On the Ordering of Sites in the Density Matrix Renormalization Group using Quantum Mutual Information

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The density matrix renormalization group (DMRG) of White 1992 remains to this day an integral component of many state-of-the-art methods for efficiently simulating strongly correlated quantum systems. In quantum chemistry, QC-DMRG became a powerful tool for \textit{ab initio} calculations with the non-relativistic Schrödinger equation. An important issue in QC-DMRG is the so-called ordering problem – the optimal ordering of DMRG sites corresponding to electronic orbitals that produces the most accurate results. To this end, a commonly used heuristic is the grouping of strongly correlated orbitals as measured via quantum mutual information (QMI). In this work, we show how such heuristics can be directly related to minimizing the entanglement entropy of matrix product states (MPS) and, consequently, to the truncation error of a fixed bond dimension approximation. Key to establishing this link is the strong subadditivity of entropy. This provides rigorous theoretical justification for the orbital ordering methods and suggests alternate ordering criteria.

Keywords: Density Matrix Renormalization Group (DMRG), Matrix Product States (MPS), Tensor Networks, Orbital Ordering, Electronic Schrödinger, Entanglement Entropy, Quantum Mutual Information

I. INTRODUCTION

Although quantum many-body systems are a priori described by the Schrödinger equation, its accurate solution is a notoriously difficult problem. Early renormalization group ideas attempted to address this issue by focusing on re-scaling transformations between energy or length scales that, ideally, in each step reduce the number of degrees of freedom while maintaining a good approximation [1]. In [2], White proposed the density matrix renormalization group (DMRG) – a breakthrough numerical algorithm that allowed for highly accurate computations of 1D quantum lattice systems. An important insight from [2,3] is that, in each renormalization step, an accurate approximation is achieved by retaining the degrees of freedom necessary for an accurate description of the entanglement structure. White achieved this by retaining the principal eigenvectors of the reduced density matrix corresponding to the largest eigenvalues – giving the method its name. To this day DMRG remains a crucial component of modern methods for quantum many-body problems.

In [1], DMRG was extended to applications in quantum chemistry (QC-DMRG) and has witnessed remarkable success ever since, see [5,6] for a review. In QC, within the Born-Oppenheimer approximation, using the full configuration interaction (FCI) ansatz and second quantization, one can represent the wave function in terms of the occupied orbitals together with a one-orbital basis set, e.g., \{ |0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle \}. After this DMRG can be applied within the occupation representation.

The efficiency of DMRG relies on an accurate representation of the entanglement structure which in turn depends on the ordering of \textit{sites}. In QC-DMRG, a site corresponds to an orbital. The correlation between different orbitals depends on the choice of the single-particle basis and the ordering of the orbitals on a 1D chain. Unlike for a 1D lattice system, a good ordering of orbitals in QC applications with non-local interactions is not a priori clear. It is by now well-known that optimizing said ordering leads to substantial gains in accuracy [6–18]. The correlation structure within DMRG is affected by the choice of the single-particle basis as well, see, e.g., [6,19].

There have been several approaches to ordering optimization. In this work, we discuss one of the most successful ones, introduced in [9,13]. It relies on an analysis of the \textit{quantum mutual information} (QMI) \(I_{i,j}\) between orbitals \(i,j\). The authors propose to order the orbitals such that the entanglement distance\[20\]

\[
\hat{I}^\pm_{\text{dist}} := \pm \sum_{i,j} I_{i,j} \times |i-j|^\eta,
\]

is minimized. This forces strongly correlated orbitals to be grouped closer together on the chain. To avoid the inevitable combinatorial complexity of testing all possible orbital configurations, the authors propose using an approximate optimization scheme. The latter point is not relevant to this discussion as we focus on \(\hat{I}^\pm_{\text{dist}}\) as a measure of entanglement and approximability. We also note the recent ordering criteria proposed in [18] which outperforms [13] in several numerical experiments. We comment on this criteria in Appendix [B].

Although minimizing \(\hat{I}^\pm_{\text{dist}}\) is based on sound entanglement principles, it does not fully explain why optimizing this criteria has such a significant effect on the approximation accuracy of DMRG. In this work, we demonstrate that the link between two-orbital entanglement and DMRG approximation accuracy can be made quite...
rigorous. DMRG can be seen as a variational ansatz over a matrix product state (MPS) [21]. It is known [22, 23] that the approximation accuracy of a fixed bond MPS is intimately related to the entanglement entropy of sub-chains. The latter, as we will demonstrate, is bounded by the two-site QMI $I_{ij}$. This holds for any entropy measure satisfying the subadditivity (SA) property. We thus provide a rigorous justification for using $f_{\text{dist}}^{\varepsilon}$ as a measure of accuracy and entanglement of 1D chains. This also suggests alternate minimization criteria that are more precise bounds for the subchain entropy, see (11). We discuss the role of different SA properties and entanglement measures.

