Correlations constitute fundamental resources for various tasks in quantum information processing [1]. In order to create the paradigmatic resource—entanglement—global operations are required. These operations come at a price: They require access to all the subsystems of the target system and precise control over their interactions. This motivates the formulation of quantum information theory as a resource theory with respect to the limitations imposed by local operations and classical communication (LOCC) [2–5].

However, there is another price to be paid for correlating quantum systems. As any amount of correlation implies extractable work [6], it follows that energy is required to establish correlations. The required energy depends on the inevitable initial entropy of the system. This establishes a link to another resource theory—(quantum) thermodynamics, where the purity of the system, as well as the available free energy constitute fundamental resources due to the restrictions of the first and second laws of thermodynamics.

Recent interest in thermodynamics in the quantum domain (see, e.g., [7–9]) is fueled by this interesting connection to (quantum) information and its implications for the very foundations of thermodynamic laws [10, 11]. Combining the limitations of both theories shows that the resources of one theory are of great significance to the other as well. Examples range from an inevitable energy cost of measurements [12], and the role of entanglement (and other quantum effects) in thermal machines [13–19], to scenarios [20] in which thermodynamic resources play a role in the formation of entanglement and other types of shared information.

This naturally leads us to ask two fundamental questions about the physical limitations of quantum information processing: What is the optimal amount of correlation and entanglement that can be generated for a given energy cost, and how does the inevitable mixedness due to finite temperatures influence these costs? For closed systems, these questions were addressed in Ref. [20]. In this work we aim to give more general answers.

Using the available control over the system, over the operations thereon, and assuming unlimited control over the system and its arbitrarily large thermal bath (see Fig. 1), we derive the ultimate limitations for any protocol to generate correlations. This top-down approach provides absolute bounds which cannot be outperformed, and we present a protocol for which these bounds can indeed be saturated.

To complement these results, we then present a bottom-up approach for the generation of genuine quantum correlations between fundamental physical systems—field modes with fermionic or bosonic statistics. Taking into account limitations like superselection rules for fermions, and using experimentally feasible and widely available techniques for bosonic modes, we provide protocols for the creation of entanglement. While we find the fermionic protocols to be optimal, we show that the practical bosonic protocols become optimal only in the limit of large input energies. Surprisingly, we find that for both the total and genuine quantum correlations, operations involving the bath may be restricted to simple thermalization processes.

I. FRAMEWORK

Let us start by defining some of the basic notions of quantum thermodynamics. The energy $E$ of any quantum system $S$ is given by the expectation value of the corresponding Hamiltonian $H_S$ in the system state $\rho$, that is, $E(\rho) = \text{Tr}(H_S \rho)$. A crucial quantity, which we will refer to throughout this work, is the free energy $F$, i.e.,
FIG. 1. **Illustration of the general setup:** Two quantum systems, $S_1$ and $S_2$, at thermal equilibrium with a bath at temperature $T$ are acted upon either by a unitary $U_S$ on the bipartite system, or by a more general unitary $U_{SB}$ that also involves the bath. The application of these unitaries, which correlate the system, requires a supply of external energy. In this general setting, we determine the optimal amount of correlations and entanglement that can be generated in the system for any given amount of energy.

The part of the energy that is extractable from a closed system at temperature $T$,

$$ F(\rho) = E(\rho) - TS(\rho), \quad (1) $$

where $S(\rho) = -\text{Tr}(\rho \ln(\rho))$ is the von Neumann entropy. For thermal states $\tau(\beta)$ of the form

$$ \tau(\beta) = \frac{e^{-\beta H_S}}{Z(\beta)}, \quad (2) $$

the free energy takes on its minimal value $F(\tau(\beta)) = -T \ln(Z)$, where $Z$ is the partition function, $\beta = 1/T$, and we work in units where $\hbar = k_B = 1$. For arbitrary states, $F(\rho)$ may be referred to as the non-equilibrium free energy. In the following, we consider the initial state of the system $S$ to be thermal, $\rho_S = \tau_S(\beta)$.

We further assume that a heat bath $B$, that is, an arbitrarily large ancillary system in thermal equilibrium, is available. The total Hamiltonian is $H = H_S + H_B$, and the initial state can be written as $\tau_{SB}(\beta) = \tau_S(\beta) \otimes \tau_B(\beta)$. The Hilbert space $\mathcal{H} = \mathcal{H}_S \otimes \mathcal{H}_B$ of $S$ is divided into two subsystems, $S_1$ and $S_2$, which we assume to be non-interacting, such that $H_S = H_{S_1} + H_{S_2}$ and, consequently, $\tau_S(\beta) = \tau_{S_1}(\beta) \otimes \tau_{S_2}(\beta)$. These initially uncorrelated subsystems are to be correlated via a global unitary operation $U_{SB}$ on the total Hilbert space $\mathcal{H} = \mathcal{H}_S \otimes \mathcal{H}_B$. The unitary $U_{SB}$ is the most general operation available, assuming that $S$ and $B$ are isolated. This assumption allows us to account for all required resources, including ancillary systems. Any such unitary can be thought of as a single cycle of a quantum machine. The associated energy cost $W$ is defined as the average overall energy change

$$ W = \text{Tr}(H [U_{SB} \tau_{SB}(\beta) U_{SB}^\dagger - \tau_{SB}(\beta)]) = \Delta E_S + \Delta E_B, \quad (3) $$

and it corresponds to the total the work that needs to be performed to correlate $S$. Since $U_{SB}$ leaves the total entropy of $\tau_{SB}$ invariant, $W$ can be identified with the total change in free energy, which is minimal for the initial thermal state. It follows that $W \geq 0$, and hence any operation $U_{SB}$ requires some energy. The aim of this work is to determine how this energy may be used most efficiently to correlate systems $S_1$ and $S_2$.

