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1994 J. Phys. A: Math. Gen. 27 609

(http://iopscience.iop.org/0305-4470/27/3/007)

J. Phys. A: Math. Gen. 27 (1994) 609–630. Printed in the UK

Dynamical invariants for time-evolution of open quantum systems in finite dimensions

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Received 9 November 1992

Abstract. Various equivalent representations of dynamical invariants (or constants of motion) are derived for N-level systems in mixed states with particular emphasis on so-called coherence vector invariants. They appear as certain homogeneous forms in the real solutions of the von Neumann equation with coefficients given by multilinear forms in the completely symmetric structure constants of the Lie-algebra of $SU(N)$. The treatment is motivated by the close analogy between Lax pairs for classical dynamical systems and pairs given by density and Hamilton operators. In both cases the underlying mathematical structure is essentially determined by the properties of symmetric functions. However, in the quantum case more theorems of general validity can be derived due to the peculiar properties of density operators. In particular, the maximum number of functionally independent invariants in relation to the spectral properties is obtained, as well as bounds on their order of magnitude, the latter from an extremal property analysis. From group-theoretical methods a complete classification of all possibilities of completely incoherent state dynamics is deduced. As a by-product a simple algorithm for the explicit determination of all $(N-1)$-dimensional irreducible representation matrices of the symmetric group S_N and an associated construction of hyperpolyhedra is worked out. Finally, the importance of invariants is stressed for the control of numerical accuracy in large-scale computations in very high dimensions which, after taking partial traces, can be used for the description of irreversible processes. In summary, the results will be of practical relevance for applications to problems of short-time dynamics in molecular laser spectroscopy, quantum optics and magnetic resonance.

1. Introduction

Time-evolution of physical systems is mostly determined by some differential or integro-differential laws whose solution has always posed difficult problems. In general, the full global time-evolution may not be known but certain particular aspects like, for instance, short-time or long-time behaviour may be accessible by some approximate treatments. In view of these difficulties those quantities which are conserved during time-evolution have always played a central role since, in many cases, they provide exact answers to some selected questions. Particularly in classical Hamiltonian mechanics, the search for transformations to action-angle variables or, else, for a complete set of constants of motion in involution has a remarkable history with many successful results [1–3]. Furthermore, even for more general dynamical systems like those obeying, for instance, equations of Korteweg–de Vries type, the pioneering ideas of Lax [4] have led to surprising progress with wide applications [5]. The developed methods have, in turn,

0305-4470/94/030609+22$07.50

become extremely fruitful in the theory of Hamiltonian systems and led to such unexpected proofs as, for instance, complete integrability of Toda-lattice problems and many others [6–11]. The central point is a possible existence of transformations of complicated nonlinear equations to a linear Lie-structure of type

$$\dot{L}=[B, L] \tag{1.1}$$

where the so-called Lax-pair L and B is given by matrices in some higher dimension N and where, in many cases, even the limit $N\to\infty$ may be included. In this formulation time-evolution appears then as isospectral deformations and the proof of complete integrability is straightforward.

It is surprising that the above ideas have not been well enough appreciated, and applied similarly for quantum dynamics since the von Neumann equation

$$\dot{\rho}=-\mathrm{i}[H, \rho] \tag{1.2}$$

where ρ is a density operator and H a Hamiltonian, has an analogous mathematical structure. The trivial difference is just that the general similarity transformations of the classical case are replaced by unitary transformations in the quantal case due to the self-adjointness of operators and the additional imaginary factor. A few comments regarding the applicability of (1.2) are necessary. In the general description of open quantum systems the appearance of irreversibility causes the dynamics to become non-unitary. In particular, in the weak or singular-coupling limits or in the more general formulation derived from the concept of complete positivity [12–15], time-evolution is given by a quantum dynamical semigroup whose infinitesimal generator structure is exactly known. After imposing the uniqueness condition $\mathrm{Tr}(H)=0$ the generator can always be written as the sum of a Hamiltonian part as in (1.2) plus a non-Hamiltonian unitarity-violating part. More than that, in most physically interesting problems the characteristic time-scale set by H for Hamiltonian evolution is appreciably shorter than that caused by the second part for dissipation. For instance, in optical problems H contains transition times of the order of femtoseconds whereas relaxation frequently occurs on a picosecond or slower scale. Moreover, the analogous separation for ESR is roughly between nano- and microseconds and for NMR between micro- and less than milliseconds, disregarding exceptional cases. Roughly speaking, a time-scale separation of about three orders of magnitude or more is almost the rule in the majority of cases. This fact, by the way, is the basis of success of the powerful pulse-sequence methods in NMR [16, 17]. In summary, there is always a first time-interval on which the dynamics is unitary to good approximation and (1.2) can be used in this form without additional terms. Note also that even for closed systems the initial state may be a mixed state in the sense of quantum statistical mechanics [18] and (1.2) cannot be circumvented by a Schrödinger equation in terms of a wavefunction description. It is under all the above circumstances that either analytical or numerical methods for the solution of the von Neumann equation may be supplemented by fully taking advantage of the existence of dynamical invariants, and it is certainly worth developing related details, as will be done in the following sections.

In sections 2, 3 and 4 various representations of the invariants are given which may be transformed into each other and which, depending on technical convenience, may be used in one or the other explicit form. However, in terms of the real solutions of (1.2) in a higher-dimensional space the natural choice is offered by the coherence-vector invariants derived in section 4. Since the importance of invariants is emphasized also in relation to numerical solutions, it is of interest to have a knowledge of their order

of magnitude or, in other words, of their extremal properties, which will be considered in section 5. Answers to questions about global dynamics like, for instance, possibilities of reaching desired final states starting from selected initial states, can be found successfully for completely incoherent states. Some attempts have been made earlier in this direction [19–24] and section 6 gives a complete account by Lie-algebraic and other group-theoretical methods for these cases. Then, section 7 is devoted to general considerations on the importance of invariants in numerical problems with a quantitative explanation of trends in terms of a low-dimensional model. Finally, the concluding section 8 provides some outlook to the use of invariants in large-scale computations for irreversible processes.

2. Polynomial invariants for unitary dynamics

The time-dependent mixed states of an N-level system are given in terms of an $(N\times N)$-density matrix $\rho(t)$ with $\mathrm{Tr}(\rho(t))=1$ and $\mathrm{Tr}(\rho^2(t))<1$. The pure states with $\mathrm{Tr}(\rho^2(t))=1$ can be described by wavefunctions and will not be of relevance in the following considerations. Time-evolution is governed by

$$\dot{\rho}(t)=-\mathrm{i}[H(t),\rho(t)] \tag{2.1}$$

where H is a self-adjoint Hamiltonian. The solution to (2.1) can always be written as

$$\rho(t)=U(t)\rho(0)U^*(t) \tag{2.2}$$

in terms of a unitary transformation

$$U(t)=\begin{cases}\exp(-\mathrm{i}Ht) & H \text{ time-independent} \qquad (2.3)\\ T\exp\left(-\mathrm{i}\displaystyle\int_0^t H(\tau)\,\mathrm{d}\tau\right) & H \text{ time-dependent} \qquad (2.4)\end{cases}$$

where T is the Dyson time-ordering operator. The diagonal form of $\rho(0)$ will be denoted by D_0 with the set of eigenvalues $\{\lambda_i\}_1^N$, $0\leqslant\lambda_i\leqslant 1$ $(\forall i)$, as obtained through a unitary transformation V according to $D_0=V^*\rho(0)V$. This gives, for (2.2)