We remark that in this work we loosely refer to entanglement even though QMI measures total correlation which includes classical correlation as well. In a recent work [24] this was addressed in detail. This does not affect the issue of estimating block entropy.

In Sections IV and V we briefly review MPS, entanglement entropy and the link to approximation accuracy. Section IV contains the main technical result. We conclude in Section V with a discussion about $f_{\text{dist}}^{\varepsilon}$, alternate criteria and different entropy measures. In the Appendix, we provide a proof of the main result (6), a brief discussion on the recent ordering criteria from [18], tree tensor networks and point out a connection between different entropy measures and notions of low-rank approximability.

II. MATRIX PRODUCT STATES

As was observed in [21], DMRG converges to a fixed point that can be written down as an MPS

$$|\Psi\rangle = \sum_{s_1 \cdots s_L} U^{s_1} \cdots U^{s_L} |s_1 \cdots s_L\rangle,$$

where $U^{s_k}$ are matrices of size $\chi_{k-1} \times \chi_k$, the numbers $\chi_k$ are referred to as bond dimensions, and $d$ is the size of the local Hilbert space. For the purposes of this work we only consider finite size MPS with open boundary conditions $\chi_0 = \chi_L = 1$. The contraction of the matrices $U^{s_k}$ is represented pictorially in a tensor diagram in Figure 1 as a 1D chain. Such representations can be easily generalized to more complex networks and so an MPS is a particular kind of a tensor network [25].

In QC-DMRG, in second quantized form, the basis is given as

$$|s_1 \cdots s_L\rangle = (a_1^\dagger)^{s_1} \cdots (a_L^\dagger)^{s_L} |0\rangle,$$

where $a_k^\dagger$ is the creation operator for the $k$-th orbital. The basis thus represents occupation numbers of the corresponding orbitals, e.g., $|s_k\rangle \in \{ |0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle\}$, and in this case $d = 4$.

An MPS can represent any state in the full tensor product Hilbert space provided the bond dimensions are sufficiently large. For a generic state, the bond dimension will grow exponentially in the system size. But it is by now well understood that for many physical states of interest – such as low-lying states of 1D Hamiltonians – a fixed bond MPS provides an accurate approximation [26]. This provides theoretical justification for using DMRG.

The truncation error incurred for a fixed bond dimension MPS can be analyzed via the Schmidt decomposition. Any pure state of a bipartite system can be decomposed as

$$|\Psi_{AB}\rangle = \sum_k \sigma_k^{[A]} |k_A\rangle \otimes |k_B\rangle,$$

with $\sigma_1^{[A]} \geq \sigma_2^{[A]} \geq \ldots \geq 0$. Retaining only the first $\chi$ states, the resulting truncation error is

$$\| |\Psi_{AB}\rangle - |\Psi_{AB}^{\text{trunc}}\rangle \|_2^2 = \sum_{k>\chi} (\sigma_k^{[A]})^2.$$  (3)

For an MPS such a truncation can be applied successively to different subchains [27-30]. In each step, $A := \{1, \ldots, j\}$ and $B := \{j + 1, \ldots, L\} \setminus A$ and we denote by $\varepsilon^2_j(\chi)$ the truncation error from (3) for bond dimension $\chi$. The total truncation error is subadditive

$$\| |\Psi\rangle - |\Psi^{\text{trunc}}\rangle \|_2^2 \leq \sum_{j=1}^{L-1} \varepsilon_j^2(\chi) = \sum_{j=1}^{L-1} \sum_{k>\chi} (\sigma_k^{[1 \cdots j]})^2 =: \varepsilon^2(\chi).$$  (4)

The efficiency of DMRG relies on $\varepsilon^2(\chi)$ remaining small for not too large $\chi$. Note that, strictly speaking, (4) is an idealized situation: DMRG does not rigorously guarantee convergence to a global minimum [31, 32], and so, in that sense, (4) is the best error one can hope for. Nonetheless, in practice – applying possibly modifications to DMRG [33, 34] – $\varepsilon^2(\chi)$ provides a good a priori estimate of its performance.

III. ENTANGLEMENT ENTROPY

The question of simulability with MPS is closely related to the deeper notion of entanglement entropy. A contiguous block $A$ as in Figure 1 may be in general entangled with its environment. The reduced state of $A$ is then described by a density operator $\rho^{[A]} : \mathcal{H}_A \rightarrow \mathcal{H}_A$

$$\rho^{[A]} = \sum_k \lambda_k^{[A]} |k_A\rangle \langle k_A|,$$

with ordered probabilities $\lambda_1^{[A]} \geq \lambda_2^{[A]} \geq \ldots \geq 0$ summing to 1.

