Intrinsic upper bound on two-qubit polarization entanglement predetermined by pump polarization correlations in parametric down-conversion

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(Dated: April 22, 2016)

We study how one-particle correlations transfer to manifest as two-particle correlations in the context of parametric down-conversion (PDC), a process in which a pump photon is annihilated to produce two entangled photons. We work in the polarization degree of freedom and show that for any two-qubit generation process that is both trace-preserving and entropy-nondecreasing the concurrence $C(\rho)$ of the generated two-qubit state $\rho$ follows an intrinsic upper bound with $C(\rho) \leq (1 + P)/2$, where $P$ is the degree of polarization of the pump photon. We also find that for the class of two qubit states that is restricted to have only two non-zero diagonal elements such that the effective dimensionality of the two-qubit state is same as the dimensionality of the pump polarization state, the upper bound on concurrence is the degree of polarization itself, that is, $C(\rho) \leq P$. Our work shows that the maximum manifestation of two-particle correlations as entanglement is dictated by one-particle correlations. The formalism developed in this work can be extended to include multiparticle systems and can thus have important implications towards deducing the upper bounds on multi-particle entanglement, for which no universally accepted measure exists.

PACS numbers:
field. In this Letter, we study correlation transfer from one-particle to two-particle systems, not in any restricted subspace, but in the complete space of the polarization degree of freedom. We quantify intrinsic one-particle correlations in terms of the degree of polarization and the two-particle correlations in terms of concurrence.

We begin by noting that the state of a normalized quasi-monochromatic pump field may be described by a $2 \times 2$ density matrix $\rho$ given by

$$J = \begin{bmatrix} \langle E_n^x E_n^x \rangle & \langle E_n^x E_{n'}^x \rangle \\ \langle E_{n'}^x E_n^x \rangle & \langle E_{n'}^x E_{n'}^x \rangle \end{bmatrix},$$

which is referred to as the ‘polarization matrix.’ The complex random variables $E_n^x$ and $E_{n'}^x$ denote the horizontal and vertical components of the electric field, respectively, and $\langle \cdots \rangle$ denotes an ensemble average. By virtue of a general property of $2 \times 2$ density matrices, $J$ has a decomposition of the form,

$$J = P |\psi_{pol}\rangle \langle \psi_{pol}| + (1 - P) \mathbb{1},$$

where $|\psi_{pol}\rangle$ is a pure state representing a completely polarized field, and $\mathbb{1}$ denotes the normalized $2 \times 2$ identity matrix representing a completely unpolarized field $\mathbb{1}$. This means that any arbitrary field can be treated as a unique weighted mixture of a completely polarized part and a completely unpolarized part. The fraction $P$ corresponding to the completely polarized part is called the degree of polarization and is a basis-invariant measure of polarization correlations in the field. If we denote the eigenvalues of $J$ as $\epsilon_1$ and $\epsilon_2$, then it can be shown that $P = |\epsilon_1 - \epsilon_2|$. Furthermore, the eigenvalues are connected to $P$ as $\epsilon_1 = (1 + P)/2$ and $\epsilon_2 = (1 - P)/2$.

We now investigate the PDC-based generation of polarization entangled two-qubit signal-idler states $\rho$ from a quasi-monochromatic pump field $J$ (see Fig. 1). The nonlinear optical process of PDC is a very low-efficiency process $\mathbb{1}$. Most of the pump photons do not get down-converted and just pass through the nonlinear medium. Only a very few pump photons do get down-converted, and in our description, only these photons constitute the ensemble containing the pump photons. We further assume that the probabilities of the higher-order down-conversion processes are negligibly small so that we do not have in our description the down-converted state containing more than two photons. With these assumptions, we represent the state of the down-converted signal and idler photons by a $4 \times 4$, two-qubit density matrix $\rho$ in the polarization basis $\{|H\rangle_s |H\rangle_i , |H\rangle_s |V\rangle_i , |V\rangle_s |H\rangle_i , |V\rangle_s |V\rangle_i \}$. In what follows, we will be applying some results from the theory of majorization $\mathbb{1}$ in order to study the propagation of correlations from the $2 \times 2$ pump density matrix $J$ to the $4 \times 4$ two-qubit density matrix $\rho$. This requires us to equalize the dimensionalities of the pump and the two-qubit states. We therefore represent the pump field by a $4 \times 4$ matrix $\sigma$, where

$$\sigma = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \otimes J.$$  

We denote the eigenvalues of $\sigma$ in non-ascending order as $(\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4) = ((1 + P)/2, (1 - P)/2, 0, 0)$ and the eigenvalues of $\rho$ in non-ascending order as $(\lambda_1, \lambda_2, \lambda_3, \lambda_4)$.

