}}{N_{\text{phot}}^{\text{tot}}} = \frac{N_{\text{HH}}^{\text{tot}} + \frac{1}{2} N_{\text{HV}}^{\text{tot}}}{N_{\text{HH}}^{\text{tot}} + N_{\text{VV}}^{\text{tot}} + N_{\text{HV}}^{\text{tot}}}, \\
  w^{(s)}_{\text{V}} &= \frac{N_{\text{V}}^{\text{tot}}}{N_{\text{phot}}^{\text{tot}}} = \frac{N_{\text{VV}}^{\text{tot}} + \frac{1}{2} N_{\text{HV}}^{\text{tot}}}{N_{\text{HH}}^{\text{tot}} + N_{\text{VV}}^{\text{tot}} + N_{\text{HV}}^{\text{tot}}}.
\end{align*}
\]

Although these equations are appropriate, unfortunately in experiments the numbers of generated pairs are not directly observable.

After the BS we obtain \( \frac{1}{4} N_{\sigma\sigma'}^{\text{tot}} \) unsplit pairs in each channel, and photons of the remaining \( \frac{1}{2} N_{\sigma\sigma'}^{\text{tot}} \) split pairs are equally distributed between channels I and II (figure A.2).

Owing to unsplit pairs, the results of photon counting by a single detector in one of the channels I or II can be rather confusing, because when an unsplit pair arrives at the detector the latter can produce one click instead of two. In contrast, coincidence measurements by two detectors in channels I and II (figure A.2(a)) register only photons of split pairs, which makes such measurements quite unambiguous and informative.

Relations between the number of single photons and the corresponding total number of pairs are different in cases when single photons arise from split HH and VV pairs and when they arise from split HV pairs. As seen from figure A.2, the number of H- or V-polarized photons arising from split HH or VV pairs equals \( \frac{1}{2} N_{\text{HH}}^{\text{tot}} \) or \( \frac{1}{2} N_{\text{VV}}^{\text{tot}} \) (figure A.2(a)), whereas the number of the same photons arising from split HV pairs is equal to \( \frac{1}{4} N_{\text{HV}}^{\text{tot}} \) (figure A.2(b)). From these results, we easily find the numbers of detector coincidence counts \( N_{\sigma\sigma'}^{\text{d(c)}} \) obtainable when \( \sigma \)- and \( \sigma' \)-polarizers are installed in front of the detectors, correspondingly, in channels...
where $\eta$ is the efficiency of detectors. Relative numbers of coincidence counts coincide with conditional probabilities $w^{(c)}_{\sigma | \sigma'}$ for a photon ‘1’ to have polarization $\sigma$ under the condition that another photon ‘2’ has polarization $\sigma'$. They are defined as ratios of specific coincidence detector counts (A.3) to the total numbers of coincidence counts:

$$w^{(c)}_{\sigma | \sigma'} = \frac{N^{d(c)}_{\sigma | \sigma'}}{\sum_{\sigma, \sigma'} N^{d(c)}_{\sigma | \sigma'}} = \eta \frac{N_{\text{tot}}^{HH} + N_{\text{tot}}^{HV} + N_{\text{tot}}^{VV}}{2 N_{\text{pair}}^{\text{tot}}}. \tag{A.5}$$

Evidently, the sum of all conditional probabilities (A.4) equals unity:

$$\sum_{\sigma, \sigma'} w^{(c)}_{\sigma | \sigma'} = w^{(c)}_{H | H} + 2 w^{(c)}_{H | V} + w^{(c)}_{V | V} = 1. \tag{A.6}$$

Equations (A.4) and (A.5) show that, in principle, experimental measurement of conditional probabilities is straightforward because they are expressed explicitly in terms of the coincidence detector counts $N^{d(c)}_{\sigma | \sigma'}$. In terms of numbers of pairs in a beam, expressions for conditional probabilities take the form

$$w^{(c)}_{\sigma | \sigma'} = \frac{N^{d(c)}_{\sigma | \sigma'}}{\sum_{\sigma, \sigma'} N^{d(c)}_{\sigma | \sigma'}} = \frac{N_{\text{tot}}^{HH}}{2 N_{\text{pair}}^{\text{tot}}}, \quad w^{(c)}_{V | V} = \frac{N_{\text{tot}}^{VV}}{2 N_{\text{pair}}^{\text{tot}}}. \tag{A.7}$$

