Visualizing operators of coupled spin systems

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The state of quantum systems, their energetics, and their time evolution is modeled by abstract operators. How can one visualize such operators for coupled spin systems? A general approach is presented which consists of several shapes representing linear combinations of spherical harmonics. It is applicable to an arbitrary number of spins and can be interpreted as a generalization of Wigner functions. The corresponding visualization transforms naturally under non-selective spin rotations as well as spin permutations. Examples and applications are illustrated for the case of three spins 1/2.

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I. INTRODUCTION

We present a technique to visualize operators acting on coupled spin systems. Their high-dimensional structure is uniquely described by several shapes (cf. Fig. 1 below), which represent linear combinations of spherical harmonics. Crucial features are directly observable and transform naturally under non-selective spin rotations as well as spin permutations. This provides a general approach to systematically analyze coupled spin systems and their time evolution. We emphasize that our approach is generally applicable and that arbitrary operators on multi-spin systems can be visualized. Examples applicable in research and education include density operators which describe the state of a quantum-mechanical system (e.g., spin systems or quantum bits from quantum information processing), Hamilton operators which specify energy terms, and unitary transformations modeling the time evolution.

Various approaches to visualize quantum systems are known. A quantum-mechanical operator for a two-level system (such as an isolated spin 1/2 particle in an external magnetic field) can always be mapped to a three-dimensional (real) vector as shown in the seminal work of Feynman et al. [1]. This vector can represent a Bloch vector, a field vector, or a rotation vector related to applications ranging from magnetic resonance imaging [2] and spectroscopy [3] to quantum optics [4].

A multi-spin operator can be displayed as a bar chart of the absolute value (or the real and imaginary parts) of its individual matrix elements. This technique is commonly used, e.g., to present experimental results of state tomography of a quantum system [5]. Alternatively, energy-level diagrams are used, e.g., in quantum optics and magnetic resonance spectroscopy. The corresponding populations can be represented by circles on energy levels, and coherences can be depicted by lines between energy levels [6]. Another visualization of a density operator is based on the non-classical vector representation based on single-transition operators [6, 7, 8]. Moreover, graphical shorthand notations for coupled-spin dynamics have been proposed in [9]. All these approaches are cumbersome for many spins, in particular if the density matrix has many non-zero entries. Frequently, non-selective spin rotations do not act naturally on these visualizations.

Our method surmounts these difficulties by relying on a map between a multi-spin basis given by tensor operators (vide infra) and multiple sets of spherical harmonics which are independently plotted in different locations. Related work can be at least traced back to Pines et al. [12] where (albeit without a formal map) selected density operator terms of a spin 1 particle and their symmetry properties are depicted using spherical harmonics. Further visualizations have been presented in [13, 14]. Dowling et al. [15] illustrated collections of (non-interacting) two-level atoms while highlighting connections to Wigner functions (which will be discussed in Sec. V). Similar figures can also be found in [16]. More recently, the usefulness of visualizing single-spin systems with spherical harmonics has been impressively demonstrated for nuclear magnetic resonance experiments of quadrupolar nuclei, including the generation of multiple-quantum coherence and multiple-quantum filters [17]. However, the authors were skeptical if this approach could be generalized to coupled spins, see the discussion in the appendix of [17]. A special class of two coupled spins was treated in [18]. Certain states of two (and three) spins could be visualized by the method of [19]; however in [19] it was also emphasized that a general method was still missing. We present in this work a versatile approach which is applicable to an arbitrary number of coupled spins.

Although our approach is completely general, we focus in the following on the most common situation typically found in the field of magnetic resonance spectroscopy and quantum information processing where all spins are distinguishable and have spin number 1/2 unless otherwise stated. This article has the following structure: First, maps between tensor operators and sets of spherical harmonics are analyzed which furnishes a general framework for our approach to visualization. Then, the LISA basis (with defined linearity, subsystem, and auxiliary criteria,
II. VISUALIZATION

Abstract objects such as quantum mechanical operators can be visualized by mapping them into vivid, three-dimensional objects (such as three-dimensional functions). For example, the state of a quantum mechanical two-level system can be mapped to the Bloch vector visualized as a three-dimensional arrow \([\mathbf{1}]\). In order to generalize this idea, the mapping from an abstract object to its visualization should ideally satisfy the following essential properties: (A) An operator should be bijectively mapped to a unique function (or object). (B) Crucial features should be directly visible. In our context, (B) can refer to (e.g.) observables, symmetries under rotations or permutations, natural transformation characteristics under rotations, as well as the set of involved spins.

In order to describe the mapping from operators to functions, we first recall a complete, orthonormal operator basis known as reducible tensor operators \(T_j\) \([20]\): The components \(T_{jm}\) of \(T_j\) with fixed rank \(j\) and varying order \(m \in \{-j, \ldots, j\}\) form a basis of a space which stays invariant under the action of the rotation group \(SO(3)\) (or any group) and which does not contain a proper invariant subspace. In the following, we usually substitute the rotation group \(SO(3)\) by the locally-isomorphic unitary group \(SU(2)\) which consists of all unitary \(2\times 2\)-matrices of determinant one \([22]\). The tensor operators form the foundation for the theory of angular momentum \([11, 23-28]\) and are part of the standard curriculum of quantum mechanics, see, e.g., \([23]\). It can be illuminating to note (as has been done by Mackey \([30, 31]\), see also \([34]\)) that the tensor operators provide an explicit form for the well-established representation theory of the Lie algebra \(su(2)\) (and more general ones), see, e.g., \([35]\).

Some readers might find it convenient to have a more explicit definition for tensor operators \(T_j\) which is provided using the conditions of Racah \([10]\):

\[
\begin{align*}
[J_x, T_{jm}] &= m T_{jm}, \\
[J_\pm, T_{jm}] &= \sqrt{j(j+1) - m(m \pm 1)} T_{jm, m \pm 1},
\end{align*}
\]

which feature the raising and lowering operators \(J_x := J_x \pm iJ_y\), the infinitesimal rotation operators \(J_x, J_y, J_z\), and the commutator \([A, B] := AB - BA\). In the case of a single-spin system with spin number \(s\), all these operators can be interpreted as \((2s+1)\times(2s+1)\)-matrices and all possible tensors have distinct ranks \(j \in \{0, 1, \ldots, 2s\}\). As an example for \(s = 1/2\), we obtain the Pauli spin matrices \(J_x := \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}/2, J_y := \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}/2, J_z := \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}/2\) as well as the tensor operator components \(T_{0,0} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}/\sqrt{2}, T_{1,-1} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}/\sqrt{2}, T_{1,1} = \begin{pmatrix} 0 & 0 \\ 0 & -1 \end{pmatrix}/\sqrt{2}\), cf. \([3]\).

Having given an operator basis by recalling tensor operators, we can complete the discussion how to map operators to functions by deciding on a suitable set of functions. Note that the tensor operators have been explicitly defined by Wigner \([20]\) and Racah \([10]\) (generalizing the vector operators discussed in \([30]\)) to mimic the properties of spherical harmonics \(Y_{jm}\) \([11]\), which map the spherical coordinates \(\theta\) and \(\phi\) to a complex value \(r(\theta, \phi) \exp[i\beta(\theta, \phi)]\) with radial part \(r(\theta, \phi)\) and phase \(\beta(\theta, \phi)\). The components \(T_{jm}\) can consequently be mapped to spherical harmonics \(Y_{jm}\), see Chap. 5 of \([37]\) or Chap. 8 of \([38]\). Hence, an operator \(A\) acting on a single spin with spin number \(s\) can be represented by a unique spherical function \(f_A\) using the straightforward mapping (in this particular case \(J = \{0, 1, \ldots, 2s\}\))

\[
A = \sum_{j \in J} \sum_{m=-j}^j c_{jm} T_{jm} \iff f_A = \sum_{j \in J} \sum_{m=-j}^j c_{jm} Y_{jm},
\]

which translates an expansion of an operator (in terms of a tensor operator basis) into an expansion of a function (in terms of spherical harmonics).

In this work, we systematically generalize this approach to systems consisting of an arbitrary number of coupled spins. A particular focus will be the case of three coupled spins. In the general case of multiple spins, the set of irreducible tensor operators contains multiple elements with the same rank \(j\). Consequently, the direct mapping as in \([22]\) would not be bijective, as distinct operators would be mapped onto the same function. For instance, the tensor basis for a system consisting of two coupled spins contains three distinct tensors of rank \(j = 1\), see, e.g., \([3]\). The tensor operators \(T_j\) of rank \(j\) are not even uniquely determined if their multiplicity \(n_j\) is larger than one. The corresponding subspace \(\mathbb{1}_{n_j} \otimes B_{2j+1} = \mathbb{1}_{n_j} \otimes B_{2j+1}\) of the tensor operator space is decomposed into \(n_j\) blocks \(B_{2j+1}\) of dimension \(2j+1\) and allows for transformations of the form \(M \otimes \mathbb{1}_{2j+1}\) (cf. \([28]\)), where \(M\) is a non-singular \(n_j \times n_j\)-matrix and \(\mathbb{1}_q\) denotes an identity matrix of dimension \(q\). Therefore, the different tensor operators \(T_j\) with identical rank \(j\) can be mixed using linear combinations.

A first idea for a generalization would be to view a multi-spin system with spin numbers equal to \(s\) as a single spin with a higher spin \(s' > s\) and visualize it using the map of \([22]\). Even though this approach would meet the uniqueness of the map as stated in property (A), it would destroy invariance properties under rotation and conceal important physical features of the system contrary to our intentions in property (B). There were even doubts if a generalization is possible at all \([17]\).

We present a possibility to distinguish the representations for the tensors \(T_j^{(\ell)} \neq T_j^{(\ell')}\) by introducing additional labels \(\ell, \ell' \in L\). For suitably chosen labels, the
irreducible tensor operators $T_{j}^{(\ell)}$ can then be grouped into subsets

$$B(\ell) := \bigcup_{j \in J(\ell)} T_{j}^{(\ell)} = \bigcup_{j \in J(\ell)} \bigcup_{m=-j}^{j} T_{jm}^{(\ell)}$$

with respect to their label $\ell \in L$ such that each index set $J(\ell)$ never contains a rank $j$ more than once. This allows us to independently apply the approach of (4) to each subset $B(\ell)$. Thus, the main idea is to introduce multiple spherical functions $f_{A}^{(\ell)}$ for an operator $A$ and visualize them in parallel:

$$A = \sum_{\ell \in L} A^{(\ell)} \leftrightarrow \bigcup_{\ell \in L} f_{A}^{(\ell)} \quad \text{with} \quad (4a)$$

$$A^{(\ell)} = \sum_{j \in J(\ell)} \sum_{m=-j}^{j} c_{jm}^{(\ell)} T_{jm}^{(\ell)} \leftrightarrow f_{A}^{(\ell)} = \sum_{j \in J(\ell)} \sum_{m=-j}^{j} c_{jm}^{(\ell)} Y_{jm}. \quad (4b)$$

In the following, such a visualization of spin operators will be denoted as DROPS (discrete representation of operators for spin systems) visualization and we will refer to each individual visualization of a spherical function $f_{A}^{(\ell)}$ as a droplet. It is essential that all $2j+1$ components $T_{jm}^{(\ell)}$ of a tensor operator $T_{j}^{(\ell)}$ are contained in the same droplet in order to ensure the invariance properties under rotation due to property (B).

Finding a suitable choice for the labels $\ell \in L$ is sometimes called the problem of missing labels (see, e.g., [40–42] and p. 145 of [43]). Note that labels are not restricted to numbers. In a more general context one aims at finding a complete set of mutually commuting operators or a set of good quantum numbers (see, e.g., Sec. 10.4 and p. 473 of [29]) which enables the analysis of a quantum system in a complete and problem-adapted basis. Although our choice of labels will only be presented in Sec. III, we refer the reader to Fig. 1 to establish ideas: Our approach is applied to a system of three spins by visualizing a randomly-chosen operator $A$ using $|L|=11$ droplets. Here, the value $f(\theta, \phi) = r(\theta, \phi) \exp[i\beta(\theta, \phi)]$ of a spherical function at a point with coordinates $(\theta, \phi)$ is mapped to its distance $r(\theta, \phi)$ from the origin (forming the shape of the droplet) and its color corresponding to the phase $\beta(\theta, \phi)$ as defined by the color key in Fig. 1 (see, e.g., [18] for an alternative technique for visualizing spherical harmonics). We will detail further aspects of Fig. 1 in the course of our presentation.

