



Tensor Train Compression of Molecular Orbitals

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Supplementary Material: Tensor Train Compression of Molecular Orbitals

Ruslan A. Mallaev,¹ Dmitry Morozov,¹ Dmitry Zheltkov,¹ Michael Perelshtein,¹ and
Roman Ellerbrock¹

Terra Quantum AG, St. Gallen 9000, Switzerland

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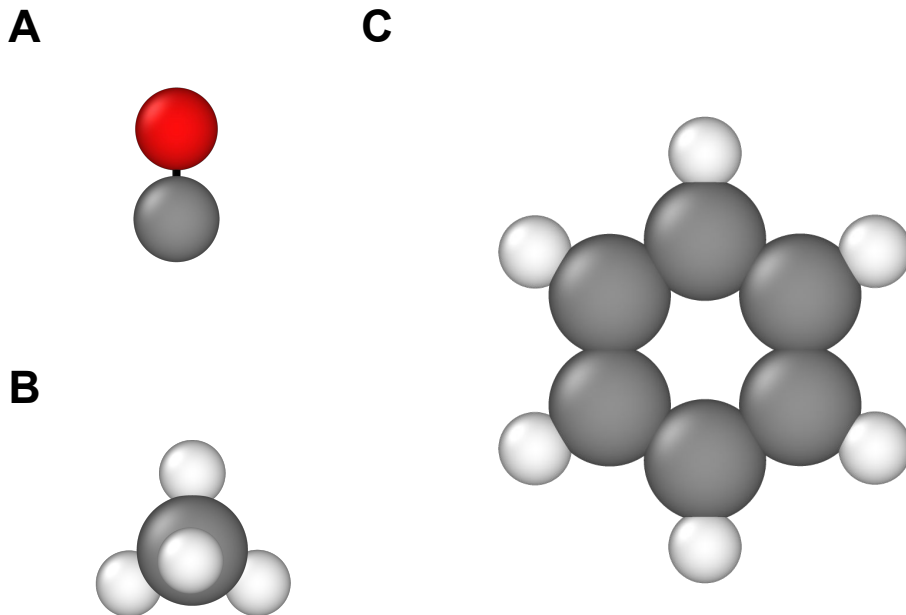


FIG. S1. Organic molecules. (A) CO, (B) CH₄, and (C) benzene.

I. STUDIED MOLECULAR SYSTEMS

We investigated several representative molecular systems. As small organic benchmarks we considered carbon monoxide (CO), methane (CH₄) and benzene; these structures are shown in Figure S1 (panels A–C).

In addition to finite molecules, quasi-periodic carbon-based systems were studied to assess the scalability of the tensor-train representations for extended structures. In particular, graphene-like sheets of increasing size were considered as finite fragments of an ideal infinite graphene lattice. A total of seven graphene-like systems were examined; their sizes and compositions are summarized in Table S1.

The graphite systems were modelled as stacks of graphene-like layers, each layer consisting of 42 atoms (28 carbon atoms). A total of seven structures with the number of layers ranging from one to seven were considered.

In addition, polyaniline chains adopting an α -helix secondary structure were studied as representative biomolecular systems. An example of the atomic structure of polyaniline containing seven amino acid residues is shown in Figure S2. The sizes of the investigated polyaniline systems are summarized in Table S2.

System number	Number of atoms	Number of carbons
1	12	6
2	36	24
3	72	54
4	120	96
5	180	150
6	252	216
7	336	294

TABLE S1. Graphene-like sheets studied.

Number of amino acids	Number of atoms
1	13
2	23
3	33
5	53
7	73
10	103

TABLE S2. Polyalanine systems studied.

Finally, the fullerene C_{60} was studied as a highly symmetric finite carbon system.

II. MEMORY REQUIREMENTS OF TENSOR TRAIN REPRESENTATIONS

One of the primary motivations for employing tensor-train representations is their ability to substantially reduce memory consumption through low-rank approximations. In this section, we quantify the memory savings provided by different tensor-train formats relative to a full real-space grid representation of the orbital wave function.

