Supplementary Information

Quantum correlations with no causal order

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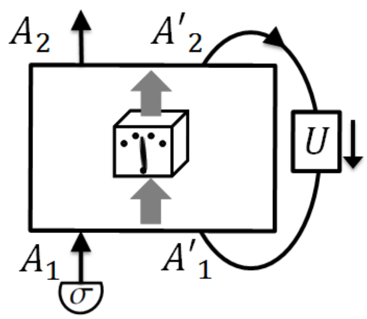
SUPPLEMENTARY FIGURES



Supplmentary Figure S1: **Nonlinear model of closed time-like curve.** In the model of closed time-like curves considered in Refs. [34, 36], a chronology-respecting system A, initially in a state σ, interacts with a second system, A', which travels back in time according to a unitary U. This model can be represented in our formalism by an “unphysical” process matrix, i.e. one for which probabilities do not sum up to one.

SUPPLEMENTARY METHODS

Formal derivation of the causal inequality

A causal structure (for instance, space-time) is a set of event locations equipped with a partial ordering relation $\leq$ that defines the possible causal relations between events at these locations. If A and B are two such locations, $A \leq B$ reads "A is in the *causal past* of B", or equivalently, "B is in the *causal future* of A" (e.g. if A and B are space-time points, $A \leq B$ corresponds to A being in the past light cone of B). Operationally, if $A \leq B$, an agent at A can *signal* to an agent at B by encoding information in events at A that get correlated with events at B which the other agent can observe. (Formally, signalling from A to B is the existence of statistical correlations between a random variable at A which can be chosen freely, and another random variable at B. By definition, a freely chosen variable is one that can be correlated only with variables in its causal future. Note that a freely chosen variable is an idealization since the result of a coin toss or any other candidate for a freely chosen variable may be correlated with initial conditions in the past or with space-like separated events, but these correlations are ignored as not relevant to the variables of interest.) The fact that the relation $\leq$ is a partial order means that it satisfies the following conditions: 1) $A \leq A$ (reflexivity); 2) if $A \leq B$ and $B \leq C$, then $A \leq C$ (transitivity); and 3) if $A \leq B$ and $B \leq A$, then $A = B$ (antisymmetry). The last condition says that if A and B are two different locations, there can either be signalling from A to B, or vice versa, but no signalling in both directions is possible (i.e. there are no causal loops). If A is *not* in the causal past of B, we will write $A \npreceq B$. Note that in a causal structure both $A \npreceq B$ and $B \npreceq A$ may hold (as in the case when A and B are space-like separated), and at least one of the two *must* hold for $A \neq B$. We will denote the situation where both $A \npreceq B$ and $B \npreceq A$ hold by $A \npreceq\!\nsucceq B$.

Since every event specifies an event location, we will use the same notation directly for events. For instance, if X and Y are two events such that the location of X is in the causal past of the location of Y, we will write $X \leq Y$ (similarly for $\npreceq$ and $\npreceq\!\nsucceq$).

The main events in our communication task are the systems entering Alice's and Bob's laboratories, which we will denote by A_1 and B_1, respectively, and the parties producing the bits a, b, b', x, and y, which we will denote by the same letters as the corresponding bits. The fact that Alice generates the bit a and produces her guess x *after* the system enters her laboratory means that $A_1 \leq a, y$. Similarly, we have $B_1 \leq b', b, y$.

The assumptions behind the causal inequality are:

Causal structure (*CS*)—The events A_1, B_1, a, b, b', x, y are localized in a causal structure.

Free choice (*FC*)—Each of the bits a, b, and b' can only be correlated with events in its causal future (this concerns only events relevant to the task). We assume also that each of them takes values 0 or 1 with probability $1/2$.

Closed laboratories (*CL*)—x can be correlated with b only if $b \leq A_1$, and y can be correlated with a only if $a \leq B_1$.

We want to show that these assumptions imply

$$p_{succ} = \frac{1}{2}p(x = b|b' = 0) + \frac{1}{2}p(y = a|b' = 1) \leq \frac{3}{4} \tag{S1}$$

for the success probability that Alice and Bob can achieve in their task.