Before presenting our choice of labels, we first elaborate on how the set $L$ of labels and their quantity $|L|$ is limited by what ranks $j$ (including their multiplicity) appear in a concrete system. These limitations also directly affect how the set of irreducible tensor operators is grouped into a complete orthonormal basis $B = \bigcup_{\ell \in L} B(\ell)$ as outlined by (3). One obtains for a coupled system of three spins that the ranks $j \in \{0, 1, 2, 3\}$ occur respectively with multiplicity five, nine, five, and one (see Table II as will be explained in Secs. III and IV). Therefore, the number $|L|$ of labels is restricted to $9 \leq |L| \leq 20$: The lower bound results from the maximal multiplicity of nine, and the upper bound is a consequence of the maximal number of distinguishable irreducible tensor operators as given by the sum of the multiplicities. Our choice of eleven different labels (and droplets) in Fig. 1 contains a few more labels than necessary but will yield further benefits, as explained below.

### III. LISA Tensor Operator Basis

Building on the discussion of how the set $L$ of labels induces a grouping of irreducible tensor operators, the specific choice of labels for the LISA tensor operator basis is introduced. We proceed in three steps and sort the irreducible tensor operators into non-overlapping classes: First, we divide them with respect to the number $g$ of spins involved (i.e. $g$-linearity). Second, we further split up the irreducible tensor operators with identical $g$ according to the set $G$ of involved spins with $|G| = g$. Third, the symmetry type $\tau$ under permutations of the set $G$ gives rise to a decomposition of the subspace of irreducible tensor operators with identical $G$. The permutations of a set of cardinality $g$ are known as the symmetric group $S_{g}$ [44–47]. The third step is suppressed for $g \leq 2$ as no rank $j$ occurs more than once for a given set $G$. In summary, a complete label is given by $(\ell) = (G, \tau)$ (if the number of spins is five or smaller as will be explained
in Sec. [VII]. We use the notations $T^{(l)}_j = T^I_1(\ell) = T^I_3(\tau)$ for a labeled irreducible tensor operator, where both $G$ and $\tau$ can be omitted at will. Next, we specify the labels (including the explicit form of $\tau$) for each $g$ while highlighting the case of three spins (see Table I).

For $g=0$, a single rank of zero appears in Table I. The corresponding label is given by Id (or $\emptyset$), and the droplet of the single irreducible tensor component $T^{(0)}_0$ is plotted in the center of the triangle in Fig. I. The three linear irreducible tensor operators of rank one acting on a single spin (i.e. $g=1$) are associated with the labels (and subsystems) $G \in \{\{1\}, \{2\}, \{3\}\}$ and are plotted at the vertices of the triangle in Fig. I. The droplets for bilinear tensor operators ($g=2$) are plotted at the edges of the triangle in Fig. I and contain the ranks $j \in \{0, 1, 2\}$ for each subsystem label $G \in \{\{1, 2\}, \{1, 3\}, \{2, 3\}\}$ (see Table I). The full structure of the labeling will emerge for trilinear operators with $g = 3$ and $G = \{1, 2, 3\}$. Here, the ranks $j = 1$ and $j = 2$ occur more than once (see Table I) and the symmetry types

$$\tau^{[3]}_1 := \begin{array}{c} 0 \\ 2 \\ 3 \end{array}, \quad \tau^{[3]}_2 := \begin{array}{c} 0 \\ 2 \\ 3 \end{array}, \quad \tau^{[3]}_3 := \begin{array}{c} 2 \\ 0 \\ 3 \end{array}, \quad \tau^{[3]}_4 := \begin{array}{c} 2 \\ 0 \\ 3 \end{array} $$

will be applied for a complete labeling which reflects the symmetries under permutations of the elements in $G$. Each $\tau_i^{[g]}$ is a standard Young tableau of size $g$ [44-47] which is a left-aligned arrangement of $g$ boxes where the number of boxes does not increase from one row to following ones and where each box contains a different number from a set $G$ such that the numbers are ordered strictly increasing from left to right and top to bottom. The standard Young tableaux $\tau^{[3]}_1$ and $\tau^{[3]}_2$ represent complete symmetrization and antisymmetrization, respectively. The four droplets for the three-linear operators are given above the triangle in Fig. I.

Note that the symmetry types are trivial for $g \in \{0, 1\}$. But it is worthwhile to discuss their explicit form for $g=2$ even though they are suppressed in the labels of Table I. The bilinear irreducible tensor operators of rank zero and two are automatically symmetric under spin permutations (i.e. have the symmetry type $\tau^{[2]}_1 := \begin{array}{c} 2 \\ 0 \\ 3 \end{array}$ for the case of $G = \{2, 3\}$), while the case of rank one is antisymmetric (i.e. $\tau^{[2]}_2 := \begin{array}{c} 0 \\ 2 \\ 3 \end{array}$).

In general, different symmetry types are mapped to different droplets. But bilinear operators are an exception where all symmetry types are combined in a single droplet (see Table I).

Evidently, the LISA tensor operator basis and the corresponding decomposition of the tensor space are based on methods perfected by Weyl [48-50] which relate the structures of the unitary group SU(2) and the symmetric group $S_n$, where $n$ denotes the total number of spins. But we symmetrize only with respect to spin permutations over tensors with defined linearity and subsystem (see Table I). Approaches which symmetrize over all tensors of fixed linearity (or even over the complete set of tensors) as in [51, 52] are more suitable for sets of indistinguishable spins (refer also to the discussion in Sec. VII). The corresponding decomposition of the tensor space has been analyzed before by [53, 54] (see also [55]) using different methods. We refer in this context also to the detailed work of Temme et al. [56-60], and references therein.

### Table I. Labels for the irreducible tensor operators corresponding to the LISA basis for three coupled spins

| # spins | subsystems | SU(2) | $S_g$ | labels |
|---------|------------|-------|-------|--------|
| 0       | $\emptyset$ | 0     | Id    | $\emptyset$ |
| 1       | {1}        | 1     | {1}   | \tau_1^{[3]} |
| 2       | {2}        | 1     | {2}   | \tau_2^{[3]} |
| 3       | {3}        | 1     | {3}   | \tau_3^{[3]} |
| 0       | {1,2}      | 0     | {1,2} | \tau_4^{[3]} |
| 0       | {1,3}      | 1     | {1,3} | \tau_5^{[3]} |
| 0       | {2,3}      | 2     | {2,3} | \tau_6^{[3]} |
| 0       | {1,2,3}    | 3     | {1,2,3} | \tau_7^{[3]} |

#### A. Phase and sign

Before we outline how to explicitly construct the irreducible tensor operators, we address the non-uniqueness of their phase and sign. The phase of a tensor component $T^{jm}_{\ell}$ is determined by observing the Condon-Shortley phase convention $T^{jm}_{\ell} = (-1)^m T^{jm}_{\ell}$ [36, 61], where $A^\dagger$ denotes the complex conjugate and transpose of a matrix $A$. Consequently, the tensor components $T^{jm}_{\ell}$ are defined up to an algebraic sign and the ones for $m = 0$ are hermitian. Moreover, visualizations of hermitian matrices feature only the colors red and cyan corresponding to phases of zero and $\pi$, respectively. More generally, only the two colors for the phases $\gamma$ and $\gamma + \pi$ appear if a matrix is hermitian up to a factor of $\exp(i\gamma)$. Although the choice of sign for each rank $j$ is completely arbitrary, the resulting visualizations can differ notably. Our choice for the LISA basis is motivated in Appendix A.
B. Iterative construction

We outline the explicit construction of the LISA tensor operator basis which is built up iteratively from the tensor operators $T_0$ and $T_1$ for one spin. For a general system consisting of $n$ spins with $n \leq 5$, the construction consists of three steps: (I) In the first step, the $g$-linear tensor operators of a $g$-spin system are constructed and symmetrized for all $g \in \{0, 1, \ldots, n\}$ such that they reflect both the symmetries of the unitary group SU(2) and the symmetric group $S_g$. (II) In the second step, the tensor operators are multiplied with suitable phase factors in order to comply with the phase and sign conventions detailed above. (III) Lastly, the tensor operators which have been constructed for a $g$-spin system are naturally embedded into the full $n$-spin system for each $g$-element subset of $\{1, \ldots, n\}$. The steps (II) and (III) are straightforward, and we provide further details for the first step which will be subdivided into two parts (Ia) and (Ib).

In part (Ia), the tensor operators $T_j(\tau_i^{[g-1]})$ for $g-1$ spins are combined with the tensor operator $T_1$ for one spin to iteratively build up tensor operators $T_j(\tau_i^{[g-1]}) \otimes T_1$ for $g$ spins. We determine the occurring tensor operators using the Clebsch-Gordan decomposition

$$T_j \otimes T_1 = T_{|j-1|} \oplus \cdots \oplus T_{j+1},$$

and their explicit form is given by the Clebsch-Gordan coefficients $[21, 26, 27]$ for which—in the case of Eq. (6)—simple closed formulas (cf. p. 635 in [26]) and tables exist (see p. 419 in [62]).

After completing part (Ia) the tensor operators observe the symmetry of SU(2) but not the one of the symmetric group $S_g$. In step (Ib), the symmetrization can be completed using one of two approaches. The first approach relies on explicit projection operators [44, 45, 47, 62, 64] which project onto subspaces with distinct symmetry type $\tau_i^{[g]}$ as given in Table I (for details refer to Appendix B). In the second approach, fractional parentage coefficients $[37, 65, 66]$ determine how the tensor operators are recombined into their permutation-symmetrized versions. Formulas for the fractional parentage coefficients are analyzed in the literature $[70, 73]$, but in this case one can conveniently rely on explicit tables from $[74]$. The explicit matrix form of the LISA basis is detailed in Appendix B. Before addressing the relation of the DROPS visualization to Wigner functions as well as the extension of our approach to an arbitrary number of spins, we provide explicit applications and examples for the LISA basis.

IV. VISUALIZATION OF TYPICAL OPERATORS IN THE LISA BASIS

A. Cartesian product operators

The LISA basis is in particular suitable for visualizing Cartesian product operators which form a widely-used orthogonal basis in spin physics [3]. For a single spin, the Cartesian product operators $I_\eta$ with $\eta \in \{x, y, z\}$ have been defined above. The one-spin operators $I_\eta$ are embedded in an $n$-spin system as $I_{\eta} := \bigotimes_{s=1}^{n} I_{a_s}$ where $a_s = \eta$ for $s = k$ and $a_s = 0$ otherwise; $I_0 := \{0, 0\}$. Cartesian product operators as $I_{2x}$, $2I_{1z}I_{3y}$ and $4I_{1z}I_{2x}I_{3y}$ have usually a prefactor of $2^{d-1}$ where $d$ denotes the number of single-spin operators. Explicit transformations between the LISA basis and the Cartesian product operators are given in Appendix B. A Cartesian product operator acts on a well-defined subsystem and can consequently be represented with very few droplets. Each of these droplets features only the colors red and cyan (see Fig. 2) as Cartesian product operators are hermitian. We discuss now the visualizations of the linear, bilinear, and trilinear cases (see Fig. 2).

The only droplet for a linear Cartesian product operator $I_{k\eta}$ on spins $k$ consists of two spheres which are colored red and cyan, while the red one corresponds to a positive sign and points into the direction of the axis $\eta \in \{x, y, z\}$ (see Fig. 2). More generally, the axis of the droplet for a Cartesian operator $a_x I_{kx} + a_y I_{ky} + a_z I_{kz}$ with $a_\eta \in \mathbb{R}$ is collinear and proportional to the vector $a_x \vec{x} + a_y \vec{y} + a_z \vec{z}$. This corresponds to a vector generalizing respectively a (magnetic) field vector for Hamiltonians and the Bloch vector for density matrices, which repre-

![FIG. 2. (Color online) Examples of characteristic droplets for Cartesian product operators. The red and cyan colors refer respectively to positive and negative values of the spherical functions. The droplets for 4I_{1y}I_{2x}I_{3z} differ from the ones for 4I_{1x}I_{2y}I_{3z} only in an inversion of color for the $\tau_3$-component.](image-url)
Bilinear Cartesian operators $2I_{kx}I_{ly}$ on spins $k$ and $l$ with $\eta_1 \neq \eta_2$ such as an anti-phase coherence operator $\vec{\eta}$ with $\eta_1 = x$ and $\eta_2 = z$ induce a droplet with a particular shape (see $2I_{kx}I_{ly}$ in Fig. 2) which is not common for atomic or molecular orbitals. This droplet consists of two bean-shaped lobes with colors red and cyan whose major axes are orthogonal to each other. The major axis of the red lobe is oriented in the direction $\vec{\eta} + \vec{\eta}'$, while the axis from the cyan lobe to the red one points in the direction $\vec{\eta} \times \vec{\eta}'$ given by the right-hand rule. The droplet transforms naturally under rotations, e.g., when both spins are simultaneously rotated by $\pi$ around the $x$-axis, the anti-phase operator $2I_{kx}I_{ly}$ is mapped to $-2I_{kx}I_{ly}$ corresponding to a droplet of the same shape but with inverted colors. More general shapes appear for the bilinear operators $2I_{kx}I_{ly}$ in the form of elongated shapes as well as antisymmetrically elongated shapes oriented along the $\eta$-direction for the trilinear operators $4I_{1\eta}I_{2\eta}I_{3\eta}$ (see Fig. 2).