We distinguish two kinds of correlations, total correlations, and genuine quantum correlations (entanglement). We quantify the former by the mutual information

$$ I_{S_1 S_2}(\rho_S) = S(\rho_{S_1}) + S(\rho_{S_2}) - S(\rho_S), \quad (4) $$

which measures the amount of global information shared among the systems $S_1$ and $S_2$, i.e., the information encoded within the state $\rho_S$ that is not accessible through its subsystems alone. Pure quantum states for which the mutual information is nonzero are entangled, but this is not necessarily the case for mixed states. To quantify genuine quantum correlations between $S_1$ and $S_2$ we employ the entanglement of formation (see, e.g., Ref. [21] for a review of available entanglement measures), which can be defined as the minimal average mutual information across all decompositions of the mixed quantum state into pure state ensembles, i.e.,

$$ E_{AF}(\rho_S) := \frac{1}{2} \inf_{D(\rho_S)} \sum_i p_i I_{S_1 S_2}(|\psi_i\rangle\langle\psi_i|), \quad (5) $$

where $D(\rho_S) = \{p_i, |\psi_i\rangle\} \sum_i p_i |\psi_i\rangle\langle\psi_i| = \rho_S$. In a finite-dimensional system, the entanglement of formation represents the number of maximally entangled states per copy that are needed asymptotically to create the state via LOCC.

**II. CORRELATING QUANTUM SYSTEMS:**

**ENERGY COST AND OPTIMAL PROTOCOLS**

We now present our main results. We start with the top–down approach, where we determine the ultimate limitations of creating correlations, as quantified by the mutual information. Using the facts that the initial thermal state is completely uncorrelated, $S(\tau_S) = S(\tau_{S_1}) + S(\tau_{S_2})$, and that the global unitary leaves the overall entropy invariant, $S(U_{SB} \tau_{SB} U_{SB}^\dagger) = S(\tau_{SB})$, we combine Eqs. (1) and (3) to express the energy cost $W$ in terms of the free energy difference as

$$ W = \Delta F_S + \Delta F_B + T I_{SB}, \quad (6) $$
obtaining a similar expression to those discussed, e.g., in Refs. [22, 23] in related contexts. A detailed derivation of Eq. (6) can be found in Section A.2. In complete analogy to (6), we may split $\Delta F_S$ into the free energy differences of its subsystems, and their correlation as

$$\Delta F_S = \Delta F_{S_1} + \Delta F_{S_2} + T I_{S_1,S_2},$$

for which a proof is also given in Section A.2. For any thermal state $\tau$, the free energy difference to another (non-equilibrium) state $\rho$ may be expressed through the relative entropy $S(\rho\|\tau) = -S(\rho) - Tr(\rho \ln \tau)$ as $\Delta F = TS(\rho\|\tau(\rho))$. This, in turn, allows us to write $W$ in the form

$$\beta W = S(\rho_{S_1}\|\tau_{S_1}) + S(\rho_{S_2}\|\tau_{S_2}) + S(\rho_B\|\tau_B) + I_{S_1,S_2} + I_{SB},$$

where $\rho_{S_1}$, $\rho_{S_2}$, and $\rho_B$ denote the final reduced states for the subsystems, $S_1$ and $S_2$, and the bath $B$, respectively. In other words, work can be invested to shift the thermal marginals away from equilibrium or to create correlations. Since all quantities on the right-hand side of Eq. (8) are non-negative, it can be immediately inferred that the following ultimate bound holds for the amount of correlation that can be generated between the subsystems for a given energy cost $W$ and temperature $T = 1/\beta$,

$$I_{S_1,S_2} \leq \beta W.$$  

Remarkably, it is possible to saturate this bound using a simple set of operations: unitary operations on $S$, and interactions with the bath to thermalize the system. These operations are enough to obtain $W = \Delta F_S$ in (6) in the limit of an arbitrarily large bath that is complex enough to thermalize the system each time they come in contact (see Ref. [24] for a proof, and Ref. [25] for a description in terms of unitary operations). We are now ready to present the protocol achieving $W = T I_{S_1,S_2}$, which can be divided into two steps.

(I) Cooling: First, the temperature of $S$ is lowered from $T$ to $T_1 \leq T$, reducing the global entropy of the system. The (minimal) energy cost for this thermalization process is $W_1 = \Delta F_S$, i.e.,

$$W_1 = F(\tau_1(\beta_1)) - F(\tau_S(\beta)),$$

where $\beta_1 = 1/T_1$.

(II) Correlating: In the second step, the system is isolated from the bath and it is correlated via a unitary operation $U_{corr}$. Following Ref. [20], the unitary is chosen such that $S_1$ and $S_2$ are locally thermal at temperature $T_2 = 1/\beta_2 \geq T_1$, i.e.,

$$Tr_{S_1,S_2}(U_{corr} \tau_1(\beta_1) U_{corr}^\dagger) = \tau_{S_1,S_2}(\beta_2).$$

This choice ensures that the systems are correlated at minimal energy cost $W_2$, see [20].

FIG. 2. Illustration of the protocol: In the first step the system is cooled down by a controlled interaction with the bath, and the heat $Q$ is transferred to the bath. The associated work cost is $W_1$. In the second step the system is isolated from the bath before it is correlated through a unitary operation, which effectively heats up the subsystems. The energy cost of the second step is $W_2$.

There is thus a tradeoff between the amount of work $W_1$, invested to cool down the system, which allows to potentially obtain larger correlations, and the work $W_2$, invested to actually correlate it. As we show in detail in Section A.3, both contributions add up to

$$W = W_1 + W_2 = T I_{S_1,S_2} + T S(\tau_1(\beta)\|\tau_2(\beta)).$$

Therefore, optimality is achieved when the local temperature of the final state marginals is identical to the initial temperature, $T_2 = T$, such that $W = T I_{S_1,S_2}$.

