Resource Theory of Superposition

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The superposition principle lies at the heart of many nonclassical properties of quantum mechanics. Motivated by this, we introduce a rigorous resource theory framework for the quantification of superposition of a finite number of linear independent states. This theory is a generalization of resource theories of coherence. We determine the general structure of operations which do not create superposition, find a fundamental connection to unambiguous state discrimination, and propose several quantitative superposition measures. Using this theory, we show that trace decreasing operations can be completed for free which, when specialized to the theory of coherence, resolves an outstanding open question and is used to address the free probabilistic transformation between pure states. Finally, we prove that linearly independent superposition is a necessary and sufficient condition for the faithful creation of entanglement in discrete settings, establishing a strong structural connection between our theory of superposition and entanglement theory.

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Introduction.—During the last decades, there has been an increasing interest in quantum technologies. The main reason for this is the operational advantages of protocols or devices working in the quantum regime over those relying on classical physics. Early examples include entanglement-based quantum cryptography [1], quantum dense coding [2], and quantum teleportation [3], where entanglement is a resource which is consumed and manipulated. Therefore the detection, manipulation, and quantification of entanglement was investigated, leading to the resource theory of entanglement [4]. Typical quantum resource theories (QRTs) are built by imposing an additional restriction to the laws of quantum mechanics [5–7]. In the case of entanglement theory, this is the restriction to local operations and classical communication (LOCC). From such a restriction, the two main ingredients of QRTs emerge: The free operations and the free states (which are LOCC and separable states in the case of entanglement theory). All states which are not free contain the resource under investigation and are considered costly. Therefore, free operations must transform free states to free states, allowing for the resource to be manipulated but not freely created. Once these main ingredients are defined, a resource theory investigates the manipulation, detection, quantification, and usage of the resource.

In principle, not only entanglement but every property of quantum mechanics not present in classical physics could lead to an operational advantage [8,9]. This motivates the considerable interest in the rigorous quantification of nonclassicality [10–15]. The superposition principle underlies many nonclassical properties of quantum mechanics including entanglement or coherence. Recently, resource theories of coherence [11,16,17] and their role in fields as diverse as quantum computation [8,18,19], quantum phase discrimination [20], and quantum thermodynamics [21] attracted considerable attention. In these settings, the free states form a finite orthonormal basis of the system under consideration and the resource is the superposition of these, called coherence. Here we present a generalization of coherence theories and relax the requirement of orthogonality of the free states to linear independence. To be precise, we construct a resource theory in which the pure free states are a finite linearly independent set and their nontrivial superpositions are resource states. Mixed states are free if and only if they can be represented as statistical mixtures of free pure states. Thus our framework contains coherence theory as a special case. For obvious reasons, we call the free states superposition-free and the resource states superposition states.

Such a generalization of coherence theory is interesting for several reasons. Linear independence relaxes the convenient but restrictive requirement of orthogonality, yet still provides a fundamental framework in which the notion of superposition is unambiguous and self-consistent. From a conceptual point of view, our theory helps to clarify the role of orthogonality versus linear independence. We show that many of the results of coherence theory are just special cases of their counterparts in our nonorthogonal setting. This indicates that linearly independent superposition, rather than the stronger requirement of orthogonality, is a major underlying factor in such quantum resource theories. In addition, superposition states can be faithfully converted into entanglement, which implies a fundamental connection between entanglement and single-system nonclassicality [12,13]. Thus, our resource theory can give new insights into the resource theory of entanglement and, vice versa, the faithful mapping between these theories allows for an investigation of the controversial notion of nonclassicality based on the well-founded principles of entanglement [22]. As an application, the theory presented here can quantify the nonclassicality in the superposition of a finite number of optical coherent states. This is not possible using the framework of coherence theory, since the optical coherent states are not orthogonal. Our theory can thus be seen as a starting point for more general resource theories with less restrictive, yet still physically meaningful constraints on the free states. Mastering these further generalizations will allow us to quantify optical and other forms of nonclassicality rigorously and to unify their description with entanglement theory (see also Refs. [6,14,23]).

