Simulating open quantum systems by applying SU(4) to quantum master equations

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We show that open quantum systems of two-level atoms symmetrically coupled to a single-mode photon field can be efficiently simulated by applying a SU(4) group theory to quantum master equations. This is important since many foundational examples in quantum optics fall into this class. We demonstrate the method by finding exact solutions for many-atom open quantum systems such as lasing and steady state superradiance.

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I. INTRODUCTION

Most physical situations to which quantum mechanics is applied are open. The open nature is necessary to treat basic irreversible processes such as energy transfer with a heat bath, particle exchange with a reservoir, and quantum measurements. Open quantum systems can be treated under the Born and Markov approximations by the quantum master equation in the Lindblad form [1], which has been applied across many fields of physics, including quantum optics and quantum information science [2, 3], atomic and molecular physics [4], solid state physics [5], and optomechanics [6]. Various approximation methods have been introduced, e.g. perturbation theories [7], mean-field approaches [8, 9], cumulant expansions [10, 11], linear response theories [12] and e-number Langevin equations [13, 14]. However, it is often necessary to benchmark approximate methods with exact numerical solutions. Existing numerical simulation approaches, such as the quantum Monte Carlo method [15], scale exponentially with the underlying dimensionality of the Hilbert space. Therefore, treating any appreciable system size is extremely difficult.

Here we present a novel group-theoretic approach to find an efficient solution of the quantum master equation, which reduces the exponential scaling of the problem to cubic. Even though we focus on an important class of quantum optical systems, the methods we present could be more generally applied. We consider the symmetric coupling of a single-mode cavity field to an ensemble of N two-level atoms (analogous to pseudo-spin-1/2 systems or qubits). The Hamiltonian that describes this situation in the interaction picture is given by

\[ H = \frac{\hbar \Delta}{2} \sum_{j=1}^{N} \sigma_j^{(3)} + \hbar \Omega \sum_{j=1}^{N} (a_j^\dagger \sigma_j^+ + a_j \sigma_j^-), \tag{1} \]

where the first term is the free energy, with \( \Delta \) being the detuning of the light field from the atomic transition, and the second term is the reversible atom-field coupling with strength \( \Omega \). The photon annihilation operator is \( a \), and \( \sigma_j^{(3)} \) and \( \sigma_j^{\pm} = (\sigma_j^z \pm i \sigma_j^x)/2 \) are Pauli operators for the \( j \)th spin-component. In the presence of decoherence, the full quantum evolution is described by the quantum master equation for the reduced density operator \( \rho \):

\[ \dot{\rho} = \mathcal{L} \rho = \frac{1}{i\hbar} [H, \rho] + \kappa \mathcal{D}(a^\dagger) \rho + \sum_{j=1}^{N} \left( \gamma \mathcal{D}(\sigma_j^x) + w \mathcal{D}(\sigma_j^y) + \frac{1}{2T_2} \mathcal{D}(\sigma_j^z) \right) \rho, \tag{2} \]

where \( \mathcal{D}(\sigma_j^x) = (2\hat{\sigma}_j^x \hat{\rho} - \hat{\rho} \hat{\sigma}_j^x)/2 \) denotes the Lindblad superoperator. We have introduced the decay rate \( \kappa \) for the cavity, and population relaxation rates for the spin components \( \gamma, w \) (for decay and pumping respectively) and dephasing rate \( 1/(2T_2) \).

II. APPLYING SU(4) TO THE QUANTUM MASTER EQUATION

Recently, it was pointed out that it is preferable to work in Liouville space rather than in Hilbert space since the Lindblad operators are invariant under SU(4) transformations [16]. This observation allows one to express all of the Lindblad operators in terms of generators of the SU(4) group. For this purpose, 18 superoperators \( \mathcal{Q}_\alpha, \mathcal{Q}_- \) and \( \mathcal{O}_3 \) where \( \mathcal{O} \in \{Q, \Sigma, M, N, \mathcal{U}, \mathcal{V} \} \) are defined