Using CO, CH₄, and benzene as representative small-molecule benchmarks, we compare three tensor-train formats employed in this work: TT, QTT, and ZQTT.^{1,2} Table S3 reports the number of double-precision numbers required to represent the HOMO wave function

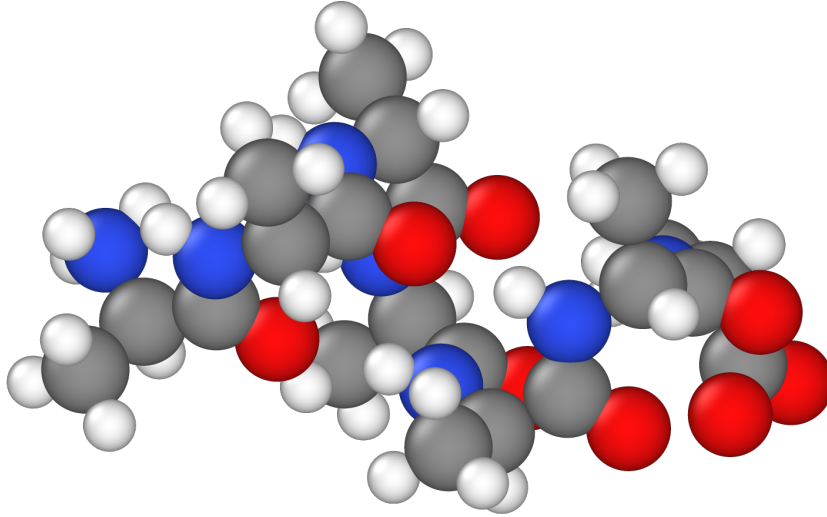


FIG. S2. Atomic structure of a polyalanine chain with seven amino acid residues in an α -helix conformation.

System	Full tensor (%)	TT (%)	QTT (%)	ZQTT (%)
CO	100	0.11	0.03	1.26
CH ₄	100	0.15	0.06	1.40
Benzene	100	0.02	0.01	1.98

TABLE S3. Memory requirements for HOMO wave functions of CO, CH₄, and benzene represented in different tensor-network formats. Values are shown as percentages of the full tensor size (2^{27} points).

with approximation accuracy $\|\psi - \psi_{\text{ref}}\|_F = 10^{-10}$. All values are given as a percentage of the storage required by the full tensor, which contains 2^{27} elements corresponding to a three-dimensional grid with 2^9 points along each spatial axis.

Among the considered representations, the QTT format consistently provides the lowest memory footprint for the studied molecules. Based on this observation, all subsequent calculations for larger systems were performed exclusively using the QTT representation.

Figure S3 illustrates how the memory required by the QTT format scales with system