First, notice that assumption *FC* implies that the bits a, b, and b' are independent of each other (*CS* is assumed throughout). Indeed, there are two general ways in which the three bits could be correlated—two of them are correlated with each other while the third one is independent, or each of them is correlated with the other two. In the first case, the free-choice assumption implies that the two correlated bits would have to be in each other's causal pasts, which is impossible. In the second case, each of the bits would have to be in the causal past of the other two, which is again impossible. Hence, the bits are uncorrelated.

Next, consider the following three possibilities that can be realized in a causal structure (*CS* is assumed throughout): $A_1 \leq B_1$, $B_1 \leq A_1$, $A_1 \npreceq\!\nsucceq B_1$. Since these possibilities are mutually exclusive and exhaustive, their probabilities satisfy $p(A_1 \leq B_1) + p(B_1 \leq A_1) + p(A_1 \npreceq\!\nsucceq B_1) = 1$. From assumption *FC* it follows that the bits a, b, and b' are independent of which of these possibilities is realized. To see this, consider for instance b'. Since $B_1 \leq b'$, we have that b' must be independent of whether A_1 takes place in the causal past of B_1 or not, i.e. $p(A_1 \leq B_1|b') = p(A_1 \leq B_1)$. Similarly, b' must be independent of whether A_1 takes place in the larger region which is a complement of the causal future of B_1, which implies $p(B_1 \npreceq A_1|b') = p(B_1 \npreceq A_1)$. But $p(B_1 \npreceq A_1|b') = p(A_1 \leq B_1|b') + p(A_1 \npreceq\!\nsucceq B_1|b') = p(A_1 \leq B_1) + p(A_1 \npreceq\!\nsucceq B_1|b')$, while $p(B_1 \npreceq A_1) = p(A_1 \leq B_1) + p(A_1 \npreceq\!\nsucceq B_1)$, which implies $p(A_1 \npreceq\!\nsucceq B_1|b') = p(A_1 \npreceq\!\nsucceq B_1)$. Finally, since $p(A_1 \leq B_1|b') + p(A_1 \npreceq\!\nsucceq B_1|b') + p(B_1 \leq A_1|b') = p(A_1 \leq B_1) + p(A_1 \npreceq\!\nsucceq B_1) + p(B_1 \leq A_1|b') = 1 = p(A_1 \leq B_1) + p(A_1 \npreceq\!\nsucceq B_1) + p(B_1 \leq A_1)$, we have $p(B_1 \leq A_1|b') = p(B_1 \leq A_1)$. An analogous argument shows that a and b are also independent of the causal relation between A_1 and B_1.

Using the above, the success probability can be written

$$p_{succ} = \frac{1}{2}p(x = b|b' = 0) + \frac{1}{2}p(y = a|b' = 1)$$
$$= \frac{1}{2}p(x = b|b' = 0; A_1 \preceq B_1)p(A_1 \preceq B_1) + \frac{1}{2}p(x = b|b' = 0; B_1 \preceq A_1)p(B_1 \preceq A_1) + \frac{1}{2}p(x = b|b' = 0; A_1 \npreceq\nsucceq B_1)p(A_1 \npreceq\nsucceq B_1)$$
$$+\frac{1}{2}p(y = a|b' = 1; A_1 \preceq B_1)p(A_1 \preceq B_1) + \frac{1}{2}p(y = a|b' = 1; B_1 \preceq A_1)p(B_1 \preceq A_1) + \frac{1}{2}p(y = a|b' = 1; A_1 \npreceq\nsucceq B_1)p(A_1 \npreceq\nsucceq B_1)$$
$$= \left(\frac{1}{2}p(x = b|b' = 0; A_1 \preceq B_1) + \frac{1}{2}p(y = a|b' = 1; A_1 \preceq B_1)\right)p(A_1 \preceq B_1)$$
$$+\left(\frac{1}{2}p(x = b|b' = 0; B_1 \preceq A_1) + \frac{1}{2}p(y = a|b' = 1; B_1 \preceq A_1)\right)p(B_1 \preceq A_1)$$
$$+\left(\frac{1}{2}p(x = b|b' = 0; A_1 \npreceq\nsucceq B_1) + \frac{1}{2}p(y = a|b' = 1; A_1 \npreceq\nsucceq B_1)\right)p(A_1 \npreceq\nsucceq B_1). \tag{S2}$$