By the definition of conditional probabilities, their sums can be constructed to give single-particle, unconditional probabilities:

$$w^{(s)}_H = w^{(c)}_H | H + w^{(c)}_H | V = \frac{N_{\text{tot}}^{HH} + \frac{1}{2} N_{\text{tot}}^{HV}}{N_{\text{pair}}^{\text{tot}}},$$

$$w^{(s)}_V = w^{(c)}_V | V + w^{(c)}_V | H = \frac{N_{\text{tot}}^{VV} + \frac{1}{2} N_{\text{tot}}^{HV}}{N_{\text{pair}}^{\text{tot}}}. \tag{A.8}$$

These expressions are absolutely identical to equations (A.2) derived simply by counting the numbers of photons in the original biphoton beam without any BS. Coincidence of equations (A.2) and (A.8) proves that, indeed, coincidence measurements after the BS can be used for obtaining information about such features of the beam before the BS as characterizing unconditional (single-particle) probabilities for photons to have horizontal or vertical polarizations. In other words, equations (A.8) show that although, owing to unsplit pairs, direct measurement of single-particle probabilities after the BS is problematic, nevertheless, probabilities $w^{(s)}_H$ and $w^{(s)}_V$ can be found from the results of coincidence measurements.

Conditional probabilities of equation (A.7) can be used for a partial reconstruction of the qutrit parameters from experimental data. Indeed, directly from equation (4.1) for the density

$$
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matrix $\rho$, we find the following relations between the absolute values of all three coefficients $C_{1,2,3}$ in wave function (3.11) and the conditional probabilities

$$w_H^{(c)} |H| = |C_1|^2, \quad w_V^{(c)} |V| = |C_3|^2.$$  \hspace{1cm} (A.9)

$$w_V^{(c)} |H| = w_H^{(c)} |V| = \frac{1}{2} |C_2|^2.$$  \hspace{1cm} (A.10)

For single-particle probabilities (A.8), relations with parameters of qutrits $C_i$ have the form

$$w_H^{(s)} = |C_1|^2 + \frac{1}{2} |C_2|^2, \quad w_V^{(s)} = |C_3|^2 + \frac{1}{2} |C_2|^2.$$  \hspace{1cm} (A.11)

### A.2. Qutrits in a turned basis

In addition to absolute values of $C_i$, we have to find their phases, $\varphi_{1,2,3}$, and this requires additional measurements, e.g. with polarizers installed at 45° to the $x$- and $y$-axes, as shown in figure A.1(b). In theory, this is equivalent to description of the biphoton wave function in a turned basis. In a general case, let $x^\alpha$ and $y^\alpha$ be axes turned an angle $\alpha$ with respect to the $x$- and $y$-axes. Let polarizations along the axes $x^\alpha$ and $y^\alpha$ be denoted as $H^\alpha$ and $V^\alpha$. The corresponding one-photon wave functions are $\left(\begin{array}{l}1 \\ 0 \end{array}\right)^\alpha$ and $\left(\begin{array}{l}0 \\ 1 \end{array}\right)^\alpha$. From these one-photon wave functions, we can construct the two-photon basis wave functions $\Psi_{HH}=\Psi_{HV}=\Psi_{VH}$ and $\Psi_{VV}$ in the same way (3.8)–(3.10) as $\Psi_{HH}$, $\Psi_{HV}$ and $\Psi_{VH}$ were constructed from the one-photon wave functions $\left(\begin{array}{l}1 \\ 0 \end{array}\right)$ and $\left(\begin{array}{l}0 \\ 1 \end{array}\right)$. Let us present the wave function (3.11) in terms of expansion in the basis wave function of the frame $(x^\alpha, y^\alpha)$:

$$\Psi = C_1(\alpha) \Psi_{HH} + C_2(\alpha) \Psi_{HV} + C_3(\alpha) \Psi_{VV} + C_3(\alpha) \Psi_{VV}.$$  \hspace{1cm} (A.11)