To quantify entanglement, we will use a measure of the entanglement entropy. The most commonly used measure is the von Neumann entropy

$$S(\rho^{[A]}) := -\text{Tr}(\rho^{[A]} \log_2 \rho^{[A]}) = -\sum_k \lambda_k^{[A]} \log_2(\lambda_k^{[A]}).$$


It is the quantum analogue of the Shannon entropy, and it quantifies the amount of uncertainty about the state $\rho[A]$. Another common measure is the Rényi entropy

$$S^\alpha(\rho[A]) := (1 - \alpha)^{-1} \log_2 \text{Tr}(\rho[A]^\alpha) = (1 - \alpha)^{-1} \log_2 \left( \sum_k (\lambda_k[A])^\alpha \right),$$

with $\alpha > 0$, $\alpha \neq 1$. For $\alpha \wedge 1$, $S^\alpha(\rho[A]) \to S(\rho[A])$ and so one can denote the von Neumann entropy as $S^{\alpha=1}(\rho[A])$. Another interesting limit is the Hartley entropy $S^{\alpha=0}(\rho[A]) = \lim_{\alpha \to 0} S^\alpha(\rho[A]) = \log_2 \text{rank}(\rho[A])$, see also Appendix D.

For the MPS from (2), applying the Schmidt decomposition for the bipartite splitting into subchain $A := \{1, \ldots, j\}$ and environment $B := \{j + 1, \ldots, L\}$ yields

$$|\Psi\rangle = \sum_k \sigma_k^{[1, \ldots, j]} |k_{1, \ldots, j}\rangle \otimes |k_{j + 1, \ldots, L}\rangle.$$

This gives the reduced density matrix

$$\rho^{[1, \ldots, j]} := \text{Tr}_{\{j + 1, \ldots, L\}}(|\Psi\rangle\langle\Psi|) = \sum_k (\sigma_k^{[1, \ldots, j]})^2 |k_{1, \ldots, j}\rangle \langle k_{1, \ldots, j}|.$$

We measure the entanglement entropy of this subchain as

$$S^\alpha_{[1, \ldots, j]} := S^\alpha(\rho^{[1, \ldots, j]}), \quad \alpha \geq 0.$$

If the entanglement entropy $S$ scales proportionally to the area $|\partial A|$, then such systems are said to satisfy an area law [22, 26]. Area laws in general do not guarantee approximability with tensor networks [35]. But for the special case $D = 1$, an area law (see Figure 1b) implies $S \sim 1$ and thereby approximability [23].

As shown in [22, 23], for any state, one can bound the truncation error $\varepsilon_j(\chi)$ by the entanglement entropy of the corresponding subchain

$$\varepsilon_j(\chi) \leq \left( \frac{\chi}{1 - \alpha} \right)^{(\alpha - 1)/\alpha} 2^{-\frac{\alpha - 1}{\alpha} S^\alpha_{[1, \ldots, j]}}, \quad 0 < \alpha < 1,$$

$$\varepsilon_j(\chi) \geq 1 - \chi^{(\alpha - 1)/\alpha} 2^{-\frac{\alpha - 1}{\alpha} S^\alpha_{[1, \ldots, j]}}, \quad \alpha > 1.$$

Note that Rényi entropies are monotonically decreasing [30]: $S^{\alpha_1} \geq S^{\alpha_2}$ for $0 \leq \alpha_1 \leq \alpha_2$. An upper bound for $\varepsilon_j(\chi)$ trivially holds using the Hartley entropy $S^{\alpha=0}$ but this is not useful for approximation.

The von Neumann entropy $S^{\alpha=1}$ represents a border case. As Schuch et al. [22] demonstrated, it can neither guarantee an upper nor a lower bound in general. However, a diverging von Neumann entropy provides a lower bound [22]. More importantly, Hastings showed [37] that, if $|\Psi\rangle$ is a ground state of a gapped local Hamiltonian, $S^{\alpha=1}$ provides an upper bound for the truncation error of a fixed bond MPS that scales roughly as $2^{-S_{1,\ldots,j}}$, similarly to $\varepsilon_j(\chi)$.

IV. QUANTUM MUTUAL INFORMATION

The entanglement entropy thus provides a good estimate of the simulability of states with MPS. For the truncation error of a fixed bond MPS the relevant quantities are the entropies of subchains $S_{[1, \ldots, j]}$. One can, in principle, estimate these entropies during a DMRG procedure by diagonalizing the reduced density matrices $\rho^{[1, \ldots, j]}$. The computational effort of computing and diagonalizing $\rho^{[1, \ldots, j]}$ will, however, grow exponentially in the length $j$ of the subchain.