$$\rho(t)=W(t)D_0W^*(t) \qquad W(t)=U(t)V \qquad W^*=W^{-1}. \tag{2.5}$$

Clearly, $\rho(t)$ and $\rho(0)$ have identical spectrum obtained from the characteristic polynomial

$$P_N(\lambda)=\mathrm{Det}[\lambda\mathbb{1}_N-\rho(t)]=\mathrm{Det}[\lambda\mathbb{1}_N-\rho(0)] \tag{2.6}$$

by setting $P_N=0$. The representation in terms of powers is then

$$P_N(\lambda)=\lambda^N-I_1\lambda^{N-1}+I_2\lambda^{N-2}-\ldots+(-1)^{N-1}I_{N-1}\lambda+(-1)^NI_N \tag{2.7a}$$

with coefficients given by

$$I_1[\rho(t)]=\mathrm{Tr}[\rho(t)]=1 \tag{2.7b}$$

$$I_2[\rho(t)]=\sum_{i<k}^{N}(\rho_{ii}(t)\rho_{kk}(t)-|\rho_{ik}(t)|^2). \tag{2.7c}$$

$I_l[\rho(t)] \equiv I_l$: sum of leading minors of lth order $(1 \leqslant l \leqslant N)$

$$I_N[\rho(t)] = \mathrm{Det}[\rho(t)]. \tag{2.7d}$$

In terms of the roots one has

$$P_N(\lambda) = \prod_{i=1}^{N} (\lambda - \lambda_i) \tag{2.8}$$

and a comparison with (2.7*a*) shows that the coefficients I_l are so-called elementary symmetric functions [25, 26] on the spectrum of ρ

$$I_l(\lambda_1, \lambda_2, \ldots, \lambda_N) = \sum_{m_1 < m_2 < \ldots < m_l}^{N} \lambda_{m_1} \lambda_{m_2} \ldots \lambda_{m_l} \tag{2.9}$$

since an arbitrary permutation of the roots leaves all I_ls invariant. The most important consequence is now that due to (2.6) every $I_l(\lambda_1, \lambda_2, \ldots, \lambda_N) = I_l[\rho(t)]$ is a dynamical invariant (or constant of motion) satisfying

$$\frac{\mathrm{d}}{\mathrm{d}t} I_l[\rho(t)] = 0 \qquad 1 \leqslant l \leqslant N. \tag{2.10}$$

The important question as to the maximum number and mutual independence of these quantities can be answered by recalling the main theorem on symmetric functions [26, 27]. It states that the elementary symmetric functions form a complete basis in the 'nonlinear' space of all entirely rational functions which are symmetric in N variables and of arbitrary degree. The notion of a 'nonlinear' space means that any of its elements can be expressed as a polynomial in the I_ls in a unique way. Thus, the maximum number $N_{\max}$ of functionally independent invariants is obtained for faithful states ρ $(\mathrm{Det}(\rho) \neq 0)$ with distinct roots, in which case $N_{\max} = N$. For non-faithful states of rank $r < N$ with all non-zero roots still distinct, one finds directly from (2.9) $N_{\max} = r$. More delicate is the case of degeneracies in the spectrum as treated in appendix 1. Here, we just summarize the results. The first trivial case concerns the central state

$$\rho = \zeta = \frac{1}{N} \mathbb{1}_N \tag{2.11}$$

for which all invariants are just a function of N and l (and of I_1, but here, $I_1 = 1$)

$$I_l = \left(\frac{N!}{N^l (N-l)!\, l!} \right) I_1 \qquad 1 \leqslant l \leqslant N. \tag{2.12}$$

Thus, there is only one free invariant I_1 and $N_{\max} = 1$. Arbitrary states with $r \leqslant N$ are characterized by $N_{\max} < r$ if all non-zero roots have the same multiplicity g with the consequence that $N_{\max} = r/g$. This follows from a reduction of the elementary symmetric functions of r variables to non-elementary but still symmetric functions all of which can be expressed as polynomials in elementary ones belonging to a lower-dimensional problem in m fewer distinct variables $(m < r)$. As a trivial corollary one finds that $N_{\max} = r$ whenever r is prime. As is clear from the treatment in appendix 1, all the above conditions are sufficient for an analysis of the maximum number of functionally independent invariants. However, no general proof could be formulated (so far) in order to show that the given conditions are also necessary. At the moment, this remains an open question although some simple explicit calculations for low N support strong evidence for necessity.

3. Trace-invariants

Since the trace of a matrix is a unitary invariant one can generate a set K_m of different integrals of motion by taking the trace of successive powers of the density matrix

$$K_m = \mathrm{Tr}\{\rho^m(t)\} = \mathrm{Tr}\{\rho^m(0)\} = \sum_{i=1}^{N} \lambda_i^m. \tag{3.1}$$

By this procedure independent invariants are obtained in the most general case for $m \leqslant N$ only. This is immediately clear from the Cayley–Hamilton theorem [28] which states that any matrix ρ with characteristic polynomial $P_N(\lambda) = 0$ fulfills ($\rho^0 \equiv \mathbb{1}_N$)

$$P_N(\rho) = \rho^N - I_1\rho^{N-1} + \ldots + (-1)^{N-1} I_{N-1}\rho + (-1)^N I_N \mathbb{1}_N = \mathbb{0}_N. \tag{3.2}$$

Again, as long as unitary dynamics is considered, the time is arbitrary. Then, from $\mathrm{Tr}(\rho P_N(\rho)) = 0$ one obtains

$$K_{N+1} = I_1 K_N - I_2 K_{N-1} + \ldots + (-1)^{N+1} I_N K_1 \tag{3.3}$$

and K_{N+1} appears, indeed, as being 'linearly' dependent upon all lower-degree invariants. The same conclusion applies to any higher degree. Strictly speaking, the notion of 'linear' dependence is misleading in the sense that the theory is nonlinear in N variables and, in fact, all K_ms ($m \leqslant N$) are again certain polynomials in the basic I-invariants. This follows directly from the main theorem on symmetric functions quoted in the preceding section since, again, any K_m in (3.1) is a completely symmetric function of the roots. In order to obtain relations between the polynomial and trace-invariants one may take the trace of (3.2) to find

$$\mathrm{Tr}\{P_N(\rho)\} = K_N - I_1 K_{N-1} + \ldots + (-1)^{N-1} I_{N-1} K_1 + (-1)^N I_N N = 0. \tag{3.4}$$