If $A_1 \preceq B_1$ (which implies $B_1 \npreceq A_1$), from the transitivity of partial order it follows that $A_1 \preceq b$ (and thus $b \npreceq A_1$). From assumption *CL*, x can only be correlated with b if b is in the causal past of A_1, thus $p(b|x; A_1 \preceq B_1) = p(b|A_1 \preceq B_1) = \frac{1}{2}$ [the last equality follows from the independence of b from the causal relations between A_1 and B_1, together with assumption *FC*]. Using also that b and b' are independent, we thus obtain $p(x = b|b' = 0; A_1 \preceq B_1) = p(b = 0; x = 0|b' = 0; A_1 \preceq B_1) + p(b = 1, x = 1|b' = 0; A_1 \preceq B_1) = p(b = 0|x = 0; b' = 0; A_1 \preceq B_1)p(x = 0|b' = 0; A_1 \preceq B_1) + p(b = 1|x = 1; b' = 0; A_1 \preceq B_1)p(x = 1|b' = 0; A_1 \preceq B_1) = \frac{1}{2}p(x = 0|b' = 0; A_1 \preceq B_1) + \frac{1}{2}p(x = 1|b' = 0; A_1 \preceq B_1) = \frac{1}{2}$.

If $B_1 \preceq A_1$ (which implies $A_1 \npreceq B_1$), by an analogous argument we obtain $p(y = a|b' = 1; B_1 \preceq A_1) = \frac{1}{2}$. Finally, if $A_1 \npreceq\nsucceq B_1$, we have both $p(y = a|b' = 1; A_1 \npreceq\nsucceq B_1) = \frac{1}{2}$ and $p(x = b|b' = 0; A_1 \npreceq\nsucceq B_1) = \frac{1}{2}$. Substituting this in Eq. (S2), we obtain

$$p_{succ} = \left(\frac{1}{4} + \frac{1}{2}p(y = a|b' = 1; A_1 \preceq B_1)\right)p(A_1 \preceq B_1) + \left(\frac{1}{2}p(x = b|b' = 0; B_1 \preceq A_1) + \frac{1}{4}\right)p(B_1 \preceq A_1) + \left(\frac{1}{4} + \frac{1}{4}\right)p(A_1 \npreceq\nsucceq B_1)$$
$$\leq \frac{3}{4}p(A_1 \preceq B_1) + \frac{3}{4}p(B_1 \preceq A_1) + \frac{3}{4}p(A_1 \npreceq\nsucceq B_1) = \frac{3}{4}. \tag{S3}$$

This completes the proof.

Characterization of process matrices

Here we derive necessary and sufficient conditions for a matrix $W^{A_1A_2B_1B_2}$ to satisfy

$$W^{A_1A_2B_1B_2} \geq 0 \tag{S4}$$

[*non-negative probabilities*]

and

$$\mathrm{Tr}\left[W^{A_1A_2B_1B_2}\left(M^{A_1A_2} \otimes M^{B_1B_2}\right)\right] = 1, \quad \forall M^{A_1A_2}, M^{B_1B_2} \geq 0, \; \mathrm{Tr}_2 M^{A_1A_2} = \mathbb{1}^{A_1}, \mathrm{Tr}_2 M^{B_1B_2} = \mathbb{1}^{B_1} \tag{S5}$$