An alternative is provided by the quantum mutual information (QMI)

$$I_{i,j} := S_i + S_j - S_{ij}.$$

For two sites $i$ and $j$, $I_{i,j}$ requires only the computation of the single-site entropies $S_i$, $S_j$ and the two-site entropy $S_{ij}$. In [9, 13] the authors propose minimizing the entanglement distance

$$\bar{F}_{\text{dist}} := \sum_{i,j} I_{i,j} \times |i - j|^n.$$

The quantity $I_{i,j}$ measures the amount of classical and quantum correlations between sites $i$ and $j$. It is equal to the relative entropy

$$I_{i,j} = S(\rho^{[ij]}||\rho^{[i]} \otimes \rho^{[j]}),$$

which is the quantum analogue of the Kullback-Leibler divergence. The quantity $S(\rho^{[ij]}||\rho^{[i]} \otimes \rho^{[j]})$ measures, in a sense, the entropic distance between the true reduced two-site state $\rho^{[ij]}$ and its separable approximation $\rho^{[i]} \otimes \rho^{[j]}$. In case the sites $i$ and $j$ are uncorrelated, we have $\rho^{[ij]} = \rho^{[i]} \otimes \rho^{[j]}$, $S_{ij} = S_i + S_j$ and $I_{i,j} = 0$. In case the correlation between $i$ and $j$ is maximal, $\rho^{[ij]} = |\Psi_{ij}\rangle \langle \Psi_{ij}|$ is a pure state, $S_{ij} = 0$ and $I_{i,j} = S_i + S_j$.

The two-point QMI can be directly related to the entanglement entropy of subchains as follows.

Theorem. For any $j = 1, \ldots, L - 1$ and any $\delta = 1, \ldots, \lfloor j/2 \rfloor$

$$S_{[1, \ldots, j]} \leq \sum_{k=1}^j S_k - \sum_{k=1}^{j-\delta} I_{k,k+\delta}.$$

This bound holds for any entropy measure satisfying the strong subadditivity property (SSA), see Appendix A for a proof and Figure 1b for an illustration.

V. DISCUSSION

From (6) and Figure 1b, we see that we can sum up $I_{i,j}$ in different ways, and one can thus ask which bound is sharper. For further reference, note that the von Neumann entropy satisfies the following subadditivity (SA)
The volume $|A|$ is the number of nodes in $A$, the area $|\partial A|$ is the number of nodes connecting to the environment, highlighted in red.

FIG. 1

and strong subadditivity (SSA) properties

$$S(\rho^{AB}) \leq S(\rho^A) + S(\rho^B), \quad \text{(SA)}$$

$$S(\rho^{AB}) + S(\rho^{BC}) \geq S(\rho^B) + S(\rho^{ABC}) \quad \text{(SSA)}$$

Consider first $\delta = 1$. In [9], one “chains” the entropy together via a series of inequalities as

$$S(\rho^{[1,\ldots,k]}) + S(\rho^{[k,k+1]}) \geq S(\rho^{[k]}) + S(\rho^{[1,\ldots,k+1]}), \quad (7)$$

using [SSA]. Denoting by $I(A : B)$ the QMI between subsystems $A$ and $B$, we can write

$$S(\rho^{[1,\ldots,k]}) + S(\rho^{[k]}) + [I([1,\ldots,k] : k + 1) - I(k : k + 1)] = S(\rho^{[1,\ldots,k+1]}).$$

The difference between the two sides of inequality (7) is the amount of entanglement the site $k + 1$ shares with the site $k$ vs. the subchain $\{1, \ldots, k\}$. This difference is bounded by the single-site entropy. Using the symmetry $S(\rho^{[1,\ldots,k]}) = S(\rho^{[k+1,\ldots,L]})$ and [SSA], we namely have

$$I([1,\ldots,k] : k + 1) - I(k : k + 1) \leq 2S(\rho^{[k+1]}).$$

The single-site entropy is itself bounded by the size of the local Hilbert space. I.e., in (7) we overestimate by at most $2\log_2 d$. However, overall we need to “chain” such inequalities $j - 1$ times. In total for $[0]$, in the worst case, one overestimates by $2(j - 2)\log_2 d$.