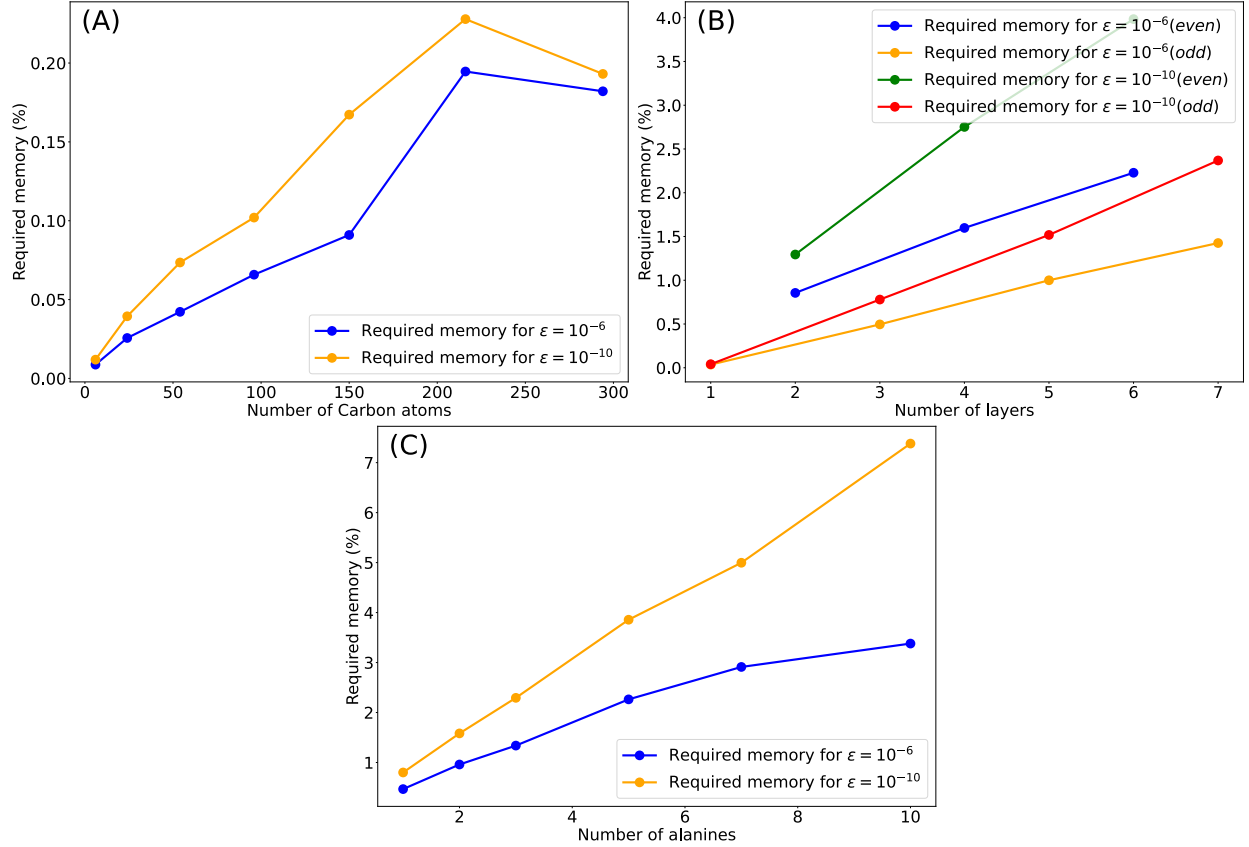


FIG. S3. Memory requirements of the QTT representation for HOMO wave functions. Graphene-like systems as a function of the number of carbon atoms for $\|\psi - \psi_{\text{ref}}\|_F = 10^{-6}$ and 10^{-10} (Panel A). Graphite systems as a function of the number of layers (Panel B). Polyaniline molecules as a function of the number of amino acid residues (Panel C).

size for different classes of systems. Panel A shows the dependence of the number of stored double-precision numbers on the number of carbon atoms in graphene-like structures for two approximation accuracies, $\|\psi - \psi_{\text{ref}}\|_F = 10^{-6}$ and $\|\psi - \psi_{\text{ref}}\|_F = 10^{-10}$. Panel B presents the corresponding results for graphite as a function of the number of layers, while panel C shows the scaling with the number of amino acid residues for polyaniline chains.

III. EFFECT OF GRID REFINEMENT ON QTT ORBITAL COMPRESSIBILITY FOR CO

To complement the discussion in the main text on grid-discretization effects, Figure S4 reports QTT decomposition errors for all CO molecular orbitals in the 6-31G basis for two

different grid resolutions.

Figure S4 compares the QTT decomposition errors for all CO molecular orbitals in the 6-31G basis using two cubic grids over $[-30, 30]^3$ Bohr: 2^9 points per axis (panel A) and 2^{10} points per axis (panel B). Increasing the grid resolution by one level raises the total number of grid points by a factor of 8.