This Letter is structured as follows. In the next section, we define our free states and operations formally. To validate the choice of linear independent free states, we prove that linear independence is a necessary and sufficient ingredient for the faithful creation of entanglement, completing earlier results from Ref. [12]. Then we characterize the free operations using the concept of reciprocal states known from unambiguous state discrimination [24,25]. This leads to a proof that any trace-decreasing operation can be completed for free to a trace-preserving operation in the theory of superposition and hence in the special case of coherence theory. We proceed to address the quantification of superposition and propose several measures. For free transformations between pure states we show that generically the maximal probability of success is the solution of a semidefinite program. Finally, we investigate states with maximal superposition and the operational advantages they allow for, before concluding with a discussion on future research directions. Proofs and some additional results are given in the Supplemental Material [26], including a game in which access to superposition turns certain loss into certain win.

Basic framework.—In this section, we give the formal definition of the free states and operations that we consider. *Definition 1.*—Let $\{|c_i\rangle\}_{i=1}^d$ be a normalized, linear independent and not necessarily orthogonal basis of the Hilbert space represented by $\mathbb{C}^d$, $d \in \mathbb{N}$. Those basis states are called pure superposition-free states. All density operators ρ of the form

$$\rho = \sum_{i=1}^{d} \rho_i |c_i\rangle\langle c_i|, \tag{1}$$

where the ρ_i form a probability distribution, are called superposition-free. The set of superposition-free density operators is denoted by $\mathcal{F}$ and forms the set of free states. All density operators which are not superposition-free are called superposition states and form the set of resource states.

For $d = 1$, the concept of superposition is empty; thus, all the following results assume $d \geq 2$. In Refs. [12,15], the *classical rank* of a state has been introduced as the minimum number of free states we need to superpose in order to represent the state. We will say that an isometry Λ is a faithful conversion operation (to and from entanglement) when the Schmidt rank of $\Lambda|\psi\rangle$ is equal to the classical rank of $|\psi\rangle$. The relevance of linear independence for our resource theory is based on the following theorem.

Theorem 2.—If the free states in a finite dimensional Hilbert space form a countable set, then linear independence of the free states is a necessary and sufficient condition for the existence of a faithful conversion operation. In case the free states are a finite set of optical coherent states, the faithful conversion can be implemented by a beam splitter [14].

Sufficiency is proved in an earlier theorem from Ref. [12] (and extended in Ref. [13]). In the Supplemental Material [26], we prove the converse result, thus completing the original theorem. *Definition 3.*—A Kraus operator K_n is called superposition-free if $K_n \rho K_n^\dagger \in \mathcal{F}$ for all $\rho \in \mathcal{F}$. Quantum operations $\Phi(\rho)$ are called superposition-free if they are trace preserving and can be written as

$$\Phi(\rho) = \sum_n K_n \rho K_n^\dagger, \tag{2}$$

where all K_n are free. The set of superposition-free operations forms the free operations and is denoted by $\mathcal{FO}$.

At this point, let us highlight that the definition of the free operations is not unique. This is a common trait of QRTs. The biggest possible class of free operations for our choice of the free states is given by those quantum operations that map the free states onto themselves which are denoted by $\mathcal{MFO}$ (maximally superposition-free operations). However, in general, these operations do not possess a representation in terms of superposition-free Kraus operators.

Proposition 4.—$\mathcal{MFO}$ is strictly larger than $\mathcal{FO}$. This is also valid in the special case of coherence theory.

Hence, someone who has access to measurement outcomes of an element of $\mathcal{MFO}$ and can thus do postselection could conclude that a superposition-free operation generated superposition from a superposition-free state. Our definition of the free operations guarantees that one cannot create resources for free by obtaining measurement results. On the other hand, it is not as restricted as other definitions demanding, for example, a free dilation [34,35]. For a discussion of alternative choices, see the Supplemental Material [26].

Free operations.—In order to describe the general structure of $\mathcal{FO}$, we need to introduce some notation. Since the pure superposition-free states form a basis of $\mathbb{C}^d$, $d \in \mathbb{N}$, there exist vectors $|c_i^\perp\rangle$, $i = 1, \ldots, d$ such that

$$\langle c_i^\perp | c_j\rangle = \delta_{i,j}, \tag{3}$$

which are not normalized but form a basis as well. In the context of unambiguous state discrimination, the states one gets by normalizing $|c_i^\perp\rangle$ are called reciprocal states [24,25]. For explicit calculations, it is convenient to express both $\{|c_i\rangle\}_{i=1}^d$ and $\{|c_i^\perp\rangle\}_{i=1}^d$ with respect to an orthonormal basis $\{|i\rangle\}_{i=1}^d$ which will be called computational. Now we can introduce two linear operators V and W such that $V|i\rangle = |c_i\rangle$ and $W|i\rangle = |c_i^\perp\rangle$. Notice that both V and W are full rank since they correspond to basis transformations. From Eq. (3), it follows that $\delta_{i,j} = \langle c_i^\perp | c_j\rangle = \langle i|W^\dagger V|j\rangle$ and thus $W = (V^\dagger)^{-1}$. With this notation at hand, the explicit form of a superposition-free Kraus operator can be given, which is done in the following theorem.