However, it may occur that this condition would require more energy to be used in the first step than is needed to reach the ground state. In such a case, the excess energy can be put to better use further correlating the final state, raising the local temperatures of the subsystems beyond $T_2 = T$. These considerations yield a more precise bound (see Section A.3), given by

$$I_{S_1,S_2} \leq \begin{cases} 
\beta W & \text{if } \beta W \leq S(\tau_2(\beta)), \\
S(\tau_1(\beta_2)) & \text{if } \beta W > S(\tau_2(\beta)). 
\end{cases}$$

where $\beta_2$ is given by the implicit relation $E(\tau_2(\beta_2)) = W + F(\tau_2(\beta))$. There are hence two distinct regimes. When an energy smaller than $TS(\tau_2(\beta))$ is supplied, the correlations scale linearly with the work input. As more energy is provided, additional work needs to be invested to move the states further out of local equilibrium, leading to noticeably different behaviour. For instance, for two bosonic modes the correlations scale logarithmically with the work input for $\beta W \gg S(\tau_2(\beta))$, as we show in Section A.4.

Finally, it is worth mentioning that our protocol is extendible to non-equilibrium initial states. One then needs to first extract the work content of the state, which leaves it in a thermal state at the temperature of the bath. Our protocol can then be readily applied using the extracted work in addition to any externally supplied energy to correlate the system.
III. ENERGY COST OF ENTANGLEMENT GENERATION

Having provided general bounds on the energy cost of correlating two arbitrary systems, we now turn to the case of genuine quantum correlations, i.e., entanglement. Here the situation is more complex. Even determining whether a given quantum state is separable or not is generally NP hard. We will therefore take a bottom-up approach, focusing our attention on the generation of entanglement in systems of two fermionic or bosonic modes.

The motivation of this choice is two-fold. On one hand, modes of quantum fields play a fundamental role in the description of nature in the context of (relativistic) quantum theory. Hence, they provide a more general framework for our analysis than systems with fixed numbers of particles, which appear as secondary quantities, i.e., as excitations of the modes in question. On the other hand, this approach allows us to analyze the interesting features of fermionic and bosonic particle statistics, and the corresponding finite and infinite-dimensional Hilbert spaces for two modes. In addition, the formulation in terms of individual mode operators naturally lends itself to the Hamiltonian structure, giving a clear interpretation to the involved energy costs.

A. Fermionic systems

We now consider a finite-dimensional system, two modes of (equal) frequency $\omega$ of an uncharged, noninteracting fermionic field. On one hand, the simplicity of this system allows us to determine the amount of entanglement that may be generated for any given amount of energy. On the other hand, several conceptually interesting features arise from the fermionic algebra, that is, the mode operators $b_1, b_1^\dagger, b_2, b_2^\dagger$ satisfy the anticommutation relations $\{ b_m, b_n^\dagger \} = \delta_{mn}$ and $[ b_m, b_n ] = 0$, where $m, n = 1, 2$. The Hamiltonian of the system is (up to a constant) given by $H_S = H_{S_1} + H_{S_2} = \omega (b_1 b_1^\dagger + b_2 b_2^\dagger)$. To distinguish the fermionic and bosonic case, we denote the fermionic Fock states by double-lined kets, e.g., the vacuum state is written as $| 0 \rangle$. The single particle states are obtained by the action of the creation operators, i.e., $| 1_m \rangle = b_m^\dagger | 0 \rangle$. We define the two-particle state via $| 1_{S_1} \rangle \langle 1_{S_2} | = b_1 b_2 b_2^\dagger | 0 \rangle$, where we have omitted the symbol for the antisymmetrized tensor product on the left-hand side (see Refs. [26] or [27], pp. 37 for more details on the notation used here and the fermionic Fock space). The system we investigate here obeys Fermi-Dirac statistics, and the partition function is hence $Z_{FD}(\beta) = (e^\beta + 1)$, and we specify temperatures in units of $\omega$ (recall, that $\hbar = k_B = 1$) from now on. The average initial particle numbers are given by $N_{S_1(S_2)} = \text{Tr}(n_{S_1(S_2)} \tau_S) = Z_{FD}^{|S_1(S_2)|}(\beta)$. The fermionic two-mode thermal state may then be expressed as

$$\tau_S = \frac{e^{\beta}}{Z_{FD}^{|S_1(S_2)|}(\beta)} \left( e^{\beta} | 0 \rangle \langle 0 | + | 1_{S_1} \rangle \langle 1_{S_1} | + | 1_{S_2} \rangle \langle 1_{S_2} | \right).$$

With these preliminaries at hand, we consider protocols along the lines of that presented in Section II to create entanglement. In the first step of such a procedure, using the interaction with the bath, the temperature of the two modes is lowered as before, which manifests in altered particle numbers $N_{S_1}^\dagger$ and $N_{S_2}^\dagger$. The energy cost $W_I$ for this step is given by the free energy difference to the transformed state.

In the second step of the protocol, unitaries on the two-mode space $S$ are applied to correlate the system. In the case of fermionic modes, these operations are further restricted by superselection rules. Since the state of any single fermion acquires a phase of $\pi$ upon a rotation around $2\pi$, rotational symmetry prohibits coherent superpositions of even and odd numbers of fermions. Moreover, the superselection rules modify the definition of the entanglement of formation of Eq. (5) in the sense that the minimization is carried out only over pure state ensembles that respect superselection [28]. We hence take as a measure of entanglement the minimum number, per copy, of maximally entangled states of the two fermionic modes, that are needed to assemble a given two-mode state. As is shown in Section A.5, this well-defined measure of entanglement can be expressed by the energy cost $W_{II}$ of the correlating step as

$$E_{oF} = \ln(2) \sqrt{\frac{W_{II}}{\omega}} \sqrt{\frac{2 e^{\beta} - 1}{e^{2\beta} + 1} - \frac{W_{II}}{\omega}}.$$  

Similar to the previous section, we determine the optimal splitting of $W$ into $W_I$ and $W_{II}$, and we express it in terms of the optimal final temperature $T_{II}$. The results of this numerical optimization are presented in Fig. 3. Although the protocol is very similar to that for the generation of mutual information, optimality is not achieved for $T_{II} = T$ but rather when $T_{II} \geq T$, see Fig. 3 (b).