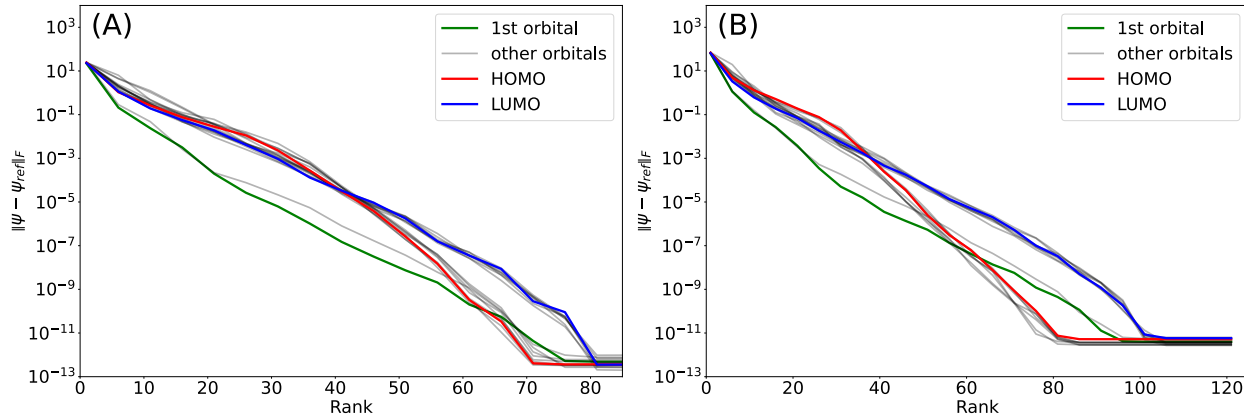


FIG. S4. Representation error as a function of rank for Hartree-Fock orbitals of CO in the 6-31G basis at two grid discretizations. Panel A shows the QTT decomposition error for all orbitals on a cubic grid of $[-30, 30]^3$ Bohr with 2^9 points per axis. Panel B shows the same data for a finer grid with 2^{10} points per axis. Grid refinement shifts the error curves toward higher ranks, but the overall convergence behavior remains similar across orbitals.

The finer discretization leads primarily to a modest rightward shift of the error curves: the same accuracy is reached at somewhat larger QTT ranks, while the overall convergence pattern remains nearly unchanged. In particular, all orbitals still exhibit similar exponential decay of the approximation error with rank, and no strong orbital-type dependence emerges. For CO, the increase in the required rank is relatively small compared to the increase in the total grid size: despite the eightfold growth in the number of grid points, the characteristic ranks increase only by about 20. This behavior is consistent with the discussion in the main text and further supports the conclusion that QTT ranks grow only weakly with grid refinement.

IV. OCTOPUS CALCULATIONS

Within the Octopus/citeoctopus calculations, we analyzed how the accuracy of the ground-state energy depends on the rank of the QTT approximation of the orbital wave functions. In addition, we investigated the relationship between the approximation errors of the orbital wave functions, the resulting electron density, and the total energy of the system.

In our calculations, the primary controlled quantity is the accuracy of the orbital wave functions. Therefore, establishing a quantitative connection between the wave-function approximation error and the resulting energy error is essential. Figure S5 presents the relative error of the electron density as a function of the QTT rank, evaluated using the L^1 , L^2 , and L^∞ norms. Panel A shows the results for graphene. In addition to the electron density error, the panel also reports the approximation errors of two selected orbital wave functions as functions of the QTT rank. These orbitals correspond to the smallest and largest approximation errors among all computed orbitals. Panels B and C present analogous results for benzene and methane (CH_4), respectively. The results demonstrate that the L^1 , L^2 , and L^∞ norms exhibit similar scaling behavior with respect to the QTT rank. This observation justifies the use of a single norm — specifically, the Frobenius (L^2) norm — for assessing wavefunction accuracy in subsequent analyses.

All panels additionally display the relative error of the ground-state energy as a function of the QTT rank. The horizontal gray line indicates an energy error threshold of 1 kcal/mol. The data show that the relative error in the total energy is consistently smaller than the corresponding error in the wave function. Since the orbital wave functions are normalized to unity in the Frobenius norm, the absolute and relative wave function errors coincide, enabling a direct estimation of the expected energy accuracy from the wave function approximation error.

A broader set of molecular and extended systems was also examined within the Octopus calculations. Figure S6 summarizes the dependence of the ground state energy error (in kcal/mol) on the QTT rank used for orbital wave-function approximation.

