Foundations of the Determinantal Formalism in Time-Dependent Quantum Mechanics

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It may look surprising that direct application of linear system theory to the Laplace transform of the time-dependent Schrödinger equation does not yield the evolution operator or density matrix in a physically reliable form, but requires a further "reduction procedure," earlier introduced in the frame of the so-called determinantal formalsim. It is shown that this procedure can in fact be viewed as a factorization of determinants intended so as to remove unexpected terms from the result.

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The so-called determinantal formalism has proved a fairly attractive method for solving typical time-dependent problems in quantum mechanics.[1,2] This formalism is mainly based on the resolution of the Laplace transform of the Schrödinger equation, conducted with use of linear system theory. The primary solution, as given by Cramer's formulas in the Laplace space, however, does not yield the result in a form quite satisfactory as to the time behavior of quantum mean values, trace conservation, etc. As a result, a somewhat empirical "reduction procedure" has been worked out, which finally leads to a much improved form of the response, as extensively discussed in Refs. 1 and 2. This Letter aims to show that this procedure, which looks intriguing at first sight, can be given a sound foundation, from a physical as well as a mathematical point of view.

Let us briefly recall the essential argument of the theory. For the purpose of demonstration it is sufficient to consider the simplest physical situation. The system, whose unperturbed Hamiltonian is H_0, with eigenstates $a,b,c,\ldots$, of eigenvalues $\hbar\omega_2,\hbar\omega_b,\ldots$, spanning the Hilbert space $\mathscr{E}_{H_0}$, is acted upon by a constant perturbation A, from $t=0$. Diagonal matrix elements of A are ignored. The Laplace transform of the Schrödinger equation of the evolution operator $U(t)$,

$$dU(t)/dt=(i\hbar)^{-1}(H_0+A)U(t),$$

with $U(0)=1$, reads

$$[\nu+i\hbar^{-1}(H_0+A)]F(\nu)=1,$$

with

$$F(\nu)=\int_0^\infty U(t)e^{-\nu t}dt.$$

This equation can be compactly rewritten as

$$(I+d^{-1}K)F=d^{-1}. \tag{1}$$

I is the identity operator, $d=\nu I+i\hbar^{-1}H_0$, and $K=i\hbar^{-1}A$. If we assume now that the system is initially in the pure state a the Cramer solution of Eq. (1), considered as a linear system, directly yields the b component of the column vector F as

$$F^b(\nu)=D_a^b/d_aD \tag{2}$$

($d_a=\nu+i\omega_a$). D denotes the determinant of the matrix $I+d^{-1}K$ and D_a^b the a-b minor.

The reduction procedure which permits us to derive an improved form of the result (2) then consists of writing

$$F^b=D_a^b(D_a^a)^{-1}/d_aD\,(D_a^a)^{-1}. \tag{3}$$

The initial-state occupation probability, for instance, will be derived from F^a, which is given by

$$F^a=1/d_aD\,(D_a^a)^{-1}. \tag{4}$$

The goal is now to elucidate the fundamental reason why, once the determinant divisons in Eqs. (3) and (4) are carried out, we obtain new series which work much better. Coming back to the initial equation (2), we first make a partition in the overall spectrum of H_0. Let $a,b,c,\ldots$ denote the whole set of N states which can be reached in elapsing time by means of A-induced transitions. If any set $a',b',c',\ldots$ is completely uncoupled from the preceding one (i.e., any matrix element of A between the $a,b,c,\ldots$ and the $a',b',c',\ldots$ is zero), then it is clear that matrix elements involving $b',c',\ldots$ in the matrix $I+d^{-1}K$ will be blocked out, leading to factorized contributions in both the upper and the lower determinants of Eq. (2). The physically expected elimination of out-of-reach states is thus properly achieved in the Cramer determinantal solution.

Once simplified, D reduces to an $N\times N$ determinant which, nevertheless, still involves products of K matrix elements, none of which is connected

with the initial a state. The presence of such products, which cannot be associated with any realistic sequence of transitions, is rather troublesome and suggests that some further factorization of D may occur, entailing elimination of these spurious terms. We notice that the latter are exactly all those which are contained in the D_a^a minor. This new elimination, however, is not so obvious as that of the uncoupled states considered above.