B. Multiple quantum coherences

Operators $A_p$ of defined coherence order $p$ play an important role in NMR spectroscopy. They are invariant under global $z$-rotations up to a phase factor:

$$\exp(-i\alpha \sum_{k=1}^{n} I_{kz}) A_p \exp(i\alpha \sum_{k=1}^{n} I_{kz}) = A_p \exp(-i\alpha).$$

This property is nicely captured in the DROPS representation as detailed in Appendix C.1 where characteristic multiple-quantum terms for linear, bilinear and trilinear operators are displayed in the LISA basis.

C. Hamiltonians

Hamiltonians can also be conveniently visualized using the DROPS representation. The cases of linear and bilinear terms of the Hamiltonian mirror the properties of Cartesian product operators as discussed above. We analyze the shape of the droplets for bilinear coupling Hamiltonians

$$\mathcal{H}_{\text{bil}} = 2\pi \sum_{kl} c_{kl}(aI_{kz}I_{lz} + aI_{ky}I_{ly} + bI_{kx}I_{lx}).$$

representing characteristic spin-spin interactions (see, e.g., Fig. 3). The cases $a = 0$ and $b = 1$ correspond to the Ising-ZZ (or Heisenberg-Ising) model $\mathcal{H}_{\text{iso}}$ or longitudinal coupling $\mathcal{H}_{\text{long}}$, which is also known as weak or longitudinal coupling and is represented by a longitudinally elongated droplet. The Heisenberg-XXX model $\mathcal{H}_{\text{iso}}$ with $a = 1$ and $b = 1$ is also denoted as strong or isotropic coupling $\mathcal{H}_{\text{iso}}$ and results in an isotropic droplet of spherical shape. For $a = 1$ and $b = 0$, we obtain the Heisenberg-XX model $\mathcal{H}_{\text{plan}}$, which is also known as the planar coupling and is represented as a planar disc-shaped droplet in the $x$-$y$ plane. The case $a = 0$ and $b = -2$ corresponds to a dipolar coupling $\mathcal{H}_{\text{dip}}$. More general coupling terms can also be visualized.

Examples as anisotropic Heisenberg XYZ or effective trilinear coupling terms could be used in visualizations of multi-spin systems such as the well-known Kitaev honeycomb lattice. Hence, even for very large spin systems the LISA basis (presented here explicitly for three spins) can be used to visualize Hamiltonians with at most trilinear terms.

FIG. 3. (Color online) Non-trivial droplets for coupling Hamiltonians: Ising-ZZ (or Heisenberg-Ising) model $\mathcal{H}_{\text{iso}}$, Heisenberg-XXX model $\mathcal{H}_{\text{long}}$, Heisenberg-XX model $\mathcal{H}_{\text{plan}}$, and dipolar coupling $\mathcal{H}_{\text{dip}}$ as detailed in the main text.

FIG. 4. (Color online) Examples of an NMR pulse sequence creating triple-quantum coherences which consists of a $\pi/2$ pulse (with phase $x$) followed by a delay and a second $\pi/2$ pulse (with phase $y$). The upper left panel shows (the traceless part of) the initial density operator $\rho(t_0) = I_{1z} + I_{2z} + I_{3z}$. The final density operator $\rho(t_3) = 4I_{1y}I_{2z}I_{3x} + 4I_{1z}I_{2y}I_{3x} + 4I_{1x}I_{2z}I_{3y}$ is given in the upper right panel. The effective Hamiltonian $\mathcal{H}_{\text{eff}}$ and the effective propagator $U_{\text{eff}}$ for the pulse sequence are shown in the lower left and lower right panels, respectively.
D. Time evolution

We provide an example visualizing density operators, Hamiltonians, and unitary transformations for a non-trivial pulse sequence in NMR spectroscopy (see Fig. 4); full details are given in Appendix C. The pulse sequence consists of two $\pi/2$ pulses separated by a delay, which is designed to excite triple-quantum coherence starting from the thermal density operator in the high-temperature limit $\mathcal{R}$. This highlights crucial information for the system and provides a better understanding of the corresponding time evolution. Note that the unitary transformation is not hermitian and therefore in general requires more colors.

E. Pure quantum states

In the field of quantum information $\mathcal{R}$, (in addition to mixed states) pure states and their entanglement measures are of particular interest. Density matrices for four mixed states, pure states and their entanglement measures are shown in Fig. 5. As an example for an entanglement measure for two spins, consider the concurrence $C = \sqrt{1 - b_1^2}$, which can be expressed as a function of $b_1$ (see also [81, p. 168] or [82, p. 50]). Hence, it can also be obtained from the maximal radius $r_1$ as $C = \sqrt{1 - 2r_1^2/3}$. The value of the concurrence $C$ for the pure state $|00\rangle$ and $|\Phi^+\rangle$ of Fig. 5 is zero and one, respectively.

V. GENERALIZED WIGNER REPRESENTATION

In this section we describe how the DROPS representation can be interpreted as a generalized Wigner function. Recall that the Wigner quasi-probability distribution (or Wigner function for short) provides an equivalent phase-space formulation for the standard Hilbert-space framework of quantum mechanics and mimics the phase-space probability distribution in classical physics $\mathcal{R}$ [83, 92]. It is formally not a probability distribution as negative values may appear. Although Wigner functions were originally developed for infinite-dimensional quantum systems with continuous degrees of freedom, they can be extended to finite-dimensional quantum systems following the work of Stratonovich [93, see, e.g., 14, 96]. For the finite-dimensional case, a different perspective is provided by a comprehensive theory of square-integrable functions on compact Lie groups (e.g., functions on the sphere for SU(2)) introduced in the seminal work of Peter and Weyl [97] (see [35]). The case of non-compact Lie groups is still an area of active research [98, 99]. However, a relatively simple example of a non-compact Lie group is the symplectic group which is widely studied in quantum optics [100] in the context of infinite-dimensional systems.

Returning to Wigner functions of finite-dimensional systems, the case of one spin was detailed in [15, 101], where each tensor operator is mapped to a unique (square-integrable) function on the sphere along the lines of Eq. (2). As discussed in [17, 19], the case of general coupled spin systems has not been solved so far. Therefore, it is important to point out that the approach introduced here (c.f. Eq. (4)) in fact provides a solution to this open problem. Rather than mapping each operator $A$ to a single function on a sphere (see Eq. (2)), it is mapped to a set $\{f_A^{(\ell)}(\theta, \phi)\}$ with $\ell \in L$ of functions on multiple spheres. This set satisfies conditions generalizing the ones of Stratonovich [93, 98, 99] and hence can be interpreted as a generalized Wigner function:

**Proposition 1** We assume that the DROPS representation of Eq. (4) observes the Condon-Shortley phase convention and that the functions $f_A^{(\ell)}(\theta, \phi)$ are correctly normalized. The following conditions are fulfilled:
(a) Linearity: \( A \rightarrow f_A^{(\ell)}(\theta, \phi) \) is linear for each \( \ell \in L \).
(b) Reality: \( f_A^{(\ell)}(\theta, \phi) = f_A^{(\ell)}(\theta, \phi)^* \) holds for each \( \ell \in L \).
(c) Norm: \( \sum_{\ell \in L} \int_{S^2} f_A^{(\ell)}(\theta, \phi) f_A^{(\ell)}(\theta, \phi) d\mu = \text{Tr}(A) \).
(d) Covariance: \( f_{R(A)}^{(\ell)}(\theta, \phi) = f_A^{(\ell)}(R^{-1}(\theta, \phi)) \) holds for each \( \ell \in L \) and all non-selective rotations \( R \in SU(2) \).
(e) Trace: \( \sum_{\ell \in L} \int_{S^2} f_A^{(\ell)}(\theta, \phi) f_B^{(\ell)}(\theta, \phi) d\mu = \text{Tr}(AB) \).

The (inversely) rotated point on the sphere has the coordinates \( (\theta', \phi') := (\pi - \theta, -\phi) \). The corresponding action \( R(A) = UAU^{-1} \) describes a non-selective spin conjugation where the unitary matrix \( U \) is a non-selective rotation operator (acting on complex column vectors, in particular on quantum-mechanical Hilbert-space vectors). The straightforward proof of Prop. \( \text{II} \) is given in Appendix \[.\] Based on these criteria, it is possible to describe the state of spin systems using droplets, i.e., sets of linear combinations of spherical harmonics. In particular, for a given set of droplets, the expectation value of an operator \( A \) can be calculated based on (c), where \( B \) is replaced by the density operator \( \rho = \langle A \rangle = \text{Tr}(A \rho) \). The relations of Prop. \( \text{II} \) are obtained from the Stratonovich conditions in a straightforward fashion by simply applying them to each droplet individually (for (a), (b), and (d)) or by summing over all the droplets (for (c) and (e)). Note that in contrast to the original Stratonovich conditions, in (c) the function \( f_B^{(\ell)}(\theta, \phi) \) appears. This is a direct consequence of (c) if \( B \) is replaced by the identity operator. It ensures that unwanted contributions from functions \( f_A^{(\ell)}(\theta, \phi) \) with traceless \( A \) are eliminated. Consider for example the traceless basis operator \( A = T^{(1,2)}_{00} \) for two spins which is mapped to \( Y_{00}(\theta, \phi) := 1/\sqrt{4\pi} \) on one droplet. Without the presence of \( f_B^{(\ell)}(\theta, \phi) \), (c) would result in \( 2\sqrt{\pi} = \int_{S^2} Y_{00}(\theta, \phi) d\mu = \text{Tr}(T^{(1,2)}_{00}) = 0 \), which is a contradiction. For the special case of a single spin, the only droplet with rank \( j = 0 \) corresponds to the identity operator and the generalized criterion (c) given above can be reduced to the original form \( \int_{S^2} f_A(\theta, \phi) d\mu = \text{Tr}(A) \) of Stratonovich. As the integrals \( \int_{S^2} Y_{jm}(\theta, \phi) d\mu \) vanish for \( j > 0 \) anyway, the only terms contributing to (c) come from the cases with \( j = 0 \) corresponding to the spherical harmonic \( Y_{00}(\theta, \phi) \). In the case of the LISA basis, the identity operator is mapped to a unique droplet as the basis operators are also characterized by particle number. Then, the sum in (c) reduces to a single term with the integral corresponding to this particular droplet while ignoring all the other ones.

VI. ARBITRARY NUMBER OF SPINS

We resume discussing the choice of labels \( \ell \in L \) which provides a partition of the irreducible tensor operators \( T_j^{(\ell)} \) into subsets never containing tensors operators of rank \( j \) more than once (see \[\text{III}\]). This partition induces also a decomposition into different droplets for the DROPS representation. Recall that the number \( |L| \) of droplets for three spins is bounded by \( 9 \leq |L| \leq 20 \). Bounds for up to twelve spins are given in Table \[\text{II}\].

In the particular case of the LISA tensor operator basis, the irreducible tensor operators \( T_j^{(\ell)} \) of rank \( j \) are divided according to the number \( g \) of spins involved, the set \( G \) of involved spins, and the symmetry type \( \tau \) under permutations of the set \( G \). All possible combinations of \( j \), \( G \), and \( \tau \) for up to three spins are shown in Table \[\text{II}\] where the notation was simplified by suppressing some trivial symmetry types. Below, we will explain the labels of Table \[\text{II}\] for an arbitrary number of spins and provide a general method for computing all possible combinations of rank \( j \) and symmetry type \( \tau \) reflecting both the symmetries of the unitary group \( SU(2) \) and the symmetric group \( S_g \). This also determines the number of droplets for the LISA basis as shown in Table \[\text{II}\].