Theorem 5.—A Kraus operator K_n is superposition-free if and only if it is of the form

$$K_n = \sum_k c_{k,n} |c_{f_n(k)}\rangle\langle c_k^\perp|, \quad (4)$$

where the $c_{k,n} \in \mathbb{C}$ and the $f_n(k)$ are index functions.

Incoherent Kraus operators $\tilde{K}_n$ as defined in the limit of coherence theory [16] are thus given by $\tilde{K}_n = \sum_k c_{k,n}|f_n(k)\rangle\langle k|$ [36,37]. If we choose the incoherent states $\{|k\rangle\}$ as the computational basis, the operator $K_n = V\tilde{K}_n V^{-1}$ has the form of a superposition-free Kraus operator. In order to have a valid trace nonincreasing quantum operation, we need

$$\mathbb{1} \geq \sum_n K_n^\dagger K_n = \sum_n (V^\dagger)^{-1} \tilde{K}_n^\dagger V^\dagger V \tilde{K}_n V^{-1}. \quad (5)$$

If the pure superposition-free states are not orthogonal, $V^\dagger \neq V^{-1}$ and, in general, it is therefore not possible to transform a trace nonincreasing set of incoherent Kraus operators by a basis transformation V into a superposition-free one.

Intuitively, the introduction of additional systems in free states is for free. With the above theorem at hand, we can show that this is indeed the case.

Proposition 6.—If both σ_B and all K_n are free, the quantum operation $\Phi(\rho_A) = \mathbf{tr}_B \sum_n K_n \rho_A \otimes \sigma_B K_n^\dagger$ is free.

When dealing with trace decreasing operations that can be decomposed into superposition-free Kraus operators, the question arises whether they are part of a (trace preserving) superposition-free operation. If this was not the case, it would imply that one cannot call the trace decreasing operation free because one disregards a part that can only be done in a nonfree way [38]. This leads us to our first main result.

Theorem 7.—Assume we have an (incomplete) set of Kraus operators $\{K_m\}$ such that $\sum_m K_m^\dagger K_m \leq \mathbb{1}$. Then there always exist superposition-free Kraus operators $\{F_n\}$ with $\sum_m K_m^\dagger K_m + \sum_n F_n^\dagger F_n = \mathbb{1}$.

From here on we will call trace-decreasing operations with a decomposition into superposition-free Kraus operators superposition free as well, since we can always complete them for free. Note that this is also valid in the special case of coherence theory.

Superposition measures.—In this section, we address the quantification of superposition, extending the method used in Ref. [16] to quantify coherence. *Definition 8.*—A function M mapping all quantum states to the non-negative real numbers is called a superposition measure if it is

($S1$) Faithful: $M(\rho) = 0$ if and only if $\rho \in \mathcal{F}$.

($S2a$) Monotonic under $\mathcal{FO}$: $M(\rho) \geq M[\Phi(\rho)]$ for all $\Phi \in \mathcal{FO}$.

($S2b$) Monotonic under superposition-free selective measurements on average: $M(\rho) \geq \sum_n p_n M(\rho_n)$: $p_n = \mathrm{tr}(K_n \rho K_n^\dagger)$, $\rho_n = (K_n \rho K_n^\dagger)/p_n$ for all $\{K_n\}$: $\sum_n K_n^\dagger K_n = \mathbb{1}$, $K_n \mathcal{F} K_n^\dagger \subset \mathcal{F}$.

($S3$) Convex: $\sum_n p_n M(\sigma_n) \geq M(\sum_n p_n \sigma_n)$ for all $\{\sigma_n\}$, $p_n \geq 0$, $\sum_n p_n = 1$.

If only condition ($S1$) and ($S2a$) or ($S2b$) are satisfied, we call M a superposition monotone.