One-photon wave functions in the $\alpha$- and original frames are related to each other by the evident transformation formulae

$$\begin{align*}
\left(\begin{array}{l}1 \\ 0 \end{array}\right)^\alpha &= \cos \alpha \left(\begin{array}{l}1 \\ 0 \end{array}\right)^{(\alpha)} - \sin \alpha \left(\begin{array}{l}0 \\ 1 \end{array}\right)^{(\alpha)}, \\
\left(\begin{array}{l}0 \\ 1 \end{array}\right)^\alpha &= \sin \alpha \left(\begin{array}{l}1 \\ 0 \end{array}\right)^{(\alpha)} + \cos \alpha \left(\begin{array}{l}0 \\ 1 \end{array}\right)^{(\alpha)}.
\end{align*}$$  \hspace{1cm} (A.12)

By applying these formulae to all terms and columns in equation (3.11) and regrouping the resulting $\alpha$-frame column products, we finally reduce the biphoton wave function (3.11) to the form (A.11) with the following relations between coefficients $C_{1,2,3}(\alpha)$ and $C_{1,2,3}$:

$$\begin{align*}
C_1(\alpha) &= \cos^2 \alpha C_1 + \sqrt{2} \cos \alpha \sin \alpha C_2 + \sin^2 \alpha C_3, \\
C_2(\alpha) &= -\sqrt{2} \cos \alpha \sin \alpha (C_1 - C_3) + \cos 2\alpha C_2, \\
C_3(\alpha) &= \sin^2 \alpha C_1 - \sqrt{2} \cos \alpha \sin \alpha C_2 + \cos^2 \alpha C_3.
\end{align*}$$  \hspace{1cm} (A.13)

Note that similar transformation formulae for constants $C_{\pm}$ (3.13) have the form

$$C_+ (\alpha) = C_+, \quad \begin{cases} C_-(\alpha) = \cos 2\alpha C_- + \sin 2\alpha C_2, \\
C_2(\alpha) = -\sin 2\alpha C_- + \cos 2\alpha C_2. \end{cases}$$  \hspace{1cm} (A.14)

This means, in particular, that

$$\begin{align*}
\Psi_+^{(\alpha)} &= \Psi_+, \\
\Psi_-^{(\alpha)} &= \cos 2\alpha \Psi_- + \sin 2\alpha \Psi_{HV}, \\
\Psi_{HV}^{(\alpha)} &= \cos 2\alpha \Psi_{HV} - \sin 2\alpha \Psi_-.
\end{align*}$$  \hspace{1cm} (A.16)
i.e. the function $\Psi_\alpha^{(\alpha)}$ has the same form in all $\alpha$-frames, whereas the functions $\Psi_-^{(\alpha)}$ and $\Psi_{HV}^{(\alpha)}$ transform into each other with changing $\alpha$.

In a special case ($\alpha = 45^\circ$), equations (A.13) are reduced to

$$C_{1,3}(45^\circ) = \frac{C_1 + C_3}{2} \pm \frac{C_2}{\sqrt{2}}, \quad C_2(45^\circ) = \frac{C_1 - C_3}{\sqrt{2}}. \quad (A.17)$$

Similar to (A.9), qutrit parameters in the basis turned by $45^\circ$ can be expressed in terms of the corresponding conditional probabilities

$$w_{IV}^{(c)}|_{IV} = |C_1(45^\circ)|^2, \quad w_{VIV}^{(c)}|_{VIV} = |C_3(45^\circ)|^2,$$

$$w_{IV}^{(c)}|_{IV} = w_{IV}^{(c)}|_{VIV} = \frac{1}{2} |C_2(45^\circ)|^2. \quad (A.18)$$

In turn, conditional probabilities $w_{\alpha|IV}$ are related by equations identical to (A.7) to the corresponding numbers of counts with polarizers installed in front of the detectors along the directions of the $x'$- and $y'$-axes in figure A.1(b):

$$w_{\alpha|IV} = \frac{N_{\alpha|IV}^{(c)}}{\sum_{\alpha|IV} N_{\alpha|IV}^{(c)}}. \quad (A.19)$$

Similar to (A.8) and (A.10), we can also define single-particle probabilities via conditional ones in a turned basis:

$$w_\sigma^{(s)} = w_\sigma^{(c)}|_IV + w_{IV}^{(c)}|_{VIV} = |C_1(45^\circ)|^2,$$

$$w_\sigma^{(s)} = w_\sigma^{(c)}|_{VIV} + w_{IV}^{(c)}|_IV = |C_3(45^\circ)|^2. \quad (A.20)$$

A.3. Concurrency and full reconstruction of qutrit parameters from experimental data