With the definition $I_0 \equiv 1$ this equation must hold for any $N \geqslant 2$ but, in a somewhat heuristic first attempt, one may even set $N = 1$ to obtain $K_1 = I_1$. Surprisingly, this is true for arbitrary N since, according to the earlier definitions, $K_1 = I_1 = \mathrm{Tr}(\rho)$. Next, set $N = 2$ to obtain

$$K_2 = I_1^2 - 2I_2. \tag{3.5}$$

However, for arbitrary N we have

$$K_2 = \sum_{i=1}^{N} \lambda_i^2 \qquad I_1^2 = \sum_{i=1}^{N} \lambda_i^2 + 2\sum_{i<k}^{N} \lambda_i\lambda_k \qquad I_2 = \sum_{i<k}^{N} \lambda_i\lambda_k$$

and find, again, that the relation (3.5) is universal despite having set $N = 2$. This remains true for any invariant of higher degree. In this way practical recursion relations are derived which can be solved, for instance, for any K_m as a function of the I_ls ($1 \leqslant l \leqslant m$). The first five equations, valid for arbitrary N, are as follows,

$$\begin{aligned} &K_1 = I_1 \qquad K_2 = I_1^2 - 2I_2 \qquad K_3 = I_1^3 - 3I_1I_2 + 3I_3 \\ &K_4 = I_1^4 - 4I_1^2I_2 + 4I_1I_3 + 2I_2^2 - 4I_4 \\ &K_5 = I_1^5 - 5I_1^3I_2 + 5I_1^2I_3 + 5I_1I_2^2 - 5I_1I_4 - 5I_2I_3 + 5I_5. \end{aligned} \tag{3.6}$$

The final general result is that, due to (3.4) and an equation like (3.3) but for $N+l=s$, one has the two relations

$$K_s - I_1 K_{s-1} + \ldots + (-1)^{s-1} I_{s-1} K_1 + (-1)^s I_s s = 0 \qquad s \leqslant N \tag{3.7}$$

$$K_s - I_1 K_{s-1} + \ldots + (-1)^{N-1} I_{N-1} K_{s-N+1} + (-1)^N I_N K_{s-N} = 0 \qquad s > N. \tag{3.8}$$

These are known as 'Newton's formulas'. For more details, general proofs and interesting historical remarks see [26, 27, 29, 30].

As is evident, particularly from (3.6), any symmetric function containing integer coefficients is expressible in terms of the I_is, again by integer coefficients [26]. On the other hand, the elementary symmetric functions can be represented in terms of non-elementary ones but by rational coefficients. An example is provided by the formulas inverse to those in (3.6):

$$\begin{aligned} &I_1 = K_1 \qquad I_2 = \tfrac{1}{2}(K_1^2 - K_2) \qquad I_3 = \tfrac{1}{6}[K_1^3 - 3K_1 K_2 + 2K_3] \\ &I_4 = \tfrac{1}{24}[K^4 - 6K_1^2 K_2 + 8K_1 K_3 + 3K_2^2 - 6K_4] \\ &I_5 = \tfrac{1}{120}[K_1^5 - 10K_1^3 K_2 + 20K_1^2 K_3 + 15K_1 K_2^2 - 30K_1 K_4 - 20K_2 K_3 + 24K_5]. \end{aligned} \tag{3.9}$$

For practical applications it seems to be easier to calculate trace-invariants and to use, if necessary, the inverse formulas to obtain the I-invariants.

4. Coherence-vector invariants

For many purposes it is advantageous to represent $\rho(t)$ by $M = N^2 - 1$ real-valued functions $v_k(t)$ considered to be the components of a so-called coherence-vector

$$v(t) = (v_1(t), v_2(t), \ldots, v_M(t))^T \in \mathbb{R}^M. \tag{4.1}$$

The decomposition

$$\rho(t) = \frac{1}{N} \mathbb{1}_N + \sum_{k=1}^{M} v_k(t) F_k \tag{4.2}$$

is then in terms of a complete orthonormalized set of $(N \times N)$ matrices $\{F_i\}_1^M$ with

$$F_i = F_i^* \qquad \mathrm{Tr}(F_i) = 0 \tag{4.3}$$

$$\mathrm{Tr}(F_i F_k) = \delta_{ik} \tag{4.4}$$

$$v_k(t) = \mathrm{Tr}(\rho(t) F_k). \tag{4.5}$$

In particular, we use a special representation of the infinitesimal generators of $SU(N)$ [14, 15] with

$$F_i F_k = \frac{1}{N} \mathbb{1}_N \delta_{ik} + \sum_{s=1}^{M} z_{iks} F_s \tag{4.6}$$

$$z_{iks} = \tfrac{1}{2}(d_{iks} + \mathrm{i} f_{iks}) \tag{4.7}$$

where the d_{iks}s are the completely symmetric and the f_{iks}s the completely antisymmetric real structure constants of the Lie-algebra. By considering the former trace-invariants

and using (4.2) one sees from

$$K_l = \mathrm{Tr}(\rho^l) = \sum_{k=0}^{l} \binom{l}{k} N^{k-l} \mathrm{Tr}\left(\sum_{i=1}^{M} v_i(t) F_i \right)^k \tag{4.8}$$

that any expression of the form

$$\mathscr{L}_k = \mathrm{Tr}\left(\sum_{n_1, n_2, \ldots, n_k = 1}^{M} v_{n_1}(t) v_{n_2}(t) \ldots v_{n_k}(t) F_{n_1} F_{n_2} \ldots F_{n_k} \right) \tag{4.9}$$

is an invariant, again. However, an evaluation of the trace by repeatedly using the contraction (4.6) decomposes $\mathscr{L}_k$ for $k \geqslant 4$ into more elementary invariants denoted by L_i. As a consequence, the trace-invariants can now be expressed as

$$K_l = P_l(L_1, L_2, \ldots, L_l) \tag{4.10}$$

where P_l is a polynomial in all L_is ($i \leqslant l$) with the highest L_l occurring in the first power with weight 1. An example is worked out in appendix 2. Here, we merely give the resulting homogeneous forms in the coherence-vector components with coefficients that are products of symmetric structure constants only,

$$\begin{aligned}
&L_1 = 1 \qquad L_2 = \|\boldsymbol{v}\|^2 = \sum_{i=1}^{M} v_i^2 \\
&L_3 = \tfrac{1}{2} \sum_{i,k,l} d_{ikl} v_i v_k v_l \\
&L_4 = \tfrac{1}{4} \sum_{\substack{i,k,l \\ m,p}} d_{ikp} d_{lmp} v_i v_k v_l v_m \\
&L_5 = \tfrac{1}{8} \sum_{\substack{i,k,l,m \\ n,p,q}} d_{ikp} d_{lmq} d_{npq} v_i v_k v_l v_m v_n .
\end{aligned} \tag{4.11}$$

The explicit polynomials (4.10) for the trace-invariants are as follows ($\alpha = 1/N$):

$$\begin{aligned}
&K_1 = L_1 \qquad K_2 = \alpha L_1^2 + L_2 \\
&K_3 = \alpha^2 L_1^3 + 3\alpha L_2 + L_3 \\
&K_4 = \alpha^3 L_1^4 + 6\alpha^2 L_2 + \alpha(L_2^2 + 4L_3) + L_4 \\
&K_5 = \alpha^4 L_1^5 + 10\alpha^3 L_2 + 5\alpha^2(L_2^2 + 2L_3) + \alpha(2L_2 L_3 + 5L_4) + L_5 .
\end{aligned} \tag{4.12}$$

Note that the explicit dependence upon L_1 has been kept to enable a possible use of (4.12) also for classical dynamical problems where $K_l = L_1$ need not be equal to 1.