Property ($S1$) demands that a state has zero superposition if and only if the state is superposition-free. As stated in ($S2a$), the application of a superposition-free operation to a state should not increase its superposition. If one does superposition-free selective measurements, one does not expect the superposition to increase on average which is exactly the point of ($S2b$). The convexity condition ($S3$) enforces that mixing states cannot increase the average superposition. It can be shown easily that ($S2a$) follows from ($S2b$) and ($S3$). As in coherence theory [16], some distance measures $\mathcal{D}$ can be used to define superposition measures and monotones. We define a candidate $M_\mathcal{D}$ by

$$M_\mathcal{D}(\rho) = \min_{\sigma \in \mathcal{F}} \mathcal{D}(\rho, \sigma). \quad (6)$$

If $\mathcal{D}$ is a metric, $M_\mathcal{D}$ fulfills ($S1$). If it is furthermore contractive under completely positive and trace preserving (CPTP) maps, it fulfills ($S2a$) [16,40] and for $\mathcal{D}$ being jointly convex [41], the induced $M_\mathcal{D}$ fulfills condition ($S3$).

In accordance with Refs. [12,15,36], we define the superposition rank $r_S(|\psi\rangle)$ for a state $|\psi\rangle = \sum_j \psi_j |c_j\rangle$ as the number of $\psi_i \neq 0$. Assume a state $|\varphi\rangle = \sum_j \varphi_j |c_j\rangle$ can be transformed (with some probability $p > 0$) to a state $|\xi\rangle = \sum_j \xi_j |c_j\rangle$ by $\mathcal{FO}$. According to Theorems 5 and 7, this is possible if and only if there exists a superposition-free Kraus operator $K = \sum_i c_i |c_{f(i)}\rangle\langle c_i^\perp|$ with the properties

$$\sqrt{p} \sum_i \xi_i |c_i\rangle = \sqrt{p}|\xi\rangle = K|\varphi\rangle = \sum_i \varphi_i c_i |c_{f(i)}\rangle, \quad (7)$$

and $K^\dagger K \leq \mathbb{1}$. Hence the number of $\xi_i \neq 0$ is at most as large as the number of $\varphi_i \neq 0$. This proves that the superposition rank cannot increase under the action of a superposition-free Kraus operator. With the definition of the superposition rank at hand, we present some explicit superposition measures.

Proposition 9.—The following functions are superposition measures as defined in Definition 8.

(1) The relative entropy of superposition

$$M_{\text{rel.ent}}(\rho) = \min_{\sigma \in \mathcal{F}} S(\rho || \sigma), \quad (8)$$

where $S(\rho||\sigma) = \mathrm{tr}[\rho \log \rho] - \mathrm{tr}[\rho \log \sigma]$ denotes the quantum relative entropy. See Ref. [16] for the case of coherence theory.

(2) The l_1 measure of superposition

$$M_{l_1}(\rho) = \sum_{i \neq j} |\rho_{ij}|, \quad (9)$$

for $\rho = \sum_{ij} \rho_{ij} |c_i\rangle\langle c_j|$. See again, Ref. [16] for the case of coherence theory.

(3) The rank-measure of superposition

$$M_{\text{rank}}(|\psi\rangle) = \log[r_S(|\psi\rangle)],$$
$$M_{\text{rank}}(\rho) = \min_{\rho = \sum_i \lambda_i |\psi_i\rangle\langle\psi_i|} \sum_i \lambda_i M_{\text{rank}}(|\psi_i\rangle). \quad (10)$$

(4) The robustness of superposition

$$M_R(\rho) = \min_{\tau \text{ density matrix}} \left\{ s \geq 0 \colon \frac{\rho + s\tau}{1+s} \in \mathcal{F} \right\}. \quad (11)$$

This quantity has an operational interpretation in the limit of coherence theory: the robustness of coherence quantifies the advantage enabled by a quantum state in a phase discrimination task [20].

State transformations and resources.—In resource theories, it is an important question to which other states a given state can be transformed under the free operations because this leads to a hierarchy of "usefulness" in protocols. Here we consider the transformation between single copies of pure states. Let us first clarify when probabilistic conversions are possible at all. As already mentioned, there is no possibility to increase the superposition rank of a pure state by applying a superposition-free Kraus operator. On the other hand, if two states $|\psi\rangle = \sum_{j\in R}\psi_j|c_j\rangle$ and $|\varphi\rangle = \sum_{j\in S}\varphi_j|c_j\rangle$ have the same superposition rank $r = |S| = |R|$, then there exists a superposition-free transformation that transforms one to the other with probability larger than zero. To see this, interpret R and S as (arbitrarily) ordered indexing sets. Define a function f that maps the nth element of R to the nth element of S and a superposition-free Kraus operator

$$K = \sqrt{p}\sum_{j\in R}\frac{\varphi_{f(j)}}{\psi_j}|c_{f(j)}\rangle\langle c_j^{\perp}|. \quad (12)$$