Let us represent the two-qubit generation process $\sigma \to \rho$ by a completely positive map $\mathcal{E}$ (see Fig. 1) such that

$$\rho = \mathcal{E}(\sigma) = \sum_i M_i \sigma M_i^\dagger,$$

where $M_i$’s are the Sudarshan-Kraus operators for the process $\mathbb{1}$. We restrict our analysis only to maps that satisfy the following two conditions for all $\sigma$: (i) No part of the system can be discarded, that is, there must be no postselection. This means that the map must be trace-preserving, which leads to the condition that $\sum_i M_i M_i^\dagger = \mathbb{1}$; (ii) Coherence may be lost to, but not gained from degrees of freedom external to the system. In other words, the von Neumann entropy cannot decrease. This condition holds if and only if the map is unital, that is, $\sum_i M_i M_i^\dagger = \mathbb{1}$. The above two conditions together imply that the process $\sigma \to \rho$ is doubly-stochastic $\mathbb{1}$. The characteristic implication of double-stochasticity is that the two-qubit state is majorized by the pump state, that is $\rho \prec \sigma$. This means that the eigenvalues of $\rho$ and $\sigma$ satisfy the following relations:

$$\lambda_1 \leq \epsilon_1,$$

$$\lambda_3 + \lambda_2 \leq \epsilon_1 + \epsilon_2,$$

$$\lambda_1 + \lambda_2 + \lambda_3 \leq \epsilon_1 + \epsilon_2 + \epsilon_3,$$

$$\lambda_1 + \lambda_2 + \lambda_3 + \lambda_4 = \epsilon_1 + \epsilon_2 + \epsilon_3 + \epsilon_4.$$  

We must note that condition (i) may seem not satisfied in some of the experimental schemes for producing polarization entangled two-qubit states. For example, in the scheme for producing a polarization Bell state using Type-II phase-matching $\mathbb{1}$, only one of the polarization components of the pump photon is allowed to engage in the down-conversion process; the other polarization component, even if present, simply gets discarded away. Nevertheless, our formalism is valid even for such two-qubit generation schemes. In such schemes, the state $\sigma$ represents that part of the pump field which undergoes the down-conversion process so that condition (i) is satisfied.

Now, for a general realization of the process $\sigma \to \rho$, the generated density matrix $\rho$ can be thought of as arising from a process $\mathcal{N}$, that can have a non-unitary part, fol-}

ollowed by a unitary-only process $\mathcal{U}$, as depicted in Fig. 1. This means that we have $\sigma \to \chi : = \mathcal{N}(\sigma) \to \rho \equiv \mathcal{U}(\chi)$. The process $\mathcal{N}$ generates the two-qubit state $\chi$ with eigenvalues $\{\lambda_1, \lambda_2, \lambda_3, \lambda_4\}$ which are different from the eigenvalues $\{\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4\}$ of $\sigma$, except when $\mathcal{N}$ consists of unitary-only transformations, in which case the eigenvalues of $\chi$ remain the same as that of $\sigma$. The unitary
part $\mathcal{U}$ transforms the two-qubit state $\chi$ to the final two-qubit state $\rho$. This action does not change the eigenvalues but can change the concurrence of the two-qubit state. The majorization relations of Eq. (4) dictate how the two sets of eigenvalues are related and thus quantify the effects due to $\mathcal{N}$. We quantify the effects due to $\mathcal{U}$ by using the result from Refs. [20, 37, 38] for the maximum concurrence achievable by a two-qubit state under unitary transformations. According to this result, for a two-qubit state $\rho$ with eigenvalues in non-ascending order denoted as $\lambda_1, \lambda_2, \lambda_3, \lambda_4$, the concurrence $C(\rho)$ obeys the inequality:

$$C(\rho) \leq \max\{0, \lambda_1 - \lambda_3 - 2\sqrt{\lambda_2 \lambda_4}\};$$  \hspace{1cm} (5)