[*probabilities sum up to 1*]

in terms of an expansion of the matrix in a Hilbert-Schmidt basis. A Hilbert-Schmidt basis of $\mathcal{L}(\mathcal{H}^X)$ is given by a set of matrices $\{\sigma_\mu^X\}_{\mu=0}^{d_X^2-1}$, with $\sigma_0^X = \mathbb{1}_X$, $\mathrm{Tr}\sigma_\mu^X\sigma_\nu^X = d_X\delta_{\mu\nu}$, and $\mathrm{Tr}\sigma_j^X = 0$ for $j = 1, \dots d_X^2 - 1$. A general element of $\mathcal{L}(\mathcal{H}^{A_1} \otimes \mathcal{H}^{A_2} \otimes \mathcal{H}^{B_1} \otimes \mathcal{H}^{B_2})$ can be expressed as

$$W^{A_1A_2B_1B_2} = \sum_{\mu\nu\lambda\gamma} w_{\mu\nu\lambda\gamma}\sigma_\mu^{A_1}\sigma_\nu^{A_2}\sigma_\lambda^{B_1}\sigma_\gamma^{B_2}, \quad w_{\mu\nu\lambda\gamma} \in \mathbb{C} \tag{S6}$$

(we omit tensor products and identity matrices whenever there is no risk of confusion). Since a process matrix has to be Hermitian, we consider only the cases

$$w_{\mu\nu\lambda\gamma} \in \mathbb{R}. \tag{S7}$$

We will refer to terms of the form $\sigma_i^{A_1}\otimes \mathbb{1}^{rest}$ $(i\geq 1)$ as of the type A_1, terms such as $\sigma_i^{A_1}\otimes\sigma_j^{A_2}\otimes \mathbb{1}^{rest}$ $(i,\, j\geq 1)$ as of the type A_1A_2, and so on. The properties of a process matrix can be analysed with respect to the terms it contains. For example, terms of the type A_1B_1 produce non-signalling correlations between the measurements, terms such as A_2B_1 correlate Alice's outputs with Bob's inputs, yielding signalling from Alice to Bob, etc., as illustrated in Fig. 3. Note that not all terms are compatible with the condition (S5). We will prove that a matrix W satisfies condition (S5) if and only if it only contains the terms listed in Fig. 3.

The CJ matrix of a local operation can be similarly written $M^{X_1X_2}=\sum_{\mu\nu} r_{\mu\nu}\sigma_\mu^{X_1}\sigma_\nu^{X_2}$, $r_{\mu\nu}\in\mathbb{R}$. The condition $\mathrm{Tr}_{X_2} M^{X_1X_2}=\mathbb{1}^{X_1}$ is equivalent to the requirement $r_{00}=\frac{1}{d_{X_2}}$, $r_{i0}=0$ for $i>0$. Thus CJ matrices corresponding to CPTP maps have the form

$$M^{X_1X_2}=\frac{1}{d_{X_2}}\left(\mathbb{1}+\sum_{i>0}a_i\sigma_i^{X_2}+\sum_{ij>0}t_{ij}\sigma_i^{X_1}\sigma_j^{X_2}\right), \qquad (S8)$$
$$a_i, t_{ij}\in\mathbb{R}.$$

Let us consider first the case of a single party, say, Alice. Since the set of matrices $M^{A_1A_2}\geq 0$ is a substantial set, condition (S5) can be equivalently imposed on arbitrary matrices of the form (S8) and, for a single party, it can be rewritten as

$$\frac{1}{d_{A_2}}\mathrm{Tr}\left[W^{A_1A_2}\left(\mathbb{1}+\sum_{i>0}a_i\sigma_i^{A_2}+\sum_{ij>0}t_{ij}\sigma_i^{A_1}\sigma_j^{A_2}\right)\right]=1,$$
$$\forall\ \ a_i, t_{ij}\in\mathbb{R}.$$

Using an expansion of the process matrix in the same basis in a similar way, $W^{A_1A_2}=\sum_{\mu\nu}w_{\mu\nu}\sigma_\mu^{A_1}\sigma_\nu^{A_2}$, $w_{\mu\nu}\in\mathbb{R}$, the above condition becomes

$$d_{A_1}\left(w_{00}+\sum_{i>0}w_{0i}a_i+\sum_{ij>0}w_{ij}t_{ij}\right)=1,$$
$$\forall\ \ a_i, t_{ij}\in\mathbb{R},$$

and one obtains $w_{00}=\frac{1}{d_{A_1}}$, $w_{0i}=w_{ij}=0$ for $i,\, j>0$. Thus the most general process matrix observed by a single party has the form

$$W^{A_1A_2}=\frac{1}{d_{A_1}}\left(\mathbb{1}+\sum_{i>0}v_i\sigma_i^{A_1}\right), \qquad (S9)$$
$$v_i\in\mathbb{R},\quad W^{A_1A_2}\geq 0,$$

which can be recognized as a state. This result—that all probabilities a single agent can observe are described by quantum states—is an extension of Gleason's theorem from POVMs [52, 53] to CP maps (note that here the linear structure of quantum operations is assumed, while in Gleason's theorem for POVMs it is derived from different hypotheses. However, by a similar argument one could derive linearity for CP maps too).