\[
\begin{align*}
\mathcal{Q}_+ \rho &= \sum_{j=1}^{N} \sigma_j^x \rho \sigma_j^x, \\
\mathcal{Q}_- \rho &= \sum_{j=1}^{N} \sigma_j^y \rho \sigma_j^y, \\
\Sigma_+ \rho &= \sum_{j=1}^{N} \sigma_j^z \rho \sigma_j^z, \\
\Sigma_- \rho &= \sum_{j=1}^{N} \sigma_j^z \sigma_j^z, \\
M_+ \rho &= \sum_{j=1}^{N} \sigma_j^z \rho \sigma_j^z, \\
M_- \rho &= \sum_{j=1}^{N} \sigma_j^z \sigma_j^z, \\
N_+ \rho &= \sum_{j=1}^{N} \sigma_j^z \rho \sigma_j^z, \\
N_- \rho &= \sum_{j=1}^{N} \sigma_j^z \sigma_j^z, \\
U_+ \rho &= \sum_{j=1}^{N} \sigma_j^x \rho \sigma_j^x, \\
U_- \rho &= \sum_{j=1}^{N} \sigma_j^x \sigma_j^x, \\
V_+ \rho &= \sum_{j=1}^{N} \sigma_j^y \rho \sigma_j^y, \\
V_- \rho &= \sum_{j=1}^{N} \sigma_j^y \sigma_j^y.
\end{align*}
\tag{3}
\]

Although this list, Eq. (3), contains 18 operator definitions, only 15 of them are independent (it is possible to write \( N_3 \),...
the number of required basis states needed to provide an exact
$SU(4)$ symmetry type of the basis is preserved under the action of the
tive spin-angular-momentum representation
basis representation can be precisely mapped to the collec-
where $S$
the Liouville space given by $4$
We show how to calculate the various basic observables of in-
act solutions to the quantum master equation in general form.
Note that only basis states with
$(\gamma = \delta = 0)$ have non-vanishing
In order to solve Eq. (2), we expand the density matrix as
$\rho = \sum_{q,q',\sigma_3,m,n} C_{q,q',\sigma_3,m,n}^m P_{q,q',\sigma_3} |m\rangle \langle n|,$
where $C_{q,q',\sigma_3,m,n}^m$ are complex coefficients, and $|n\rangle$ is the photon
Fock state. The Lindblad operators can be written compactly:
$\Delta[a_j^+] = -\frac{N}{2} \pm Q_3 + Q_z,$
$\Delta[a_j^-(3)] = 4M_3 - 2Q_3 - 2\Sigma_3 - N.$

The completeness of $O_{r,-,3}$ and $a$ implies that an arbitrary
Hamiltonian can be expressed by them, e.g. from Eq. (11),
$\frac{1}{i\hbar} [H, \rho] = -2i \Delta \Sigma \rho - i\Omega \left[ a(M_+ + N_+) \rho + a^\dagger (M_- + N_-) \rho \right]$
$\quad + i\Omega \left[ (U_+ + V_+) \rho a^\dagger + (U_- + V_-) \rho a \right].$ (7)

Combining Eqs. (6) and (7) with the action rules of the $SU(4)$
and photon operators on the basis states (see Appendix A)
gives a closed solution of Eq. (2). In general, this can be
solved analytically or numerically with standard methods.

III. OBSERVABLES

Having established the procedure for determining the time
evolution of $\rho$, it is now important to describe how to calculate
physical observables. We begin with the trace given by:
$\text{Tr}[\rho] = \sum_{m,q} e_{m,q}^{m,n} C_{N/2,q,0} = 1,$
which is an invariant during evolution to represent probability
conservation. Average values $\langle a \rangle$ and $\langle a^\dagger a \rangle$ are found analogously.
For the spin-operators, we provide the following ex-
amples up to quadratic order:
$\langle a_j^{(3)} \rangle = 2\text{Tr}(Q_j \rho)/N,$
$\langle a_j^{(3)} a_k^{(3)} \rangle = (4\text{Tr}[(Q_j^2 - \Sigma_j^2) \rho] - N)/[N(N - 1)],$
$\langle a_j^\dagger \rangle = \text{Tr}[(M_+ + N_+ \rho)]/N,$
$\langle a_j^\dagger a_k \rangle = \text{Tr}[V_-(M_- + N_-) \rho - Q_- \rho]/[N(N - 1)],$
where $j \neq k$.

