A PERTURBATIVE APPROACH TO THE SOLUTION FOR EXPECTATION VALUE BASED QUANTUM OPTIMAL CONTROL OF MULTIHARMONIC OSCILLATORS UNDER LINEAR CONTROL AGENTS

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ABSTRACT. Matrix ODE boundary value problem of optimally controlled quantum multiharmonic oscillators are considered in perturbation expansion perspective. Expansion is shown to be convergent for all finite control times. A scalar recursion is contructed for practial applications. The convergence can be accelerated by changing the perturbation term appropriately.

Keywords: matrix ordinary differential equations, boundary value problems, perturbation expansion.

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1. INTRODUCTION

Recent years, especially last two decades, brought many interesting ideas about the control of the quantum systems and their applications to physical and chemical problems. The fundamental consideration in modelling was the utilization of a controllable external field like lasers or magnetic fields such that the quantum systems under consideration interacts with this field through a finite time duration such that its state after interaction becomes or gets closer to a desired state. The basic goal is to find the external field amplitude which is temporal under the weak field assumption where only dipole polarization is noticable in magnitude. The governing equations are obtained by using an appropriate cost functional composed of an objective term and certain penalty terms together with the dynamical constraint which enters the Schrödinger equation and therefore quantum dynamics via a Lagrange multiplier which can be considered defining a costate to the wave function's one. Optimization gives nonlinear and somehow cubic partial differential equations, one over the wave function to describe the forward evolution and one over the costate function and the wave function, describing the backward equation. An algebraic equation containing integration over the wave and costate function and certain operators like position and momentum gives the external field amplitude as long as the equations are solved at least numerically.

There are many works about quantum optimal control. Reader can make a literature survey to get what it wants to have. We cite certain works from Rabitz and Demiralp group here because those works are the motivating and the directioning agents for this work.

First two references are for the numerical solutions of the problems related to quantum dynamics [38] and expected values [21]. The next six references are about the first step works of Rabitz group on quantum optimal control [25, 34, 46, 32, 17, 26]. Next four references are the first works of the second author. Those papers have given important fundamental issues about the existence and multiplicities of the solutions in optimal control problems under weak fields [6, 7, 8, 9]. In these works, certain bounds are also constructed beside dealing with the classical control. Following twenty six articles are about the works of Rabitz group including basic ideas

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and some applications [27, 23, 28, 4, 42, 48, 14, 33, 41, 16, 3, 29, 19, 47, 43, 15, 31, 36, 44, 45, 2, 49, 24, 20, 37, 35]. The following eight articles are about the contributions coming from the Demiralp group [18, 40, 5, 13] and from the collaboration of Rabitz and Demiralp group [10, 11, 12, 30]. The following two articles [1, 39] are just some examples to give idea about the other group works.

In a recent paper [22] we have shown that the expectation values of, and transition values over, certain position and momentum operators can be used to construct temporal ordinary differential equations for the determination of external field amplitude, E(t,T), and the auxiliary concept deviation parameter, $\eta(T)$. We have defined following entities

$$p_{i}(t) \equiv \left\langle \psi(t) \left| -i\hbar \frac{\partial}{\partial y_{i}} \right| \psi(t) \right\rangle, \qquad 1 \le i \le N, \tag{1}$$

$$q_i(t) \equiv \langle \psi(t) | y_i | \psi(t) \rangle, \qquad 1 \le i \le N, \tag{2}$$

$$r_i(t) \equiv 2 \operatorname{I\!R} e\left(\left\langle \lambda(t) \left| -i\hbar \frac{\partial}{\partial y_i} \right| \psi(t) \right\rangle \right), \qquad 1 \le i \le N, \tag{3}$$

$$s_i(t) \equiv 2\mathbb{R}e\left(\langle \lambda(t) | y_i | \psi(t) \rangle\right), \qquad 1 \le i \le N, \qquad (4)$$