We infer from Table \[\text{II}\] that the number of droplets is significantly smaller than the dimension \( 4^n \) of the operator space for \( n \) spins. Moreover, the LISA basis uses more droplets than strictly necessary. But meaningful labels are essential as even the minimum number of droplets grows very fast. Therefore, each droplet of the LISA basis has a unique symmetry type \( \tau \) except for bilinear operators, where the two occurring symmetry types (e.g., \( \tau_1^{(2)} \) and \( \tau_2^{(2)} \)) are combined as no \( j \)-value appears more than once. Based on the values in Table \[\text{II}\] we consider the LISA basis as an efficient and informative visualization for a moderate number of spins. In the following, we first present a method for computing the minimum and maximal number of droplets for a DROPS representation. Secondly, we describe the explicit form of the labels for the LISA basis and thereby determine the corresponding number of droplets.
TABLE III. We provide for each appearing $j$-value the multiplicities $n_j$ for the $n$-linear operators in an $n$-spin system. 

| $n$ | $j$ | $n_j$ | $\bar{n}_j$ | $n$ | $j$ | $n_j$ | $\bar{n}_j$ |
|-----|-----|-------|------------|-----|-----|-------|------------|
| 0   | 0   | 1     | 5          | 0   | 1   | 1     | 42         |
| 1   | 0   | 1     | 5          | 1   | 1   | 2     | 7          |
| 2   | 0   | 1     | 3          | 2   | 1   | 4     | 17         |
| 3   | 0   | 1     | 6          | 3   | 1   | 5     | 9          |
| 4   | 1   | 1     | 1          | 5   | 1   | 7     | 1          |

The multiplicity $\bar{n}_j$ represents the number of all rank-$j$ tensors considering all possible subsets $G$ of the $n$ spins (including the $n$-linear operators). The example values discussed in the text are highlighted.

A. The minimum and maximal number of droplets

Recall that the set of infinitesimal rotation operators (or equivalently the Lie algebra $\mathfrak{su}(2)$) acts on the irreducible tensor operators $T_0$ and $T_1$ of a single spin via $[\mathbf{1}]$. This means that the unitary group $SU(2)$ (and its Lie algebra $\mathfrak{su}(2)$) acts for a single spin non-trivially on a three-dimensional (complex) space $\mathbb{C}^3$ via its three-dimensional irreducible representation $\varphi_1$. Here, an irreducible representation $\varphi_j$ of $SU(2)$ denotes in the language of representation theory of $SU(2)$ an action of $SU(2)$ on the abstract space $\mathbb{C}^{j+1}$ (for which the irreducible tensor operator $T_j$ provides an explicit model) and maps an element $g \in SU(2)$ to a $(2j+1)\times(2j+1)$-matrix $\varphi_j(g)$.

For multiple spins, a simultaneous action of $SU(2)$ on (e.g.) all $n$-linear operators of an $n$-spin system arises and the tensor product representation $\varphi_1 \otimes \cdots \otimes \varphi_1$ of $SU(2)$ naturally acts on the $n$-fold tensor product $\otimes^n \mathbb{C}^3 := \mathbb{C}^3 \otimes \cdots \otimes \mathbb{C}^3$ of a three-dimensional (complex) space. The tensor product representation $\varphi_1 \otimes \cdots \otimes \varphi_1$ is known as the $n$th tensor power of $\varphi_1$ and decomposes into a sum of representations $\varphi_j$ with multiplicities $n_j$ by means of the well-known technique of Eq. (6) (for general methods, cf. pp. 424–429 of [102] or pp. 135–142 of [103]). The explicit values for $n_j$ in Table III have been computed using the computer algebra system MAGMA [104]. The corresponding multiplicities $\bar{n}_j$ for the full $n$-spin system are obtained by summing the multiplicities $n_j$ for $g$-linear operators with $g \in \{0, \ldots, n\}$ which have to be multiplied with the number $\binom{n}{g} = n!/[g!(n-g)!]$ of possible sets $\mathbb{G} \subseteq \{1, \ldots, n\}$ of $|\mathbb{G}|=g$ spins. For example consider the number $\bar{n}_j$ for $n = 3$ spins with rank $j = 2$. Here, for the set $G = \{1, 2, 3\}$ we find $n_2 = 2$ and for each of the sets $\{1, 2\}$, $\{1, 3\}$, and $\{2, 3\}$ we have $n_2 = 1$ which can be inferred from the corresponding case of two spins. None of the subsets $G \subseteq \{\emptyset, \{1\}, \{2\}, \{3\}\}$ yields an operator with rank $j = 2$ and the corresponding $n_2$-values are zero. Thus, $\bar{n}_j = 1 \times 2 + 3 \times 1 + 3 \times 0 + 1 \times 0 = 5$. For a given $n$, the minimum and maximal number of droplets in Table III are now given by the maximum of the multiplicities $\bar{n}_j$ for all ranks $j$ and the sum $\sum_j \bar{n}_j$, respectively.

B. All combinations of symmetry types

We determine all possible combinations of rank $j$ and symmetry type $\tau$ by refining our symmetry analysis of $g$-linear operators. Before, we identified the symmetries of $g$-linear operators for rank $j$ which are modeled by a $g$-fold tensor product $\otimes^g \mathbb{C}^3$ and acted on by the unitary group $SU(2)$. We extend this action on $\otimes^g \mathbb{C}^3$ to an action of the direct product $SU(2) \times S_g$, where the symmetric group $S_g$ acts by permuting spins from a set $G \subseteq \{1, \ldots, n\}$ with $|G| = g$.

The corresponding symmetry analysis for the number of spins $n \in \{1, \ldots, 8\}$ is summarized in Table IV, where we consider $g$-linear operators with $g = n$ in an $n$-spin system. Table IV states all possible combinations of rank $j$ and partition $\lambda$ from which the corresponding symmetry types $\tau$ can be easily determined. A partition $\lambda := [\lambda_1, \ldots, \lambda_{|\lambda|}]$ of length $\kappa(\lambda)$ and de-

TABLE IV. All combinations of partitions $\lambda$ and ranks $j$ for $n$-linear operators in an $n$-spin system with $1 \leq n \leq 8$. The third column shows the number of symmetry types $\tau$ for each $\lambda$.

| $n$ | $\lambda$ | $\#\tau$ | $j$ |
|-----|-----------|----------|-----|
| 1   | [1]       | 7        | 7   |
| 2   | [1,1]     | 6        | 1, 2, 3, 4 |
| 3   | [1,1,1]   | 15       | 1, 2, 3, 4 |
| 4   | [1,1,1,1] | 21       | 1, 2, 3, 4 |
| 5   | [1,1,1,1,1] | 21    | 1, 2, 3, 4 |
| 6   | [1,1,1,1,1,1] | 21   | 1, 2, 3, 4 |
| 7   | [1,1,1,1,1,1,1] | 21  | 1, 2, 3, 4 |
| 8   | [1,1,1,1,1,1,1,1] | 21 | 1, 2, 3, 4 |
is illustrated in Fig. 5 and can be computed as the number of different symmetry types \( \tau \) for each partition \( \lambda \) is given in the third column \( (\#\tau) \) of Table IV. The results for \( n \in \{1, 2, 3, 4\} \) and some partial results for \( n \in \{5, 6\} \) can also be found in Table 12 of [102] (cf. Table 2 on p. 294 of [67]).

Combinations of \( \lambda \) and \( j \) which appear more than once are highlighted in Table IV. The case of \( \lambda = [4, 2] \) and \( j = 2 \) for \( n = 6 \) has been known at least since [106] (see also [43, 108–110]). If no combination of \( \lambda \) and \( j \) appears more than once (as for \( 1 \leq n \leq 5 \)), the LISA basis is uniquely defined without any additional labels. Additional labels are required in the general case, but ad-hoc labels as in [106] are usually sufficient. The resulting symmetry types \( \tau \) for a partition \( \lambda \) are given by the standard Young tableaux of shape \( \lambda \) [44, 47].

The employed method for the computation of Table IV is a combination of the Schur-Weyl duality [107] and a technique known as plethysm. The results in Table IV were obtained using the computer algebra system MAGMA [104] and details are given in Appendix E.

VII. DISCUSSION

Before concluding, we discuss alternative DROPS visualizations which complement the LISA representation. A suitable choice can reflect the considered system and application. One possibility arises from partitioning the tensor operators from the Clebsch-Gordan decomposition of Eq. (6) into droplets without symmetrizing with respect to spin permutations as in the LISA basis. As before, one has to ensure that no rank \( j \) appears more than once in any droplet. Many different partitions are possible, and applying the Clebsch-Gordan decomposition recursively could provide a natural partition.

Indistinguishable spins utilize only a proper subspace of all tensor operators. The corresponding symmetry-adapted tensor basis can be obtained by symmetrizing tensor operators with respect to the relevant spin permutations. This usually reduces the number of droplets. In particular, one can discard all droplets with incompatible symmetry types. For example, the Hamiltonian and the density operator in a three-spin system of type \( I_3 S \) are both invariant with respect to permutations of the first two spins. Consequently, the LISA basis can be restricted to a 40-dimensional space consisting of the twelve irreducible tensor operators \( T_0, \frac{1}{2}(T_1^{(1)} + T_1^{(2)}), T_1, T_0^{(1,2)} = T_2, \frac{1}{2}(T_0^{(1,3)} + T_0^{(2,3)}), \frac{1}{2}(T_1^{(1,3)} + T_1^{(2,3)}), \frac{1}{2}(T_0^{(1,3)} + T_0^{(2,3)}), T_1(\tau_1^3), T_3(\tau_1^3), T_1(\tau_2^3), \) and \( T_2(\tau_2^3) \). This allows us to reduce the number of droplets from eleven to seven. For a three-spin system which is totally symmetric with respect to spin permutations, the LISA basis can be limited to a 20-dimensional space spanned by the tensor components of the six irreducible tensor operators \( T_0^0, \frac{1}{2}(T_1^{(1)} + T_1^{(2)} + T_1^{(3)}), \frac{1}{4}(T_0^{(1+2)} + T_0^{(1,3)} + T_0^{(2,3)}), \frac{1}{3}(T_2^{(1+2)} + T_2^{(1,3)} + T_2^{(2,3)}) \), \( T_1(\tau_1^3) \), and \( T_3(\tau_1^3) \). Thus, one obtains four droplets.

One further variant of the DROPS visualization is based on multipole tensor operators of the density matrices for the pure states of Fig. 5.
VIII. CONCLUSION

We introduced a general approach for representing arbitrary operators by a finite set of functions. Their properties make this representation particularly appealing and useful for the visualization of important quantum mechanical concepts and properties, which are conventionally represented by abstract operators or plain matrices. There are many possible bases on which such a mapping between operators and sets of functions can be based. Here we focused on the LISA basis which transforms naturally under non-selective spin rotations as well as spin permutations and which is particularly suitable for distinguishable spins. However, depending on the application, other bases can be more appropriate. It is noteworthy that the DROPS visualization can be seen as a generalization of the Bloch vector representation of simple two-level quantum systems, such as uncoupled spin 1/2 particles. On the other hand, the DROPS method can also be seen as a natural (albeit not obvious) generalization of Wigner functions that have been studied extensively for distinguishable spins. Although we focussed on the relatively simple but non-trivial example of three coupled spins 1/2, our method can be applied to more than three spins and is also not limited to spins 1/2.

We illustrated applications which benefit from the DROPS approach, such as the visualization of mixed quantum states. It can also be used to represent the density matrix of pure quantum states with or without entanglement. Furthermore, the DROPS representation can be applied to arbitrary operators, including Hamilton operators and time evolution operators. This approach is also well suited to show the time evolution of quantum mechanical operators as animations, rather than static figures. The DROPS visualization lends itself to building intuition about the dynamics of coupled spins and is expected to become a valuable tool both in education and research. Potential applications range from theoretical and experimental quantum information theory, where quantum bits (corresponding to spins 1/2) can be realized by trapped ions, quantum dots, superconducting circuits, and spin systems, to electron and nuclear magnetic resonance applications in physics, chemistry, biology, and medicine. A Mathematica package [117] and a mobile application software [118] are made available so that readers may apply the DROPS mapping to new areas and interactively explore the DROPS visualization of coupled spin dynamics in real time.

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Appendix A: Motivation for the choice of signs in the LISA basis

The phase of the irreducible tensor operators for the LISA basis is fixed by the Condon-Shortley convention up to a sign. The specific choice of signs made in the LISA basis will be presented in Appendix [133] during the construction of the LISA basis via projectors (see, e.g., Table [1]). Here, we present desirable properties which motivate this specific choice of signs:

The droplet of the identity operator has a positive value (i.e. it is red in Fig. 2). The droplets of the linear Cartesian operators \( I_{\eta_1} \) with \( \eta \in \{x, y, z\} \) for each spin \( k \) are consistent with its Bloch vector representation, i.e., the positive lobe in the LISA visualization of \( I_{\eta_1} \) is pointing in the \( \eta \)-direction (see Fig. 2).