In terms of absolute values of the qutrit parameters $C_{1,2,3}$ and their phases $\varphi_{1,2,3}$, the squared concurrence $C$ equation (5.3) has the form

$$C^2 = 4|C_1|^2|C_3|^2 + |C_2|^4 - 4|C_1||C_3||C_2|^2 \cos(\varphi_1 + \varphi_3 - 2\varphi_2). \quad (A.21)$$

As is well known and mentioned above, the common phase of the biphoton wave function does not affect any measurements. This means that multiplication of $\Psi$ by an arbitrary phase factor $e^{i\psi_0}$ does not change any physical results. Let us take, for example, $e^{i\psi_0} = -\varphi_2$, which makes the parameter $e^{i\psi_0}C_2$ real, with corresponding changes of phases in two other parameters, $C_1$ and $C_3$. Equivalently, keeping in mind this procedure we can simply take $C_2$ real and, in a general case, $C_{1,3}$ complex—$C_1 = |C_1|e^{i\psi_1}$ and $C_3 = |C_3|e^{i\psi_3}$. With real $C_2$ ($\varphi_2 = 0$), equation (A.21) takes the form

$$C^2 = 2C_1C_3 - C_2^2 \left| 2C_1C_3 - C_2^2 \right|^2 = 4|C_1|^2|C_3|^2 + C_2^4 - 4|C_1||C_3||C_2|^2 \cos(\varphi_1 + \varphi_3). \quad (A.22)$$

For finding phases $\varphi_1$ and $\varphi_3$, we can use equations (A.17), the second of which gives

$$|C_2(45^\circ)|^2 = \frac{|C_1 - C_3|^2}{2} = \frac{|C_1|^2 + |C_3|^2}{2} - |C_1||C_3|\cos(\varphi_1 - \varphi_3). \quad (A.23)$$
One equation for two unknown phases $\varphi_1$ and $\varphi_3$ can be obtained from the difference of squared absolute values of the parameters $C_i(45^\circ)$ and $C_i(45^\circ)$ in equations (A.17):

$$|C_1(45^\circ)|^2 - |C_3(45^\circ)|^2 = \sqrt{2} C_2 \text{Re}(C_1 + C_3)$$

$$= \sqrt{2} C_2 \left| [C_1 \cos \varphi_1 + C_3 \cos \varphi_3] \right|.$$  \quad \text{(A.24)}

In a general case, equations (A.23) and (A.24) can hardly be further simplified to yield a simple analytical formula for the squared concurrence (A.21) in terms of the experimentally measurable conditional probabilities. But numerical solution of equations (A.23) and (A.24) with all parameters $|C_i|$ and $|C_i(45^\circ)|$ known does not present any problems. Thus, by obtaining a full set of measurements of coincidence counts in two bases $(x,y)$ and $(x'y')$ (figure A.2(b)), one can find all conditional probabilities $w^{(s)}_{\sigma|x}$ (A.7) and $w^{(s)}_{\sigma'|x'=y'}$ (A.19). Then, from equations (A.9) and (A.18) one finds all absolute values of parameters $C_i$ and $C_i(45^\circ)$. And, finally, with known values of $|C_i|$ and $|C_i(45^\circ)|$, one solves numerically equations (A.23) and (A.24) and finds phases $\varphi_i$ and $\varphi_3$ with $\varphi_2 = 0$. This procedure permits us to reconstruct completely all qutrit parameters and to find the degree of entanglement (A.22) from experimental coincidence measurements. The procedure might be referred to as an alternative protocol of quantum state tomography of biphoton-based qutrits rather than the one described, e.g., by Bogdanov et al [29, 30].

Note that if it is known in advance that all parameters $C_i$ are real, the procedure of finding the degree of entanglement from experimental data significantly simplifies. In this case, it is more convenient to begin with equation (5.1) for the inverse Schmidt entanglement parameter, which is easily expressed in terms of single-particle probabilities (A.10). For the upper line on the right-hand side of equation (5.1), we obtain

$$\left( \left| C_1 \right|^2 + \left| C_2 \right|^2 \right) \left( \left| C_3 \right|^2 + \left| C_2 \right|^2 \right) = w_H^{(s)} + w_V^{(s)} \equiv \frac{1}{2} \left\{ 1 + \left( w_H^{(s)} - w_V^{(s)} \right)^2 \right\}. \quad \text{(A.25)}$$