For coherence properties it is necessary to calculate produc-
ts of operators evaluated at different times. Of particular
interest are the first-order and second-order correlations, which
which can be found by applying the quantum regression theorem:
$\langle \hat{O}_1(t + \tau) \hat{O}_2(t) \rangle = \text{Tr} \left[ \hat{O}_1 e^{\mathcal{L}_\tau} \hat{O}_2 \rho(t) \right],$
$\langle \hat{O}_1(t) \hat{O}_2(t + \tau) \hat{O}_2(t) \rangle = \text{Tr} \left[ \hat{O}_2 e^{\mathcal{L}_\tau} \hat{O}_1 \rho(t) \hat{O}_1 \right].$ (10)

IV. TRANSFORM TO THE $|S, M\rangle |S', M'\rangle$ REPRESENTATION

Although at this point we have provided a theoretical
framework that is complete and provides exact and efficient
solutions to the general quantum master equation, it is often
inconvenient to work in the $P_{q,q',\sigma_3}$ representation of the
density operator. For example, it can be a nontrivial
procedure to characterize the many-body spin-state in this
representation by quantifying the degree of entanglement, which
is derived from a functional (i.e. $\text{Tr}[\rho \log(\rho)]$). For this reason, we illustrate now the procedure for efficiently projecting the density operator from the $SU(4)$ basis representation onto the usual representation of density matrices formed from the Hilbert space basis vectors. These Hilbert space basis vectors are specified by the angular momentum eigenket $|S,M\rangle$, where $S = N/2, N/2 - 1, \ldots, (1/2 \text{ or } 0)$ is the total spin and $M = -S, -S+1, \ldots, S$ is the spin-projection. Note that $S$ also labels the symmetry of the states, e.g. $S = N/2$ corresponds to the fully symmetrical Dicke states.

In order to illustrate how this projection is done, it is instructive for us to first examine explicitly the $N=2$ case where the Hilbert space is 4 dimensional. Two spins form a symmetric triplet state and an antisymmetric singlet state, corresponding to total spin $S = 1$ and $S = 0$ respectively. In this case, the complete density matrix from Eq. (5) for given $m,n$ is

$$
\begin{pmatrix}
(1, 1) & (1, 0) & (1, -1) & (0, 0) \\
1, 1 & C_{1,1,1,1}^{m,n} & C_{1,1,1,1}^{m,n} & 0 \\
1, 0 & C_{1,0,1,0}^{m,n} & C_{1,0,1,0}^{m,n} & 0 \\
1, -1 & C_{1,-1,1,1}^{m,n} & C_{1,-1,1,1}^{m,n} & 0 \\
0, 0 & 0 & 0 & 0
\end{pmatrix}
$$

Notice that the resulting matrix is block diagonal in the $S = 1$ and $S = 0$ subspaces (a $3 \times 3$ block and a $1 \times 1$ block). In addition, the complex coefficients contributing to the matrix element for $|S,M\rangle\langle S,M'|$ all satisfy $q_3 + \sigma_3 = M$ and $q_3 - \sigma_3 = M'$. Finally, the trace is simply $\sum_{q_3=0}^{1} C_{1,q_3,0}^{m,n} = 1$.

These results can be systematically extended to higher $N$. For any $N$, the density matrix is block diagonal in $S$, with each block given by

$$
\rho_{S}^{m,n} = \sum_{M,M'} D_{S,M,M'}^{m,n} |S,M\rangle\langle S,M'|,
$$

where $D_{S,M,M'}$ are density matrix elements for the symmetry type $S$. There are $n_{S}$ ways for $N$ spins to construct the basis for each $S$, so that $\sum_{S}(2S+1)n_{S} = 2^{N}$, i.e. the Hilbert space dimension $2^{N}$. To find $n_{S}$, we note that $|S,M\rangle$ forms a basis of the $(2S+1)$-dimensional irreducible representation of the $SU(2)$ group. Determining $n_{S}$ is accomplished with the help of the Young tableau of the $SU(2)$ group, where one can obtain the number of equivalent representations iteratively. Fig. 1(a) shows the Young tableau for the $N = 4$ case. A corresponding tabular method for evaluating $n_{S}$ for any $N$ is shown in Fig. 1(b), which contains about one half of Pascal’s triangle.