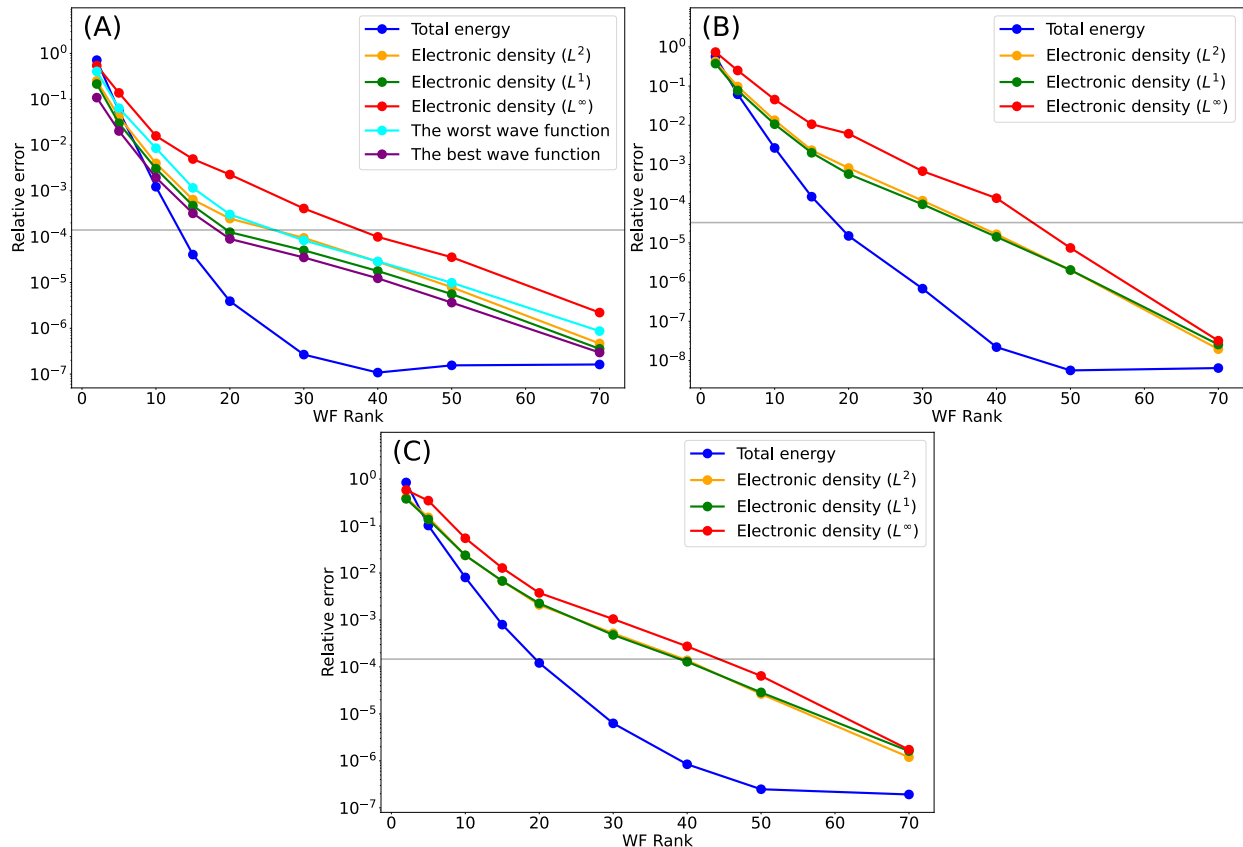


FIG. S5. Comparison of approximation errors for orbital wave functions, electron density, and ground-state energy as functions of the QTT rank. Panel A shows graphene results, including electron density errors evaluated in the L^1 , L^2 , and L^∞ norms, as well as errors for two representative orbital wave functions with the smallest and largest approximation errors. Panels B and C present analogous results for benzene and methane (CH_4), respectively. The gray horizontal line indicates an energy error of 1 kcal/mol.

V. WATER CLUSTERS

Calculations were additionally performed for finite water clusters in order to assess the scalability of the QTT representation for amorphous molecular systems. The generation of water cluster geometries was carried out using GROMACS³. The sizes of the investigated clusters are summarized in Table S4.

Figure S7 presents the results of the QTT approximation for the HOMO of the water clusters. Panel A shows the relative approximation error as a function of the QTT rank for clusters of different sizes. Panel B demonstrates the dependence of the QTT rank required