Hence, $K|\psi\rangle = \sqrt{p}|\varphi\rangle$ and since $\psi_j \neq 0$ for all $j \in R$ and the pure superposition-free states $\{|c_j\rangle\}$ are linear independent, p can always be chosen such that $p > 0$ and $K^\dagger K \leq \mathbb{1}$. With the help of theorem 7, this proves that there exists a probabilistic superposition-free transformation. Different orderings of S leads to $r!$ different functions f_n, and thus Kraus operators K_n. For convenience, we define

$$F_n = \sum_j \frac{\varphi_{f_n(j)}}{\psi_j}|c_{f_n(j)}\rangle\langle c_j^{\perp}|, \quad (13)$$

with $F_n|\psi\rangle = |\phi\rangle$ and $K_n = \sqrt{p_n}F_n$. This allows us to state our second main result: The optimum free conversion probability between two pure states of the same superposition rank is the solution of the semidefinite program

$$\begin{aligned} &\text{maximize} && \sum_n p_n \\ &\text{subject to} && \sum_n p_n F_n^\dagger F_n \leq \mathbb{1}, \quad p_n \geq 0 \quad \text{for all } n, \end{aligned} \quad (14)$$

which can be solved efficiently using numerical algorithms [42,43]. Doing so, our investigations indicate that deterministic superposition-free transformations are rare in the case of nonorthogonal bases. Already for qubits, the probability for the existence of a deterministic transformation between two randomly picked states seems to be zero. For qubits, this is investigated analytically for a specific initial state in the Supplemental Material [26]. If we consider superposition-free transformations to a target state with lower superposition rank than the initial state, a probabilistic transformation is still possible by the same arguments. The optimization problem, however, is more troublesome since we have to include Kraus operators where different pure superposition-free states are mapped to the same superposition-free target state. Therefore, the optimization problem is no longer semidefinite.

If a d-dimensional superposition state can be used to generate all other d-dimensional states deterministically by means of $\mathcal{FO}$, it can be used for all applications. These states are said to have maximal superposition. This definition is independent of a specific superposition measure and can serve to normalize measures. Such golden units exist in coherence theory for all dimensions [16], but only for qubits in our case.

Proposition 10.—For qubit systems with $\langle c_1|c_2\rangle \neq 0$, there exists a single state with maximal superposition. For higher dimensions, there exists no state with maximal superposition in general.

This is different to coherence theory where in dimension d, all states of the form $|m_d\rangle = 1/\sqrt{d}\sum_{n=1}^{d}\exp(i\phi_n)|n\rangle$, $(\phi_n \in \mathbb{R})$ are maximally coherent [16]. A reason for this seems to be that in our more general setting, one loses entire classes of deterministic free transformations, for example, diagonal unitaries which change the phases ϕ_n.

On the other hand, as in coherence theory [16], the consumption of a qubit state with maximal superposition allows us to implement any unitary qubit gate by means of $\mathcal{FO}$.

Theorem 11.—Any unitary operation U on a qubit can be implemented by means of $\mathcal{FO}$ and the consumption of an additional qubit state with maximal superposition $|m_2\rangle$ provided both qubits possess the same superposition-free basis. This means that for every U there exists a fixed $\Psi \in \mathcal{FO}$ independent of ρ_s acting on two qubits such that

$$\Psi(\rho_s \otimes |m_2\rangle\langle m_2|) = (U\rho_s U^\dagger) \otimes \rho_h, \quad (15)$$

where ρ_h is a superposition-free qubit state.

This means that consuming enough qubits with maximal superposition, one can perform any unitary and thus any operation [44].

Conclusions.—We introduced a resource theory of superposition, which is a generalization of coherence theory [16] and we showed that in a noncontinuous setting, this is the only generalization that allows for a faithful conversion to entanglement. Using the tools of quantum resource theories, we defined superposition-free states and operations. This allowed us to prove that several measures are good quantifiers of superposition, in particular, the relative entropy of superposition and the easy to compute l_1 measure of superposition. We also uncovered an important partial order structure for pure superposition states: a state can be probabilistically converted to another target state via superposition-free operations only when the target has an equal or lower superposition rank. The maximal probability for successful transformations between states of the same superposition rank is the solution of a semidefinite program. Contrasting with coherence theory, we find that only in two dimensions is there a state with maximal superposition content which can be consumed to implement an arbitrary unitary using only free operations.