On the other hand, with the help of equations (A.17) and (A.20), the lower line on the right-hand side of equation (5.1) can be easily expressed in terms of qutrit parameters and single-particle probabilities in the basis turned by $45^\circ$:

$$|C_2^* C_1 + C_2 C_3|^2 = C_2^2 (C_1 + C_3)^2$$

$$= \frac{\left\{ \left| C_1(45^\circ) \right|^2 - \left| C_3(45^\circ) \right|^2 \right\}^2}{2} = \frac{\left( w_H^{(s)} - w_V^{(s)} \right)^2}{2}. \quad \text{(A.26)}$$

Altogether this gives

$$K^{-1} = \frac{1}{2} \left\{ 1 + \left( w_H^{(s)} - w_V^{(s)} \right)^2 + \left( w_H^{(s)} - w_V^{(s)} \right)^2 \right\}$$

and

$$C = \sqrt{1 - \left( w_H^{(s)} - w_V^{(s)} \right)^2 - \left( w_H^{(s)} - w_V^{(s)} \right)^2}. \quad \text{(A.27)}$$

**Appendix B. Measuring the ququart’s parameters**

One possible way of measuring directly the parameters of ququarts, and in particular their degree of entanglement, is similar to that described above for qutrits and illustrated by figure A.2: the original biphoton beam has to be split into two channels by a non-selective BS and a full set of coincidence measurements has to be carried out in the $xy$ and $x'y'$ bases of figure A.2(b).
Differently from qutrits, in addition to polarizers one has to install frequency filters in front of the detectors to count the numbers of photons in the given frequency–polarization Hh, Hl, Vh and Vl. Because in ququarts all the basis biphoton states are of type $a_i^\dagger a_j^\dagger |0\rangle$ with $i \neq j$ (where $i$ and $j$ numerate above indicated modes), the distribution of photons of split pairs in channels I and II corresponds to the type shown in figure A.2(b). Hence, if $N_{i,j}^{\text{tot}}$ are the total numbers of generated pairs with photons in modes $i$ and $j$, the numbers of corresponding coincidence detector counts are determined by equations similar to that of the last formula in equation (A.3):

$$N_j^{d(c)}|_j = N_i^{d(c)}|_i = \frac{\eta}{4} N_{i,j}^{\text{tot}},$$

(B.1)

with $\eta$ denoting the efficiency of detectors. The full set of data on coincidence numbers of counts can be used further to find conditional probabilities for a photon ‘1’ to be in some mode $i$ under the condition that the second photon of the same pair is in some other mode $j$ (similar to equation (A.7)):

$$w_j^{(c)}|_j = \frac{N_j^{d(c)}|_j}{\sum_{i,j} N_i^{d(c)}|_j},$$

(B.2)

with the normalization condition $\sum_{i,j} w_j^{(c)}|_j = 1$.

On the other hand, conditional probabilities are determined by diagonal elements of the full density matrix with respect to both polarization and frequency variables and of both photons. Directly from expressions (7.9)–(7.12) for the basis wave functions of ququarts, from the general expression (7.14) for the ququart wave function and from the definition of the density matrix $\rho = \Psi \Psi^\dagger$, we find the following series of relations between the conditional probabilities and the parameters $C_i$ of ququarts:

$$|C_1|^2 = 2w_{Hh}^{(c)}|_{Hh} = 2w_{Hh}^{(c)}|_{Hh}, \quad |C_2|^2 = 2w_{Hh}^{(c)}|_{Vl} = 2w_{Vl}^{(c)}|_{Hh},$$

$$|C_3|^2 = 2w_{Hh}^{(c)}|_{Vh} = 2w_{Vh}^{(c)}|_{Hh}, \quad |C_4|^2 = 2w_{Vh}^{(c)}|_{Vl} = 2w_{Vl}^{(c)}|_{Vh}.$$

(B.3)