Consider now instead $\delta = 2$. Here one uses both [SSA] in the form of

$$S(\rho^{[1,3,\ldots,k]}) + S(\rho^{[k,k+2]}) \geq S(\rho^{[k]}) + S(\rho^{[1,3,\ldots,k+2]}), \quad (8)$$

as well as [SA]

$$S(\rho^{[1,3,\ldots,j-1]}) + S(\rho^{[2,4,\ldots,j]}) \geq S(\rho^{[1,2,3,\ldots,j]}). \quad (9)$$

In (8) we once again overestimate by at most $2S(\rho^{[k+2]}) \leq 2\log_2(d)$. However, the number of such chained inequalities is now reduced to roughly half $j/2$. On the other hand, we apply (9) once and we overestimate by $I([1,3,\ldots,j-1] : [2,4,\ldots,j])$ which, in the worst case, is of the order $(j/2)\log_2(d)$. Thus, in total we once again overestimate by at most $2(j - 2)\log_2(d)$.

The same reasoning applies to any $\delta$ with the overestimation always being $2(j - 2)\log_2(d)$. The question which upper bound provides a sharper estimate is then related to comparing two-point QMI $I_{i,j}$ to higher order QMI such as $I([1,3,5,\ldots,j-1] : [2,4,\ldots,j])$ and similarly for different $\delta$. In other words, we cannot reliably estimate this information by using two-point QMI $I_{i,j}$ only.

Using two-point QMI can be seen as a “first order” approximation to the block entropy. Specifically, estimating block entropy by the total entropy $\sum_k S_k$, we overestimate in the order of $\log d$. Correcting total entropy with two-point QMI as in (6) we overestimate in the order of $\log 3$. Using higher-order corrections with correlations between blocks with up to 3 sites, we overestimate by $\log 3$ and so on.

One can exploit the information provided by the bounds for different $\delta$ by combining them into a weighted average, e.g., for $1/c_j = \sum_{k=1}^{j/2} \delta^{-2}$

$$\tilde{I}_j := \sum_{k=1}^{j} S_k - c_j \sum_{\delta=1}^{j/2} \delta^{-2} \sum_{k=1}^{j-\delta} I_{k,k+\delta}. \quad (10)$$

This corresponds conceptually to $\tilde{I}_{\text{dist}}$ of [9] from (1) with $\eta = -2$.

Combining (10) with the discussion in Section [11] and [3] suggests that an appropriate cost function to minimize for control of the total truncation error and control
of the entropy of the MPS, is

$$\hat{I}_{\text{MPS}} = \log_2 \sum_{j=1}^{L-1} 2^{\hat{I}_j}. \quad (11)$$

The above quantity is the LogSumExp function and can be approximated via

$$\hat{I}_{\text{MPS}} = \max_{j=1,\ldots,L-1} \hat{I}_j,$$

with $\hat{I}_{\text{MPS}} < \hat{I}_{\text{MPS}} + \log_2(L - 1)$.

### A. Other Entropy Measures

For a more rigorous error control, one could consider the Rényi entropy $S^\alpha$ for $0 < \alpha < 1$ instead. However, Rényi satisfies only a weak subadditivity property (WSA)

$$S^\alpha(\rho^{[A]}) - S^0(\rho^{[B]}) \leq S^\alpha(\rho^{[AB]}) \leq S^\alpha(\rho^{[A]}) + S^0(\rho^{[B]}).$$

In particular, QMI $I_{i,j}$ defined for $S^0$ with $\alpha \neq 1$ is not guaranteed to be positive and does not necessarily make sense as a measure of correlation. The WSA can still be used to estimate $S_{\text{MPS}}^\alpha$ by chaining differences of two-point entanglement entropies. However, the resulting upper bound is too crude to be useful.

One can also replace $I_{i,j}$ with an analogous quantity for the Rényi divergence. The sandwiched quantum Rényi relative entropy satisfies a version of the SSA as shown in [38]. This could provide a tighter control of the subchain entropy $S_{\text{MPS}}^\alpha$, though we have not analyzed this further.

Furthermore, Rényi was numerically shown [39] to satisfy SSA for $0 \leq \alpha \leq 2$ in the case of Gaussian bosons and $0 \leq \alpha \leq 1.3$ in the case of Gaussian fermions. This suggests that the same orbital optimization as above with $S^0$ replacing the von Neumann entropy may still provide good results for important classes of physical states. However, overall, we expect the von Neumann entropy will provide better control of the total DMRG error and information loss, albeit not with the same generality as the Rényi entropy.