To proceed further, we will make use of the exponential form of determinants. For any matrix B, we have

$$\det B = \exp(\mathrm{Tr}\ln B).$$

It is straightforward to verify this equation, for instance on the eigenstate basis of $\ln B$. For $B = I + d^{-1}K$, by further expanding the logarithm, we obtain

$$D = \exp\left[-\sum_{n=1}^{\infty}(-1)^n\frac{S^{(n)}}{n}\right],$$

where $S^{(n)}$ is the trace of the nth power of $d^{-1}K$, usually called the n-times iterated kernel, in the theory of integral equations.[3] Explicitly

$$S^{(n)} = K_l^k K_m^l \cdots K_k^r / d_k d_l \cdots d_r, \tag{5}$$

where summation on repeated indices over all allowed states is implicit. Next, we separate out terms containing the a state in the $S^{(n)}$, say $S_a^{(n)}$:

$$S^{(n)} = S_a'^{(n)} + S_a^{(n)}.$$

$S_a'^{(n)}$ no longer contains any a. This splitting gives

$$D = \exp\left[-\sum_{n=1}^{\infty}(-1)^n\frac{S_a'^{(n)}}{n}\right]\exp\left[-\sum_{n=1}^{\infty}(-1)^n\frac{S_a^{(a)}}{n}\right]. \tag{6}$$

In the expansion of the second exponential, all terms containing a more than once as a row and a column index cancel one another since such terms are exactly those which appear in the overall expansion of D. For this reason the expansion of the second exponential in Eq. (6) can be limited to first order, provided that repetition of the subscript-superscript pair aa is forbidden in the iterative kernels of the remaining sum. D will therefore be finally factorized as

$$D = D_a^a\left[1-\sum_{n=1}^{\infty}(-1)^n S_a^{(n)}\right], \tag{7}$$

with the slightly modified definition

$$S_a^{(n)} = \frac{K_k^a K_l^k \cdots K_a^m}{d_a d_k \cdots d_m} \quad (k,l \neq a).$$

The aa couple is now fixed in the sequence of K's matrix elements and the n in the denominator [Eq. (6)] consequently removed.

The bracket on the right in Eq. (7) is an alternative expression of the quotient $D(D_a^a)^{-1}$ first given in Ref. 1 as

$$D(D_a^a)^{-1} = 1 + d^{-1}\langle a|K(I+Q_a d^{-1}K)^{-1}|a\rangle,$$

where Q_a denotes the projector on the space orthogonal to the a state. Substituting for D from Eq. (7) into Eq. (2) taken for $b = a$ gives the following exact expression for F^a:

$$F^a = \left\{d_a\left[1-\sum_{n=1}^{\infty}(-1)^n S_a^{(n)}\right]\right\}^{-1}. \tag{8}$$

A similar factorization can be carried out for the minor D_a^b so as to remove transitions which do not include the initial a state. it is straightforward to see that D_a^b can be split into two determinants such that in the first one $K_b^a/d_a = 1$ and $K_l^a = 0$ if $l \neq b$, whereas the second one is identically D_a^a. If, in addition, the former is expanded about the a column, we obtain

$$\begin{aligned} D_a^b &= \exp\left[-\sum_{n=1}^{\infty}(-1)^n(-S_{ba}^{(n)} + \frac{S_a'^{(n)}}{n}\right] - \exp\left[-\sum_{n=1}^{\infty}(-1)^n\frac{S_a'^{(n)}}{n}\right], \\ &= D_a^a\left\{\exp\left[\sum_{n=1}^{\infty}(-1)^n S_{ba}^{(n)}\right]-1\right\}, \end{aligned} \tag{9}$$

with

$$S_{ba}^{(n)} = K_k^b K_l^k \cdots K_a^m / d_b d_l \cdots d_m$$

$(k,l,\ldots,m \neq a)$. Notice the change of sign in front of $S_{ba}^{(n)}$ due to the lowering of the order by 1 after taking $K_b^a/d_a = 1$. The expansion of $S_{ba}^{(n)}$ now shows well every possibility of allowed transitions from a to b.

Again the exponential expansion in Eq. (9) reduces to the first term (repetition of aa entails cancellation):

$$D = D_a^a \sum_{n=1}^{\infty} (-1)^n S_{ba}^{(n)}. \tag{10}$$

The factorizations given by Eqs. (7) and (10) finally yield the following expression of F:

$$F^b = \sum_{n=1}^{\infty} (-1)^n S_{ba}^{(n)} \left\{ d_a \left[1 - \sum_{n=1}^{\infty} (-1)^n S_a^n \right] \right\}^{-1}, \tag{11}$$

which is an alternative form of the expression for F^b given in Ref. 1.