5. Extremal properties

Since the spectrum of any state ρ is restricted to the interval [0, 1] all invariants will be subject to corresponding lower and upper bounds. Note first that for the otherwise uninteresting pure states one has $I_1 = 1$, $I_l = 0$ ($2 \leqslant l \leqslant N$) and $K_l = 1$ ($1 \leqslant l \leqslant N$). In the following we will always presuppose genuine mixed states with $\mathrm{Tr}(\rho^2) < \mathrm{Tr}(\rho) = 1$. Then,

from (2.9) one finds

$$0<I_l<\binom{N}{l} \qquad 2\leqslant l\leqslant N \tag{5.1}$$

and from (3.1)

$$0<K_l<1 \qquad 2\leqslant l\leqslant N. \tag{5.2}$$

It is immediately clear that the upper limit in (5.1) is much too high since it follows from (3.6) for I_2, for instance, that $0<I_2<\frac{1}{2}$ independent of N. Furthermore, from methods described in [15] one can deduce exact bounds, for instance, for $L_2=\|v\|^2$,

$$0<L_2<1-\frac{1}{N}. \tag{5.3}$$

In any case, rough estimates may be too unsatisfactory. Fortunately, due to the very peculiar properties of the symmetric functions it will be possible to derive exact bounds for all polynomial invariants and, consequently, also for all other invariants via the reciprocal formulas given earlier. Thus, one has to determine under what conditions on the spectrum of ρ, or else, on the particular distribution of eigenvalues $\{\lambda_i\}_1^N$ do the I_l-invariants attain their maximum values.

Consider the variational functions

$$\phi_l=I_l+\mu_l\varphi \tag{5.4a}$$

where φ is the trace constraint

$$\varphi=\sum_{i=1}^{N}\lambda_i-c=I_1-c=0 \tag{5.4b}$$

and μ_l, the Lagrange multipliers. Of course, for density matrices, $c=1$ ($I_1=1$) and $\varphi=I_1-1=0$ but we will keep a general value of I_1 in all formulas to enable a possible use also in those problems of classical dynamical systems for which one can construct Lax-pairs (L, B) with $L>0$ where the trace need not be equal to 1. A general recursion relation is derived from the extremal conditions

$$\frac{\partial\phi_l}{\partial\lambda_k}=\frac{\partial I_l}{\partial\lambda_k}+\mu_l=0 \qquad 1\leqslant k\leqslant N \tag{5.5}$$

together with the homogeneity relations. In fact, (2.9) implies that I_l is a homogeneous function of degree l in N variables satisfying the Euler condition

$$\sum_{k=1}^{N}\lambda_k\frac{\partial I_l}{\partial\lambda_k}=lI_l. \tag{5.6}$$

As a first result, (5.5) and (5.6) yield

$$I_l=-\frac{\mu_l}{l}I_1. \tag{5.7}$$

A determination of μ_l is possible by symmetry considerations applied to the N equations (5.5). Note, for instance, that the fixed sequence with $l-1$ consecutive indices and a last arbitrary one like $\lambda_1\lambda_2\ldots\lambda_{l-1}\lambda_{m_l}$ occurs $N-l+1$ times since $m_l=l, l+1, \ldots, N$. Because all possible choices of any l indices out of N occur in the summation for I_l the

above argument is true for any fixed sequence $\lambda_{m_1}\lambda_{m_2}\ldots\lambda_{m_l}$ with non-consecutive indices, too. A selected $\partial I_l/\partial\lambda_k$ is no longer symmetric in the variables but upon summation of all derivatives, a completely symmetric function is restored, in fact a multipole of I_{l-1}, that is

$$\sum_{k=1}^{N}\frac{\partial I_l}{\partial\lambda_k}=(N-l+1)I_{l-1}. \tag{5.8}$$

From (5.5) we obtain

$$\mu_l=-\frac{N-l+1}{N}I_{l-1} \tag{5.9}$$

and from (5.7) the final recursion relation

$$I_l=\left(\frac{N-l+1}{Nl}I_1\right)I_{l-1} \tag{5.10}$$

valid under extremal conditions. Within the manifold of genuine mixed states the extremum must be a maximum since the minimum ($l\geqslant 2$) cannot be less than zero and this value is attained by the pure states only. Therefore, one arrives at the important conclusion that all invariants are simultaneously maximized under spectral conditions still to be derived. For this purpose consider the solution of (5.5) for I_2. By subtracting pairwise any two equations from each other one finds the unique solution

$$\lambda_1=\lambda_2=\ldots=\lambda_N \tag{5.11}$$

and, therefore, all invariants take on their maximum value for the central state

$$\zeta=\frac{1}{N}\mathbb{1}_N. \tag{5.12}$$

This reminds one of the property of the von Neumann entropy [18]

$$S[\rho]=-\mathrm{Tr}(\rho\ln\rho)=-\sum_{i=1}^{N}\lambda_i\ln\lambda_i \tag{5.13}$$

also with unique maximum for ζ given by

$$S[\zeta]=\ln N. \tag{5.14}$$

In conclusion, we have derived the exact upper bound for all invariants, realized uniquely by the central state and given explicitly by

$$I_l=\frac{N!}{N^l(N-l)!\,l!}. \tag{5.15}$$

6. Group-theoretical analysis of completely incoherent states

As is obvious from the symmetric functions, invariants imply certain symmetries with associated groups and these set all restrictions on the availability of states by the dynamical transformations. A particularly distinguished set of states is given by all diagonal density matrices, which we will denote as 'completely incoherent states' with respect to a chosen basis. The latter is usually determined by the eigenstates of an

appropriate time-independent unperturbed Hamiltonian. The reason why particular attention has been drawn to this selected set of states in applications to problems of population transfer in quantum optics and magnetic resonance is mainly due to the fact that, on a short time-scale, they are somewhat 'longer-lived' than coherent states and are, therefore, possibly more accessible to experimental manipulations. This is quickly seen by expanding $\rho(t)$ in (2.2) yielding, for the occupation probabilities, for instance,

$$\rho_{ii}(t) \simeq \rho_{ii}(0) + it \sum_{l=1}^{N} \{\rho_{il}(0)H_{li} - H_{il}\rho_{li}(0)\} + O(t^2). \tag{6.1}$$

Thus, the first-order contribution vanishes for diagonal initial states and this explains what is meant by 'longer-lived' on a short time-scale. In particular, spin ensembles exposed to a static field in thermodynamic equilibrium only show magnetization parallel to the field and this corresponds to stationary diagonal states [31].

Starting from a given diagonal initial state $\rho(0)$ with populations $\{\lambda_1^{(0)}, \lambda_2^{(0)}, \ldots, \lambda_N^{(0)}\}$ one can ask the question as to the possible completely incoherent states ever achievable by any unitary dynamics and the associated changes in populations. The answer is almost trivial on the basis of the theory developed so far. Since it follows from the uniqueness theorem for the product decomposition in (2.8) that there is a one-to-one correspondence between all eigenvalues $\{\lambda_i^{(0)}\}_1^N$ and all invariants $\{I_l\}_1^N$ no final population sequence $\{\lambda_i^{(T)}\}_1^N$ at any later time T is ever possible except for those obtained from some reordering of the given values. In other words, the only freedom left for population transfer is given by the set of all permutations. The corresponding symmetry group in the case of a completely distinct spectrum is, consequently, the symmetric group S_N of order $N!$. In the case of degeneracies the possibilities are reduced accordingly. It must be stressed that this result is independent of any details of the Hamiltonian or, in physical terms, of special pulse forms, strengths, durations or frequencies of applied pumping fields. It is a pure consequence of unitary dynamics valid as long as irreversible processes do not yet come into play.