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### Appendix A: Proof of (6)

For any $\delta = 1, \ldots, \lfloor j/2 \rfloor$, we can write

$$\sum_{k=1}^{\delta} S(\rho[k]) + \sum_{k=j-\delta+1}^{j} S(\rho[k]) + 2 \sum_{k=\delta+1}^{j-\delta} S(\rho[k]) \geq S(\rho[j])$$

We chain together the terms with a common site using SSA

$$\sum_{k=1}^{\delta} S(\rho[k,k+\delta]) \geq \sum_{k=1}^{\delta} S(\rho[k,k+\delta,k+2\delta,\ldots,k+\lfloor (j-k)/\delta \rfloor \delta]) + \sum_{k=\delta+1}^{j-\delta} S(\rho[k]).$$

We chain together the disjoint many-site entropies using SSA

$$\sum_{k=1}^{\delta} S(\rho[k,k+\delta,k+2\delta,\ldots,k+\lfloor (j-k)/\delta \rfloor \delta]) \geq S(\rho[j])$$

The single-site entropies cancel with the single-site entropies in (A1). And thus we obtain

$$S(\rho[1,\ldots,j]) \leq \sum_{k=1}^{j} S_k - \sum_{k=1}^{j-\delta} I(k : k + \delta).$$

### Appendix B: Criteria of [18]

Dupuy & Friesecce [18] observed that, if $|\Psi\rangle$ is the second quantization of a Slater determinant, e.g., it is a non-interacting fermionic state, then the singular values of a representation in any a priori fixed single particle basis obey the relation

$$\sigma_{M-k}^{[1,\ldots,j]} \sigma_{M-k}^{[1,\ldots,j]} = p_j, \quad (B1)$$

where $\sigma_{M-k}^{[1,\ldots,j]}$ is the last nonzero singular value, and the factor $p_j$ is a constant depending only on the subchain $\{1, \ldots, j\}$. The authors [18] then suggest optimizing the orbital structure by minimizing $p_j$. This scheme is tested on up to 3-term FCI expansions and outperforms the orbital ordering scheme of [13] by several orders of magnitude.
Is there a relationship to optimizing the entanglement distance \( \hat{I}_{\text{dist}}^{\pm} \)? The symmetry in \( B_1 \) provides implicitly a restriction on the entropy values \( S_{i[1,\cdots,j]} \) can attain. This by itself, however, is not sufficient to justify small entropy or, equivalently, fast decay of singular values. It is not difficult to see that one can construct an entropy maximizing sequence, obeying \( B_1 \), that will have entropy roughly in the order of \( \log_2(M/2) \), i.e., this restriction reduces the maximal entropy content by at most 1.

In that sense, \( \hat{I}_{\text{dist}}^{\pm} \) from \( 1 \) or \( \hat{I}_{\text{MPS}} \) from \( 11 \) provides a more rigorous control of the truncation error and entropy due to \( 6 \). However, using \( B_1 \) (and modifications provided in \( 13 \)) as a criteria may still perform better in practice.

### Appendix C: Tree Tensor Networks

We can replace an MPS with a tree tensor network and repeat the above considerations. E.g., consider a perfectly balanced binary tree as in Figure 2. An idealized optimization scheme would order the orbitals in each layer with \( \log_2(L) \) layers in total. Using only two-point QMI, however, does not provide enough information to reliably optimize the entire tree topology. In principle, one can use the same ordering optimization as for MPS – ordering orbitals on the lowest level only – as an approximation.

Alternatively, we can perform a finer analysis of the relevant entanglement entropies in Figure 2 using only two-point QMI. Similarly to \( 10 \), define for \( 1/c_{ij} = \sum_{\delta=1}^{(j-i+1)/2} \delta^{-2} \)

\[
\hat{I}_{i,j} := \sum_{k=i}^{j} S_k - \sum_{\delta=1}^{(j-i+1)/2} c_{ij} \delta^{-2} \sum_{k=i}^{j-\delta} I_{k,k+\delta}.
\]

Then, for a perfectly balanced binary tree with \( L = 2^M \), set

\[
\hat{I}_{\text{tree}} = \log_2 \sum_{i=1}^{M-1} \sum_{l=1}^{2^{M-1}} 2^{2l-1,2l+1},
\]

as a measure of entropy or approximability of the tree.

### Appendix D: Exact vs. Approximate Low-Rank

In multilinear algebra, an important concept in the theory of tensor networks are so-called minimal subspaces \( 10 \): for a subsystem \( A \subset \Omega \), a pure state \( |\Psi\rangle \) and a corresponding reduced density matrix \( \rho^{[A]} = \text{Tr}_{\Omega\setminus A}(|\Psi\rangle\langle \Psi|) \)

\[
U_{\min}^{[A]} := \text{range}(\rho^{[A]}).
\]

A well-known hierarchy property states that for any disjoint subsystems \( A, B \subset \Omega \), one has

\[
U_{\min}^{[AB]} \subset U_{\min}^{[A]} \otimes U_{\min}^{[B]}.
\]

This is also a well-known property in the theory of renormilation groups (RG) – after an RG transformation to the coarser scale \( AB \) (or simply coarse graining), the corresponding coarse grained Hilbert space is contained in the tensor product of the Hilbert spaces on the finer scales.