One can further improve upon these results by taking advantage of the peculiar properties of fermionic entanglement, in particular the existence of mixed, maximally entangled states [29]. These particularities may occur because the subspaces of even and odd fermion numbers decouple. Consequently, no unitaries may introduce correlations between these subspaces. The optimally correlating unitary $U_{corr}$ can therefore be decomposed into two independent rotations. Furthermore, we find that altering the temperatures of the subsystems asymmetrically, i.e., cooling one mode while heating the other, can be beneficial. Allowing for such asymmetric temperatures, we numerically optimize the fermionic entanglement of formation generated at a fixed energy cost. The results are discussed in detail in Section A.5.
and the particle states, which are obtained by applying the creation operators $a_1^\dagger$ and $a_2^\dagger$ to the vacuum. The bosonic excitations obey Bose-Einstein statistics, where the partition function is given by $Z_{\text{BE}}(\beta) = (e^\beta - 1)^{-1}$. Note that the temperatures are again given in units of $\omega$ and we have set $\hbar = k_B = 1$.

To handle this infinite-dimensional system we will restrict our analysis of entanglement generation to Gaussian states, which commonly feature in applications in quantum information [30] and quantum computing [31], to name but a few. The correlations of two-mode Gaussian states can be completely described by a real, $4 \times 4$ covariance matrix $\sigma_S$. This matrix collects the expectation values of quadratic combinations of the mode operators—the second moments—and we may assume that the expectation values of all linear combinations of mode operators—the first moments—and we may assume that the expectation values of all linear combinations of mode operators vanish. For a given state $\rho_S$, the components of $\sigma_S$ are $(\sigma_S)_{mn} = \text{Tr}(\hat X_m \hat X_n) \rho_S$, with the quadrature operators $\hat X_{2n-1} = (a_n + a_n^\dagger)/\sqrt{2}$ and $\hat X_{2n} = -i(a_n - a_n^\dagger)/\sqrt{2}$, and $m, n = 1, 2$. For the initial thermal state at temperature $T$ that we consider here, the covariance matrix is proportional to the identity operator, $\sigma_S = \nu(T) \mathbb{I}_4$, where the symplectic eigenvalue $\nu$ is given by $\nu(T) = \coth(\beta/2)$.

In the first step of the protocol to optimally generate entanglement, the initial temperature is lowered from $T$ to $T_1 < T$, after which the state is represented by $\sigma^i_0 = \nu^i \mathbb{I}_4$, where $\nu^i = \nu(T_1)$. The energy cost for this step is given by

$$\frac{W_1}{\omega} = \nu^i - \nu(T) - 2\beta^{-1} \left[ f(\nu^i) - f(\nu(T)) \right],$$

where the entropy of a two-mode thermal state represented by $\sigma$ is expressed as $S(\sigma) = 2f(\nu) = (\nu + 1) \ln(\nu + 1) - (\nu - 1) \ln(\nu - 1)$.

In the second step of the protocol, we restrict the entangling unitaries to Gaussian operations, which may be represented as linear transformations of the mode operators. Since the initial covariance matrix is proportional to that of the vacuum, the final covariance matrix must be proportional to that of a pure, two-mode Gaussian state, which is locally equivalent to a two-mode squeezed state. We may therefore conclude that the optimal Gaussian entangling operations for this situation are two-mode squeezing transformations. Moreover, throughout the protocol the state remains symmetric with respect to the two subsystems, that is, their entropies are identical. For such states, all entanglement measures depend on a single parameter $\tilde \nu_-$, the smallest symplectic eigenvalue of the partial transpose. In terms of $\tilde \nu_-$ the entanglement of formation takes the form

$$E_{\text{OF}} = \begin{cases} \mathfrak{h}(\tilde \nu_-), & \text{if } 0 \leq \tilde \nu_- < 1, \\ 0, & \text{if } \tilde \nu_- \geq 1, \end{cases}$$

where $\mathfrak{h}(x) = h_+(x) \ln(h_+(x)) - h_-(x) \ln(h_-(x))$, and $h_\pm(x) = \frac{(x \pm 1)^2}{4x}$. One may also relate $\tilde \nu_-$ to the squeezing

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**B. Bosonic systems**

Let us now investigate the optimal generation of entanglement for a bosonic system. Analogously to the fermionic case, we consider two modes of an uncharged, noninteracting bosonic field. We assume that these modes, again labelled $S_1$ and $S_2$, have the same frequency $\omega$. The corresponding annihilation and creation operators $a_1$, $a_2^\dagger$, and $a_2$ satisfy the commutation relations $[a_m, a_n^\dagger] = \delta_{mn}$ and $[a_m, a_n] = 0$, where $m, n = 1, 2$. The system Hamiltonian may be written in terms of these operators (up to a constant) as $H = H_{S_1} + H_{S_2} = \omega(a_2^\dagger a_1 a_1^\dagger a_2)$. The infinite-dimensional Fock space of these two modes is spanned by the vacuum state $|0\rangle$, which is annihilated by $a_1$ and $a_2$, and the corresponding effective final temperature $T_F$ is greater than or equal to $T$ of the marginals after the protocol.
parameter $r$ of the thermal two-mode squeezed state after step II via $e^{-2r} = \tilde{\nu} / \nu^2$, while the final state energy is given by $\omega (\nu^4 \cosh(2r) - 1)$. With this, the energy cost for step II can be expressed as

$$\frac{W_{II}}{\omega} = \frac{\nu^4}{2\tilde{\nu}} [\nu^4 - 1]^2. \quad (18)$$

Conversely, Eq. (18) allows us to express $\tilde{\nu}$, and hence $E_{\text{of}}$, in terms of $\nu^4$ and $W_{II} = W - W_{I}$. The results of the numerical optimization of the entanglement of formation over $\nu^4$ are shown in Fig. 4. Note that in contrast to the fermionic case we here find $T_{II} < T$. Another interesting feature of the bosonic system is that for nonzero initial temperatures entanglement cannot be generated for arbitrarily small amounts of supplied energy [32]. Instead, entanglement is only created when the constraint $(\nu^4 - 1)^2 < 2W_{II}/\omega$ is satisfied.