the bound is saturable in the sense that there always exists a unitary transformation $U(\chi) = \rho$ for which the equality holds true [38]. Now, from Eq. (5), we clearly have $C(\rho) \leq \lambda_1$. And, from the majorization relation of Eq. (4a), we find that $\lambda_1 \leq \epsilon_1 = (1 + P)/2$. Therefore, for a general doubly-stochastic process $\mathcal{E}$, we arrive at the inequality:

$$C(\rho) \leq \frac{1 + P}{2}. \hspace{1cm} (6)$$

We stress that this bound is tight, in the sense that there always exists a pair of $\mathcal{N}$ and $\mathcal{U}$ for which the equality in the above equation holds true. In fact, the saturation of Eq.(5) is achieved when $\mathcal{N}$ consists of unitary-only process and when $\mathcal{U}$ is such that it yields the maximum concurrence for $\rho$ as allowed by Eq. (5). This can be verified, first, by noting that when $\mathcal{N}$ is unitary the process $\chi = N(\sigma)$ preserves the eigenvalues to yield $(\lambda_1, \lambda_2, \lambda_3, \lambda_4) = ((1 + P)/2, (1 - P)/2, 0, 0)$, and second, by substituting these eigenvalues in Eq. (5) which then yields $(1 + P)/2$ as the maximum achievable concurrence. Eq. (6) is the central result of this Letter which clearly states that the intrinsic polarization correlations of the pump field in PDC predetermine the maximum entanglement that can be achieved by the generated two-qubit signal-idler states. We note that while Eq. (6) has been derived keeping in mind the physical context of parametric down-conversion, the derivation does not make any specific reference to the PDC process or to any explicit details of the two-qubit generation scheme. As a result, Eq. (6) is also applicable to processes other than PDC that would produce a two-qubit state from a single source qubit state via a doubly stochastic process.

We now recall that our present work is directly motivated by previous studies in the spatial degree of freedom for two-qubit states with only two nonzero diagonal entries in the computational basis [27]. Therefore, we next consider this special class of two-qubit states in the polarization degree of freedom. We refer to such states as ‘2D states’ in this Letter and represent the corresponding density matrix as $\rho^{(2D)}$. Since such states can only have two nonzero eigenvalues, the majorization relations of Eq. (4) reduce to: $\lambda_1 \leq \epsilon_1$ and $\lambda_1 + \lambda_2 = \epsilon_1 + \epsilon_2 = 1$. Owing to its $2 \times 2$ structure, the state $\rho^{(2D)}$ has a decomposition of the form [1],

$$\rho^{(2D)} = \bar{P}|\psi^{(2D)}\rangle\langle\psi^{(2D)}| + (1 - \bar{P})I^{(2D)}, \hspace{1cm} (7)$$

where $|\psi^{(2D)}\rangle$ is a pure state and $I^{(2D)}$ is a normalized $2 \times 2$ identity matrix. As in Eq. (2), the pure state weightage $\bar{P}$ can be shown to be related to the eigenvalues as $\bar{P} = \lambda_1 - \lambda_2$. It is known that the concurrence is a convex function on the space of density matrices [22], that is, $C(\sum_i p_i \rho_i) \leq \sum_i p_i C(\rho_i)$, where $0 \leq p_i \leq 1$ and $\sum p_i = 1$. Applying this property to Eq. (4) along with the fact that $C(I^{(2D)}) = 0$, we obtain that the concurrence $C(\rho^{(2D)})$ of a 2D state satisfies $C(\rho^{(2D)}) \leq \bar{P}$. Now since $\bar{P} = \lambda_1 - \lambda_2 = 2\lambda_1 - 1$, and $\lambda_1 \leq \epsilon_1$, we get $\bar{P} \leq 2\epsilon_1 - 1 = \epsilon_1 - \epsilon_2 = P$, or $\bar{P} \leq P$. We therefore arrive at the inequality:

$$C(\rho^{(2D)}) \leq P. \hspace{1cm} (8)$$

Thus, for 2D states the upper bound on concurrence is the degree of polarization itself. This particular result is in exact analogy with the result shown previously for 2D states in the spatial degree of freedom that the maximum achievable concurrence is bounded by the degree of spatial correlations of the pump field itself [27].