In the following we will derive some important connections between the group S_N, some of its irreducible representations and the coherence-vector representation of diagonal density matrices. It is convenient to decompose ρ into a sum of its dynamically invariant plus its traceless part, as in (4.2)

$$\rho = \zeta + \sigma \tag{6.2}$$

but only diagonal matrices σ with $\mathrm{Tr}(\sigma) = 0$ are considered and we write

$$\sigma = \begin{pmatrix} \mu_1 & & & 0 \\ & \mu_2 & & \\ & & \ddots & \\ 0 & & & \mu_N \end{pmatrix} \qquad \sum_{i=1}^{N} \mu_i = 0 \tag{6.3}$$

with the restriction

$$-\frac{1}{N} \leqslant \mu_i \leqslant 1 - \frac{1}{N} \qquad 1 \leqslant i \leqslant N. \tag{6.4}$$

Since $\mathrm{Tr}(\sigma) = 0$ one may choose μ_N, for instance, as redundant and introduce a vector in terms of the $N-1$ independent quantities

$$\mu = (\mu_1, \mu_2, \ldots, \mu_{N-1})^T. \tag{6.5}$$

In the decomposition (4.2) only the subset of diagonal generators of $SU(N)$ is needed. They form the Abelian Cartan subalgebra [32] of dimension $N-1$ and will be denoted by $\{Q_k\}_1^{N-1}$. According to the general construction their explicit form is given by [14]

$$Q_k=[(N-k+1)(N-k)]^{-1/2}\begin{pmatrix} 1 & & & & & \\ & 1 & & & & \\ & & \ddots & & & \\ & & & 1 & & \\ & & & & -(N-k) & \\ & & & & & 0 \\ & & & & & & \ddots \\ & & & & & & & 0 \end{pmatrix} \qquad (6.6)$$

where the numbering has been chosen with respect to the increasing last non-zero diagonal element. The corresponding coherence-vector will be denoted by

$$w=(w_1, w_2, \ldots, w_{N-1})^T \qquad (6.7)$$

such that we have a decomposition

$$\sigma=\sum_{k=1}^{N-1} w_k Q_k. \qquad (6.8)$$

Note that for given w equation (6.8) yields N equations to determine μ, one of them being redundant. Thus, there is a one-to-one correspondence between w and μ and one can write in terms of a non-singular $(N-1)\times(N-1)$ matrix A

$$\mu=Aw \qquad (6.9)$$

with matrix elements

$$A_{ik}=\begin{cases} [(N-k)(N-k+1)]^{-1/2} & 1\leqslant i\leqslant N-1 \quad 1\leqslant k\leqslant N-i \\ -(i-1)[(N-i)(N-i+1)]^{-1/2} & 2\leqslant i\leqslant N-1 \quad k=N-i+1 \\ 0 & \text{otherwise.} \end{cases} \qquad (6.10)$$

Detailed inspection of (6.8) shows that for given μ, on the other hand, w can be obtained by successive backward recursion. Due to the particular structure of the generators also the inverse A^{-1} is found in analytical form with matrix elements

$$(A^{-1})_{ik}=\begin{cases} \left(\dfrac{N}{N-1}\right)^{1/2} & i=1 \quad 1\leqslant k\leqslant N-1 \\ [(N-i)(N-i+1)]^{-1/2} & 2\leqslant i\leqslant N-1 \quad 1\leqslant k\leqslant N-i \\ -(N-i)[(N-i)(N-i)(N-i+1)]^{-1/2} & 2\leqslant i\leqslant N-1 \quad k=N-i+1 \\ 0 & \text{otherwise.} \end{cases} \qquad (6.11)$$

Consider now an entirely distinct spectrum of σ or, equivalently, of ρ. Then, any permutation of roots within the sequence

$$\{\mu_1, \mu_2, \ldots, \mu_N\} \tag{6.12}$$

will generate a new coherence-vector giving rise to identical invariants. For the reduced μ-vector the above operation is effected by a permutation, too, but if the latter involves the redundant μ_N one just has to replace the partner in the corresponding transposition by

$$-\sum_{i=1}^{N-1} \mu_i.$$

To analyse the details a few notions and theorems from the theory of the symmetric group S_N [33, 34] will be needed. Any permutation π from S_N is uniquely determined by a product of disjoint cyclic factors and any cycle can be written as a product of transpositions (ik). Furthermore, any transposition can be decomposed into factors of neighbouring transpositions $(l, l+1)$ and, consequently, the group is entirely determined by the minimal generating set

$$G_{N-1} = \{(12), (23), \ldots, (N-1, N)\} \tag{6.13}$$

consisting of all transpositions of successive numbers. Therefore, the following considerations are restricted to the $N-1$ elements of G_{N-1} and any general permutation can be obtained by appropriate multiplication rules. For a given initial condition $\rho(0)$ we denote the sequence by $\mu^{(0)}$ and the corresponding coherence-vector by $w^{(0)}$ where

$$w^{(0)} = A^{-1}\mu^{(0)}. \tag{6.14}$$

Then, for any $\pi \in S_N$

$$w^{(\pi)} = A^{-1}\mu^{(\pi)} \tag{6.15}$$

and in this way $N!$ different vectors are generated. The transformation $w^{(0)} \xrightarrow{\pi} w^{(\pi)}$ can also be written as

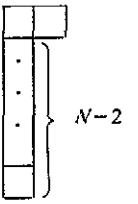
$$w^{(\pi)} = D(\pi)w^{(0)}. \tag{6.16}$$

Note that a representation of all elements of S_N necessarily involves all $N-1$ vector components and we conclude that D must constitute an $(N-1)$-dimensional irreducible representation. This is another way of proving that a representation of this dimension must exist for arbitrary N, whereas common proofs follow directly from the partitions and corresponding Young tableaux. In fact, according to the general theory there is always an irreducible representation characterized by

$$[N-1, 1] \qquad \underbrace{\begin{array}{|c|c|c|c|}\hline & \cdots & \\ \hline \\ \cline{1-1}\end{array}}_{N-2} \tag{6.17}$$

whose dimension is determined by the number of ways to place the numbers $2, 3, \ldots, N$ into the single box in the second row of the graph. For completeness note that for

$N \geqslant 4$ there exists also the inverted tableau

$[2, 1^{N-2}]$ 

(6.18)

belonging to a second irreducible representation of the same dimension $N-1$ but not equivalent to the first one. This case is compatible with a corresponding inverted enumeration of the generators $\{Q_k\}_1^{N-1}$ obtained by replacing k by $N-k$. We do not analyse the further exceptional case for $N=6$ with two additional representations of dimension 5 characterized by the partitions [3, 3] and [2, 2, 2] since, according to our choice of generators, only the case displayed in (6.17) will be needed.