Our results can help to investigate phenomena such as catalytic transformations [46–50], and act as a starting point for the investigation of mixed state transformations, transformations in the asymptotic limit [36] or approximate transformations [51]. Akin to developments in coherence theory, we can also incorporate further physical restrictions [11] such as conservation of energy [52], or restrictions for distributed scenarios such as local superposition-free operations and classical communication [53–56]. As in coherence theory [36,54], there are also connections to entanglement theory [12,13] to be further understood. As potential next steps, our results could be extended to infinite dimensional states, continuous settings, or linearly dependent free states (like those found in magic state quantum computation [57,58]). This leads towards the ultimate goal of a fully general theory of nonclassicality which puts superposition, coherence, entanglement, and quantum optical coherence on a unified standing.

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[1] A. K. Ekert, Phys. Rev. Lett. **67**, 661 (1991).
[2] C. H. Bennett and S. J. Wiesner, Phys. Rev. Lett. **69**, 2881 (1992).
[3] C. H. Bennett, G. Brassard, C. Crépeau, R. Jozsa, A. Peres, and W. K. Wootters, Phys. Rev. Lett. **70**, 1895 (1993).
[4] M. B. Plenio and S. Virmani, Quantum Inf. Comput. **7**, 1 (2007).
[5] G. Gour and R. W. Spekkens, New J. Phys. **10**, 033023 (2008).
[6] F. G. S. L. Brandão and M. B. Plenio, Nat. Phys. **4**, 873 (2008).
[7] M. Horodecki and J. Oppenheim, Int. J. Mod. Phys. B **27**, 1345019 (2013).
[8] J. M. Matera, D. Egloff, N. Killoran, and M. B. Plenio, Quantum Sci. Technol. **1**, 01LT01 (2016).
[9] M. Hillery, Phys. Rev. A **93**, 012111 (2016).
[10] G. Adesso, T. R. Bromley, and M. Cianciaruso, J. Phys. A **49**, 473001 (2016).
[11] A. Streltsov, G. Adesso, and M. B. Plenio, Rev. Mod. Phys. **89**, 041003 (2017).
[12] N. Killoran, F. E. S. Steinhoff, and M. B. Plenio, Phys. Rev. Lett. **116**, 080402 (2016).
[13] B. Regula, M. Piani, M. Cianciaruso, T. R. Bromley, A. Streltsov, and G. Adesso, arXiv:1704.04153.
[14] W. Vogel and J. Sperling, Phys. Rev. A **89**, 052302 (2014).
[15] J. Sperling and W. Vogel, Phys. Scr. **90**, 074024 (2015).
[16] T. Baumgratz, M. Cramer, and M. B. Plenio, Phys. Rev. Lett. **113**, 140401 (2014).
[17] J. Aberg, arXiv:quant-ph/0612146.
[18] M. Hillery, Phys. Rev. A **93**, 012111 (2016).
[19] J. Ma, B. Yadin, D. Girolami, V. Vedral, and M. Gu, Phys. Rev. Lett. **116**, 160407 (2016).
[20] C. Napoli, T. R. Bromley, M. Cianciaruso, M. Piani, N. Johnston, and G. Adesso, Phys. Rev. Lett. **116**, 150502 (2016).
[21] A. Misra, U. Singh, S. Bhattacharya, and A. K. Pati, Phys. Rev. A **93**, 052335 (2016).
[22] J. K. Asbóth, J. Calsamiglia, and H. Ritsch, Phys. Rev. Lett. **94**, 173602 (2005).
[23] K. C. Tan, T. Volkoff, H. Kwon, and H. Jeong, Phys. Rev. Lett. **119**, 190405 (2017).
[24] A. Chefles, Phys. Lett. A **239**, 339 (1998).
[25] A. Chefles, Contemp. Phys. **41**, 401 (2000).
[26] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.119.230401, which includes Refs. [27–33] for further details, proofs and examples.
[27] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, Rev. Mod. Phys. **81**, 865 (2009).
[28] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England, 2010).
[29] A. Pasieka, D. W. Kribs, R. Laflamme, and R. Pereira, Acta Appl. Math. **108**, 697 (2009).
[30] M.-D. Choi, Linear Algebra Appl. **10**, 285 (1975).
[31] M. Jiang, S. Luo, and S. Fu, Phys. Rev. A **87**, 022310 (2013).
[32] G. Vidal, Phys. Rev. Lett. **83**, 1046 (1999).
[33] V. Vedral and M. B. Plenio, Phys. Rev. A **57**, 1619 (1998).
[34] E. Chitambar and G. Gour, Phys. Rev. Lett. **117**, 030401 (2016).
[35] I. Marvian and R. W. Spekkens, Phys. Rev. A **94**, 052324 (2016).
[36] A. Winter and D. Yang, Phys. Rev. Lett. **116**, 120404 (2016).
[37] Y. Yao, X. Xiao, L. Ge, and C. P. Sun, Phys. Rev. A **92**, 022112 (2015).
[38] This can happen in the case of entanglement theory. Let $\{|\psi_i\rangle = |\phi_i\rangle \otimes |\xi_i\rangle\}_i$ be an unextendable separable product basis [39] of a bipartite system and define a trace-decreasing separable operation $\Lambda[\rho] = \sum_i |0\rangle\langle\phi_i| \otimes |0\rangle\langle\xi_i|\rho|\phi_i\rangle\langle 0| \otimes |\xi_i\rangle\langle 0|$ where $|0\rangle$ is an arbitrary reference state. This operation cannot be completed by separable Kraus operators by construction.
[39] C. H. Bennett, D. P. DiVincenzo, T. Mor, P. W. Shor, J. A. Smolin, and B. M. Terhal, Phys. Rev. Lett. **82**, 5385 (1999).
[40] V. Vedral, M. B. Plenio, M. A. Rippin, and P. L. Knight, Phys. Rev. Lett. **78**, 2275 (1997).
[41] A. Wehrl, Rev. Mod. Phys. **50**, 221 (1978).
[42] S. Boyd and L. Vandenberghe, *Convex Optimization* (Cambridge University Press, Cambridge, England, 2004).