where Dirac's bra and ket notation is used. $p_i(t)$ and $q_i(t)$ (i = 1, ..., N) are expectation values of the momentum and position operators for a system with N degree of freedom. In the previous paper[16] we had started with N interacting oscillator system whose degree of freedom is 3N and at the end we had emphasized that we do not need to take the degree of freedom as multiples of 3. Hence, we use N here instead of 3N. Expectation values are evaluated by using wave function ψ 's bra and ket. $r_i(t)$ and $s_i(t)$ (i = 1, ..., N) are transition values evaluated over momentum and position operators. They are evaluated via the bras and kets of both wave and costate function denoted by λ . Wave function describes the forward evolution of the system because its value is specified at the beginning of the evolution. Whereas costate function is specified at the end of the control and is responsible for the backward evolution. This is the reason why we obtain a boundary value problem in time. Indeed, during the control, wave function takes the system towards the end of the control while the costate function takes it back to the beginning of the control. By forming vectors from these four indexed entities we can construct the following 4Nelement vector.

$$\boldsymbol{z}(t)^{T} \equiv \left[\boldsymbol{p}(t)^{T}, \boldsymbol{q}(t)^{T}, \boldsymbol{r}(t)^{T}, \boldsymbol{s}(t)^{T} \right],$$
(5)

where boldfaced entities at the right hand side denote vectors composed of related unknowns defined in (1), (2),(3), and, (4). The T dependence of the entities above are not shown explicitly and the vector $\boldsymbol{z}(t)$ can be uniquely expressed through the following equation via an evolution matrix $\boldsymbol{Z}(t)$

$$\boldsymbol{z}(t) \equiv \boldsymbol{Z}(t)\boldsymbol{c},\tag{6}$$

where c is a constant vector whose value is determined by using boundary conditions and the evolution matrix satisfies the following equation and the accompanying initial condition

$$\boldsymbol{Z}(t) = \boldsymbol{A}(t)\boldsymbol{Z}(t), \qquad \boldsymbol{Z}(0) = \boldsymbol{I}_{4N}, \tag{7}$$

where

$$\boldsymbol{A}(t) = \begin{bmatrix} \boldsymbol{B} & W_E(t)^{-1}\boldsymbol{u}_1\boldsymbol{u}_2^T \\ -W_p(t)\boldsymbol{u}_3\boldsymbol{u}_4^T & \boldsymbol{B} \end{bmatrix},$$
(8)

$$\boldsymbol{B} = \begin{bmatrix} \boldsymbol{0} & -\boldsymbol{\kappa} \\ \frac{1}{m_e} \boldsymbol{I}_N & \boldsymbol{0} \end{bmatrix}, \quad \boldsymbol{u}_1 = \begin{bmatrix} \boldsymbol{\mu} \\ \boldsymbol{0} \end{bmatrix}, \quad \boldsymbol{u}_2 = \begin{bmatrix} \boldsymbol{0} \\ \boldsymbol{\mu} \end{bmatrix}, \quad \boldsymbol{u}_3 = \begin{bmatrix} \boldsymbol{\alpha}' \\ -\boldsymbol{\beta}' \end{bmatrix}, \quad \boldsymbol{u}_4 = \begin{bmatrix} \boldsymbol{\beta}' \\ \boldsymbol{\alpha}' \end{bmatrix}. \quad (9)$$

In the above formulae I_{4N} and I_N stand for the $4N \times 4N$ and $N \times N$ type unit matrices. m_e denotes the effective mass parameter of the system. κ is a diagonal matrix of $N \times N$ type. Its diagonal elements can be considered as the effective force constants of the system. That is, the formulae above describe a system as if composed of N oscillators with identical masses and interacting with the origin with different elastic force constants. $W_E(t)$ and $W_p(t)$ are the

weight functions appearing in the penalty terms of the cost functional. They have to be taken positive everywhere perhaps except a finite number of points. The vectors μ , α' , β' has the corresponding indexed elements taken from the following definitions

$$\mu \equiv \sum_{j=1}^{N} \mu_j x_j, \qquad \widehat{O}' \equiv \sum_{j=1}^{N} \left[\alpha'_j x_j + \beta'_j \left(-\hbar \frac{\partial}{\partial x_j} \right) \right], \tag{10}$$

where μ represents the dipole function which can vary spatially only and \hat{O}' stands for the penalty operator whose expectation value to be suppressed during the control. Since the detailed derivation of these equalities were given in the previous paper[22] we are not going to get into details here.