Characteristic coupling Hamiltonians, such as longitudinal and planar couplings have elongated and disc-shaped droplets (see last column of Fig. 6). For spins \( k \) and \( l \), different shapes appear if \( T_0^{(k,l)} \) and \( T_{2}^{(k,l)} \) have different relative signs (Fig. 7). If the sign of \( T_{1}^{(k,l)} \) is negative, the positive lobe of the droplet representing \( 2I_{\eta_1}I_{\eta_2} \) with \( \eta_1 \neq \eta_2 \) (refer to case c) in Fig. 7 is displaced in the \( \eta_3 \)-direction with respect to the center of the droplet, where \( \eta_3 = \eta_1 \times \eta_2 \) is given by the right hand rule. The droplet for the fully symmetric Cartesian tensor \( 4I_{\eta_1}I_{\eta_2}I_{\eta_3} \) has an elongated shape and its positive lobe points in the \( \eta \) direction (see Fig. 2).

FIG. 7. (Color online) Different choices \((a_0, a_1, a_2)\) for the signs of the bilinear tensors \( T_0^{(k,l)} \), \( T_{1}^{(k,l)} \), and \( T_{2}^{(k,l)} \) with rank 0, 1, and 2 result in different visualizations for the operators \( 2I_{kz}I_{lz} \) in a), \( 2I_{kz}I_{lz} + 2I_{kx}I_{lz} \) in b), and \( 2I_{kz}I_{lz} \) in c). The red and the cyan color refers to the positive and the negative value of the droplet function \( f^{(k,l)}(A) \), respectively. The signs for the LISA basis are given in the last column which is highlighted by a dashed rectangle.
Appendix B: Construction of the LISA basis via projectors

We provide here the details for the iterative construction of the LISA basis which is applicable to general spin systems with \( n \leq 5 \) spins and is based on explicit projection operators.

1. The symmetric group and the standard Young tableaux

We start by recalling some basic notation, see, e.g., [44][47]. The \( q! \) permutations of a set \( G = \{1, \ldots, g\} \) of cardinality \( g \) are known as the symmetric group \( S_g \). They form a finite group whose group multiplication is defined by the composition \( (\sigma \circ \sigma')(p) := (\sigma_2 \circ \sigma_1)(p) = \sigma_2(\sigma_1(p)) \) for \( \sigma_1, \sigma_2 \in S_g \) and \( p \in G \). An element \( \sigma \in S_g \) maps \( p \in G \) to \( \sigma(p) \in G \) such that \( \sigma(p_1) \neq \sigma(p_2) \) for \( p_1 \neq p_2 \). Permutations can be compactly specified as products of disjoint cycles. A cycle \( c := (c_1 \cdots c_{|c|}) \equiv (c_1, \ldots, c_{|c|}) \) of length \( |c| \) (where \( c_p \in G \) and \( c_p \neq c_q \) for \( p \neq q \)) represents a permutation \( \sigma \in S_g \) where \( \sigma(c_p) = c_{p+1} \) for \( p < |c| \) and \( \sigma(c_{|c|}) = c_1 \). Two cycles \( c \) and \( \hat{c} \) are disjoint if \( c_p \neq \hat{c}_q \) for all \( p \) and \( q \). A transposition \((p, q)\) with \( p \neq q \) is a cycle of length two which permutes \( p \) and \( q \). Note that the symmetric group acts naturally on the set of Cartesian product operators (for notation refer to Sec. 11A) by permuting particles (and their labels), e.g., \( \sigma(I_{1x}I_{2y}I_{3z}) = I_{\sigma(1)x}I_{\sigma(2)y}I_{\sigma(3)z} = I_{1x}I_{y}I_{z} = I_{1y}I_{y}I_{z} = I_{1y}I_{z}I_{z} \) for \( \sigma = (123) \). This is equivalent to setting \( \sigma(v_1 \otimes \cdots \otimes v_g) := v_{\sigma^{-1}(1)} \otimes \cdots \otimes v_{\sigma^{-1}(g)} \) for \( g \) spins with \( v_p \in \{I_x, I_y, I_z\} \).

Recall that a standard Young tableau of size \( g \) is a left-aligned arrangement of \( g \) boxes where the number of boxes does not increase from one row to following rows and where each box contains a different number from the set \( G \) such that the numbers are ordered strictly increasing from left to right and top to bottom. For every standard Young tableau \( \tau \), one introduces a parameter \( \lambda = \lambda(\tau) = [\lambda_1, \ldots, \lambda_\kappa(\lambda)] \) of length \( \kappa(\lambda) \) where the positive integers \( \lambda_p \) with \( \lambda_p \geq \lambda_{p+1} \) are equal to the number of boxes in row \( p \) of \( \tau \) and \( \kappa(\lambda) \) agrees with the number of rows of \( \tau \). The number of columns of \( \tau \) is equal to \( \lambda_1 \). Also, the filling pattern \( w(\tau) = [w(\tau)_1, \ldots, w(\tau)_g] \) of \( \tau \) consists of the entries of \( \tau \) which are strung together from left to right in each row and from top to bottom for all rows. It is convenient to introduce a total order on the set of standard Young tableaux of size \( g \) where \( \tau < \tilde{\tau} \) if either \( \lambda(\tau) > \lambda(\tilde{\tau}) \) or if \( \lambda(\tau) = \lambda(\tilde{\tau}) \) and \( w(\tau) < w(\tilde{\tau}) \). Here, we imply that \( a = [a_1, \ldots, a_{|a|}] < b = [b_1, \ldots, b_{|b|}] \) if there exists an index \( q \) such that \( a_p = b_p \) for \( p < q \) and \( a_q < b_q \) while setting \( a_p := 0 \) for \( p > |a| \) and \( b_p := 0 \) for \( p > |b| \). This total order for standard Young tableaux is reflected by the subscript \( p \) in the notation \( \tau_p^{[a]} \) introduced in Eq. (3), e.g.,

\[
\tau_1^{[3]} = \begin{pmatrix} \ast & \ast & \ast \\ \ast & \ast & \ast \\ \ast & \ast & \ast \end{pmatrix} < \tau_2^{[3]} = \begin{pmatrix} \ast & \ast & \ast \\ \ast & \ast & \ast \\ \ast & \ast & \ast \end{pmatrix} < \tau_3^{[3]} = \begin{pmatrix} \ast & \ast & \ast \\ \ast & \ast & \ast \\ \ast & \ast & \ast \end{pmatrix} < \tau_4^{[3]} = \begin{pmatrix} \ast & \ast & \ast \\ \ast & \ast & \ast \\ \ast & \ast & \ast \end{pmatrix} \quad (B1)
\]

2. Young symmetrizers and projectors

We detail now the construction for Young symmetrizers and the corresponding projectors with symmetry type \( \tau \). Let \( \{\tau_g\} \) denote the set of all (formal) real linear combinations of permutations \( \sigma \in S_g \). This means that every element \( x \in \mathbb{R}[S_g] \) can be decomposed as \( x = \sum_{\sigma \in S_g} x_\sigma \sigma \) with coefficients \( x_\sigma \in \mathbb{R} \). Given two elements \( x, y \in \mathbb{R}[S_g] \), the sum in \( \mathbb{R}[S_g] \) is naturally defined as \( x + y = \sum_{\sigma \in S_g} (x_\sigma + y_\sigma) \sigma \) and the product is given by \( xy = \sum_{\sigma, \delta \in S_g} (x_\sigma y_\delta) \sigma \delta \).

Given a standard Young tableau \( \tau \), let \( R(\tau, p) \) [resp. \( C(\tau, q) \)] denote the set of all entries in the \( p \)th row [resp. \( q \)th column] of \( \tau \). Let \( S_M \) denote the permutations of a set \( M \) and let us introduce the permutations \( S_R(\tau, p) \) of the elements \( R(\tau, p) \) in the \( p \)th row of the standard Young tableau \( \tau \) where \( 1 \leq p \leq \kappa(\tau) \). In the example of \( \tau = \tau_3^{[3]} \), one obtains \( \tau(1) = \{1, 3\} \) and \( \tau(2) = \{2\} \) as well as \( S_R(\tau, 1) = \{e, (1,3)\} \) and \( S_R(\tau, 2) = \{e\} \) where \( e \) denotes the identity permutation. The set \( S_R(\tau) \) of row-wise permutations is now defined as \( S_R(\tau) = S_R(\tau, 1) \times \cdots \times S_R(\tau, \kappa(\tau)) = \{\prod_{p=1}^{\kappa(\tau)} \sigma_p\} \) for all possibilities of \( \sigma_p \in S_R(\tau, p) \). Note that the order in the product is irrelevant as the different permutations act on non-overlapping subsets of \( G = \{1, \ldots, g\} \). For \( \tau = \tau_3^{[3]} \), we have \( R(\tau) = \{e, (1,3)\} \). Similarly, we obtain the set of column-wise permutations \( C(\tau) := S_C(\tau, 1) \times \cdots \times S_C(\tau, \kappa(\lambda)) \) where \( \lambda = \lambda(\tau) \). One introduces \( H_\tau := \sum_{\sigma \in S_R(\tau)} \sigma \) and \( V_\tau := \sum_{\sigma \in C(\tau)} (-1)^{|\sigma|} \sigma \) where \( |\sigma| \) denotes the minimal number of transpositions \((p, q)\) necessary to write \( \sigma \) as product thereof. Finally, we can write the Young symmetrizer for \( \tau \) as the product \( e_\tau := f_\tau H_\tau V_\tau \) where the normalization factor \( f_\tau \in \mathbb{R} \) is equal to the number of standard Young tableaux of shape \( \tau \) divided by \( g! \) which ensures that \( e_\tau e_\tau = e_\tau \). For instance, the Young symmetrizers for the standard Young tableaux of Eq. (3) are

\[
e_1^{[3]} = e_{\tau_3^{[3]}} = \frac{1}{6}[e(12)(13) + (23)(123) + (132)], \quad (B2a)
\]
\[
e_2^{[3]} = e_{\tau_3^{[3]}} = \frac{1}{6}[e(2)(13) - (12)(13)], \quad (B2b)
\]
\[
e_3^{[3]} = e_{\tau_3^{[3]}} = \frac{1}{6}[e(12)(13) - (123)], \quad (B2c)
\]
\[
e_4^{[3]} = e_{\tau_3^{[3]}} = \frac{1}{6}[e(12)(13) - (23)(123) + (132)], \quad (B2d)
\]
where \( e \) denotes the identity in \( S_g \) and one directly verifies that \( e_1^{[3]} + e_2^{[3]} + e_3^{[3]} + e_4^{[3]} = e \). The Young symmetrizer can be computed (e.g.) for Eq. (B2c) by multiplying \( f_\tau = 1/3 \), \( H_\tau = e + (13) \), and \( V_\tau = e - (12) \) where \( \tau = \tau_3^{[3]} \).
We determine now projectors $P^3_p$ (i.e. $P^3_p P^3_p = P^3_p$) which can be interpreted as orthogonalized versions of the operators $e^3_p$ in Eq. (B2). These projectors $P_p = P^3_p$ model the symmetries for a standard Young tableau $\tau = \tau^3_p$ and will be used to identify tensor operators $T_j$ which are left invariant under their action, i.e. $P_p T_{jm} = T_{jm}$ for all $m \in \{-j, \ldots, j\}$. Our approach for determining the projectors $P^3_p$ is similar to the one detailed on pp. 114–124 of [109] and the formulas can be inferred from the matrices of Young’s seminormal or orthogonal representation [14, 45, 120, 121]. But we stress that the employed formulas differ widely in the literature due to varying conventions. Note that we obtain a basis of projection operators which differs from the so-called seminormal basis (cf. pp. 109–114 of [124]).

Let us consider the ordered sequence $\tau^2_g, \ldots, \tau^1_g$ of all standard Young tableaux of fixed shape $\lambda$. In particular, one has $r = s = 1$ for $\lambda = [3, r = 2$ and $s = 3$ for $\lambda = [2, 1]$, as well as $r = s = 4$ for $\lambda = [1, 1, 1]$ (cf. Eq. (B1)). One defines the projection operators $P^g_p$ recursively using the operators $e^g_p$. For $p = r$, one has $P^g_p := e^g_p$. It immediately follows that

$$P^3_1 = e^3_1, P^3_2 = e^3_2, \text{ and } P^3_3 = e^3_3.$$  \hfill (B3)

For $r < p \leq s$, there exists $q \in \{r, \ldots, p-1\}$ such that $\tau^q_g$ differs from $\tau^g_p$ only by the position of two boxes in $\tau^q_g$ and $\tau^g_p$ with consecutive labels $a$ and $b := a+1$. Let $d \in \mathbb{Z}$ denote the signed axial distance from the box in $\tau^q_g$, i.e., the number of steps from $\tau^q_g$ to $\tau^g_p$ while count Mitt steps down or to the left positively and steps up or to the right negatively. After these preparations, let

$$P^g_p := f \left[ d(ab) + e \right] P^g_q \in \mathbb{R}[S_d], \quad (B4)$$

where the scalar factor $f \in \mathbb{R}$ is chosen such that $P^g_p P^g_p = P^g_p$. For $f^g_3 := e^g_3$, one obtains $q = 2, a = 2, b = 3, d = 2$, and Eq. (B3) implies that

$$P^3_3 = [2(23) + e] P^3_3 = [2(23) + e] e^3_2 = [e-(12)-(13)+2(23)-(213)+2(132)]/3.$$  \hfill (B5)

3. Details of the iterative construction

The iterative construction of the LISA basis is now described and exemplified for the example of $n = 3$ spins. In the Sec. [III.B] this construction was divided into three steps: (I) We start by building and symmetrizing $g$-linear tensor operators of a $g$-spin system for each $g \in \{1, \ldots, n\}$ which consists in applying the Clebsch-Gordan decomposition and the just-introduced projection operators. (II) Next, the tensor operators will be phase and sign corrected. (III) Lastly, the tensor operators are embedded into the full $n$-spin system for each $g$-element subset of $\{1, \ldots, n\}$.