Let us now consider a bipartite process matrix, $W^{A_1A_2B_1B_2}=\sum_{\mu\nu\lambda\gamma}w_{\mu\nu\lambda\gamma}\sigma_\mu^{A_1}\sigma_\nu^{A_2}\sigma_\lambda^{B_1}\sigma_\gamma^{B_2}$, $w_{\mu\nu\lambda\gamma}\in\mathbb{R}$. We have to impose (S5) for arbitrary matrices $M^{A_1A_2}$, $M^{B_1B_2}$ of the form (S8). First, if we fix $M^{B_1B_2}=\frac{\mathbb{1}^{B_1B_2}}{d_{B_2}}$, we obtain

$$d_{A_1}d_{B_1}\left(w_{0000}+\sum_{i>0}w_{0i00}a_i+\sum_{ij>0}w_{ij00}t_{ij}\right)=1$$
$$\forall\ \ a_i, t_{ij}\in\mathbb{R},$$

which imposes $w_{0000}=\frac{1}{d_{A_1}d_{B_1}}$ and $w_{0i00}=w_{ij00}=0$ for $i,\, j>0$. Similarly, by fixing $M^{A_1A_2}=\frac{\mathbb{1}^{A_1A_2}}{d_{A_2}}$, we can derive $w_{000i}=w_{00ij}=0$ for $i,\, j>0$. Finally, imposing (S5) for arbitrary

$$M^{A_1A_2}=\frac{1}{d_{A_2}}\left(\mathbb{1}+\sum_{i>0}a_i\sigma_i^{A_2}+\sum_{ij>0}t_{ij}\sigma_i^{A_1}\sigma_j^{A_2}\right),$$
$$M^{B_1B_2}=\frac{1}{d_{B_2}}\left(\mathbb{1}+\sum_{k>0}b_k\sigma_k^{B_2}+\sum_{kl>0}s_{kl}\sigma_k^{B_1}\sigma_l^{B_2}\right),$$

we obtain

$$\sum_{ik>0} w_{0i0k} a_i b_k + \sum_{ikl>0} w_{0ikl} a_i s_{kl} + \sum_{ijk>0} w_{ij0k} t_{ij} b_k + \sum_{ijkl>0} w_{ijkl} t_{ij} s_{kl} = 0, \quad \forall\ a_i, t_{ij}, b_k, s_{kl} \in \mathbb{R},$$

from which we conclude that the most general matrix that satisfies (S5) has the form

$$\begin{aligned} W^{A_1A_2B_1B_2} &= \frac{1}{d_{A_1}d_{B_1}}\left(\mathbb{1} + \sigma^{B\preceq A} + \sigma^{A\preceq B} + \sigma^{A\npreceq\nsucceq B}\right), \\ \sigma^{B\preceq A} &:= \sum_{ij>0} c_{ij}\sigma_i^{A_1}\sigma_j^{B_2} + \sum_{ijk>0} d_{ijk}\sigma_i^{A_1}\sigma_j^{B_1}\sigma_k^{B_2}, \\ \sigma^{A\preceq B} &:= \sum_{ij>0} e_{ij}\sigma_i^{A_2}\sigma_j^{B_1} + \sum_{ijk>0} f_{ijk}\sigma_i^{A_1}\sigma_j^{A_2}\sigma_k^{B_1}, \\ \sigma^{A\npreceq\nsucceq B} &:= \sum_{i>0} v_i\sigma_i^{A_1} + \sum_{i>0} x_i\sigma_i^{B_1} + \sum_{ij>0} g_{ij}\sigma_i^{A_1}\sigma_j^{B_1}, \\ &\text{where } c_{ij}, d_{ijk}, e_{ij}, f_{ijk}, g_{ij}, v_i, x_i \in \mathbb{R}. \end{aligned}$$

This form, together with the condition $W^{A_1A_2B_1B_2} \geq 0$, completely characterizes the most general bipartite process matrix.