[43] F. Jarre and J. Stoer, *Optimierung* (Springer-Verlag, Berlin, 2013).

[44] We can express the unitary operation as a matrix with respect to the orthonormal basis obtained when applying the Gram-Schmidt process on the pure superposition-free states. As shown in Ref. [45], we can then decompose the unitary into unitaries U_2 acting on two-dimensional subspaces spanned by two pure free states. With the help of a qubit state with maximal superposition (with respect to the two free states spanning the two-dimensional subspace under consideration) every U_2 can be implemented.

[45] M. Reck, A. Zeilinger, H. J. Bernstein, and P. Bertani, Phys. Rev. Lett. **73**, 58 (1994).

[46] D. Jonathan and M. B. Plenio, Phys. Rev. Lett. **83**, 3566 (1999).

[47] S. Du, Z. Bai, and Y. Guo, Phys. Rev. A **91**, 052120 (2015).

[48] J. Åberg, Phys. Rev. Lett. **113**, 150402 (2014).

[49] C. Duarte, R. C. Drumond, and M. T. Cunha, J. Phys. A **49**, 145303 (2016).

[50] K. Bu, U. Singh, and J. Wu, Phys. Rev. A **93**, 042326 (2016).

[51] J. M. Renes, J. Math. Phys. **57**, 122202 (2016).

[52] G. Chiribella and Y. Yang, Phys. Rev. A **96**, 022327 (2017).

[53] E. Chitambar and M.-H. Hsieh, Phys. Rev. Lett. **117**, 020402 (2016).

[54] E. Chitambar, A. Streltsov, S. Rana, M. N. Bera, G. Adesso, and M. Lewenstein, Phys. Rev. Lett. **116**, 070402 (2016).

[55] A. Streltsov, S. Rana, M. N. Bera, and M. Lewenstein, Phys. Rev. X **7**, 011024 (2017).

[56] A. Streltsov, E. Chitambar, S. Rana, M. N. Bera, A. Winter, and M. Lewenstein, Phys. Rev. Lett. **116**, 240405 (2016).

[57] V. Veitch, S. A. H. Mousavian, D. Gottesman, and J. Emerson, New J. Phys. **16**, 013009 (2014).

[58] M. Howard and E. Campbell, Phys. Rev. Lett. **118**, 090501 (2017).