In step (I), we begin by specifying the form of the $g$-linear tensor operators for a $g$-spin system in the particular simple cases of $g \in \{0, 1\}$. The symmetries with respect to particle permutations are trivial in both of these cases. The corresponding symmetry types are given by the standard Young tableaux $\tau^0_1$ and $\tau^1_1 = [1]$ where the first one is empty and the second one consists of a single box. For $g = 0$, we only have the tensor operator $T_0(\tau^0_0) := T_0$ whose only tensor component is given by $T_0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}/\sqrt{2}$. There is one single linear tensor operator $T_1(\tau^1_1) := T_1$ for $g = 1$ whose components are $T_{1,-1} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}/\sqrt{2}$ and $T_{1,1} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$. For $g \geq 2$, the tensor operators $T_j(\tau^g_p) = T^p_{g-1}$ for $g-1$ spins are combined with the tensor operator $T_1$ for one spin in order to iteratively build up the tensor operators $T^p_g$ with $p \in \{-j, \ldots, j+1\}$ for $g$ spins using the Clebsch-Gordan coefficients [21, 26, 27]. The three bilinear tensor operators $T_{2,1}, T_{2,1}$, and $T_{2,2}$ for $g = 2$ are obtained via $T_1 \otimes T_1 = T^{2}_{2} \oplus T^{2}_{1} \oplus T^{2}_{2}$ and their components are given by (see, e.g., p. 419 in [22])

$$T^{2}_{1} = \frac{1}{\sqrt{3}}[T_{1,-1} \otimes T_{1,1} - T_{1,0} \otimes T_{1,0} + T_{1,1} \otimes T_{1,-1}], \quad T^{2}_{-1} = \frac{1}{\sqrt{3}}[-T_{1,-1} \otimes T_{1,0} + T_{1,0} \otimes T_{1,-1}], \quad T^{2}_{0} = \frac{1}{\sqrt{2}}[-T_{1,-1} \otimes T_{1,1} + T_{1,1} \otimes T_{1,-1}],$$

$$T^{2}_{1,1} = \frac{1}{\sqrt{2}}[-T_{1,0} \otimes T_{1,1} + T_{1,1} \otimes T_{1,0}], \quad T^{2}_{2} = T_{1,1} \otimes T_{1,1}.$$  

The projectors for $g = 2$ are

$$P^2_1 = e^2_1 = e_{\tau^2_1} = [e+(12)]/2 \text{ where } \tau^2_1 := [12]$$

and

$$P^2_2 = e^2_2 = e_{\tau^2_1} = [e-(12)]/2 \text{ where } \tau^2_1 := [2].$$

It is obvious that $P^2_1 P^2_0 = P^2_0$ and $P^2_1 P^2_2 = P^2_1$ for $m \in \{-2, \ldots, 2\}$. Moreover, $P^2_1 T^2_{1,m} = T^2_{1,m}$ for $m \in \{-1, 0, 1\}$. Thus, both $T^2_{0}$ and $T^2_{2}$ have symmetry type $\tau^2_1$ and $T^2_{1}$ has symmetry type $\tau^2_2$ (cf. Table [IV]). We set

$$T_0(\tau^2_1) := T^2_0, T_1(\tau^2_1) := T^2_1, T_2(\tau^2_1) := T^2_2,$$

but use both variants synonymously.
For $g = 3$, the Clebsch-Gordan decomposition results in various trilinear tensors and the multiplicities of their ranks agree with Tables [III] and [V].

$$T_0(T_1^{[2]} \otimes T_1^{[1]}) = T_0^{[2]} \otimes T_1^{[1]} = T'_1 \quad \text{(B6a)}$$

$$T_1(T_2^{[2]} \otimes T_1^{[1]}) = T_1^{[2]} \otimes T_1^{[1]} = T_0^c \oplus T_2^{[2]} \oplus T_2'' \quad \text{(B6b)}$$

$$T_2(T_1^{[2]} \otimes T_1^{[1]}) = T_2^{[2]} \otimes T_1^{[1]} = T_1^{[1]} \oplus T_2'' \oplus T_3'' \quad \text{(B6c)}$$

Referring again to p. 419 in [62], the corresponding components for Eq. (B6a) are given by

$$T'_{1,m} = T_{0,0}^{[2]} \otimes T_{1,m}^{[1]} \text{ for } m \in \{-1, 0, 1\}$$

for Eq. (B6b) one obtains

$$T''_{0,0} = \frac{1}{\sqrt{3}} [T_{1,-1}^{[2]} \otimes T_{1,-1}^{[1]} - T_{1,0}^{[2]} \otimes T_{1,0}^{[1]} + T_{1,-1}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T'_{1,-1} = \frac{1}{\sqrt{2}} [T_{1,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{1,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{1,0} = \frac{1}{\sqrt{2}} [T_{1,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{1,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{1,1} = \frac{1}{\sqrt{2}} [T_{1,0}^{[2]} \otimes T_{1,0}^{[1]} + T_{1,0}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{2,-2} = T_{2,-1}^{[2]} \otimes T_{1,-1}^{[1]}$$

$$T''_{2,-1} = \frac{1}{\sqrt{2}} [T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{2,0} = \frac{1}{\sqrt{2}} [T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{2,1} = \frac{1}{\sqrt{2}} [T_{2,0}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{2,2} = T_{1,1}^{[2]} \otimes T_{1,1}^{[1]}$$

The case of Eq. (B6c) results in

$$T''_{1,-1} = \frac{1}{\sqrt{10}} [\sqrt{5} T_{2,-2}^{[2]} \otimes T_{1,-1}^{[1]} - \sqrt{3} T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{1,0} = \frac{1}{\sqrt{10}} [\sqrt{3} T_{2,-2}^{[2]} \otimes T_{1,-1}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{1,1} = \frac{1}{\sqrt{10}} [T_{2,0}^{[2]} \otimes T_{1,0}^{[1]} - \sqrt{5} T_{2,1}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{6} T_{2,2}^{[2]} \otimes T_{1,-1}^{[1]}]$$

for $j = 1$,

$$T''_{2,-2} = \frac{1}{\sqrt{6}} [- \sqrt{2} T_{2,-2}^{[2]} \otimes T_{1,-1}^{[1]} + T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{2,-1} = \frac{1}{\sqrt{6}} [- \sqrt{2} T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} - T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{3} T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{2,0} = \frac{1}{\sqrt{6}} [- T_{2,0}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{2} T_{2,1}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{2,1} = \frac{1}{\sqrt{6}} [- T_{2,1}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{2} T_{2,2}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{2,2} = \frac{1}{\sqrt{6}} [- T_{2,2}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{2} T_{2,2}^{[2]} \otimes T_{1,0}^{[1]}]$$

for $j = 2$, and

$$T''_{3,-3} = T_{2,-2}^{[2]} \otimes T_{1,-1}^{[1]}$$

$$T''_{3,-2} = \frac{1}{\sqrt{2}} [T_{2,-2}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{2} T_{2,-1}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{3,-1} = \frac{1}{\sqrt{2}} [T_{2,-2} \otimes T_{1,1}^{[1]} + \sqrt{2} T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + \sqrt{6} T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{3,0} = \frac{1}{\sqrt{2}} [T_{2,-2} \otimes T_{1,1}^{[1]} + \sqrt{2} T_{2,-1}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,0}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{3,1} = \frac{1}{\sqrt{2}} [\sqrt{2} T_{2,-1}^{[2]} \otimes T_{1,1}^{[1]} + \sqrt{2} T_{2,0}^{[2]} \otimes T_{1,0}^{[1]} + T_{2,2}^{[2]} \otimes T_{1,-1}^{[1]}]$$

$$T''_{3,2} = \frac{1}{\sqrt{2}} [\sqrt{2} T_{2,0}^{[2]} \otimes T_{1,1}^{[1]} + T_{2,2}^{[2]} \otimes T_{1,0}^{[1]}]$$

$$T''_{3,3} = T_{2,2}^{[2]} \otimes T_{1,1}^{[1]}$$

for the rank of $j = 3$.

We apply now the projectors of Eqs. (B3) and (B5) in order to obtain the permutation-symmetrized versions of the tensor operators. These permutation-symmetrized versions can be obtained by tedious but straightforward linear algebra. For example, the zero-rank tensor operator $T''_{0,0}$ is unchanged by the action of $P_4^{[3]}$, i.e.

$$T''_{0,0} = P_4^{[3]} T''_{0,0}.$$ It also holds that $P_4^{[3]} T''_{0,0} = P_3^{[3]} T''_{0,0} = 0$. Consequently, $T''_{0,0}$ is completely antisymmetric which also follows from the expansion

$$\text{for } j = \{1, 2, 3\} \text{ in } T_{0,0}^{[4]} := T''_{0,0} = (P_4^{[3]} T''_{0,0}).$$

These computations can be in general quite unwieldy but are easily automated. The permutation-symmetrized tensor components of rank one are computed as (where $m \in \{-1, 0, 1\}$)

$$T_{1,m}^{[3]} := \frac{3}{\sqrt{2}} T_{1,m}^{[1]} + \frac{6}{\sqrt{5}} T''_{1,m}^{[1]} \quad \text{(B7)}$$

The rank-two case results in (where $m \in \{-2, \ldots, 2\}$)

$$T_{2,m}^{[3]} := T''_{2,m} \quad \text{(B8)}$$

and one obtains for the case of rank three that

$$T_{3,m}^{[3]} := T''_{3,m} \quad \text{(B8)}.$$
where \( m \in \{-3, \ldots, 3\} \). All tensor components \( T_{j,m}(\tau) \) have been normalized such that \( \text{Tr}[T_{j,m}(\tau) T_{j,m}^\dagger(\tau)] = 1 \), where \( \text{Tr}(A) = \sum_p A_{pp} \) denotes the trace of a matrix \( A \). Note that non-trivial reoccurrences occur for trilinear tensor operators only in Eqs. (133) and (135).

In step (II), we apply the transformations of Table V. This ensures that the phases and signs of the tensor operators are set according to the conventions discussed and motivated before. We emphasize that the transformations of Table V lead only to the correct form of the tensor operators if one has executed step (I) exactly as described. By abuse of notation, the tensor operators after the transformation are denoted by the same symbol \( T_j(\tau) \). In the following, we assume that the tensor operators \( T_j(\tau) \) have been phase and sign corrected.