Terms not appearing in process matrices

The not-allowed terms are listed in Fig. 4, along with possible interpretations. Particularly interesting are the cases involving terms of the type A_1A_2. These would correlate Alice's output with her input and not give unit probabilities for some CPTP maps that she can choose to perform. This kind of correlations resemble a "backward in time" transmission of information: one can imagine that they can be generated by a quantum channel "in the inverse order", from the output A_2 to the input A_1. It is worth noting that a recently proposed model of closed time-like curves [34, 36] can be expressed precisely in this way. Using our terminology, such a model considers an agent receiving two quantum systems in her laboratory: a chronology-respecting system A and a second system A' which, after leaving the laboratory, is sent back in time to the laboratory's entrance (see Supplementary Fig. **S1**). This can be described by the process matrix $W^{A_1A_1'A_2A_2'} = \sigma^{A_1} \otimes \mathbb{1}^{A_2} \otimes \left(U \otimes \mathbb{1}|\phi^+\rangle\langle\phi^+|^{A_1'A_2'}U^\dagger \otimes \mathbb{1}\right)$, where σ^{A_1} is the state of the chronology-respecting system when it enters the laboratory and $\left(U \otimes \mathbb{1}|\phi^+\rangle\langle\phi^+|^{A_1'A_2'}U^\dagger \otimes \mathbb{1}\right)$ is a process matrix corresponding to a unitary U from A_2' to A_1', describing the evolution back in time of the chronology-violating system. (The labels A_1, A_1' represent the two systems entering the laboratory, while A_2, A_2' represent the systems going out. Note that here the two systems belong to the same laboratory and they can undergo any joint operation.) In this model, probabilities have to be renormalized in order to sum up to one, which introduces a non-linearity that violates our original assumptions (in particular, as opposed to quantum mechanics, probabilities are contextual in this model, since it is necessary to specify the events that did not occur in order to perform the renormalization step). The same can be said for Deutsch's model of closed time-like curves [33], which is also non-linear (although it uses a different mechanism to obtain well-defined probabilities) and thus violates our premise that ordinary quantum mechanics holds locally in each laboratory.

Casual order in the classical limit

Let us now show that in the classical limit all correlations are causally ordered. Classical operations can be described by transition matrices $M_j^{(ki)} = P(k, j|i)$, where $P(k, j|i)$ is the conditional probability that the measurement outcome j is observed and the classical output state k is prepared given that the input state is i. They can be expressed in the quantum formalism as CP maps diagonal in a fixed ("pointer") basis, and the corresponding CJ matrices are $M_j = \sum_{ki} M_j^{(ki)}|i\rangle\langle i|^{A_1} \otimes |k\rangle\langle k|^{A_2}$. In order to express arbitrary bipartite probabilities of classical operations, it is sufficient to consider process matrices of the standard form

$$W^{A_1A_2B_1B_2} = \frac{1}{d_{A_1}d_{B_1}}\left(\mathbb{1} + \sigma^{B\npreceq A} + \sigma^{A\npreceq B}\right), \tag{S10}$$

where $\sigma^{B\not\leq A}$ and $\sigma^{A\not\leq B}$ are diagonal in the pointer basis. Probabilities are still given by

$$P\left(\mathcal{M}_i^A, \mathcal{M}_j^B\right) = \mathrm{Tr}\left[W^{A_1A_2B_1B_2}\left(M_i^{A_1A_2} \otimes M_j^{B_1B_2}\right)\right]. \tag{S11}$$