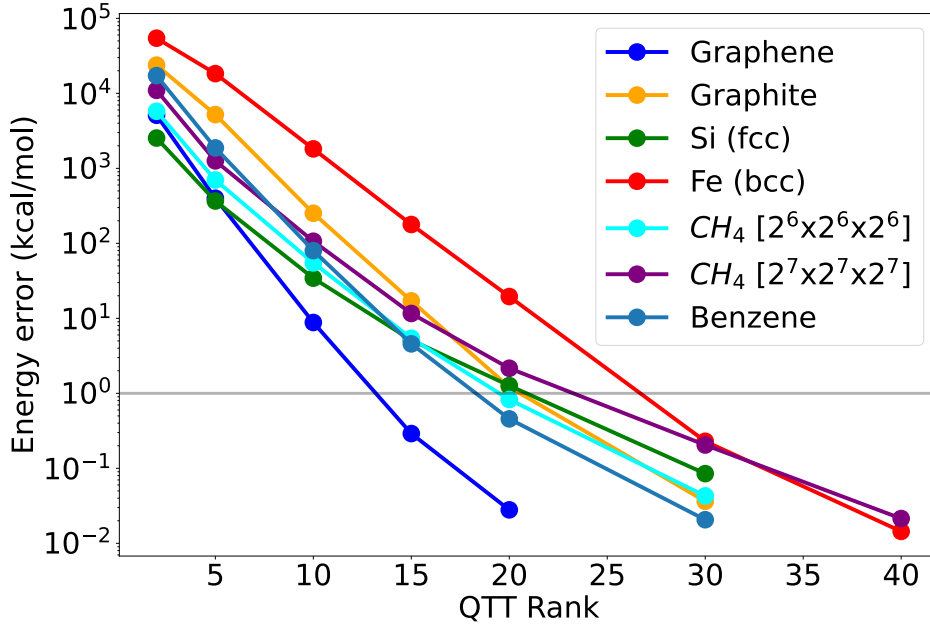


FIG. S6. Ground state energy error as a function of the QTT rank for various systems, as obtained from Octopus calculations. Energy errors are reported in kcal/mol.

Number of H_2O molecules	Number of atoms
5	15
12	36
16	48
21	63
30	90
43	129

TABLE S4. Water clusters investigated in this work.

to achieve a fixed Hartree-Fock orbital accuracy on the number of water molecules. For each target accuracy, the corresponding curve in panel B was fitted using a linear regression of the form $\ln(r) = m \cdot \ln(N) + b$ where r is the QTT rank and N denotes the number of water molecules. The observed trends indicate a sublinear growth of the QTT rank with respect to the system size for a fixed approximation accuracy, confirming favorable scalability properties of the QTT representation for molecular clusters.

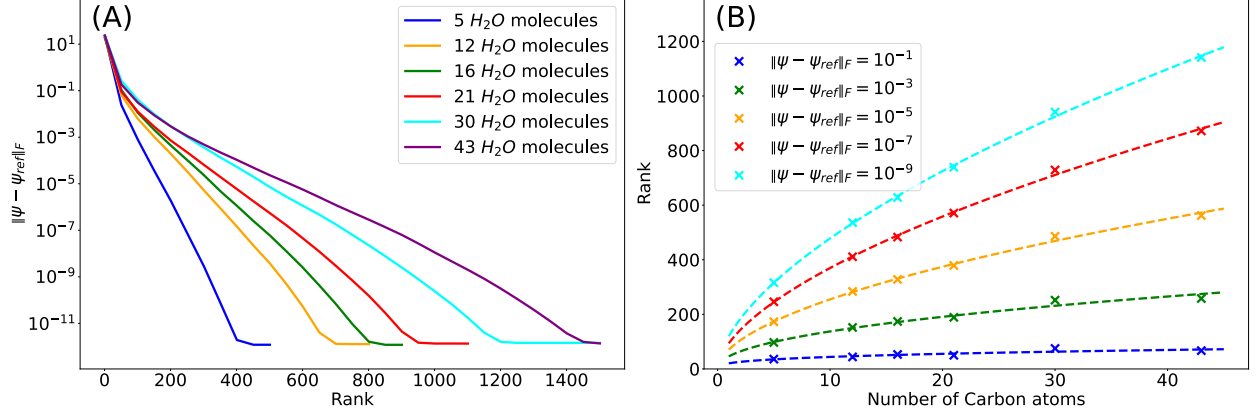


FIG. S7. Scalability of water clusters. Panel (A) shows the overall picture of the QTT decomposition of the orbital wave function (HOMO) for water clusters of different sizes. Panel (B) shows the dependence of the rank of the QTT with a fixed error as a function of the size of the system (number of water molecules). The results demonstrate sublinear growth of the rank with increasing system size.

	m	b
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-1}$	0.46	1.48
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-3}$	0.46	2.01
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-5}$	0.47	2.16
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-7}$	0.48	2.31
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-9}$	0.46	2.56

TABLE S5. Fitted rank scaling coefficients for graphene.