Our construction is now completed by step (III) where each \( g \)-linear tensor operator of a \( g \)-spin system is embedded into \( n \)-spin systems for \( n \geq g \). In particular, we detail the three-spin case with \( n = 3 \). The zero-linear tensor component \( T_{0,0}(\tau_0^{[0]}) = T_{0,0} = (1, 0, 0) / \sqrt{2} \) (i.e. \( g = 0 \)) is embedded as

\[
T^0_{0,0}(\tau_0^{[0]}) := \left( \begin{array}{c} \sqrt{2} \end{array} \right) \tau_0^{[0]} \left( \begin{array}{c} 1 \end{array} \right) = T_{0,0} \otimes \cdots \otimes T_{0,0},
\]

where \( \mathbb{1}_q \) denotes the \( q \times q \)-dimensional identity matrix. The embedding of linear tensor components \( T_{1,1}(\tau_1^{[1]}) \) (i.e. \( g = 1 \)) is given for \( n = 3 \) as follows (where \( m \in \{-1, 0, 1\} \))

\[
T^{(1)}_{1,1}(\tau_1^{[1]}) := T_{1,1}(\tau_1^{[1]}) \otimes T_{0,0} \otimes T_{0,0}
\]

\[
T^{(2)}_{1,1}(\tau_1^{[1]}) := T_{0,0} \otimes T_{1,1}(\tau_1^{[1]}) \otimes T_{0,0}
\]

\[
T^{(3)}_{1,1}(\tau_1^{[1]}) := T_{0,0} \otimes T_{0,0} \otimes T_{1,1}(\tau_1^{[1]})
\]

More generally, one applies for \( g \geq 1 \) a transposition (1k) to \( T^{(1)}_{1,1}(\tau_1^{[1]}) \) which permutes the first and the \( k \)th particle:

\[
T^{(k)}_{1,1}(\tau_1^{[1]}) := T_{1,1}(\tau_1^{[1]}) \otimes T_{0,0} \otimes \cdots \otimes T_{0,0}
\]

\[
T^{(k)}_{1,1}(\tau_1^{[1]}) := (1k) T^{(1)}_{1,1}(\tau_1^{[1]}) \quad \text{for} \quad k \neq 1.
\]

This technique can be easily generalized to bilinear tensor components \( T_{j,m}(\tau_p^{[2]}) \) with \( m \in \{-j, \ldots, j\} \) (and beyond) which are embedded into a system of \( n \geq 2 \) spins:

\[
T^{(1,2)}_{j,m}(\tau_p^{[2]}) := T_{j,m}(\tau_p^{[2]}) \otimes T_{0,0} \otimes \cdots \otimes T_{0,0}
\]

\[
T^{(1,1)}_{j,m}(\tau_p^{[2]}) := (2l) T^{(1,2)}_{j,m}(\tau_p^{[2]}) \quad \text{for} \quad l > 2
\]

\[
T^{(k,l)}_{j,m}(\tau_p^{[2]}) := (1k) (2l) T^{(1,2)}_{j,m}(\tau_p^{[2]}) \quad \text{for} \quad l > k > 1.
\]

Finally, trilinear tensor components are embedded into a three-spin system by (where \( m \in \{-j, \ldots, j\} \))

\[
T^{(1,2,3)}_{j,m}(\tau_p^{[3]}) := T_{j,m}(\tau_p^{[3]}).
\]

### 4. From the LISA basis to Cartesian product operators

Before we close this section, our computations are summarized by providing explicit basis transformations from Cartesian product operators (for definitions refer to Sec. IV A) to the LISA basis and vice versa. The basis transformations for the linear tensor components of an \( n \)-spin system are

\[
\begin{pmatrix}
\tau_1^{(k)} \\
\tau_2^{(k)} \\
\tau_3^{(k)}
\end{pmatrix} = \left( \begin{array}{c}
\sqrt{2} \end{array} \right)^{n-1} \begin{pmatrix}
1 & -i & 0 \\
0 & 0 & \sqrt{2} \\
-1 & -i & 0
\end{pmatrix} \begin{pmatrix}
I_{kx} \\
I_{ky} \\
I_{kz}
\end{pmatrix}
\]

\[
\begin{pmatrix}
I_{kx} \\
I_{ky} \\
I_{kz}
\end{pmatrix} = \left( \sqrt{2} \right)^{-n-1} \begin{pmatrix}
1/2 & 0 & -1/2 \\
0 & i/2 & 0 \\
0 & 1/\sqrt{2} & 0
\end{pmatrix} \begin{pmatrix}
\tau_1^{(k)} \\
\tau_2^{(k)} \\
\tau_3^{(k)}
\end{pmatrix}.
\]

Similarly, the transformations in the bilinear case are

\[
\begin{pmatrix}
\tau_1^{(k,l)} \\
\tau_2^{(k,l)} \\
\tau_3^{(k,l)} \\
\tau_4^{(k,l)} \\
\tau_5^{(k,l)} \\
\tau_6^{(k,l)}
\end{pmatrix} = A_n \begin{pmatrix}
\tau_1^{(k,l)} \\
\tau_2^{(k,l)} \\
\tau_3^{(k,l)} \\
\tau_4^{(k,l)} \\
\tau_5^{(k,l)} \\
\tau_6^{(k,l)}
\end{pmatrix}
\]

\[
\begin{pmatrix}
\tau_1^{(k,l)} \\
\tau_2^{(k,l)} \\
\tau_3^{(k,l)} \\
\tau_4^{(k,l)} \\
\tau_5^{(k,l)} \\
\tau_6^{(k,l)}
\end{pmatrix}
\]

with \( A_n = \left( \sqrt{2} \right)^{-n-2} \)

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

\[
\begin{pmatrix}
1/\sqrt{3} & 0 & 0 & 1/\sqrt{6} \\
0 & 0 & 0 & 1/\sqrt{2} & -1/2 \\
1/\sqrt{6} & 0 & 0 & 0 & 0
\end{pmatrix}
\]
correspond to mixtures of multiple quantum coherences.

Different examples of multiple-quantum coherences are visualized in the third and fourth columns of Fig. 11, for $p \geq 0$ and $p \leq 0$, respectively.

As discussed in Sec. 11.1, an operator $A_p$ has a well-defined coherence order $p$ if a rotation around the $z$ axis by an arbitrary angle $\alpha$ reproduces the operator $A_p$ up to an additional phase factor $\exp(-i\alpha)$:

$$\exp(-i\alpha) \sum_{k=1}^{n} I_{kz} A_p \exp(i\alpha) \sum_{k=1}^{n} I_{kz} = A_p \exp(-i\alpha).$$

Similarly, a droplet representing a function $J^{(l)}_A$ (c.f. Eq. 41) corresponds to an operator term $A^{(l)}$ with well-
defined coherence order \( p \), if a rotation around the \( z \)-axis by an arbitrary angle \( \alpha \) reproduces the droplet (and the function \( f_A^{(t)} \) up to an additional phase factor \( \exp(-ipa) \). Hence, a droplet with coherence order \( p \) as well as the corresponding function \( f_A^{(t)}(\theta, \phi) = \psi_A^{(t)}(\theta, \phi) \exp[i\varphi_A^{(t)}(\theta, \phi)] \) are transformed by a \( z \)-rotation with angle \( \alpha \) to \( \tilde{f}_A^{(t)}(\theta, \phi) = \psi_A^{(t)}(\theta, \phi) \exp[i\varphi_A^{(t)}(\theta, \phi)] \) with \( \varphi_A^{(t)}(\theta, \phi) = \varphi_A^{(t)}(\theta, \phi) - \alpha a \). In order to illustrate this point, consider how the operator \( I^x_3 \) in Fig. 111 with \( p = +1 \) changes under a \( z \)-rotation with angle \( \alpha = \pi/2 \). A \( z \)-rotation of the corresponding droplet only changes its color (representing the phase of the function \( f_A^{(t)}(\theta, \phi) \) in Fig. 111, with \( p = +1 \) changes under a \( z \)-rotation with angle \( \alpha = \pi/2 \).

**TABLE VII.** Decomposition of trilinear Cartesian-product operators \( I_{abc} \) in terms of LISA tensor operators. This form is valid for a three-spin system, for a general system with \( n \geq 3 \) spins one has to multiply \( 4I_{abc} \) with \( \sqrt{2} \).
but not its shape. For example, for the azimuthal angle \( \phi = 0 \), the droplet is red (corresponding to a phase \( \phi_A(\theta, 0) = 0 \). After the rotation by \( \alpha = \pi/2 \), the droplet has turned dark blue at the azimuthal angle \( \phi = 0 \) (corresponding to a phase \( \phi_A(\theta, 0) = -\pi/2 \)), which is exactly what is expected from the general formula given above: \( \phi_A(\theta, \phi) = \phi_A(\theta, \phi) - p\alpha = 0 - (1+1)\pi/2 = -\pi/2 \) for \( p = +1 \) and \( \alpha = \pi/2 \).

Based on these properties, the droplets of an operator with unique coherence order \( p \) can be easily recognized using the following criteria: (1) Disregarding the color, the shape of the droplet is rotationally invariant under rotations around the \( z \)-axis, i.e. the shape is not changed by a \( z \)-rotation (c.f. third and fourth column in Fig. 11). (2) The coherence order \( p \) of a droplet can be identified based on its color: (2a) A droplet of coherence order \( p = 0 \) does not change its color if it is rotated by an arbitrary angle \( \alpha \) around the \( z \)-axis, as illustrated by the operators \( I_x^0 I_z^1, I_y^0 I_z^1, I_y^0 I_z^2, \) and \( I_y^0 I_z^3 \) in Fig. 11. (2b) A droplet of unique coherence order \( p \neq 0 \) (where \( p \) is a non-zero integer with either positive or negative sign) is rainbow-colored. For positive coherence order \( p > 0 \), the colors change from red to yellow to green to blue when moving counter-clockwise around the \( z \)-axis. For negative coherence order, the colors change in the opposite direction. (2c) For a non-zero unique coherence order \( p \), the absolute value \( |p| \) of the coherence order of a droplet is reflected by the number of rainbows encountered when moving once around the \( z \)-axis. (2d) A droplet of unique coherence order \( p \neq 0 \) is invariant under a rotation by integer multiples of \( 2\pi/|p| \) around the \( z \)-axis. This is illustrated for characteristic operators with unique coherence orders in the third column and fourth column of Fig. 11. (2e) Even if operators do not contain a unique coherence order \( p \), but a mixture of coherence orders \( \pm p \) with \( |p| \neq 0 \), it is still true that the corresponding droplets do not change their appearance if they (or the corresponding operators) are rotated by integer multiples of \( 2\pi/|p| \) around the \( z \)-axis. This is illustrated by the examples in the first and second column of Fig. 11.

Note that all tensor operators \( T_{jm} \) have the unique coherence order \( p = m \). Furthermore, the bean-shaped droplet of the Cartesian product operator \( 2I_k x I_y \) (c.f. Fig. 8) is an example of an operator that is composed of terms with different coherence orders \(-2, 0, \) and \(+2\). The operator is a superposition of the double-quantum operator \( (DQ_y)_{kl} = I_k x I_y + I_y I_k x I_y \) with rank \( j = 2 \) and quantum orders \( p = \pm 2 \) and the zero-quantum operator \( (ZQ_y)_{kl} = -I_k x I_y + I_y I_k x I_y \) with rank \( j = 1 \) and quantum order \( p = 0 \).

2. Extended NMR example

The example shown in Fig. 12 represents a common experiment in NMR spectroscopy which is designed to create triple-quantum coherences from the polarization of three coupled spins [2]. The system consists of three spins in the weak-coupling limit (i.e. longitudinal or Ising-type coupling; see Fig. 3) with identical coupling constants \( J_{12} = J_{13} = J_{23} = 10 \) Hz. The building blocks of the experiment are visualized in Fig. 12 in the LISA basis. A triple-quantum coherence consists of combinations of tensor operators of rank \( j = 3 \) and order \( m = \pm 3 \). At the initial time \( t_0 \), the system is in thermal equilibrium, which corresponds in the high-temperature limit to the density matrix

\[
\rho(t_0) = I_{1x} + I_{2x} + I_{3x} = \sqrt{2}(T_{10}^{(1)} + T_{10}^{(2)} + T_{10}^{(3)})
\]

(where for simplicity only the traceless part of the density operator is considered here).