An important practical result of this analysis is that the representation matrices can now easily be constructed in the desired explicit form. To do this introduce simple permutation matrices $P(l, l+1)$ for all neighbouring transpositions such that

$$\mu^{(l,l+1)} = P(l, l+1)\mu^{(0)}. \tag{6.19}$$

The matrix elements for $l \leqslant N-2$ are explicitly given by

$$\begin{aligned} &P_{ik}(l, l+1) = \delta_{ik} \qquad i \neq l, l+1 \qquad k \neq l, l+1 \\ &P_{ll}(l, l+1) = P_{l+1,l+1}(l, l+1) = 0 \\ &P_{l,l+1}(l, l+1) = P_{l+1,l}(l, l+1) = 1. \end{aligned} \tag{6.20}$$

Since for $l = N-1$ the result of exchanging μ_{N-1} with μ_N must be written in terms of the independent μ_i-values the last matrix missing in (6.20) has the special form

$$P_{ik}(N-1, N) = \begin{cases} \delta_{ik} & 1 \leqslant i, k \leqslant N-2 \\ -1 & i = N-1 \qquad 1 \leqslant k \leqslant N-1. \end{cases} \tag{6.21}$$

Finally, in terms of the analytically constructed matrices A, A^{-1} and $P(l, l+1)$ one finds the $(N-1)$-dimensional irreducible representation matrices $D(l, l+1)$ of S_N from (6.19) and (6.14), (6.15) by multiplication

$$D(l, l+1) = A^{-1}P(l, l+1)A \qquad 1 \leqslant l \leqslant N-1. \tag{6.22}$$

It is interesting to note that the particular choice of the diagonal generators $\{Q_k\}$ determines the structure of the A-matrices in such a way that all representation matrices appear in exactly the same orthogonal form as given by Hamermesh [33]. As an illustration the example for $N=4$ is worked out in detail in appendix 3. Note also that degeneracies in the spectrum of the initial state are automatically accounted for by this procedure. In such a situation the components of $w^{(0)}$ will be restricted to a particular form such that some of the transformations lead to the same final coherence-vectors. In particular, the transpositions of equal roots must leave $w^{(0)}$ invariant, of course.

In fact, the generated set of vectors shows a rich geometrical structure in that all patterns can be viewed as hyperpolyhedra inscribed into spheres of radius $\|w^{(0)}\|$. The symmetry is higher or lower depending on details of the spectrum. For instance, for the simple special case of a three-level system with $\{\lambda_1 = \frac{2}{3}, \lambda_2 = 0, \lambda_3 = \frac{1}{3}\}$ one obtains a

regular hexagon in the (w_1, w_2)-plane whereas for $\{\lambda_1=\lambda_2=\frac{1}{2}, \lambda_3=0\}$ one obtains an equilateral triangle, etc. The situation reminds one very much of the patterns encountered in the classification of simple and semisimple Lie-algebras in terms of root- and weight diagrams [32, 35, 36]. In addition, important contributions by Stiefel [37] should be mentioned regarding the connection between discontinuous transformation groups of Euclidean spaces and compact Lie groups [38] and the crystallographic determination of their characters [39]. However, a closer look into the details shows that the analogies are less direct than one might expect and going further in this direction is definitely beyond the scope of this paper.

7. Importance for numerical calculations

In most cases the solutions to (2.1) must be computed numerically. From the decomposition (4.2) a set of $M=N^2-1$ coupled ordinary linear first-order differential equations is generated in the form [14]

$$\dot{v}(t)=Q(t)v(t) \tag{7.1}$$

where $Q(t)$ is a skew-symmetric $(M\times M)$ matrix, its elements being real-valued functions of time, in general. Even the most sophisticated numerical methods can impose only local error control criteria [40] on the integration procedure. This provides reliable estimates on relatively short time-intervals whereas global control on larger intervals is only possible if (7.1) implies certain time-conserved quantities for the solutions. This puts in clear evidence how important it is to have a complete set of dynamical invariants available for this purpose, the latter being known very accurately almost to machine precision δ_m from the chosen initial state $\rho(0)$ or, equivalently, from $v(0)$. As an instructive example showing already typical trends it will be sufficient to consider a modest case in four dimensions but with a rather strongly time-dependent Hamiltonian. After choosing an initial state $v(0)$ the 15 coupled equations (7.1) are solved by a procedure due to Hindmarsh [40]. The standard program 'lsode' has been modified in order to allow for an optional value of the local relative error ε over a wide range of orders of magnitude. The meaning of ε is roughly as follows. If any coherence-vector component has been determined up to accuracy ε a further decrease of the integration step lengths will change this result by a relative amount less than ε at any point of the integration mesh within the chosen entire interval. Even for an optimal choice of ε compatible with δ_m the errors tend to accumulate more and more in long-time runs. This means that the numerically computed trace-invariants, for instance, deviate from their reference values K_i at $t=0$. For a calculation over an interval

$$T=[0, \tau] \tag{7.2}$$

we write $K_i(t)$ and denote by K_i' the value with the largest deviation in T, that is

$$K_i'=\max_{t\in T} K_i(t). \tag{7.3}$$

As control parameter we therefore choose

$$\Delta K_i=|K_i-K_i'|. \tag{7.4}$$

The ordinary trace $(i=1)$ need not be considered since it is always accurate almost to precision δ_m.

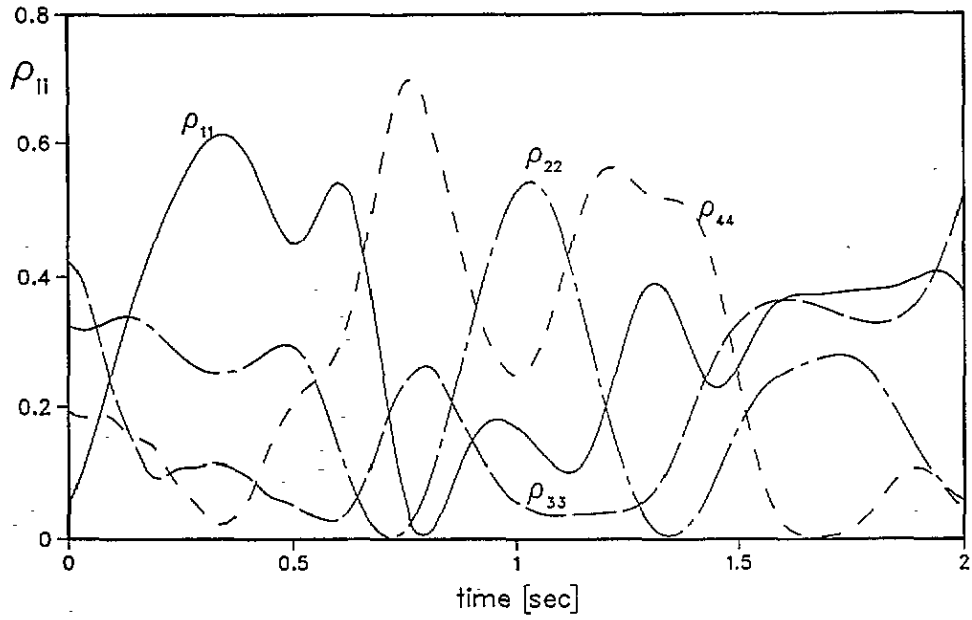


Figure 1. Time-evolution of the occupation probabilities of a four-level system with strongly time-dependent Hamiltonian. The details will be found in appendix 4.

The relevant details of the model are briefly summarized in appendix 4. All parameters have been adjusted in such a way that the characteristic strong time-variations can be displayed in the interval $\tau = 2$ s as shown in figure 1 for the level populations. All computations have been performed in double precision with $\delta_m < 10^{-15}$. Table 1 then summarizes the resulting data for different choices of ε and shows, in particular, the consequences for the values of ΔK_i. Increasing τ by a factor 5 causes a further loss of accuracy, as indicated in table 2.

8. Conclusions

The effects presented in the last section cannot be said to be dramatic and are certainly much less pronounced than one is used to, for instance, in problems of nonlinear

Table 1. Largest numerical deviations of the three relevant trace-invariants on the interval $[0, 2]$ *for the model displayed in figure 1 (see also appendix 4) as a function of a local* control parameter ε explained in the text. The numbers for the ΔK_i-values are rounded to one figure with the exponent to the basis 10 in round brackets.