Our entire analysis leading upto Eq. (6) and Eq. (8) describes the transfer of one-particle correlations, as quantified by $P$, to two-particle correlations and their eventual manifestation as entanglement, as quantified by concurrence. For 2D states, which have a restricted Hilbert space available to them, the maximum concurrence that can get manifested is $P$. Thus, restricting the Hilbert space appears to restrict the degree to which pump correlations can manifest as the entanglement of the generated two-qubit state. However, when there are no restrictions on the available Hilbert space, the maximum concurrence that can get manifested is $(1 + P)/2$.

Next, for conceptual clarity, we illustrate the bounds derived in this Letter in an example experimental scheme shown in Fig. [2a]. This scheme can produce a wide range of two-qubit states in a doubly-stochastic manner. A pump field with the degree of polarization $P$ is split into two arms by a non-polarizing beam-splitter (BS) with splitting ratio $t : 1 - t$. We represent the
horizontal and vertical polarization components of the field hitting the PDC crystals in arm (1) as $E_{H1}$ and $E_{V1}$, respectively. The phase retarder (PR1) introduces a phase difference $\alpha_1$ between $E_{H1}$ and $E_{V1}$. The rotation plate (RP1) rotates the polarization vector by angle $\theta_1$. The corresponding quantities in arm (2) have similar representations. The stochastic variable $\gamma$ introduces a decoherence between the pump fields in the two arms. Its action is described as $\langle e^{i\gamma} \rangle = \mu e^{i\gamma_0}$, where $\langle \cdots \rangle$ represents the ensemble average, $\mu$ is the degree of coherence and $\gamma_0$ is the mean value of $\gamma$. The entangled photons in each arm are produced using type-I PDC in a two-crystal geometry [36]. The purpose of the half-wave plate (HP) is to convert the two-photon state vectors $|H\rangle_s|H\rangle_i$ and $|V\rangle_s|V\rangle_i$, into $|V\rangle_s|H\rangle_i$ and $|H\rangle_s|V\rangle_i$, respectively. Therefore, a typical realization $|\psi_\gamma\rangle$ of the two-qubit state in the ensemble detected at $D_s$ and $D_i$ can be represented as $|\psi_\gamma\rangle = E_{V1}|H\rangle_s|H\rangle_i + E_{H1}|V\rangle_s|V\rangle_i + e^{i\gamma} (E_{V2}|H\rangle_s|V\rangle_i + E_{H2}|V\rangle_s|H\rangle_i)$. The two-qubit density matrix is then $\rho = \langle |\psi_\gamma\rangle \rangle = \langle \psi_\gamma | \psi_\gamma \rangle$.

For calculating the matrix elements of $\rho$, we represent the polarization vector of the pump field before the BS as $(E_H, E_V)$ and thus write $E_{H1}$ and $E_{V1}$ as

$$
\begin{bmatrix}
E_{H1} \\
E_{V1}
\end{bmatrix} = \eta_1 \begin{bmatrix}
\cos \theta_1 & \sin \theta_1 \\
-\sin \theta_1 & \cos \theta_1
\end{bmatrix} \begin{bmatrix}
E_H \\
E_V
\end{bmatrix},
$$

(9)

where $\eta_1 = \sqrt{t}$, and the two matrices represent the transformations by PR1 and RP1. $E_{H2}$ and $E_{V2}$ are calculated in a similar manner, with the corresponding quantity $\eta_2 = \sqrt{1-t} e^{i\gamma}$. Without the loss of generality, we assume $(E_{H1}, E_{V1}) = (E_H, E_V) = 1/2$ and $(E_{H2}, E_{V2}) = P/2$, and calculate the matrix elements to be