Finally, a comment about the optimality of Gaussian operations is in order. As we show in detail in Section A.6, there are two energy regimes. In the low-energy regime, Gaussian operations may be outperformed by non-Gaussian operations in generating entanglement. We provide a protocol which achieves this, and allows leaving the separable states even for arbitrarily small amounts of supplied energy. In the high-energy regime, on the other hand, Gaussian operations are shown to be optimal for the generation of entanglement.

### IV. CONCLUSION

We have investigated the equivalence between free energy and the ability to create correlations in quantum systems. Any amount of correlation implies that extractable work is present in the system. Conversely, the creation of any amount of correlation comes at the price of investing work. Following this premise, we have introduced protocols that are optimal for the generation of total correlations, as well as genuine quantum correlations. For total correlations, as quantified by the mutual information, we have presented a protocol that we have shown to be optimal for arbitrary physical systems. For entanglement, we have pursued a bottom-up approach, where we have shown optimality for creating entanglement between two fermionic, or two bosonic modes.

For the case of the paradigmatic quantum resource, entanglement, this is only strictly true in an asymptotic sense of a large available energies. On the other hand, for low energies one encounters limitations depending on the initial entropy and the temperature of the thermal bath. In infinite dimensional systems, entanglement may not be created for arbitrarily small energies if one restricts to Gaussian operations. We have countermanded this issue by introducing also a non-Gaussian protocol, which does not suffer from this problem. A common feature of all the mentioned protocols is their remarkably simple structure.

They make use of the interaction with a thermal bath to cool (or heat) the (sub)system, which, remarkably, requires only elementary thermalization processes, before introducing correlations.

Our results connect two important resource theories, revealing the implicit thermodynamical cost and value of quantum correlations. While we have focused our efforts on bipartite quantum systems, the results concerning correlations have the potential for a straightforward generalization to the multipartite case when considering correlations quantified by $S(\rho) - \sum_i S(\rho_i)$ where $\rho_i = Tr_j \rho$. Such considerations are possible extensions of our work, especially when connected to cases of multipartite entanglement generation. Here, the focus on bipartite entanglement has guaranteed the utility of the created resources for quantum communication, whereas
future work concerning multipartite entanglement should be approached with great care, as generic generation of entanglement may be less useful than previously believed [33]. Other possible directions inspired by our work include similar considerations for single-shot scenarios as, e.g., in Refs. [10, 24, 34], which effectively means focusing on different entropies in the mutual information, the inclusion of catalytic systems [10], or even the interesting connection with the thermodynamic properties of transformations induced by non-uniform motion [35, 36].

APPENDIX

A.1: Preliminaries

Before we present detailed proofs for the main results, let us review some preliminary concepts. First, recall that the free energy of a state $\rho$ is given by

$$F(\rho) = E(\rho) - TS(\rho) = \text{Tr}(\rho H) + T \text{Tr}(\rho \ln \rho).$$

(A.11)

For a thermal state, $\tau(\beta) = e^{-\beta H}/Z$, with the partition function $Z \in \mathbb{R}$, and $\beta = 1/T$, where we have set $k_B = 1$, the free energy reduces to

$$F(\tau(\beta)) = -T \ln Z.$$  

(A.12)

Moving a thermal state away from equilibrium always requires work, which is given by the free energy difference

$$\Delta F(\tau(\beta) \rightarrow \rho) = F(\rho) - F(\tau)$$

(A.13)

to the final state $\rho$. An elementary way to see that $\Delta F \geq 0 \ \forall \ \rho$ for initial thermal states is via the relative entropy $S(\rho||\tau)$, defined as

$$S(\rho||\tau) = -S(\rho) - \text{Tr}(\rho \ln \tau).$$

(A.14)

For thermal states $\tau(\beta)$ we may then write

$$T S(\rho||\tau(\beta)) = -T S(\rho) - T \text{Tr}(\rho \ln \tau(\beta))$$

$$= -T S(\rho) + \text{Tr}(\rho H) + T \ln Z \text{Tr}(\rho)$$

$$= F(\rho) - F(\tau(\beta))$$

$$= \Delta F(\tau(\beta) \rightarrow \rho).$$

(A.15)

By virtue of Klein’s inequality (see, e.g., Ref. [37]) the quantum relative entropy is non-negative, $S(\rho||\tau) \geq 0$, and vanishes if and only if $\rho = \tau$. Consequently, we can conclude that $\Delta F(\tau(\beta) \rightarrow \rho) \geq 0$.

A.2: Energy cost of a general unitary

We now give a detailed proof of Eq. (6), where we denote the transformed states of the system, the subsystems, and the bath as $\rho_s$, $\rho_{s_1}$, $\rho_{s_2}$, and $\rho_B$, respectively. Starting from Eq. (3), the energy differences are rewritten in terms of the changes in free energy and entropy as

$$W = \Delta F_\rho + \Delta E_\rho$$

$$= \Delta F_\rho + \Delta F_B + T \left[ S(\rho_s) + S(\rho_B) - S(\tau_B) - S(\tau_B) \right]$$

$$= \Delta F_\rho + \Delta F_B + T \left[ S(\rho_s) + S(\rho_B) - S(\tau_{SB}) \right]$$

$$= \Delta F_\rho + \Delta F_B + T I_{SB},$$

(A.21)

where we have made use of the fact that the global unitary leaves the overall entropy unchanged, $S(\rho_{SB}) = S(\tau_{SB})$. To prove the similar result of Eq. (7) for the partition of the system $S$ into its subsystems we first write

$$\Delta F_S = \Delta F_\rho - T \Delta S_S$$

$$= \Delta E_{s_1} + \Delta E_{s_2} - T \left[ S(\rho_s) - S(\tau_s) \right].$$

(A.22)

The energy differences of the subsystems may then be expressed as

$$\Delta E_{s_1} = \Delta F_{s_1} + T \left[ S(\rho_{s_1}) - S(\tau_{s_1}) \right],$$

(A.23a)

$$\Delta E_{s_2} = \Delta F_{s_2} + T \left[ S(\rho_{s_2}) - S(\tau_{s_2}) \right].$$

(A.23b)

Finally, noting that $S(\tau_{s_1}) + S(\tau_{s_2}) = S(\tau_s)$, one arrives at

$$\Delta F_S = \Delta F_{s_1} + \Delta F_{s_2} + T \left[ S(\rho_{s_1}) + S(\rho_{s_2}) - S(\rho_s) \right]$$

$$= \Delta F_{s_1} + \Delta F_{s_2} + T I_{s_1} s_2,$$

(A.24)

which concludes the proof.