With this in mind, one can now derive a systematic algorithm for obtaining density matrix elements $D_{S,M,M'}^{m,n}$ given $SU(4)$ expansion coefficients $C_{q_3,q_3',\sigma_3'}^{m,n}$. The procedure is outlined as follows. For each layer of the pyramid (cf. Fig. 1(c)), one may start with a corner element ($M$ and $M'$ maximal) and fill out the matrix by successive application of the angular momentum lowering operator $\hat{J}_{-} = \sum_{j=1}^{N} \sigma_{j}^{-}$ (noting that $\rho J_{-} = (|U_{+},+V_{+}\rangle \rho$) to recursively fill out each row, and $\hat{J}_{+}$ (or hermiticity of $\rho$) to fill out each column. The layers are filled upwards from the base, starting with $D_{N/2,N/2}^{m,n} = C_{N/2,N/2}^{m,n}$ as the corner element of the lowest layer, and finding the corner element of higher layers by Gaussian elimination from the trace constraint Eq. (6). In Appendix C, we demonstrate explicit application to 3 atoms, with extrapolation to higher $N$ straightforward.

Being able to express the density operator in the $|S,M\rangle$ representation makes easy the calculation of functionals, such as the purity $\text{Tr}[\rho^{2}]$, or the von Neumann entropy

$$
S = -\text{Tr}(\rho \log(\rho)) = - \sum_{j} \lambda_{j} \log \lambda_{j},
$$

where $\lambda_{j}$ are eigenvalues of $\rho$. The point is that, because the density matrix is block diagonal in the $|S,M\rangle$ representation, we do not need to diagonalize the whole density matrix, which would be a daunting task. Instead, we only need to diagonalize a series of $[N/2]+1$ blocks of dimension $2S+1$.

V. APPLICATION TO LASING

In the following, we demonstrate the method by solving many-atom open quantum systems such as lasing and steady state superradiance. We show the capability for finding exact solutions of large systems and are able to obtain full information about both the transient and steady-state density matrix.

First, let us consider a single-mode laser consisting of an ensemble of two-level atoms coupled to an optical cavity, which can be modeled by the general quantum master equation Eq. (2). In this model we will ignore $T_{2}$ dephasing for simplicity. The laser system is difficult to solve without approximation since it involves both many atoms and large numbers of photons when above threshold. Therefore, it constitutes an interesting test-case to illustrate the capability of the $SU(4)$ approach.

Fig. 2(a) shows the average intracavity photon number of the laser as a function of the repumping rate, where the threshold is evident. This result confirms the conventional laser theory prediction [3]. Interestingly, the spin-spin correlation $\langle \sigma_{1}^{z} \sigma_{2}^{z} \rangle$ above the threshold is directly proportional to the photon number, which shows that the collective photon emission
FIG. 2. (color online). Calculations of laser behaviors described by Eq. (2) with $\Omega = 1, \gamma = 5, \kappa = 1/2T_c^2 = 0$ and $N = 30$. (a) The average intracavity photon number (red dots) and spin-spin correlation (blue squares) as a function of the repumping rate $w$. The blue line is the laser theory prediction of the average photon number [3]. (b) Photon statistics of the laser below threshold $w = 4$ (red dots) and above threshold $w = 8$ (blue squares). (c) Normalized spectra of the laser below threshold $w = 4$ (squares) and above threshold $w = 8$ (dots). The red dashed line and green solid line are fitted Lorentzian lineshapes. (d) Threshold behavior illustrated by the intensity correlation $g^{(2)}(0)$ and the entropy of the whole system. It can be seen that $g^{(2)}(0)$ jumps from two below threshold to one above threshold with the entropy increasing and saturating. It is remarkable to have an exact solution to this fundamental system and to be able to rigorously confirm standard laser theory results. As discussed earlier, those results are typically based on various kinds of analytic approximations necessary to make the problem tractable.