A first \( 90^\circ \) pulse (with phase \( x \)) is applied to the system with an amplitude of \( 10 \) kHz for a time \( t_1 - t_0 = 25 \) \mu s and flips the three magnetization vectors into the transverse plane. The corresponding linear control Hamiltonian is

\[
\mathcal{H}(t_0, t_1) = 2\pi 10 \text{ kHz} \left( I_{1x} + I_{2x} + I_{3x} \right)
\]

and the density operator of the system at time \( t_1 \) is

\[
\rho(t_1) = -I_{1y} - I_{2y} - I_{3y} = -i[T_{1,-1,-1} + T_{1,1,1} + T_{1,-1,-1} + T_{1,1,1} + T_{1,1,1} + T_{1,1,1}]
\]

The next step consists in letting the bilinear coupling Hamiltonian act on the system in order to create trilinear terms in the density operator. The coupling Hamiltonian is applied for a time \( t_2 - t_1 = 50 \) ms and has the form

\[
\mathcal{H}(t_1, t_2) = 2\pi 10 \text{ Hz} \left( I_{1z} I_{2z} + I_{1z} I_{3z} + I_{2z} I_{3z} \right)
\]

At time \( t_2 \), the systems is in the state

\[
\rho(t_2) = 4I_{1y} I_{2z} I_{3z} + 4I_{1y} I_{2y} I_{3z} + 4I_{1y} I_{2z} I_{3y} \\
+ 0.78 [T_{1,-1,-1}(T_{1}^{(1)}) + T_{1,1,1}(T_{1}^{(1)})] \\
+ 1.55i [T_{3,-1,-1}(T_{1}^{(1)}) + T_{3,1,1}(T_{1}^{(1)})]
\]

Finally, a second \( 90^\circ \) pulse (with phase \( y \)) is applied in order to create terms of order \( m = \pm 3 \). The corresponding linear control Hamiltonian

\[
\mathcal{H}(t_2, t_3) = 2\pi 10 \text{ kHz} \left( I_{1y} + I_{2y} + I_{3y} \right)
\]
is applied for a time $t_3 - t_2 = t_1 - t_0$. At time $t_3$, the density operator of the system is (where $I_{abc} := I_{1a}I_{2b}I_{3c}$)
\[
\rho(t_3) = 4I_{yxz} + 4I_{xyz} + 4I_{xzy} \\
\cong 0.78 [T_{1,-1}(\tau_{1}^{[3]}),+T_{1,1}(\tau_{1}^{[3]})] \\
- 0.39i [T_{3,-1}(\tau_{1}^{[3]}),+T_{3,1}(\tau_{1}^{[3]})] \\
+ 1.5i [T_{3,-3}(\tau_{1}^{[3]}),+T_{3,3}(\tau_{1}^{[3]})].
\]
At this point, the desired triple-quantum coherence term $T_{3,-3}(\tau_{1}^{[3]}) + T_{3,3}(\tau_{1}^{[3]})$ has been created. Note that the shape of the droplet corresponding to the term $\tau_{1}^{[3]}$ of $\rho(t_3)$ in Fig. 12 also exhibits the partial content of the triple-quantum coherence $(TQ_{1})$ (see Fig. 11). The remaining undesired terms of the density operator can be removed by applying a triple quantum filter 3 to $\rho(t_3)$ (not shown for simplicity). The density operators $\rho(t_i)$ for this experiment are depicted in the middle row of Fig. 12.

The Hamiltonians $\mathcal{H}(t_i, t_{i+1})$ (which are re-scaled for better visibility) are shown in the second row and the effective Hamiltonian
\[
\mathcal{H}_{eff} \cong -18.1 \text{Hz}(I_{1z}+I_{2z}+I_{3z}) \\
-24.2 \text{Hz}(I_{xzx} + I_{yyz} + I_{zzz}) \\
-72.5 \text{Hz}(I_{xxy} + I_{yyx} + I_{xyz} + I_{xxy} + I_{xyy})
\]
of the experiment is given at the top. In the fourth row, the DROPS representations of the propagators ($\mathbf{1}_{q}$ denotes the $q \times q$-dimensional identity matrix)
\[
U(t_0, t_1) \cong +0.35 \mathbf{1}_8 - 0.71i(I_{1x} + I_{2x} + I_{3x}) \\
-1.41(I_{1y}I_{2y} + I_{1y}I_{3y} + I_{2y}I_{3y}) \\
+ 2.83I_{1z}I_{2z}I_{3z},
\]
\[
U(t_1, t_2) \cong +0.35(1+i) \mathbf{1}_8 - 1.41(1+i)(I_{1x}I_{2x} + I_{1x}I_{3x} + I_{2x}I_{3x}),
\]
and
\[
U(t_2, t_3) \cong +0.35 \mathbf{1}_8 - 0.71i(I_{1y} + I_{2y} + I_{3y}) \\
-1.41(I_{1y}I_{2y} + I_{1y}I_{3y} + I_{2y}I_{3y}) \\
+ 2.83I_{1y}I_{2y}I_{3y},
\]
are shown. The overall effective propagator
\[
U_{eff} \cong +0.18(1+i) \mathbf{1}_8 - 0.35(1+i)(I_{1x} + I_{2x} + I_{3x}) \\
-0.71(1+i)(I_{1z}I_{2z} + I_{1z}I_{3z} + I_{2z}I_{3z}) - 1.41(1-i)\times
(I_{xzx} + I_{yyz} + I_{zzz} + I_{xxy} + I_{yyx} + I_{xyz} + I_{xxy} + I_{xyy})
\]
of the pulse sequence is represented at the bottom of Fig. 12.

### 3. Pure quantum states

The examples of Fig. 9 display density matrices for entangled pure states of two spins and three spins in the

\[
|\Phi^{\pm}\rangle = \frac{|00\rangle + |11\rangle}{\sqrt{2}} \\
|\Psi^{\pm}\rangle = \frac{|01\rangle \pm |10\rangle}{\sqrt{2}}
\]
DROPS representation corresponding to the LISA basis. The four Bell states $|\Phi^{\pm}\rangle = (|00\rangle \pm |11\rangle)/\sqrt{2}$ and $|\Psi^{\pm}\rangle = (|01\rangle \pm |10\rangle)/\sqrt{2}$ correspond to maximally entangled states of a two-spin system. For comparison, a separable state $(|00\rangle)$ and a partially entangled state $[(|00\rangle + |11\rangle)/\sqrt{2} + 3|\Psi^{-}\rangle]/4$ are depicted in the third row of Fig. 9. The last row of Fig. 9 shows the Werner state $|W\rangle = (|00\rangle + |01\rangle + |10\rangle)/\sqrt{3}$ and the Greenberger-Horne-Zeilinger state $|\text{GHZ}\rangle = (|000\rangle + |111\rangle)/\sqrt{2}$, which are both entangled quantum states of three spins. For comparison, refer also to Fig. 10 below.

### Appendix D: Wigner representation: Proof of Proposition 1

Property (a) is a direct consequence of the definition of the DROPS mapping. Proving property (b), we deduce
directly from Eq. (41) that
\[
\begin{align*}
  f^{(\ell)^*}_{A} &= \sum_{j \in \ell} \sum_{m=-j}^{j} \epsilon^{(\ell)^*}_{jm} Y_{jm} \\
  A^{(\ell)^*} &= \sum_{j \in \ell} \sum_{m=-j}^{j} \epsilon^{(\ell)^*}_{jm} T_{jm}^{(\ell)^*}.
\end{align*}
\]

Consequently, (b) follows from the Condon-Shortley phase convention \(T_{jm} = (-1)^m T_{jm}^{\dagger}\) and a similar relation \(Y_{jm} = (-1)^m Y_{jm}^{*}\) for spherical harmonics. Property (c) is a special case of (e), which we prove below. Property (d) uses the fact that irreducible tensor operators and spherical harmonics are an explicit form of irreducible representations for SU(2). These properties are preserved by the DROPS mapping, i.e., the components \(T_{jm}\) with \(j \leq m \leq j\) corresponding to the same tensor define an invariant subspace under rotations. Furthermore, all of these components are part of the same droplet. The proof of property (e) requires a more detailed analysis. We expand both sides of (e) in order to show that they are equal. To simplify the notation, the dependence of the spherical harmonics on the variables \(\theta\) and \(\phi\) is suppressed.

The left-hand side of the equality is expanded as
\[
\sum_{\ell \in L} \sum_{jm} \text{Tr}[T_{jm}^{(\ell)^*}] A[Y_{jm} \cdot \sum_{j' m'} \text{Tr}[T_{j' m'}^{(\ell)^*}] B[Y_{j' m'}] d\mu.
\]

Applying the orthonormality of the spherical harmonics as well as the property \(Y_{jm} = (-1)^m Y_{jm}^{*}\), we obtain the expression
\[
\sum_{\ell \in L} \sum_{jm} \text{Tr}[T_{jm}^{(\ell)^*}] A \cdot \text{Tr}[T_{jm}^{(\ell)^*}] B(-1)^m.
\]

Applying \(T_{jm} = (-1)^m T_{jm}^{\dagger}\), this can be further simplified to
\[
\sum_{\ell \in L} \sum_{jm} \text{Tr}[T_{jm}^{(\ell)} A] \cdot \text{Tr}[T_{jm}^{(\ell)} B].
\]

On the right-hand side, we have \(\text{Tr}(A)\) for the multipole tensors operators \([63, 112–116]\) are defined by building on a state-space basis of the quantum system which reflects its angular momentum properties \(\{\kappa, j, m\}\), where \(\kappa \in \{\kappa_1, \ldots, \kappa_t\}\) are suitable-chosen auxiliary labels distinguishing angular momentum states with identical rank \(j\) and order \(m\). Given \(\kappa\) and \(j\), the symbol \(|\kappa, j\rangle\) with \(-j \leq m \leq j\) denotes the ordered set of states with angular momentum \(j\) and auxiliary label \(\kappa\). Moreover, the state space of coupled
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TABLE VIII. Labels for the DROPS representation based on multipole operators.

| transition | rank |
|------------|------|
| $|3/2\rangle\rightarrow|3/2\rangle$ | $0, 1, 2, 3$ |
| $|\kappa_1, 1/2\rangle\rightarrow|\kappa_1, 1/2\rangle$ | $1$ |
| $|\kappa_2, 1/2\rangle\rightarrow|\kappa_2, 1/2\rangle$ | $1$ |
| $|3/2\rangle\rightarrow|\kappa_1, 1/2\rangle$ | $1, 2$ |
| $|\kappa_1, 1/2\rangle\rightarrow|3/2\rangle$ | $1, 2$ |
| $|\kappa_2, 1/2\rangle\rightarrow|3/2\rangle$ | $1, 2$ |

TABLE IX. All compatible combinations of transitions and ranks for multipole tensor operators

The angular momentum state basis is given by $B_1 := |1/2\rangle := \Phi^+ \rangle$, $B_2 := |1/2\rangle := \Psi^+ \rangle$, $B_3 := |\Psi^-\rangle = |0\rangle + |1\rangle$, $B_4 := |\Psi^-\rangle = |0\rangle + |1\rangle$. For two coupled spin-1/2 particles, we use the Clebsch-Gordan decomposition of $B_1 \otimes B_2$ and build the basis $B_2 := |\Psi^-\rangle$ consisting of singlet and triplet states (see, e.g., pp. 430–431 of [29]). No auxiliary labels $\kappa$ are necessary as each rank appears only once in $B_2$. For three coupled spin-1/2 particles, one obtains the basis $B_3 := |3/2\rangle \cup |\kappa_1, 1/2\rangle \cup |\kappa_2, 1/2\rangle$ (see Table VIII). The auxiliary labels $\kappa_1 := 1$ and $\kappa_2 := 0$ of the state refer to the parent rank of the element in $B_2$ involved in its generation. All compatible combinations of the nine possible transitions $|j_1, \kappa_1\rangle \rightarrow |j_2, \kappa_2\rangle$ with ranks $j$ are given in Table IX.

The multipole tensor operators differ from the tensor operators in the LISA basis in not having a defined particle number. The explicit decomposition of multipole tensor operators in the LISA basis can be found in Table IX.

Examples for the DROPS representation based on multipole tensor operators are illustrated in Fig. [10], where the density matrices of different pure states are given (cf. Fig. [9]), some of which are entangled and most of them have only one single non-empty droplet. Interestingly, both $|W\rangle$ and $|GHZ\rangle$ exhibit a symmetry under $z$-rotations, i.e. they are respectively rotationally invariant or invariant under rotations of $2\pi/3$, see also Fig. [9].
TABLE X. Decomposition of the multipole tensor operators for a system of three spins in terms of the LISA tensors.
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FIG. 11. (Color online) DROPS visualization in the LISA basis of characteristic multiple quantum coherences for three coupled spins. The operators are classified according to their linearity and their coherence order $p \in \mathbb{Z}$. For visualization purposes, the droplets which are always empty are not displayed. The four trilinear droplets are ordered from left to right according to the symmetry of each droplet, i.e., from $\tau_{3}^{[3]}$ to $\tau_{3}^{[0]}$ (see Eq. (B1)). The above pictures correspond to the DROPS visualization of the tensors after normalization, where the droplets for the trilinear operators are depicted in a smaller size. For definitions of the zero, double, and triple quantum operators $(ZQ_{\eta})_{kl}$, $(DQ_{\eta})_{kl}$, and $(TQ_{\eta})_{kl}$ with $\eta \in \{x, y\}$ refer to [3, 12].
FIG. 12. (Color online) Experimental NMR pulse sequence to create triple-quantum coherences starting from the thermal equilibrium density operator in the high temperature limit \[3\]. The pulse sequence consists of a 90° pulse (with phase \(x\)) followed by a delay \((t_2 - t_1)\) and a second 90° pulse (with phase \(y\)). The density operators \(\rho(t_i)\) are depicted in the middle row. The Hamiltonians \(\mathcal{H}(t_i, t_{i+1})\) (which are re-scaled for better visibility) are given in the second row and the effective Hamiltonian \[3\] of the experiment is shown at the top. In the fourth row, DROPS representations of the propagators associated to the individual time steps are displayed. The effective propagator is visualized at the bottom.