A.3: Optimal protocol for generating mutual information

Let us now turn the attention to the protocol for the optimal generation of correlations. We proof here that the ultimate bound $W = T I_{s_1 s_2}$ can be achieved, by first proving Eq. (12). The (minimal) energy cost $W_1$ for the first step, reducing the system temperature from $T$ to $T_1 \leq T$, is given by

$$W_1 = \Delta F_S(\tau_S(\beta) \rightarrow \tau_S(\beta_1)) = E(\tau_S(\beta_1)) - E(\tau_S(\beta))$$

$$- T \left[ S(\tau_S(\beta_1)) - S(\tau_S(\beta)) \right].$$

(A.31)

For the second step we use a unitary operation, which leaves the system entropy invariant, while the subsystems become locally thermal at temperature $T_{1 B} = 1/\beta_1$. The average energy of the system after the transformation is hence identical to that of a thermal state $\tau_S(\beta_1)$. The
minimal energy cost $W_H$ is hence given by

$$W_H = E(\tau_5(\beta_H)) - E(\tau_5(\beta_1)). \quad (\text{A.32})$$

The correlations of the final state, as measured by the mutual information, are then

$$\mathcal{I}_{s_1 s_2} = S(\tau_5(\beta_H)) + S(\tau_5(\beta_1)) - S(\tau_5(\beta_1)).$$

Using Eq. (A.33), the energy costs for both steps can be combined to arrive at

$$W = W_1 + W_H = E(\tau_5(\beta_H)) - E(\tau_5(\beta))$$
$$- T \left[ S(\tau_5(\beta_H)) - S(\tau_5(\beta)) + \mathcal{I}_{s_1 s_2} \right]$$
$$= \Delta F(\tau_5(\beta) - \tau_5(\beta_1)) + T \mathcal{I}_{s_1 s_2}$$
$$= T \left[ S(\tau_5(\beta_1)) \right] + \mathcal{I}_{s_1 s_2}. \quad (\text{A.34})$$

Now, if $W$ is split into the contributions $W_1$ and $W_H$ such that $\beta_H = \beta$, one obtains $T \mathcal{I}_{s_1 s_2} = W$, as desired. Interestingly, this is not always achievable. Setting $\beta_H = \beta$ may require $W_1$ to become larger than the energy that is necessary to cool down to the ground state. This leads to a surplus of energy for the correlation step. In such a case $T \mathcal{I}_{s_1 s_2}$ is larger than the initial temperature $T$. The transition to this regime occurs when,

$$W = \tilde{W} = \tilde{W}_1 + \tilde{W}_H = TS(\tau_5(\beta)),$$  \quad (\text{A.35})$$

where $\tilde{W}_1 = -F(\tau_5(\beta))$ corresponds to the energy necessary to cool down to the ground state and $W_H = E(\tau_5(\beta))$ is the work necessary to correlate the systems such that $\beta_H = \beta$. After some rearranging, one obtains

$$\mathcal{I}_{s_1 s_2} \leq \begin{cases} \beta W & \text{if } \beta W \leq S(\tau_5(\beta)), \\ S(\tau_5(\beta_H)) & \text{if } \beta W > S(\tau_5(\beta)), \end{cases} \quad (\text{A.36})$$

where $\beta_H$ is given by the implicit relation

$$E(\tau_5(\beta_H)) = W + F(\tau_5(\beta)). \quad (\text{A.37})$$

There are thus two fundamentally different regimes for the generation of mutual information.

A.4: Generation of mutual information between two bosonic modes

Let us examine more closely the scaling of the generated correlations with the input energy. Since the amount of energy that may be used to correlate two fermionic modes is finite, we will focus on the system of two bosonic modes as described in Section III B. Recall, the system Hamiltonian is given by $H_s = H_{s_1} + H_{s_2}$. Up to a constant, the subsystem Hamiltonians may be expressed in terms of the Fock states $|n_{s_1(s_2)}\rangle = (1/\sqrt{n!})(a_{s_2}^\dagger)^n |0\rangle$ as

$$H_{s_1(s_2)} = \sum_{n=0}^{\infty} n\omega |n_{s_1(s_2)}\rangle \langle n_{s_1(s_2)}|,$$  \quad (\text{A.41})$$

and we use units where $\hbar = 1$. Likewise, the initial thermal state $\tau_5(\beta) = \tau_{s_1}(\beta) \otimes \tau_{s_2}(\beta)$ can be expressed in this way, i.e.

$$\tau_{s_1(s_2)}(\beta) = \sum_{n=0}^{\infty} p_n(\beta) |n_{s_1(s_2)}\rangle \langle n_{s_1(s_2)}|,$$  \quad (\text{A.42})$$

where $p_n = (1 - e^{-\beta}) e^{-n\beta}$, with $\beta = 1/T$, and temperatures in units of $\omega$. The energy and entropy of the thermal state evaluates to

$$E(\tau_5(\beta)) = \text{Tr}(H_5 \tau_5(\beta)) = \omega \left[ \coth(\beta/2) - 1 \right], \quad (\text{A.43})$$

$$S(\tau_5(\beta)) = -\text{Tr}(\tau_5 \ln(\tau_5)) = 2 f \left( \coth(\beta/2) \right), \quad (\text{A.44})$$

where $f(x)$ is the entropic function

$$f(x) = \frac{x + 1}{2} \ln \left( \frac{x + 1}{2} \right) - \frac{x - 1}{2} \ln \left( \frac{x - 1}{2} \right). \quad (\text{A.45})$$

As we have argued in Eq. (13), the optimal mutual information that may be generated from such a thermal state using energies $W$ smaller than $S(\tau_5(\beta))/\beta$, scales linearly with $W$.