VI. RANK SCALING COEFFICIENTS FOR PERIODIC AND MOLECULAR SYSTEMS

The coefficients characterizing the dependence of the QTT rank on the system size for a fixed wave function approximation accuracy were extracted for several classes of systems. In all cases, the dependence was fitted using the logarithmic scaling law $\ln(r) = m \cdot \ln(pr) + b$. Table S5 reports the fitted coefficients for graphene, Table S6 for water clusters, and Table S7 for polyaniline chains. The obtained values demonstrate that the scaling exponent m remains below unity for all studied systems, confirming sublinear growth of the QTT rank with increasing system size.

	m	b
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-1}$	0.33	3.02
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-3}$	0.48	3.83
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-5}$	0.56	4.26
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-7}$	0.59	4.54
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-9}$	0.60	4.79

TABLE S6. Fitted rank scaling coefficients for water clusters.

	m	b
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-1}$	0.18	3.64
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-3}$	0.35	4.59
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-5}$	0.44	5.11
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-7}$	0.48	5.43
$\ \psi - \psi_{\text{ref}}\ _F = 10^{-9}$	0.49	5.67

TABLE S7. Fitted rank scaling coefficients for polyanines.

VII. DIPOLE MOMENTS OF GRAPHITE LAYERS

During the analysis of the QTT ranks for graphite multilayer systems, it was observed that structures with an odd number of layers systematically exhibit lower ranks compared to those with an even number of layers. This behavior suggests the presence of additional symmetry in odd-layer systems.

To verify this hypothesis, the dipole moments of graphite structures with varying numbers of layers were computed using the PySCF package⁴. The Cartesian components of the dipole moment, expressed in atomic units, are reported in Table S8. The results indicate that systems with an odd number of layers possess non-zero in-plane dipole components, while even-layer systems retain higher symmetry and exhibit vanishing dipole moments along y -axis. This symmetry difference provides a plausible explanation for the observed variation in QTT ranks.

Number of graphite layers	μ_x	μ_y	μ_z
1	-0.061	0.000	0.000
2	-0.088	0.000	0.000
3	-0.122	0.210	0.000
4	-0.156	0.000	0.000
5	-0.189	0.252	0.000
6	-0.222	0.000	0.000
7	-0.254	0.264	0.000

TABLE S8. Dipole moment components for graphite structures with different numbers of layers.

VIII. RELATION BETWEEN WAVEFUNCTION RANK AND ATOMIC POSITIONS

At an early stage of this work, we hypothesized that the rank of an orbital wave function might correlate with the rank of a tensor-train representation of atomic positions. We define the tensor of atomic positions as a tensor that takes the value one at grid points closest to atomic nuclei (i.e., discrete delta functions) and zero elsewhere.

In the QTT representation, a single delta function has rank one. Since the rank of a sum of tensors is bounded by the sum of their ranks, the rank of such an atomic-position tensor is equal to the maximum number of atoms aligned along any spatial axis. Consequently, for a cubic lattice of dimensions $n_x \times n_y \times n_z$, the QTT rank of the atomic-position tensor scales as $\max(n_x, n_y, n_z)$.

To examine whether a similar behavior is observed for molecular orbitals, we analyzed the QTT rank of the HOMO wave function for a series of linear alkenes $C(=CC)_n$. The results are shown in Figure S8. For each molecule, the HOMO was approximated in the QTT format with a target accuracy of $\|\psi - \psi_{\text{ref}}\|_F = 10^{-9}$. Prior to the decomposition, all molecular geometries were aligned along the z -axis to ensure consistent comparison.

The analysis of the QTT ranks of HOMO wave functions for the series of linear alkenes $C(=CC)_n$, together with the results presented in the main text, demonstrates that the initial hypothesis is not supported. Specifically, the expected linear relationship between the rank of the molecular wave function and the rank of the corresponding atomic-position