FIG. 3. (color online). (a) Comparison of the second order intensity correlation $g^{(2)}(0) = \langle a^\dagger a^\dagger a a \rangle / \langle a^\dagger a \rangle^2$ as a function of the repump rate in the steady state superradiance. The green symbols show the Monte Carlo results including the statistical errors for $N = 10$ atoms from Ref. [11]. The red squares show the present calculation using the SU(4) theory. And the blue solid line shows the semiclassical results from Ref. [11]. (b) Atom statistics for $w = 0.1T_c$. The length of the bars represents populations of the $|S, M\rangle$ states.

VI. APPLICATION TO STEADY STATE SUPERRADIANCE

As a second example, we apply our approach to steady-state superradiance as previously proposed [10] and demonstrated in a recent experiment [19]. The steady-state superradiance represents a novel regime of cavity quantum electrodynamics, where the highly coherent collective atomic dipole induces an extremely narrow linewidth for the generated light. The bad-cavity mode only plays a role as the source of collective coupling for the atoms and the definition of the spatial mode for the output light [11]. The behavior of this system is also de-
scribed by a master equation Eq. (2), but in a completely different parameter regime to the conventional laser. For steady-state superradiance, the vacuum Rabi splitting is much less than the cavity linewidth, $\sqrt{N\Omega} \ll \kappa$, and equivalently the photon number per atom in the cavity is much less than unity.

We present here calculations of the second order intensity correlation $g^{(2)}(0)$ in steady-state as a function of the repump rate. As shown in Fig.3(a), the agreement of the present calculation and the quantum Monte Carlo result from Ref. [11] is within error bars. The quantum Monte Carlo simulations were significantly more numerically intensive. In the weak pumping limit, the light exhibits strongly super-Poissonian fluctuations and deviates remarkably from the semiclassical prediction (blue line in Fig. 3(a)). The failure of the semiclassical prediction in the weak pumping limit indicates that the atoms are in a highly-correlated state. To reveal the atomic states in this case, we apply the techniques of projecting the density operator in the $P_{q,s}$ representation onto the $|S,M\rangle$ representation and obtain the atomic populations. The inset of Fig. 3 shows explicitly that the atoms are mainly pumped into long-lived collective subradiant states [20] $|S=0, M=0\rangle$ and $|S=1, M=-1\rangle$. From $|S=0, M=0\rangle$, the atoms can only be repumped to $|S=1, M=1\rangle$, from which they rapidly emit two photons and relax to $|S=1, M=-1\rangle$. Therefore, our methods have enabled us to reveal detailed information about the underlying quantum dynamics.

VII. CONCLUSION

In conclusion, we have formulated and applied a $SU(4)$ theory to numerically solve the quantum master equation, which has reduced the exponential scaling of the problem to cubic in $N$. We have developed powerful methods to transform the density operator in the $SU(4)$ basis representation to the $|S,M\rangle$ representation. This has enabled us to efficiently diagonalize the whole density matrix and thus provided complete information about the system, including state information and functional properties of the density operator. We have included lasing and steady-state superradiance as examples in order to illustrate the potential for this method. The method described here will find numerous applications for simulating open quantum systems with large system size.

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Appendix A: SU(4) Algebra

In order to see how the superoperators [Eq. (3)] are related to generators of the $SU(4)$ group (Gell-Mann matrices), consider first the fundamental one atom case. We interpret the $2 \times 2$ density matrix as a $4 \times 1$ vector in the representing vector space (i.e. Liouville space).

$$\begin{pmatrix} a & c \\ d & b \end{pmatrix} \rightarrow \begin{pmatrix} a \\ c \\ d \\ b \end{pmatrix}. \quad (A1)$$