Let us now consider the regime where the supplied energy $W$ is much larger than $S(\tau_5(\beta))/\beta$. After reaching the ground state in the first step of the protocol, all of the excess energy increases the correlations. The energy of the final state is equal to the work invested into the correlation step, i.e., $E(\tau_5(\beta_H)) = W_H$. From Eq. (A.43) we hence find

$$\coth(\beta_H/2) = \frac{W_H}{\omega} + 1. \quad (\text{A.46})$$

From Eq. (A.36) we infer that the mutual information is given by $\mathcal{I}_{s_1 s_2} = S(\tau_5(\beta_H))$. Inserting into Eq. (A.44) and expanding $f((W_H/\omega) + 1)$ into a Taylor-Maclaurin series for $(\omega/W_H) \ll 1$ we find

$$\mathcal{I}_{s_1 s_2} = 2 + 2 \ln \left( \frac{1}{2} \frac{W_H}{\omega} \right) + \mathcal{O} \left( \frac{\omega}{W_H} \right), \quad (\text{A.47})$$

where $\mathcal{O}(x)$ is a quantity such that $\mathcal{O}(x)/x$ remains finite in the limit $x \to 0$. We conclude that for large energy supply the optimally generated correlations increase only logarithmically with increasing energy, in stark contrast to the linear increase at small energies, see Fig. 4 (a).
A.5: Optimal protocol for fermionic entanglement of formation

We now present a modification of our previous protocol for the generation of entanglement between two fermionic modes. To optimally convert the supplied energy into fermionic entanglement of formation, the temperatures of the two modes are allowed to change independently of each other in the first step of the protocol. In particular, this entails heating as well as cooling of the individual modes, and the average particle numbers \(N_{s_1}^I\) and \(N_{s_2}^I\) may be different from each other. As before, the energy cost \(W_I\) for this step is given by the free energy difference of the initial thermal and the transformed state.

For step II of the protocol, the two modes are correlated using unitary operations on the system only. As mentioned before, the superselection rules forbid coherent superpositions between even and odd numbers of fermions. In particular, the maximally entangled two-mode pure states for the even parity subspace, \(|\phi^\pm\rangle = \frac{1}{\sqrt{2}} (|0\rangle \pm |1\rangle_1 |0\rangle_2)\), and those for the odd parity subspace, \(|\psi^\pm\rangle = \frac{1}{\sqrt{2}} (|1\rangle_1 |0\rangle_2 \pm |0\rangle_1 |1\rangle_2)\), may not be interconverted by parity conserving operations. These states may hence be regarded as forming a maximally entangled set [39]. Consequently, the optimally correlating unitary \(U_{\text{corr}}\) for two modes decomposes into a direct sum of two \(SU(2)\) rotations. For each, only one real parameter, denoted by \(\theta_{\text{even}}\) and \(\theta_{\text{odd}}\), respectively, is relevant for the amount of generated entanglement. We quantify the entanglement by the superselected entanglement of formation, \(\mathcal{E}_{\text{corr}}\), for a given two-mode state.

However, note that the imposed superselection rules also prevent local changes of basis for each fermionic mode. The states \(|\phi^\pm\rangle\) and \(|\psi^\pm\rangle\) could therefore be considered to be entangled only in a mathematical sense, that is, the entanglement may not be directly used, for instance, to violate a Bell inequality. Nonetheless, if the entanglement is extracted by swapping it to a bosonic system, it becomes useful in the conventional sense. Since a swap using local unitaries cannot create entanglement, its origin must lie in the original fermionic entanglement. Keeping this argument in mind, a pure state decomposition of the transformed state that requires the fewest copies of the maximally entangled pure states \(|\phi^\pm\rangle\) and \(|\psi^\pm\rangle\) may easily be found, yielding the entanglement of formation

\[
E_{\text{ofF}} = \ln(2) \left[ |1 - N_{s_1}^I - N_{s_2}^I| \sin(2\theta_{\text{even}}) \right. \\
+ \left. |N_{s_1}^I - N_{s_2}^I| \sin(2\theta_{\text{odd}}) \right],
\]

where \(0 \leq \theta_{\text{even}}, \theta_{\text{odd}} \leq \pi/4\). Since the odd-subspace rotation shifts excitations of equal frequency, \(\theta_{\text{odd}}\) does not contribute to the energy cost of the second step, which

is given by

\[
\frac{W_{\text{II}}}{\omega} = 2(1 - N_{s_1}^I - N_{s_2}^I) \sin^2(\theta_{\text{even}}). \quad (A.52)
\]

We may hence set \(\theta_{\text{odd}} = \pi/4\) at no additional expense in energy. We note that this suggests a trade-off between creating entanglement in the even and odd subspace, by heating one mode, while the other is cooled. The entanglement of formation becomes maximal when enough energy is supplied to cool one mode, we assume here \(S_1\), to the ground state, whilst \(\theta_{\text{even}} = \pi/4\). The minimum energy \(W_{\text{opt}}\) for which this is the case is obtained when the reduced state of the second mode \(S_2\) is maximally mixed. If less energy than \(W_{\text{opt}}\) is supplied, it is split between cooling and heating the modes \(S_1\) and \(S_2\), respectively, in step I, before correlating them in step II. The resulting state is a mixed state that is entangled both in the even and odd subspace. When \(W = W_{\text{opt}}\), the weights of the even and odd subspace entangled states are equal.

As more energy is provided, it may be used to shift the entanglement to one of the subspaces, obtaining a final state with higher purity. When \(W = W_{\text{max}}\), where \(W_{\text{max}} = W_{\text{opt}} + T \ln(2) = 2T \ln(Z_{\text{FP}}) - \omega\), the final overall state is pure, but the entropy of both subsystems is maximal. The exact values of \(N_{s_1}^1, N_{s_2}^1\), and \(\theta_{\text{even}}\) may be determined by numerical optimization for fixed values of \(W\) and \(T\). In Fig. 5 the protocol is illustrated for various temperatures, where the excess energy between \(W_{\text{opt}}\) and \(W_{\text{max}}\) is used to shift the entanglement towards the even subspace.