We will show that any such diagonal process matrix can be written in the form

$$W^{A_1A_2B_1B_2} = \frac{1}{d_{A_1}d_{B_1}}\left(\rho^{A_1A_2B_1} + \rho^{A_1B_1B_2}\right), \tag{S12}$$

where $\rho^{A_1A_2B_1}$ and $\rho^{A_1B_1B_2}$ are positive semidefinite matrices. This is sufficient to conclude that $W^{A_1A_2B_1B_2}$ is causally separable. Indeed, if $W^{A_1A_2B_1B_2}$ could be written in the form (S12), we know that $\rho^{A_1A_2B_1}$ would not contain Hilbert-Schmidt terms of the types A_1A_2 or A_2 (which are not allowed in a process matrix), since by assumption these terms are not part of $W^{A_1A_2B_1B_2}$. Therefore, the matrix

$$W^{B\not\leq A} \equiv \frac{\rho^{A_1A_2B_1}}{\mathrm{Tr}\rho^{A_1A_2B_1}} d_{A_2}d_{B_2}, \tag{S13}$$

which is positive semidefinite, has trace $d_{A_2}d_{B_2}$, and contains only terms of the allowed types, would be a valid process matrix with no signalling from B to A. Similarly,

$$W^{A\not\leq B} \equiv \frac{\rho^{A_1B_1B_2}}{\mathrm{Tr}\rho^{A_1B_1B_2}} d_{A_2}d_{B_2} \tag{S14}$$

would be a valid process matrix with no signalling from A to B. The whole process matrix could then be written in the causally separable form

$$W^{A_1A_2B_1B_2} = qW^{B\not\leq A} + (1-q)W^{A\not\leq B}, \tag{S15}$$

where

$$q \equiv \frac{\mathrm{Tr}\rho^{A_1A_2B_1}}{d_{A_1}d_{A_2}d_{B_1}d_{B_2}}. \tag{S16}$$

Note that $0 \leq q \leq 1$ since $\rho^{A_1A_2B_1}$ and $\rho^{A_1B_1B_2}$ in Eq. (S12) are positive semidefinite and $\mathrm{Tr}W^{A_1A_2B_1B_2} = d_{A_2}d_{B_2}$.

To prove Eq. (S12), we will construct $\rho^{A_1A_2B_1}$ and $\rho^{A_1B_1B_2}$ from the general form in Eq. (S10). Let the minimum eigenvalue of $\sigma^{B\not\leq A} + \sigma^{A\not\leq B}$ be m. Since $W^{A_1A_2B_1B_2}$ is positive semidefinite and $\sigma^{B\not\leq A} + \sigma^{A\not\leq B}$ is traceless, we have $m \in [-1, 0]$. Define the matrices

$$\kappa^{A_1A_2B_1} = -m\mathbb{1} + \sigma^{B\not\leq A}, \tag{S17}$$

$$\kappa^{A_1B_1B_2} = \sigma^{A\not\leq B}. \tag{S18}$$

The full process matrix can then be written

$$W^{A_1A_2B_1B_2} = \frac{1}{d_{A_1}d_{B_1}}\left((1+m)\mathbb{1} + \kappa^{A_1A_2B_1} + \kappa^{A_1B_1B_2}\right), \tag{S19}$$

where $\kappa^{A_1A_2B_1} + \kappa^{A_1B_1B_2}$ is positive semidefinite.

We are now going to modify $\kappa^{A_1A_2B_1}$ and $\kappa^{A_1B_1B_2}$ by adding matrices of the form $\kappa^{A_1B_1}$ to $\kappa^{A_1A_2B_1}$ and subtracting them from $\kappa^{A_1B_1B_2}$ (therefore leaving $\kappa^{A_1A_2B_1} + \kappa^{A_1B_1B_2}$ unchanged), until we transform both $\kappa^{A_1A_2B_1}$ and $\kappa^{A_1B_1B_2}$ in Eq. (S19) into positive semidefinite matrices.