The relations are then given by

$$Q_x = \frac{1}{2}(\lambda_9 \pm i\lambda_{12}), \quad Q_y = \frac{1}{4}I_3 + \frac{1}{4}\sqrt{3}I_8 + \frac{\sqrt{5}}{6}I_{15},$$

$$\Sigma_x = \frac{1}{2}(\lambda_6 \pm i\lambda_7), \quad \Sigma_y = \frac{1}{4}I_3 + \frac{\sqrt{3}}{4}I_8,$$

$$M_x = \frac{1}{2}(\lambda_4 \pm i\lambda_5), \quad M_y = \frac{1}{4}I_3 + \frac{\sqrt{3}}{4}I_8,$$

$$N_x = \frac{1}{2}(\lambda_{11} \pm i\lambda_{12}), \quad N_y = \frac{1}{4}I_3 + \frac{\sqrt{3}}{4}I_8 + \frac{\sqrt{5}}{6}I_{15},$$

$$U_x = \frac{1}{2}(\lambda_1 \pm i\lambda_2), \quad U_y = \frac{1}{2}I_3,$$

$$V_x = \frac{1}{2}(\lambda_{13} \pm i\lambda_{14}), \quad V_y = -\frac{1}{2}I_8 + \frac{\sqrt{5}}{6}I_{15}. \quad (A2)$$

The commutation relations of the superoperators are given in both Ref. [16] and [21]. We can also identify six $SU(2)$ subalgebras,

$$[O_x, O_-] = 2O_3, \quad [O_3, O_\pm] = \pm O_x, \quad (A3)$$

so that it is useful to define six corresponding quadratic superoperators $O^2 = O_+ O_- + O_3^2 + O_3$, which commute with $O_3$. The $SU(4)$ group has 3 Casimir operators, one of which is quadratic in the generators, and the others are of higher order. The quadratic Casimir operator $C_1$ can be expressed in terms of superoperators

$$C_1 = \sum_{\alpha}(O_+O_-+O_3)+U_3^2+\frac{1}{3}(U_3+2\Sigma)^2+\frac{1}{6}(3Q_3-2U_3-\Sigma)^2. \quad (A4)$$

Appendix B: Fully symmetrical basis for $SU(4)$ group

The fundamental representation of the $SU(4)$ group, adapted to serve as basis of the single-spin density matrix, is given by $\eta = |1\rangle\langle 1|, d = |0\rangle\langle 0|, s = |1\rangle\langle 0|, c = |0\rangle\langle 1|$. Higher order representations can then be obtained from the fundamental representation and the symmetry type, which is described by the Young tableau. The basis for the fully symmetrical case is defined as

$$P_{q,s}\approx = S(u^a d^b s^c e^d), \quad (B1)$$

which are eigenstates of both $O^2$ and $O_3$, with eigenvalues

$$O^2_{q,s}\approx = o(o+1)_{q,s}\approx, \quad O_3 P_{q,s}\approx = o_3 P_{q,s}\approx, \quad (B2)$$
where $o \in \{q, \sigma, m, n, u, v\}$. The eigenvalues are not independent, but can be expressed in terms of $\alpha, \gamma, \beta, \delta$:

\[
q = (\alpha + \beta)/2, \quad q_3 = (\alpha - \beta)/2, \\
\sigma = (\gamma + \delta)/2, \quad \sigma_3 = (\gamma - \delta)/2, \\
m = (\alpha + \delta)/2, \quad m_3 = (\alpha - \delta)/2, \\
n = (\gamma + \beta)/2, \quad n_3 = (\gamma - \beta)/2, \\
u = (\alpha + \gamma)/2, \quad u_3 = (\alpha - \gamma)/2, \\
v = (\delta + \beta)/2, \quad v_3 = (\delta - \beta)/2.
\]

(B3)

Then it is straightforward to determine actions of all the raising and lowering superoperators on $P_{q,q',\sigma',\tau'}$:

\[
\begin{align*}
Q_+ P_{q,q',\sigma',\tau'} &= (q \mp q_3) P_{q,q',\sigma',\tau'}, \\
\Sigma_+ P_{q,q',\sigma',\tau'} &= (\sigma \mp \sigma_3) P_{q,q',\sigma',\tau'}, \\
M_+ P_{q,q',\sigma',\tau'} &= (m \mp m_3) P_{q+1/2,q_3+1/2,\sigma_3+1/2}, \\
N_+ P_{q,q',\sigma',\tau'} &= (n \mp n_3) P_{q+1/2,q_3+1/2,\sigma_3+1/2}, \\
U_+ P_{q,q',\sigma',\tau'} &= (u \mp u_3) P_{q+1/2,q_3+1/2,\tau_3+1/2}, \\
V_+ P_{q,q',\sigma',\tau'} &= (v \mp v_3) P_{q+1/2,q_3+1/2,\tau_3+1/2}.
\end{align*}
\]

(B4)

We note that the fully symmetrical basis are also eigenstates of the quadratic Casimir operator $C_1$ with common eigenvalue $3(N+4)/8$.