$$
\begin{align*}
\langle E_{V1} E_{V1} \rangle &= |\eta_1|^2 (1 - P \cos \alpha_1 \sin 2 \theta_1)/2, \\
\langle E_{H1} E_{H1} \rangle &= |\eta_1|^2 (1 + P \cos \alpha_1 \sin 2 \theta_1)/2, \\
\langle E_{V1} E_{H1} \rangle &= |\eta_1|^2 P (\cos \alpha_1 \cos 2 \theta_1 + i \sin \alpha_1)/2, \\
\langle E_{V1} E_{V2} \rangle &= \mu |\eta_1|^2 (\sin \theta_1 \sin \theta_2 + \cos \theta_1 \cos \theta_2 e^{i(\alpha_1 - \alpha_2)}) - P \cos \theta_1 \sin \theta_2 e^{i(\alpha_1 - \alpha_2)} - P \sin \theta_1 \cos \theta_2 e^{-i(\alpha_1 - \alpha_2)} e^{-i\gamma_0}/2, \\
\langle E_{H1} E_{H2} \rangle &= \mu |\eta_1|^2 (\sin \theta_1 \cos \theta_2 + \cos \theta_1 \sin \theta_2 e^{i(\alpha_1 - \alpha_2)}) + P \sin \theta_1 \cos \theta_2 e^{i(\alpha_1 - \alpha_2)} - P \sin \theta_1 \sin \theta_2 e^{-i(\alpha_1 - \alpha_2)} e^{-i\gamma_0}/2, \\
\langle E_{V1} E_{H2} \rangle &= \mu |\eta_1|^2 (\sin \theta_1 \cos \theta_2 + \cos \theta_1 \sin \theta_2 e^{i(\alpha_1 - \alpha_2)}) - P \sin \theta_1 \sin \theta_2 e^{-i(\alpha_1 - \alpha_2)} - P \cos \theta_1 \cos \theta_2 e^{i(\alpha_1 - \alpha_2)} e^{i\gamma_0}/2, \\
\langle E_{H1} E_{H2} \rangle &= \mu |\eta_1|^2 (\cos \theta_1 \cos \theta_2 + \sin \theta_1 \sin \theta_2 e^{-i(\alpha_1 - \alpha_2)}) + P \sin \theta_1 \cos \theta_2 e^{-i(\alpha_1 - \alpha_2)} + P \cos \theta_1 \sin \theta_2 e^{i(\alpha_1 - \alpha_2)} e^{i\gamma_0}/2.
\end{align*}
$$

Here, $t$, $\alpha_1$, $\alpha_2$, $\theta_1$, $\theta_2$, $\mu$, and $\gamma_0$ are the tunable parameters. We numerically vary these parameters with a uniform random sampling and simulate a large number of two-qubit states. Fig.(b) and Fig.(c) are the scatter plots of concurrences of 5 x 10^5 and 5 x 10^6 two-qubit states, respectively, numerically generated by varying all the tunable parameters. (d) and (e) are the scatter plots of concurrence of 2D states, numerically generated by keeping $t = 1$ and varying all the remaining tunable parameters. The solid black lines are the general upper bound $C(\rho) = (1+P)/2$ and the dashed black lines are the upper bound $C(\rho) = P$ for 2D states. The unfilled gaps in the scatter plots can be filled in either by sampling more data points or by adopting a different sampling strategy. To this end, we note that one possible setting for which the general upper bound can be achieved is: $t = 0.5, \theta_1 = -\pi/4, \theta_2 = 0, \alpha_1 = \pi/2, \alpha_2 = \pi, \mu = 1$ and $\gamma_0 = 0$.

In conclusion, we have investigated how one-particle correlations transfer to manifest as two-particle correlations in the physical context of PDC. We have shown that...
if the generation process is trace-preserving and entropy-nondecreasing, the concurrence $C(\rho)$ of the generated two-qubit state $\rho$ follows an intrinsic upper bound with $C(\rho) \leq (1 + P)/2$, where $P$ is the degree of polarization of the pump photon. For the special class of two-qubit states $\rho^{(2D)}$ that is restricted to have only two nonzero diagonal elements, the upper bound on concurrence is the degree of polarization itself, that is, $C(\rho^{(2D)}) \leq P$. The surplus of $(1 + P)/2 - P = (1 - P)/2$ in the maximum achievable concurrence for arbitrary two-qubit states can be attributed to the availability of the entire $4 \times 4$ computational space, as opposed to $2$D states which only have a $2 \times 2$ computational block available to them. We believe these results can have two important implications. The first one is to understand from a fundamental perspective, whether or not correlations too follow a quantifiable conservation principle just as physical observables such as energy, momentum do. The second one is that this formalism provides a systematic method of deducing an upper bound on the correlations in a generated quantum system, purely from the knowledge of the correlations of the source. In the light of the recent experiment on generation of three-photon entangled states from a single source photon \cite{40}, this formalism may prove useful in determining upper bounds on the entanglement of such multipartite systems, for which no well-accepted measure exists. This alternative approach based on intrinsic source correlations could complement the existing information-theoretic approaches \cite{23,24} towards quantifying entanglement.

GK acknowledges helpful discussions on the Physics StackExchange online forum. AKJ acknowledges financial support through an initiation grant from IIT Kanpur.

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