Denote the pointer basis of system X by $|i\rangle^X$, $i = 1, ..., d_X$, $X = A_1, A_2, B_1, B_2$. All matrices we consider are diagonal in the basis $\{|i\rangle^{A_1}|j\rangle^{A_2}|k\rangle^{B_1}|l\rangle^{B_2}\}$. Let $m_1(i, j, k, l)$ denote the eigenvalues of $\kappa^{A_1A_2B_1}$ corresponding to the eigenvectors $|i\rangle^{A_1}|j\rangle^{A_2}|k\rangle^{B_1}|l\rangle^{B_2}$, and let $m_2(i, j, k, l)$ be the eigenvalues of $\kappa^{A_1B_1B_2}$ corresponding to the same vectors. For every i and k, we do the following. Define

$$\tilde{m}_1(i, k) = \min_{j,l} m_1(i, j, k, l), \tag{S20}$$

$$\tilde{m}_2(i, k) = \min_{j,l} m_2(i, j, k, l). \tag{S21}$$

Note that $m_1(i, j, k, l)$ do not depend on l since $\kappa^{A_1A_2B_1}$ acts trivially on B_2, and similarly $m_2(i, j, k, l)$ do not depend on j. This means that for given i and k, the minimum of the eigenvalues of $\kappa^{A_1A_2B_1}+\kappa^{A_1B_1B_2}$ for all eigenvectors of the type $|i\rangle^{A_1}|j\rangle^{A_2}|k\rangle^{B_1}|l\rangle^{B_2}$ is equal to $\tilde{m}_1(i, k) + \tilde{m}_2(i, k)$. But by construction $\kappa^{A_1A_2B_1} + \kappa^{A_1B_1B_2}$ is positive semidefinite, so we have

$$\tilde{m}_1(i, k) + \tilde{m}_2(i, k) \geq 0. \tag{S22}$$

Now, if both $\tilde{m}_1(i, k)$ and $\tilde{m}_2(i, k)\}$ are non-negative, we will not modify $\kappa^{A_1A_2B_1}$ and $\kappa^{A_1B_1B_2}$. However, if one of these numbers is negative, say $\tilde{m}_1(i, k) < 0$ (both cannot be negative due to (S22)), we will add the term $-\tilde{m}_1(i, k)|i\rangle\langle i|^{A_1} \otimes \mathbb{1}^{A_2} \otimes |k\rangle\langle k|^{B_1} \otimes \mathbb{1}^{B_2}$ to $\kappa^{A_1A_2B_1}$ and subtract the same term from $\kappa^{A_1B_1B_2}$. After this step, the modified $\kappa^{A_1A_2B_1}$ is such that the eigenvalues $m_1(i, j, k, l)$ have been changed to $m_1(i, j, k, l) - \tilde{m}_1(i, k) \geq \tilde{m}_1(i, k) - \tilde{m}_1(i, k) = 0$, i.e. $\kappa^{A_1A_2B_1}$ does not have any more negative eigenvalues $m_1(i, j, k, l)$ for the given i and k. The same holds for $\kappa^{A_1B_1B_2}$ since the eigenvalues $m_2(i, j, k, l)$ change to $m_2(i, j, k, l) + \tilde{m}_1(i, k) \geq \tilde{m}_2(i, k) + \tilde{m}_1(i, k) \geq 0$. In other words, the eigenvalues of the modified $\kappa^{A_1A_2B_1}$ and $\kappa^{A_1B_1B_2}$ satisfy

$$m_1(i, j, k, l),\ m_2(i, j, k, l) \geq 0, \quad \forall j, l. \tag{S23}$$

By performing this procedure for all i and k, we eventually transform $\kappa^{A_1A_2B_1}$ and $\kappa^{A_1B_1B_2}$ into matrices all of whose eigenvalues are non-negative. Denote the resultant positive semidefinite matrices by $\tilde{\kappa}^{A_1A_2B_1}$ and $\tilde{\kappa}^{A_1B_1B_2}$. We can now add the term $(1 + m)\mathbb{1}$ in Eq. (S19) for instance to $\tilde{\kappa}^{A_1A_2B_1}$ (recall that $m \in [-1, 0]$), defining the positive semidefinite matrices

$$\rho^{A_1A_2B_1} \equiv (1 + m)\mathbb{1} + \tilde{\kappa}^{A_1A_2B_1}, \tag{S24}$$

$$\rho^{A_1B_1B_2} \equiv \tilde{\kappa}^{A_1B_1B_2}. \tag{S25}$$

We thus arrive at the desired form (S12) which implies (S15) as argued above.

SUPPLEMENTARY REFERENCES

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