Analogous actions for the photon part are the simple harmonic oscillator relations:

\[
a |n\rangle = \sqrt{n} |n-1\rangle,
\]

\[
a^\dagger |n\rangle = \sqrt{n+1} |n+1\rangle.
\]

(B5)

**Appendix C: $|S, M\rangle$ $|S, M'\rangle$ representation**

In order to project the density operator from the SU(4) basis onto the $|S, M\rangle$ $|S, M'\rangle$ representation, let us first show that $M$ and $M'$ are related to the $P_{q,q',\sigma',\tau'}^{(s)}$ by $q_3 + \sigma_3 = M$ and $q_3 - \sigma_3 = M'$. To see this, defining $\hat{J}_3 = \sum_{j=1}^N \sigma_j^{(3)}/2$, we could get

\[
\begin{align*}
\hat{J}_3 P_{q,q',\sigma',\tau'}^{(s)} &= \frac{1}{2} (\alpha + \gamma - \beta - \delta) P_{q,q',\sigma',\tau'}^{(s)} = (q_3 + \sigma_3) P_{q,q',\sigma',\tau'}^{(s)}, \\
P_{q,q',\sigma',\tau'}^{(s)} \hat{J}_3 &= \frac{1}{2} (\alpha + \delta - \beta - \gamma) P_{q,q',\sigma',\tau'}^{(s)} = (q_3 - \sigma_3) P_{q,q',\sigma',\tau'}^{(s)},
\end{align*}
\]

(C1)

and by definition, we have

\[
\begin{align*}
\hat{J}_3 |S, M\rangle |S, M'\rangle &= M |S, M\rangle |S, M'\rangle, \\
|S, M\rangle |S, M'\rangle \hat{J}_3 &= M' |S, M\rangle |S, M'\rangle.
\end{align*}
\]

(C2)

Therefore, the complex coefficients from the $P_{q,q',\sigma',\tau'}^{(s)}$ basis contributing to the matrix element for $|S, M\rangle |S, M'\rangle$ all satisfy $q_3 + \sigma_3 = M$ and $q_3 - \sigma_3 = M'$.

With this in mind, we now describe a systematic algorithm to obtain the density matrix elements $D_{S,M,M'}^{m,n}$ from the SU(4) expansion coefficients $C_{m,n}^{q,q',\sigma',\tau'}$. We illustrate our method by considering in detail the elementary case of three atoms. The density matrix in the $|S, M\rangle |S, M'\rangle$ representation is block diagonal in $S$; the block matrices for all $S$ can be arranged in the shape of a pyramid as shown in Fig. 1(c). For instance, the base layer corresponds to $S = N/2$, with the matrix dimension being $(N + 1)^2$. The second layer has $S = N/2 - 1$ and dimension $(N - 1)^2$, and so on. Furthermore there are $n_S$ copies associated with each layer, so that $\sum_S (2S + 1)n_S = 2^N$. Taking $N = 3$ for example, there are two layers, $S = 3/2$ and $S = 1/2$ with $n_{3/2} = 1$ and $n_{1/2} = 2$, so that the Hilbert space dimension is $(3 + 1) + 2(1 + 1) = 2^3$.