To determine the unknown constant vector \boldsymbol{c} we can partition the matrix $\boldsymbol{Z}(t)$ and \boldsymbol{c} as follows

$$\boldsymbol{Z}(t) \equiv \begin{bmatrix} \boldsymbol{Z}_{11}(t) & \boldsymbol{Z}_{12}(t) \\ \boldsymbol{Z}_{21}(t) & \boldsymbol{Z}_{22}^{T}(t) \end{bmatrix}, \qquad \boldsymbol{c} \equiv \begin{bmatrix} \boldsymbol{c}_{1} \\ \boldsymbol{c}_{2} \end{bmatrix},$$
(11)

where c_1 and c_2 are (2N)-element vectors, while $Z_{11}(t)$, $Z_{12}(t)$, $Z_{21}(t)$, and $Z_{22}(t)$ are denoting $(2N) \times (2N)$ blocks. These blocks satisfy the following initial conditions

$$Z_{ij}(0) = \delta_{ij} I_{2N}, \qquad i, j = 1, 2,$$
 (12)

where δ_{ij} and I_{2N} stand for the Kroenecker's symbol and $2N \times 2N$ type unit matrix respectively. A careful look at $\mathbf{z}(0)$ reveals that the first half of its elements can be determined from the initial conditions since the momentum and position vectors, $\mathbf{p}(t)$ and $\mathbf{q}(t)$, are defined as expectation values and therefore depend on wave function which can have initial condition only. Thus, when t is set equal to zero in (6) we can obtain the following equation from the first half of the resulting equation

$$\boldsymbol{c}_{1}^{T} = \left[p_{1}^{(in)}, \cdots, p_{N}^{(in)}, q_{1}^{(in)}, \cdots, q_{N}^{(in)} \right] \equiv \boldsymbol{v}_{1}^{T},$$
(13)

where $p_j^{(in)}$ and $q_j^{(in)}$ $(1 \le j \le N)$ denote the initial values of $p_j(t)$ and $q_j(t)$ $(1 \le j \le N)$ respectively. They are explicitly given in terms of the expectation values of the momentum and position operators evaluated by the given initial form of the wave function as follows

$$p_{j}^{(in)} \equiv \left\langle in \left| -i\hbar \frac{\partial}{\partial y_{i}} \right| in \right\rangle, \qquad 1 \le j \le N,$$
(14)

$$q_j^{(in)} \equiv \langle in | y_i | in \rangle, \qquad 1 \le j \le N, \qquad (15)$$

where the bra and ket symbolized by *in* stand for the initial form of wavebra and waveket.

The determination of c_2 is related to the conditions imposed on the vectors $\mathbf{r}(t)$ and $\mathbf{s}(t)$. Since they contain the costate function in their definitions we can not use the initial conditions. Instead, we need final conditions. Hence the determination of c_2 can be accomplished by setting t equal to T in (6). The second part of the resulting partitioned equations can be solved to give the following equality by skipping the intermediate steps which can be referred to the previous work[22]

$$\boldsymbol{c}_{2} = \eta(T)\boldsymbol{Z}_{22}(T)^{-1}\boldsymbol{v}_{2} - \boldsymbol{Z}_{22}(T)^{-1}\boldsymbol{Z}_{21}(T)\boldsymbol{v}_{1}, \qquad (16)$$