It is worthwhile to outline the reason why expressions (8) and (11) are the best from among other ones which are apparently equivalent. Let us rewrite

$$F^a = D_a^a / d_a D = D_a^a \left\{ d_a \left[D_a^a \exp\left(- \sum_{n=1}^{\infty} (-1)^n \frac{S_a^{(n)}}{n} \right) \right] \right\}^{-1} \tag{12a}$$

$$= \left\{ D_a^a \exp\left[\sum_{n=1}^{\infty} (-1)^n \frac{S_a^{(n)}}{n} \right] \right\} / d_a D_a^a \tag{12b}$$

$$= d_a^{-1} \exp\left[\sum_{n=1}^{\infty} (-1)^n \frac{S_a^{(n)}}{n} \right]. \tag{12c}$$

Performing the expansion in (12c) leads to the usual pertrubation series which is not as good as (8) derived from (12a). The reason is that the exponential in Eqs. (12) must be viewed as a series (stopped in practice at a selected order). When we perform the expansions and their subsequent products in the square brackets of Eq. (12a), the determinant D and thereby the initial expression of F^a are recovered at any order. if instead the same is done in the curly brackets of Eq. (12b), we obtain an only approximate expression valid at large ν, i.e., small t.

This can be best appreciated by considering a simple case. Assume that transitions from a to any of the $N-1$ states $b,c, \ldots, l, \ldots$ are allowed alone. Then

$$S_a^{(1)} = 0; \quad S_a^{(2)} = \frac{K_l^a K_a^l}{d_a d_l} = - \sum_{l \neq a} \frac{\hbar^{-2} |A_a^l|^2}{d_a d_l},$$

$$S_{ba}^{(1)} = K_a^b / d_b = i\hbar^{-1} A_a^b / d_b,$$

$S_a^{(n)} = 0$ if $n > 2$, and $S_{ba}^{(n)} = 0$ if $n > 1$. Since in addition $D_a^a = 1$, with use of (11), we obtain the *exact* results

$$F^a = \left[d_a + \sum_{l \neq a} \frac{\hbar^{-2} |A_2^l|^2}{d_l} \right]^{-1},$$

$$F^b = - i\hbar^{-1} A_a^b \left\{ d_b \left[d_a + \sum_{l \neq a} \frac{\hbar^{-2} |A_a^l|^2}{d_l} \right] \right\}^{-1}.$$

If instead we make use of the perturbation expansion (12c), the result will be

$$F^a = d_a^{-1} \left[1 + \sum_{p=1}^{\infty} (-1)^p \left(\sum_{l \neq a} \frac{\hbar^{-2} |A_a^l|^2}{d_a d_l} \right)^p \right],$$

and a similar expression for F^b, valid at small A and large ν only ($t \to 0$).

We now turn to extend the above results to the density-matrix problem. Let $\rho(t)$ be the density matrix satisfying the well-known evolution equation

$$d\rho/dt = (i\hbar)^{-1} [H_0 + A, \rho(t)]. \tag{13}$$

In Ref. 2, the Laplace transform of this equation is given a form similar to (1) in the tensorial product $\mathscr{E}_{H_0 H_0} = \mathscr{E}_{H_0} \otimes \mathscr{E}^*_{H_0}$ of the Hilbert space by its own dual. Let us introduce the appropriate commutator kernel operating in that space,

$$\hat{K} = i\hbar^{-1} (A\,\delta - \delta A),$$

whose $c_2 b_2 - c_1 b_1$ matrix elements are given by

$$\hat{K}_{c_1 b_2}^{c_2 b_1} = i\hbar^{-1} (A_{c_1}^{c_2} \delta_{b_2}^{b_1} - \delta_{c_1}^{c_2} A_{b_2}^{b_1}),$$

and the diagonal operator $\hat{d} = \nu \hat{I} + i\hbar^{-1} \hat{H}_0$, whose cb matrix element is

$$\hat{d}_{cb} = \nu + i\omega_{cb}$$

($\omega_{cb} = \omega_c - \omega_b$). Equation (13) can be rewritten in the $\mathscr{E}_{H_0 H_0}$ space as

$$(\hat{I} + \hat{d}^{-1} \hat{K}) \hat{R} = \hat{d}^{-1} \hat{\rho}(0), \tag{14}$$

with

$$\hat{R}(\nu) = \int_0^{\infty} \hat{\rho}(t) e^{-\nu t} dt.$$

$\hat{R}(\nu)$ denotes the vector of the $\mathscr{E}_{H_0 H_0}$ space equivalent with the operator $R(\nu)$ of $\mathscr{E}_{H_0}$, i.e.,

$$\hat{R}_b^c(\nu) = \langle cb | \hat{R}(\nu) \rangle = R_b^c(\nu).$$

Assuming again the system in a pure state a, i.e., the state aa in $\mathscr{E}_{H_0H_0}$, we can now write the Cramer solution of Eq. (14) as follows:

$$\hat{R}_a^a(\nu)=\hat{D}_{aa}^{aa}\rho_a^a(0)/\hat{d}_{aa}\hat{D}, \tag{15}$$

$$\hat{R}_b^c(\nu)=\hat{D}_{ab}^{ca}\rho_a^a(0)/\hat{d}_{aa}\hat{D}, \tag{16}$$

with $\rho_a^a(0)=1$. $\hat{D}$ stands for the determinant of $\hat{I}+\hat{d}^{-1}\hat{K}$, $\hat{D}_{ab}^{ca}$ for the aa-cb minor, etc. Transitions from state to state in $\mathscr{E}_{H_0H_0}$ are now induced by the kernel $\hat{K}$. The foregoing arguments leading to a factorization of $\hat{D}$, so as to remove any set of transitions which do not involve the initial aa state, still hold. The result can be written as

$$\hat{R}_a^a=\left\{\hat{d}_{aa}\left[1-\sum_{n=1}^{\infty}(-1)^n\hat{S}_{aa}^{(n)}\right]\right\}^{-1}, \tag{17}$$

and

$$\hat{R}_b^c=\sum_{n=1}^{\infty}(-1)^n\hat{S}_{cbaa}^{(n)}\left\{\hat{d}_{aa}\left[1-\sum_{n=1}^{\infty}(-1)^n\hat{S}_{aa}^{(n)}\right]\right\}^{-1} \tag{18}$$

($cb\neq aa$), with the extended definitions

$$\hat{S}_{aa}^{(n)}=\hat{K}_{c_1a}^{ab_1}\hat{K}_{c_2b_1}^{c_1b_2}\cdots\hat{K}_{ab_{n-1}}^{c_{n-1}a}/\hat{d}_{aa}\hat{d}_{c_1b_1}\cdots\hat{d}_{c_{n-1}b_{n-1}},$$

$$\hat{S}_{cbaa}^{(n)}=\hat{K}_{c_1b}^{cb_1}\hat{K}_{c_2b_1}^{c_1b_2}\cdots\hat{K}_{ab_{n-1}}^{c_{n-1}a}/\hat{d}_{cb}\hat{d}_{c_1b_1}\cdots\hat{d}_{c_{n-1}b_{n-1}} \quad (c_ib_i\neq aa).$$

When applied to the simple case considered before, we obtain the following results for the initial- and any final-state occupation probability:

$$\hat{R}_a^a=1/[\nu+\hbar^{-2}|A_a^k|^2(\hat{d}_{ka}^{-1}+\hat{d}_{ak}^{-1})]; \tag{19}$$

$$\hat{R}_b^b=\hbar^{-2}A_a^bA_b^2(\hat{d}_{cb}^{-1}+\hat{d}_{ba}^{-1})/\nu[\nu+\hbar^{-2}|A_a^k|^2(\hat{d}_{ka}^{-1}+\hat{d}_{ak}^{-1})]. \tag{20}$$

As shown in Refs. 1 and 2, the series appearing in Eq. (11) or (18) can in turn be regarded as solutions of linear systems, written in the form of iterative expansions. Thus the series

$$X=\sum_{n=1}^{\infty}(-1)^nS_{ba}^{(n)}$$

is the iterative expansion of the b component of the vector X obeying the equation

$$(I+d^{-1}KQ_a)|X\rangle=d^{-1}K|a\rangle.$$

Hence

$$X_a^b=(D_k'^b/d_kD')K_a^b/d_b,$$

where D' now stands for determinants and minor pertaining to the matrix $I+d^{-1}KQ_a$. If we consider then X_a^b as the solution of a problem in which the initial state is k, the above factorization can be performed again. If the same is done for the series arising in the denominator of (11), the result will be finally expressed in a new and more detailed form. It is readily realized that the process can be continued.

This may be of importance in applications whenever the first stage is not sufficient, e.g., for solving particular divergences, as often occurs in the case of density matrix. The long-term limit ($t\rightarrow\infty$) of expressions (19) and (20), for instance, is not well defined, whereas it is unambiguous in the twice-reduced expressions.[2]

In conclusion, the reduction procedure leading to the determinantal form of the response in the evolution-operator or the density-matrix problem can be understood as resulting from necessary factorizations in the primary Cramer's solution, by which all physically unexpected terms are eliminated. Let us recall that the main interest of the determinantal formalism is to yield tractable expressions, including energy shift and damping effects, in which unitarity or trace conservation can be carefully monitored at any order of the kernel.

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