ε	ΔK_2	ΔK_3	ΔK_4
1 (−13)	3 (−13)	4 (−13)	6 (−13)
1 (−11)	5 (−11)	8 (−11)	1 (−10)
1 (−9)	3 (−9)	4 (−9)	5 (−9)
1 (−7)	2 (−7)	4 (−7)	5 (−7)
1 (−6)	2 (−6)	2 (−6)	3 (−6)
1 (−5)	4 (−5)	6 (−5)	8 (−5)
1 (−4)	1 (−3)	2 (−3)	3 (−3)
1 (−3)	3 (−3)	4 (−3)	6 (−3)

Table 2. Largest deviations of invariants (as in table 1) for two selected ε-values but on the interval [0, 10].

ε	ΔK_2	ΔK_3	ΔK_4
1 (−13)	2 (−12)	2 (−12)	3 (−12)
1 (−4)	8 (−3)	1 (−2)	2 (−2)

classical dynamical systems. Nevertheless, the trends do exist and control by invariants will acquire its real significance for very large dimensions and, ultimately, in the notoriously tricky problems of irreversible processes. The latter appear in the dynamics of open systems considered as small subsystems of very large closed systems which undergo unitary dynamics. In contrast to (2.1) the density matrix $\rho_s(t)$ describing the open subsystem obeys a complicated integro-differential equation. It is only under many restrictive assumptions that the latter can be cast in a mathematically satisfactory and tractable form [12, 15]. Although the resulting so-called 'Markovian' master equations cover a wide field of applications there are left too many relevant problems which cannot be treated in this simplified way. Despite many respectable attempts towards a generalization to the 'non-Markovian' case, one must admit that it has been impossible to develop the theory on a level which even approaches todays standards of mathematical physics. Therefore, it is obvious and challenging to take advantage of the numerical solutions of (2.1) for the entire large systems including the control by invariants, and to project out to some physically relevant subspace afterwards in order to obtain $\rho_s(t)$. Note that for $\rho_s(t)$ no invariant may exist at all due to the unitarity-violating irreversible behaviour. This is already the case for uniquely relaxing semigroups [15]. Some interesting molecular problems with N of the order of 10^4 have already been treated in the above spirit [41–43] and one may hope that more progress will be made along this direction. Finally, some results similar to those presented in section 6 for completely incoherent states have been used in recent investigations on the possibilities of magnetization transfer in NMR [44, 45].

Appendix 1. N_{max} for a degenerate spectrum

Consider first the trivial case for $N=2$:

$$I_1=\lambda_1+\lambda_2 \qquad I_2=\lambda_1\lambda_2 \tag{A1.1}$$

by setting $\lambda_1=\lambda_2=x$. Then

$$I_1=2x \qquad I_2=x^2=\tfrac{1}{4}I_1^2. \tag{A1.2}$$

In the next example for $N=3$:

$$\begin{aligned} &I_1=\lambda_1+\lambda_2+\lambda_3 \\ &I_2=\lambda_1\lambda_2+\lambda_1\lambda_3+\lambda_2\lambda_3 \qquad I_3=\lambda_1\lambda_2\lambda_3 \end{aligned} \tag{A1.3}$$

we set, for instance, $\lambda_1=\lambda_2=x$ and $\lambda_3=y$ to obtain

$$I_1=2x+y \qquad I_2=x^2+2xy \qquad I_3=x^2y. \tag{A1.4}$$

Obviously, neither I_2 nor I_3 can be represented as any polynomial in the lower invariants and remain, therefore, independent. The only reducible situation occurs for $\lambda_1=\lambda_2=$

λ_3. Note also that, in the reduced variables x and y, the invariants (A1.4) are no longer symmetric functions, which turns out to be of crucial importance. Next we proceed to $N=4$ where

$$\begin{aligned} I_1&=\lambda_1+\lambda_2+\lambda_3+\lambda_4\\ I_2&=\lambda_1\lambda_2+\lambda_1\lambda_3+\lambda_1\lambda_4+\lambda_2\lambda_3+\lambda_2\lambda_4+\lambda_3\lambda_4\\ I_3&=\lambda_1\lambda_2\lambda_3+\lambda_1\lambda_2\lambda_4+\lambda_1\lambda_3\lambda_4+\lambda_2\lambda_3\lambda_4\\ I_4&=\lambda_1\lambda_2\lambda_3\lambda_4. \end{aligned} \tag{A1.5}$$

The only reducible case is for

$$\lambda_1=\lambda_2=x \qquad \lambda_3=\lambda_4=y \tag{A1.6}$$

$$\begin{aligned} &I_1=2(x+y) \qquad I_2=x^2+4xy+y^2=(x+y)^2+2xy\\ &I_3=2(x^2y+xy^2)=(x+y)(2xy) \qquad I_4=(xy)^2. \end{aligned} \tag{A1.7}$$

I_1 and I_2 are independent but

$$\begin{aligned} I_3&=\tfrac{1}{2}I_1I_2-\tfrac{1}{8}I_1^3\\ I_4&=\tfrac{1}{64}I_1^4-\tfrac{1}{8}I_1^2I_2+\tfrac{1}{4}I_2^2. \end{aligned} \tag{A1.8}$$

From these elementary considerations one can draw the following general conclusions. If there are $m<r$ ($r=\mathrm{rank}(\rho)$) distinct roots with exactly the same multiplicity g there are also exactly m independent invariants ($N_{\max}=m$) and the remaining $r-m$ invariants can be expressed as suitable polynomials in the first m invariants. Obviously, $N_{\max}$ is a divisor of r ($r\leqslant N$).

Appendix 2. Evaluation of L_5

By repeated multiplication of (4.2) one obtains

$$\begin{aligned} \rho^5=\frac{1}{N^5}\mathbb{1}_5+\frac{5}{N^4}\sum_i v_iF_i+\frac{10}{N^3}\sum_{i,k}v_iv_kF_iF_k+\frac{10}{N^2}\sum_{i,k,l}v_iv_kv_lF_iF_kF_l\\ +\frac{5}{N}\sum_{i,k,l,m}v_iv_kv_lv_mF_iF_kF_lF_m+\sum_{i,k,l,m,n}v_iv_kv_lv_mv_nF_iF_kF_lF_mF_n. \end{aligned} \tag{A2.1}$$

As an example we evaluate the last term of (A2.1) which will be denoted by G_5. According to (4.9) we have

$$\mathscr{L}_5=\mathrm{Tr}(G_5). \tag{A2.2}$$

By use of (4.6) and (4.7) for the pairs (i,k), (l,m) and, afterwards, for (q,p) we find

$$G_5=A+B+C \tag{A2.3}$$

$$A=\frac{1}{N^2}\sum_{i,k,l,m,n}v_iv_kv_lv_mv_n\delta_{ik}\delta_{lm}F_n \tag{A2.4}$$

$$B=\frac{1}{N}\sum_{i,k,l,m,n} v_i v_k v_l v_m v_n \left\{\delta_{ik}\sum_p z_{lmp}F_pF_n+\delta_{ln}\sum_q z_{ikq}F_qF_n\right\} \tag{A2.5}$$

$$C=\sum_{\substack{i,k,l,m,n\\q,p}} v_i v_k v_l v_m v_n z_{ikq}z_{lmp}\left\{\frac{1}{N}\mathbb{1}_N\delta_{qp}+\sum_s z_{qps}F_s\right\}F_n. \tag{A2.6}$$