where

$$\boldsymbol{v}_2^T \equiv \frac{1}{\eta(T)} \left[\boldsymbol{r}(T)^T, \boldsymbol{s}(T)^T \right] \equiv \left[-\alpha_1, \cdots, -\alpha_N, \beta_1, \cdots, \beta_N \right] = \left[-\alpha, \beta \right], \tag{17}$$

and the parameters α_j and β_j are the linear combination coefficients of the objective operator which is assumed to be purely linear in momentum and position as follows

$$\widehat{O} \equiv \sum_{j=1}^{N} \left[\alpha_j x_j + \beta_j \left(-i\hbar \frac{\partial}{\partial x_j} \right) \right].$$
(18)

The deviation parameter, $\eta(T)$ is defined by using final control moment wave function as follows

$$\eta(T) \equiv \left\langle \psi(T) \left| \widehat{O} \right| \psi(T) \right\rangle - \widetilde{O}, \tag{19}$$

where \widetilde{O} stands for the given target value of objective operator's expectation value. If the first part of $\boldsymbol{z}(t)$ is denoted by $\boldsymbol{z}_1(t)$ when it is partitioned into two same type subvectors then we can express the above result as follows

$$\eta(T) = \boldsymbol{v}_3^T \boldsymbol{z}_1(T) - \tilde{O},\tag{20}$$

where

$$\boldsymbol{v}_3^T \equiv \left[\boldsymbol{\beta}^T, \boldsymbol{\alpha}^T\right]. \tag{21}$$

This equality urges us to write (20) more explicitly and then to solve $\eta(T)$ as below

$$\eta(T) = \frac{\boldsymbol{v}_3^T \boldsymbol{Z}_{11}(T) \boldsymbol{v}_1 - \boldsymbol{v}_3^T \boldsymbol{Z}_{12}(T) \boldsymbol{Z}_{22}(T)^{-1} \boldsymbol{Z}_{21}(T) \boldsymbol{v}_1 - \tilde{O}}{1 - \boldsymbol{v}_3^T \boldsymbol{Z}_{12}(T) \boldsymbol{Z}_{22}(T)^{-1} \boldsymbol{v}_2}.$$
(22)

The external field amplitude is given by the following equation

$$E(t) = \frac{2}{W_E(t)} \mathbb{R}e\left(\left\langle \lambda(t) \left| \mu \right| \psi(t) \right\rangle\right), \tag{23}$$

which can be rewritten as follows

$$W_E(t)E(t) = \sum_{i=1}^{3N} \mu_i s_i(t) = \boldsymbol{\mu}^T \boldsymbol{s}(t), \qquad 1 \le i \le N,$$
(24)

whose solution can be expressed as

$$E(t) = W_E(t)^{-1} \boldsymbol{u}_2^T \boldsymbol{z}_2(t)^T = W_E(t)^{-1} \boldsymbol{u}_2^T \left[\boldsymbol{Z}_{11}(t) \boldsymbol{c}_1 + \boldsymbol{Z}_{12}(t) \boldsymbol{c}_2 \right].$$
(25)

Now all these formulae mean that the determination of the evolution matrix is sufficient for the evaluation of the deviation parameter and external field amplitude.

2. A perturbative approach to determine external field amplitude and deviation parameter

One of the possible difficulties arising in the solution of the ordinary matrix differential equation to be satisfied by the evolution matrix under given initial condition was the dependence of at least one of the weight functions on time. Otherwise the analytical solution was possible via the use of exponential matrix structures. In the case of time dependent weight functions the level of the difficulty for the solution is determined by how the weight functions depend on time. Certain series expansions or factorization methods can be used to obtain the solution and finite truncations of these entities can be used as approximants to the solution.

When the analytical solutions either in rather simple forms or in series expansions or infinite products are not available, or their utilization is very expensive, one can use purely numerical approximations or certain divide–and–conquer type approximations whose each step has analytical formulae. To this end we can use a perturbative approach.