The density matrix needs to be built from the bottom layer upwards. In the bottom layer, we find that the only element contributing to $|N/2, N/2\rangle \langle N/2, N/2\rangle$ is $P_{S_{2},N/2}^{m,n}$. So the top left corner is $D_{S_{2},N/2}^{m,n} = C_{m,n}^{S_{2},N/2}$. We next apply the lowering operator $\hat{J}_2 = \sum_{j=1}^N \sigma_j^{(2)}$ to iteratively generate $D_{S_{2},N/2,M}^{m,n}$ with $M = N/2 - 1, \ldots, -N/2$. To do this, we need the recursion relation

\[
D_{S_{2},N/2,M}^{m,n} = \langle S, M | \hat{J}_2 | S, M'\rangle = \frac{\langle S, M | (\hat{U}_2 + \hat{V}_2) P_{S_{2},N/2}^{m,n} | S, M'\rangle}{\sqrt{(S + M')(S - M' + 1)}}.
\]

(C3)

Therefore, with the actions of the raising and lowering operators [Eq. (B4)], we can derive all $D_{S_{2},N/2,M}^{m,n}$, i.e. the first row of the bottom layer. Using the fact that the density matrix is Hermitian and $C_{m,n}^{q,q',\sigma',\tau'} = (C_{m,n}^{q,q',\sigma',\tau'})^*$, we could get all the elements for the first column by $D_{S_{2},N/2,M}^{m,n} = (D_{S_{2},N/2,M}^{m,n})^*$. By repeatedly applying the recursion relation [Eq. (C3)] in each row, we then construct the full base layer. As an explicit example, we have constructed the bottom layer, i.e. $S = 3/2$ for the three atom case, and

\[
\begin{align*}
|S, M\rangle &|S, M'\rangle \\
\begin{pmatrix}
C_{m,n}^{S_{2},3/2} & 0 \\
0 & C_{m,n}^{S_{2},1/2}
\end{pmatrix}
\end{align*}
\]

(C4)

In order to illustrate the use of the recursion relation, we now show how we get $D_{3/2,1/2}^{m,n}$ from $D_{3/2,1/2}^{m,n}$. Because $\hat{U}_2 P_{3/2,1/2}^{(s)} = P_{1,1-1/2}^{(s)}$ and $\hat{V}_2 P_{1,1-1/2}^{(s)} = P_{1,1-1/2}^{(s)}$, we have

\[
D_{3/2,1/2}^{m,n} = (C_{m,n}^{3/2,1/2} + C_{m,n}^{1/2,1/2})/\sqrt{3}.
\]

To construct the next layer, we thus find out the top left matrix element first, and then apply the same procedure as before to determine the rest of the matrix elements. Let us first examine the three atom case. The $S = 1/2$ layer has two copies, each of which is a $2 \times 2$ matrix. To find the top left element $D_{3/2,1/2}^{m,n}$, noticing the constraint imposed by the trace of the density matrix, we derive $2D_{3/2,1/2}^{m,n} + D_{3/2,1/2}^{m,n} = 2(2C_{m,n}^{3/2,1/2} - 1)C_{m,n}^{1/2,1/2}/6. By applying the same method as in the bottom layer, we construct the block...
matrix for $S = 1/2$ layer

\[
\begin{pmatrix}
|\frac{1}{2}, \frac{1}{2}\rangle \\
|\frac{1}{2}, -\frac{1}{2}\rangle
\end{pmatrix}
= \begin{pmatrix}
\langle \frac{1}{2}, \frac{1}{2} | 
\frac{2C_{m,1}^{m,0}}{6} - C_{m,1/2}^{m,0/2} & C_{m,1/2}^{m,0/2} \\
C_{m,1/2}^{m,0/2} & \frac{2C_{m,0}^{m,1}}{6} - C_{m,1/2}^{m,0/2}
\end{pmatrix}
\begin{pmatrix}
|\frac{1}{2}, \frac{1}{2}\rangle \\
|\frac{1}{2}, -\frac{1}{2}\rangle
\end{pmatrix}
\]  \hspace{1cm} (C5)

Therefore in general, if we suppose that we have constructed the block matrix for $S' > S$, the formula to find the top left matrix element $D_{S,S,S}^{m,n}$ for layer $S$ is

\[
\sum_{S' \leq S} n_{S'} D_{S',S,S}^{m,n} = C_{m,n/2,S,0}^{m,n}.
\]  \hspace{1cm} (C6)

Having the top left matrix element for each layer $S$, we can easily construct the $(2S + 1) \times (2S + 1)$ block matrix by applying the recursion relation based on the angular momentum lowering operator. Repeated iteration of these steps systematically fills in all sites of the pyramid.

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