The same relations (B.1)–(B.3) between numbers of detector counts, conditional probabilities and expansion coefficients of the ququart wave function can also be written in the $x'y'$ basis turned by $45^\circ$ with respect to the horizontal–vertical basis. Thus, all sets of coincidence measurements in two bases can be used to determine all absolute values of ququart parameters in both bases, $|C_i|^2$ and $|C_i(45^\circ)|^2$. Relations between $C_i(45^\circ)$ and $C_i$ are easily found in the same way as in the case of qutrits, and in the case of ququarts they are given by

$$C_1(45^\circ) = \frac{1}{2} \left( C_1 + C_2 + C_3 + C_4 \right),$$

$$C_2(45^\circ) = \frac{1}{2} \left( -C_1 + C_2 - C_3 + C_4 \right),$$

$$C_3(45^\circ) = \frac{1}{2} \left( -C_1 - C_2 + C_3 + C_4 \right),$$

$$C_4(45^\circ) = \frac{1}{2} \left( C_1 - C_2 - C_3 + C_4 \right).$$

(B.4)

The squared absolute values of expressions on the left- and right-hand sides of equations (B.4) take the form of equations for the phases $\varphi_{1,2,3,4}$ of constants $C_i$. For shortening formulae, these equations can be grouped in pairs to give

$$|C_1(45^\circ)|^2 + |C_2(45^\circ)|^2 - \frac{1}{2} = |C_1| |C_3| \cos(\varphi_1 - \varphi_3) + |C_2| |C_4| \cos(\varphi_2 - \varphi_4),$$

(B.5)

$$|C_1(45^\circ)|^2 + |C_3(45^\circ)|^2 - \frac{1}{2} = |C_1| |C_2| \cos(\varphi_1 - \varphi_2) + |C_3| |C_4| \cos(\varphi_2 - \varphi_4),$$

(B.6)
\[ |C_1(45^\circ)|^2 + |C_4(45^\circ)|^2 - \frac{1}{2} = |C_1||C_4| \cos(\varphi_1 - \varphi_4) + |C_2||C_3| \cos(\varphi_2 - \varphi_3). \] (B.7)

Because of normalization \( \sum_i |C_i(45^\circ)|^2 = 1 \), one can obtain only three independent equations of type (B.5)–(B.7) from four equations (B.4). The fourth equation for finding four phases \( \varphi_{1,2,3,4} \) follows from the above-mentioned fact that the phase of the wave function \( \Psi^{(qqrt)} \) (7.14) as a whole does not affect any observable quantities and can be arbitrarily chosen. For this reason, one can put some additional condition restriction for phases, such as

\[ \varphi_1 + \varphi_2 + \varphi_3 + \varphi_4 = 0. \] (B.8)

Four equations (B.5)–(B.8) are sufficient for finding numerically all phases \( \varphi_{1,2,3,4} \) as soon as the squared absolute values \( |C_i|^2 \) and \( |C_i(45^\circ)|^2 \) are found from coincidence measurements. Thus the procedure described above provides a full reconstruction of the ququart wave function \( \Psi^{(qqrt)} \) (7.14) and, in particular, can be used for determining the degree of entanglement. This procedure serves as an alternative protocol of quantum state tomography of ququarts described in the work [31].

Note that, in analogy with the case of qutrits, the determination of entanglement quantities of ququarts from experimental data is simplified if the parameters \( C_{1,2,3,4} \) are known in advance to be real. In this case, the key element of equations (7.16) and (7.17) for the Schmidt entanglement parameter and \( I \)-concurrence can be written as

\[ |C_1C_4 - C_2C_3|^2 = C_1^2C_4^2 + C_2^2C_3^2 - 2C_1C_4C_2C_3. \] (B.9)

On the other hand, by summing the squared first and last lines of equations (B.4) we obtain

\[ C_1C_4 + C_2C_3 = C_1^2(45^\circ) + C_4^2(45^\circ)^2 - \frac{1}{2}. \] (B.10)

Now, with a simple algebra, we find from equations (B.9) and (B.10) the following final expression for \( |C_1C_4 - C_2C_3|^2 \):

\[ |C_1C_4 - C_2C_3|^2 = 2 \left( |C_1|^2|C_4|^2 + |C_2|^2|C_3|^2 \right) - \left[ |C_1(45^\circ)|^2 + |C_4(45^\circ)|^2 - \frac{1}{2} \right]^2, \] (B.11)

where all terms on the right-hand side are expressible via conditional probabilities either in the original (\( xy \)) basis or in the basis turned by 45\(^\circ\). This solves the problem of determining the entanglement quantities \( K \) or \( C_I \).

References

[16] Schweber S S 1961 An Introduction to Relativistic Quantum Field Theory (Evaston, IL: Row, Peterson and Co) section 6, equation (34)