One consequence of this hierarchy is the relationship between the corresponding exact bond dimensions (or ranks) of \( |\Psi\rangle \)

\[
\chi^{[AB]} \leq \chi^{[A]} \chi^{[B]}.
\]

(D1)

The ratio of the two quantities above is sometimes taken as a measure of the efficiency of an exact tensor network representation. Note that \( D1 \) is equivalent to the SA property of the Hartley entropy

\[
S(\rho^{[AB]}) \leq S^0(\rho^{[A]}) + S^0(\rho^{[B]}).
\]

For \( \alpha = 0 \), \( S^0 \) quantifies “low-rankness” for exact low-rank representations, while \( \alpha > 0 \) provides a finer grading of low-rank approximability.

\[1\] K. G. Wilson, The renormalization group: Critical phenomena and the Kondo problem, Rev. Mod. Phys. 47, 773 (1975).

\[2\] S. R. White, Density matrix formulation for quantum
renormalization groups, Phys. Rev. Lett. 69, 2863 (1992).
[3] U. Schollwöck, The density-matrix renormalization group in the
wedge of matrix product states, Annals of Physics 326, 96 (2011) january 2011 Special Issue.
[4] S. R. White and R. L. Martin, Ab initio quantum chemistry using the density matrix renormalization group, 
The Journal of Chemical Physics 110, 4127 (1999), https://doi.org/10.1063/1.478295.
[5] S. Wouters and D. V. Neck, The density matrix renormalization group for ab initio quantum chemistry, 
The European Physical Journal D 68, 10.1140/epjd/e2014-50500-1 (2014).
[6] S. Szalay, M. Pfeffer, V. Murg, G. Barcza, F. Verstraete, R. Schneider, and Ö. Legeza, Tensor product methods and entanglement optimization for ab initio quantum chemistry, International Journal of Quantum Chemistry 115, 1342 (2015).
https://onlinelibrary.wiley.com/doi/pdf/10.1002/qua.24898