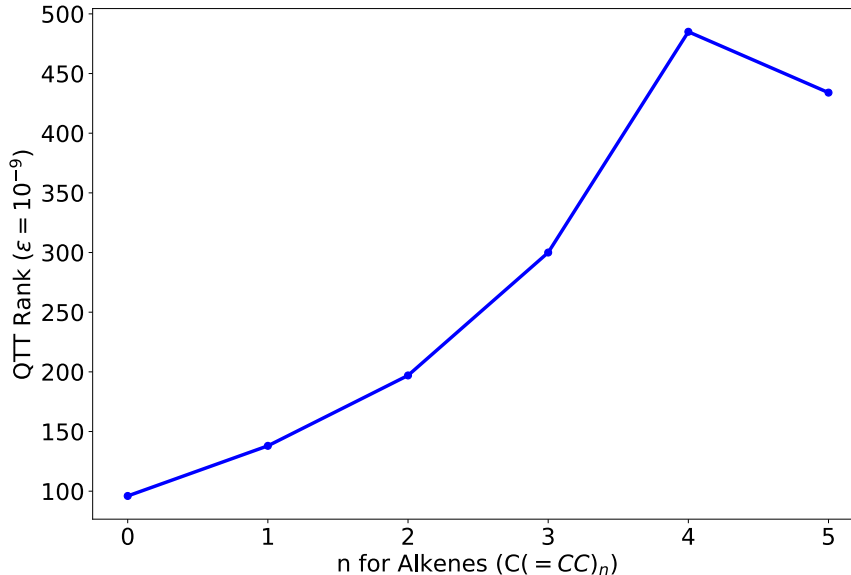


FIG. S8. Scalability of alkenes. The figure illustrates how the rank of the alkene’s molecular orbital wavefunction (HOMO) varies depending on the number of carbon atoms in the molecule. A QTT approximation with target error 10^{-9} has been calculated for each molecule, and the rank is presented in the figure. The molecules were pre-aligned along the z-axis.

tensor is not observed for any of the investigated systems. This indicates that factors beyond the simple spatial arrangement of atoms determine the tensor rank of molecular orbitals in the QTT representation.

IX. ORTHOGONAL TENSOR TRAIN CROSS APPROXIMATION (TT-CROSS-ORTH)

Temporary objects and index sets used in Algorithm 1 of the main text are listed in Algorithm S1.

X. COMPRESSIBILITY AND SCALING OF MOLECULAR SCF WAVEFUNCTIONS IN QTT FORMAT: POWER LAW PARAMETERS

At the studied system sizes, the QTT rank divided by the number of atomic orbitals (at a QTT approximation error of 10^{-6}) as a function of the number of atoms in the sys-

Algorithm S1 Temporary objects and index sets used in Algorithm 1 of the main text.

$\hat{X}^{(k)} \in \mathbb{C}^{r_{k-1} \times n_k \times \hat{r}_k}$: intermediate contracted tensor at sweep position k
 $\hat{\mathcal{J}}_k \subseteq \overline{1..n_{k+1}} \times \cdots \times \overline{1..n_d}$: suffix index sets obtained from TT-MaxVol
 $\mathcal{J}_k \subseteq \overline{1..n_{k+1}} \times \cdots \times \overline{1..n_d}$: augmented suffix index set used for sampling
 $\mathcal{S}_k \subseteq \overline{1..n_1} \times \cdots \times \overline{1..n_k}$: prefix multi-index set
 $B_k \in \mathbb{C}^{r_{k-1} n_k \times \hat{r}_k}$: reshape of $\hat{X}^{(k)}$
 $C_k \in \mathbb{C}^{r_{k-1} n_k \times \hat{r}_k}$: matrix $C_k = (I_{n_k} \otimes R_{k-1}^{-1}) B_k$
 $D_k \in \mathbb{C}^{r_{k-1} n_k \times |\mathcal{J}_k|}$: sampled tensor slices
 $E_k = (I_{n_k} \otimes R_{k-1}) D_k$
 $F_k = [E_k / \|E_k\|_F \quad B_k / \|B_k\|_F]$
 $H_k = [D_k / \|E_k\|_F \quad C_k / \|B_k\|_F]$
 P_k : row indices selected by OMP
 $Z_k \in \mathbb{C}^{r_{k-1} n_k \times r_k}$: OMP coefficient matrix
 $Z_k = Q_k R_k$: QR factorization defining new TT rank r_k
 $T^{(k)} \in \mathbb{C}^{r_{k-1} \times n_k \times r_k}$: TT-core obtained from reshaped Q_k

System	m	b
Graphene	-0.48	-0.11
Graphite odd layers (C_{2v})	-0.23	-0.25
Graphite even layers (C_s)	-0.55	1.85
Polyalanine (α -helix)	-0.50	2.34
Water clusters (amorphous)	-0.42	2.32

TABLE S9. Power law coefficients used for showing the scaling of molecular SCF wavefunctions in QTT format.

tem follows a power law for all studied scalable systems. We performed a power law fit ($\ln(Rank/NAOs) = m \cdot \ln(NAtoms) + b$) which are included in the Figure of the main work. Table S9 shows the obtained fitting parameters.

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