Note that the single-mode marginals of the superselected fermionic modes after step I of the protocol are fully determined by the corresponding average particle numbers. In principle, one may therefore consider the first step to involve the preparation of more general, uncorrelated states, for which \(1/2 < N_{s_1}(s_2) \leq 1\). However, we find that optimality is achieved for particles numbers that are compatible with thermal marginals.

A.6: Optimality of Gaussian operations

Finally, we investigate the optimality of Gaussian operations for the generation of entanglement. As for the mutual information, we identify two energy regimes with qualitatively different behaviour. In a certain low-energy regime, we are able to show that Gaussian operations are not optimal. To achieve this, we construct a protocol using specific non-Gaussian unitaries, which outperforms our previously established protocol for Gaussian operations. Nevertheless, in the high-energy regime, Gaussian operations perform better. Indeed, we show that the entanglement generated by the Gaussian protocol scales optimally with the available energy in this case.
Low-energy regime

Instead of the previously established protocol based on Gaussian operations, we now introduce a scheme to generate entanglement using non-Gaussian operations in the correlation step. That is, after cooling the temperature to the temperature $T_1 = 1/\beta_1$ using the energy $W_1$, we perform a unitary transformation that rotates in the subspace of the two-mode Fock space that is spanned by $|0_S⟩|0_S⟩$ and $|n_{S_1}⟩|n_{S_2}⟩$, where we recall the notation of Section A.4. One may think of this operation as generating Bell states in the 4-dimensional subspace. We conveniently parameterize this rotation by a single, real parameter $\alpha$, where $0 \leq \alpha \leq \pi/4$, such that

\[
|0_{S_1}⟩|0_{S_2}⟩ \mapsto \cos(\alpha)|0_{S_1}⟩|0_{S_2}⟩ + \sin(\alpha)|n_{S_1}⟩|n_{S_2}⟩, \quad (A.61)
\]

\[
|n_{S_1}⟩|n_{S_2}⟩ \mapsto \cos(\alpha)|n_{S_1}⟩|n_{S_2}⟩ - \sin(\alpha)|0_{S_1}⟩|0_{S_2}⟩. \quad (A.62)
\]

The energy cost $W_\Pi$ of this rotation is given by

\[
W_\Pi = 2n\omega(p_0^2 - p_n^2)\sin^2(\alpha), \quad (A.63)
\]

where we now have $p_n = (1 - e^{-\beta_1})e^{-\beta_1}$, with $\beta_1 = 1/T_1$, and temperatures in units of $\omega$. Here, the entanglement of formation of the transformed state can be quantified by way of the concurrence[38] of the (unnormalized) state of the subspace spanned by $|0_{S_1}⟩|0_{S_2}⟩$, $|0_{S_1}⟩|n_{S_2}⟩$, $|n_{S_1}⟩|0_{S_2}⟩$, and $|n_{S_1}⟩|n_{S_2}⟩$, see Refs. [40, 41]. For the concurrence $C$, we obtain the expression

\[
C = (p_0^2 - p_n^2)\sin(2\alpha) - 2p_0p_n \quad (A.64)
\]

\[
= \sqrt{\frac{2}{n}}\frac{W_\Pi}{\omega}\sqrt{2(p_0^2 - p_n^2) - \frac{1}{2}W_\Pi}\geq 2np_0^2p_n. \quad (A.65)
\]

Whenever $C > 0$, entanglement is present, which translates to the condition

\[
\frac{W_\Pi}{\omega}\left(p_0^2 - p_n^2 - \frac{1}{2}\frac{W_\Pi}{\omega}\right) > 2np_0^2p_n^2. \quad (A.65)
\]

It can easily be seen that this condition can always be satisfied by choosing $n$ to be large enough. Therefore, some entanglement can be generated at an arbitrary low energy cost given two infinite-dimensional systems. Recall that Gaussian operations require at least the energy $\frac{1}{2}(\nu^1 - 1)^2$ to leave the separable set. Consequently, Gaussian operations cannot be optimal for entanglement generation in all regimes, although they are optimal for the generation of total correlations. Specifically, the unitary of Eq. (11) can be implemented with Gaussian operations. On the other hand, the amount of entanglement generated by the non-Gaussian protocol we have presented here is bounded. For fixed $n$, the maximal amount of energy useful for this protocol is $n(p_0^2 - p_n^2)$, and the corresponding maximal concurrence is given by

\[
C_{\text{max}} = (p_0 - p_n)^2. \quad (A.66)
\]

In contrast, the entanglement that may be generated by Gaussian operations is unbounded. Our considerations are illustrated in Fig. 6.

High-energy regime

To study the regime of large energies, we first show that Gaussian operations are optimal to generate entanglement from the ground state. If the state is pure, the entanglement of formation is simply given by the entropy of the local state. For a given amount of work, the unitary maximizing $E_{\text{of}}$ will then be precisely the expression of Eq. (11), as the thermal state maximizes the en-
entropy for a given energy. Given two bosonic modes, this operation can be implemented by a two-mode squeezing operation. In the protocols that we have considered, the first step consists of cooling. Whenever the ground state is reached, the Gaussian correlating operation is optimal. This occurs when \( W_1 > -F(\tau_S(\beta)) \), and we conclude that the protocol will is certainly optimal when \( W > -F(\tau_S(\beta)) \).

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FIG. 6. Comparison of Gaussian and non-Gaussian operations: The plot shows the amount of entanglement (of formation) that can maximally be generated between two bosonic modes in step II of the protocol. Both modes are assumed to have been cooled to temperature \( T_I \) in the first step. Using the energy \( W_{II} \), the solid curves show the optimal entanglement generated by Gaussian operations, while the dashed curves show the amount of entanglement generated by the non-Gaussian protocol. In both cases the curves are plotted for temperatures varying from \( T_I = 0 \) to \( T_I = 1 \) in steps 0.1 (top to bottom) in units \( \hbar \omega / k_B \).

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