[7] Ö. Legeza and J. Sólyom, Optimizing the density-matrix renormalization group method using quantum information entropy, Phys. Rev. B 68, 195116 (2003).
[8] G. Moritz, B. A. Hess, and M. Reiher, Convergence behavior of the density-matrix renormalization group algorithm for optimized orbital orderings, The Journal of Chemical Physics 122, 024107 (2005).
https://doi.org/10.1063/1.1828891.
[9] L. Grasedyck, Hierarchical singular value decomposition, in Tensor-Train Format, SIAM Journal on Scientific Computing 31, 2029 (2010), https://doi.org/10.1137/090752286.
[10] D. Ghosh, J. Hachmann, T. Yanai, and G. K.-L. Chan, Orbitval optimization in the density matrix renormalization group with applications to polyenes and β-carotene, The Journal of Chemical Physics 128, 144117 (2008).
https://doi.org/10.1063/1.2883976.
[11] V. Murg, F. Verstraete, O. Legeza, and R. M. Noack, Simulating strongly correlated quantum systems with tree tensor networks, Phys. Rev. B 82, 205105 (2010).
[12] K. H. Marti, B. Bauer, M. Reiher, M. Troyer, and F. Verstraete, Complete-graph tensor network states: a new fermionic wave function ansatz for molecules, New Journal of Physics 12, 103008 (2010).
[13] G. Barcza, Ö. Legeza, K. H. Marti, and M. Reiher, Quantum-information analysis of electronic states of different molecular structures, Phys. Rev. A 83, 012508 (2011).
[14] A. O. Mitrushchenkov, G. Fano, R. Linguerri, and P. Palmieri, On the importance of orbital localization in QC-DMRG calculations, International Journal of Quantum Chemistry 112, 1606 (2012).
https://onlinelibrary.wiley.com/doi/pdf/10.1002/qua.24173
[15] S. F. Keller and M. Reiher, Determining factors for the accuracy of DMRG in chemistry, CHIMIA International Journal for Chemistry 68, 200 (2014).
[16] V. Murg, F. Verstraete, R. Schneider, P. R. Nagy, and Ö. Legeza, Tree tensor network state with variable tensor order: An efficient multireference method for strongly correlated systems, Journal of Chemical Theory and Computation 11, 1027 (2015) pMID: 25884072, https://doi.org/10.1021/ct501187.
[17] C. Krumnow, L. Veis, O. Legeza, and J. Eisert, Fermionic orbital optimization in tensor network states, Phys. Rev. Lett. 117, 210402 (2016).
[18] M.-S. Dupuy and G. Friesecke, Inversion symmetry of singular values and a new orbital ordering method in tensor train approximations for quantum chemistry, SIAM Journal on Scientific Computing 43, B108 (2021).
https://doi.org/10.10137/2006320122.
[19] S. R. White and E. M. Stoudenmire, Multiscale gausslet basis sets for electronic structure, Phys. Rev. B 99, 081110 (2019).
[20] In [9], the authors minimize $I_{\text{dist}}$ with $\eta = -2$. In [13], the authors minimize $I_{\text{dist}}^+$ with $\eta = 1, 2$.
[21] S. Östlund and S. Rommer, Thermodynamic limit of density matrix renormalization, Phys. Rev. Lett. 75, 3537 (1995).
[22] N. Schuch, M. M. Wolf, F. Verstraete, and J. I. Cirac, Entropy scaling and simulability by matrix product states, Phys. Rev. Lett. 100, 030504 (2008).
[23] F. Verstraete and J. I. Cirac, Matrix product states represent ground states faithfully, Phys. Rev. B 73, 094423 (2006).
[24] L. Ding, S. Mardasad, S. Das, S. Szalay, U. Schollwöck, Z. Zimborás, and C. Schilling, Concept of orbital entanglement and correlation in quantum chemistry, Journal of Chemical Theory and Computation 17, 79 (2021), pMID: 33430597, https://doi.org/10.1021/acs.jctc.0c00559.
[25] R. Orús, Tensor networks for complex quantum systems, Nature Reviews Physics 1, 358 (2019).
[26] J. Eisert, M. Cramer, and M. B. Plenio, Colloquium: Area laws for the entanglement entropy, Rev. Mod. Phys. 82, 277 (2010).
[27] G. Vidal, Efficient simulation of one-dimensional quantum many-body systems, Phys. Rev. Lett. 93, 040502 (2004).
[28] I. V. Osledeets, Tensor-train decomposition, SIAM Journal on Scientific Computing 33, 2295 (2011).
https://doi.org/10.1137/090752286.
[29] G. Vidal, Efficient classical simulation of slightly entangled quantum computations, Phys. Rev. Lett. 91, 147902 (2003).
[30] L. Grasedyck, Hierarchical singular value decomposition of tensors, SIAM Journal on Matrix Analysis and Applications 31, 2029 (2010).
https://doi.org/10.1137/090764189.
[31] G. K.-L. Chan and M. Head-Gordon, Highly correlated calculations with a polynomial cost algorithm: A study of the density matrix renormalization group, The Journal of Chemical Physics 116, 4462 (2002).
https://doi.org/10.1063/1.149459.
[32] S. Holtz, T. Rohwedder, and R. Schneider, The alternating linear scheme for tensor optimization in the tensor train format, SIAM Journal on Scientific Computing 34, A683 (2012), https://doi.org/10.1137/100818893.
[33] G. Moritz and M. Reiher, Construction of environment states in quantum-chemical density-matrix renormalization group calculations, The Journal of Chemical Physics 124, 034103 (2006), https://doi.org/10.1063/1.2139998.
[34] S. V. Dolgov and D. V. Savostyanov, Alternating minimal energy methods for linear systems in higher dimensions, SIAM Journal on Scientific Computing 36, A2248 (2014).
https://doi.org/10.1137/140953289.
[35] Y. Ge and J. Eisert, Area laws and efficient descriptions of quantum many-body states, New Journal of Physics 18, 083026 (2016).
[36] A. Rényi et al., On measures of entropy and information, in Proceedings of the Fourth Berkeley Symposium on Mathematical Statistics and Probability, Volume 1: Con-
tributions to the Theory of Statistics (The Regents of the University of California, 1961).

[37] M. B. Hastings, An area law for one-dimensional quantum systems, Journal of Statistical Mechanics: Theory and Experiment 2007, P08024 (2007).

[38] M. Müller-Lennert, F. Dupuis, O. Szehr, S. Fehr, and M. Tomamichel, On quantum Rényi entropies: A new generalization and some properties, Journal of Mathematical Physics 54, 122203 (2013), https://doi.org/10.1063/1.4838856.

[39] G. Camilo, G. T. Landi, and S. Eliëns, Strong subadditivity of the Rényi entropies for bosonic and fermionic Gaussian states, Phys. Rev. B 99, 045155 (2019).

[40] A. Falcó and W. Hackbusch, On minimal subspaces in tensor representations, Foundations of Computational Mathematics 12, 765 (2012).