From (4.3) one obtains

$$\mathrm{Tr}(A)=0 \tag{A2.7}$$

and, after using (4.4) and renaming summation indices,

$$\mathrm{Tr}(B)=\frac{2}{N}\|v\|^2\sum_{i,k,n} v_i v_k v_n z_{ikn}. \tag{A2.8}$$

The sum is real since, according to (4.7)

$$z_{ikn}=\bar{z}_{kin} \tag{A2.9}$$

and, consequently, in terms of the definitions in (4.11)

$$\mathrm{Tr}(B)=\frac{2}{N}L_2L_3. \tag{A2.10}$$

As a first result for C one obtains

$$\mathrm{Tr}(C)=\sum_{\substack{i,k,l,m\\n,p,q}} v_i v_k v_l v_m v_n z_{ikq}z_{lmp}z_{qpn}. \tag{A2.11}$$

Due to the above arguments all contributions involving products of one d and two f-constants or three f-constants vanish. Furthermore, because of summation over all indices every term of the form

$$v_i v_k v_l v_m v_n d_{ikq}d_{lmp}f_{qpn} \tag{A2.12}$$

has a counterpart obtained by simultaneously interchanging q with p and the pair (i, k) with (l, m), resulting just in a change of sign and therefore cancelling each other. What remains is

$$\mathrm{Tr}(C)=L_5 \tag{A2.13}$$

and, finally,

$$\mathrm{Tr}(G_5)=\frac{2}{N}L_2L_3+L_5. \tag{A2.14}$$

All summands in (A2.1) containing products of fewer coherence-vector components are calculated similarly. In this way all relations in (4.12) can be obtained straightforwardly.

Appendix 3. Explicit matrices for $N=4$

$SU(4)$ is of rank 3 with corresponding diagonal generators given according to (6.6) by

$$Q_1=\frac{1}{\sqrt{12}}\begin{pmatrix}1&&&\\&1&&\\&&1&\\&&&-3\end{pmatrix}\qquad Q_2=\frac{1}{\sqrt{6}}\begin{pmatrix}1&&&\\&1&&\\&&-2&\\&&&0\end{pmatrix}$$
$$Q_3=\frac{1}{\sqrt{2}}\begin{pmatrix}1&&&\\&-1&&\\&&0&\\&&&0\end{pmatrix}\qquad \mathrm{Tr}(Q_i)=0\qquad \mathrm{Tr}(Q_i^2)=1\qquad i=1,2,3 \tag{A3.1}$$

with an associated structure matrix

$$A=\frac{1}{\sqrt{12}}\begin{pmatrix}1&\sqrt{2}&\sqrt{6}\\1&\sqrt{2}&-\sqrt{6}\\1&-\sqrt{8}&0\end{pmatrix} \tag{A3.2}$$

and its inverse

$$A^{-1}=\frac{1}{\sqrt{6}}\begin{pmatrix}\sqrt{8}&\sqrt{8}&\sqrt{8}\\1&1&-2\\\sqrt{3}&-\sqrt{3}&0\end{pmatrix}. \tag{A3.3}$$

The reduced permutation matrices for the generating transpositions (see (6.19) and (6.20)) are then

$$P(12)=\begin{pmatrix}0&1&0\\1&0&0\\0&0&1\end{pmatrix}\qquad P(23)=\begin{pmatrix}1&0&0\\0&0&1\\0&1&0\end{pmatrix}\qquad P(34)=\begin{pmatrix}1&0&0\\0&1&0\\-1&-1&-1\end{pmatrix}. \tag{A3.4}$$

From (6.22) the irreducible representation matrices are found in the form

$$D(12)=\begin{pmatrix}1&0&0\\0&1&0\\0&0&-1\end{pmatrix}\qquad D(23)=\frac{1}{2}\begin{pmatrix}1&0&0\\0&-1&\sqrt{3}\\0&\sqrt{3}&1\end{pmatrix}$$
$$D(34)=\frac{1}{3}\begin{pmatrix}-1&\sqrt{8}&0\\\sqrt{8}&1&0\\0&0&3\end{pmatrix} \tag{A3.5}$$

To generate any further matrix as, for instance, the one for the cycle (124) one uses the cycle decomposition [34]

$$(i_1 i_2 \ldots i_r) = (i_1 i_2)(i_2 i_3) \ldots (i_{r-1} i_r) \qquad 1 \leqslant r, i_r \leqslant N \tag{A3.6}$$

together with repeated application of the shift rule

$$(j, k+1) = (k, k+1)(jk)(k, k+1) \qquad 1 \leqslant j < k < N \tag{A3.7}$$

in order to obtain $(124) = (12)(34)(23)(34)$ in terms of the generating elements and, accordingly,

$$D(124) = -\frac{1}{6}\begin{pmatrix} 2 & \sqrt{8} & -\sqrt{24} \\ 8 & -5 & -\sqrt{3} \\ \sqrt{24} & \sqrt{3} & 3 \end{pmatrix}. \tag{A3.8}$$

In an analogous way all 24 representation matrices of S_4 are readily derived.

Appendix 4. Details of the numerical test case for $N = 4$

Initial state (rounded):

$$\rho(0) = 10^{-3}\begin{pmatrix} 55 & -113-102\mathrm{i} & 100+27\mathrm{i} & 11+133\mathrm{i} \\ & 425 & -256+131\mathrm{i} & -271-254\mathrm{i} \\ & & 195 & 85+237\mathrm{i} \\ \text{h.c.} & & & 325 \end{pmatrix}. \tag{A4.1}$$

For the Hamiltonian the time-independent choice is (units in s^{-1} omitted throughout)

$$H = \begin{pmatrix} 6 & 5-2\mathrm{i} & 4+2\mathrm{i} & 3+\mathrm{i} \\ & 4 & 3-3\mathrm{i} & 2+\mathrm{i} \\ & & -3 & 1+2\mathrm{i} \\ \text{h.c.} & & & -7 \end{pmatrix} \tag{A4.2}$$

with eigenvalues (rounded)

$$E_1 = -8.87 \qquad E_2 = -6.13 \qquad E_3 = 2.24 \qquad E_4 = 12.76.$$

The full time-dependent Hamiltonian is obtained by replacing

$$H_{ik} \rightarrow H_{ik}(t) = H_{ik} f_{ik}(t) \qquad 1 \leqslant i < k \leqslant 4 \tag{A4.3}$$

with the following real time-functions:

$$f_{12}(t)=\cos\omega_a t \qquad f_{13}(t)=\sin\omega_b t$$
$$f_{14}(t)=\operatorname{sech}(\omega_c t-s) \qquad f_{23}(t)=e^{-\omega_d t}\cos\omega_e t \tag{A4.4}$$
$$f_{24}(t)=(1+\omega_f t^2)^{-1} \qquad f_{34}(t)=2\tanh(\omega_g t)$$

$$\omega_a=4.354 \qquad \omega_b=7$$
$$\omega_c=10 \qquad s=5 \qquad \omega_d=3 \tag{A4.5}$$
$$\omega_e=60 \qquad \omega_f=5 \qquad \omega_g=3.$$

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