Let us now rewrite (7) as follows

$$\frac{d\mathbf{Z}(t)}{dt} = \mathbf{A}^{(1)}\mathbf{Z}(t) + \mathbf{A}^{(2)}(t)\mathbf{Z}(t), \qquad \mathbf{Z}(0) = \mathbf{I}_{4N},$$
(26)

where the coefficient matrices are defined as below

$$\boldsymbol{A}^{(1)} \equiv \begin{bmatrix} \boldsymbol{B} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{B} \end{bmatrix}, \qquad \boldsymbol{A}^{(2)}(t) \equiv \begin{bmatrix} \boldsymbol{0} & W_E(t)^{-1}\boldsymbol{u}_1\boldsymbol{u}_2^T \\ -W_p(t)\boldsymbol{u}_3\boldsymbol{u}_4^T & \boldsymbol{0} \end{bmatrix}.$$
(27)

All entities appearing in this equations have been defined before. As can be noticed immediately the time dependence in the structure is originated from $A^{(2)}(t)$ matrix and the solution of the problem can be expressed in terms of exponential matrices in the nonexistence of this matrix. Hence, it is reasonable to construct an iterative determination scheme which primarily ignores the contribution of this matrix and then evaluates its contribution secondarily. Therefore we can insert a dummy parameter ν , whose value will be set equal to 1 later, into the equation and accompanying initial condition given by (26). The method is based on the expansion of all entities, known or unknown, in nonnegative integer powers of this dummy parameter. The equation (26) can be rewritten as follows

$$\frac{d\mathbf{Z}(t,\nu)}{dt} = \mathbf{A}^{(1)}\mathbf{Z}(t) + \nu\mathbf{A}^{(2)}(t)\mathbf{Z}(t), \qquad \mathbf{Z}(0,\nu) = \mathbf{I}_{4N},$$
(28)

whose solution can be expressed as follows

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$$\boldsymbol{Z}(t,\nu) \equiv \sum_{k=0}^{\infty} \nu^k \boldsymbol{Z}_k(t), \qquad (29)$$

where $\mathbf{Z}_k(t)$ terms stand for unknown matrix valued functions which do not depend on ν . If we use this expansion in the equation and accompanying initial condition given in (26) then the following recursion and initial conditions are obtained

$$\frac{d\mathbf{Z}_{k}(t)}{dt} = \mathbf{A}^{(1)}\mathbf{Z}_{k}(t) + \mathbf{A}^{(2)}(t)\mathbf{Z}_{k-1}(t), \qquad \mathbf{Z}_{k}(0) = \delta_{k,0}\mathbf{I}_{4N}, \\ 0 \le k < \infty,$$
(30)

where $\delta_{k,0}$ denotes Kroenecker's Delta symbol and negative index in matrices implies the nonexistence of those matrices. The corresponding forms of this equation and accompanying initial condition for k = 0 can be easily and analytically solved since $\mathbf{A}^{(1)}$ is a constant matrix, and the following result is obtained.

$$\boldsymbol{Z}_{0}(t) = e^{t\boldsymbol{A}^{(1)}} = \begin{bmatrix} e^{t\boldsymbol{B}} & \boldsymbol{0} \\ \boldsymbol{0} & e^{t\boldsymbol{B}} \end{bmatrix}.$$
(31)

This result can be expressed more explicitly. For this purpose one can use the following equality

$$e^{t\boldsymbol{B}} = \sum_{k=0}^{\infty} \frac{t^{2k}}{(2k)!} \boldsymbol{B}^{2k} + \sum_{k=0}^{\infty} \frac{t^{2k+1}}{(2k+1)!} \boldsymbol{B}^{2k+1}.$$
(32)

To proceed we can use the following equality

$$\boldsymbol{B}^{2} = \begin{bmatrix} \boldsymbol{0} & -\boldsymbol{\kappa} \\ \frac{1}{m_{e}}\boldsymbol{I}_{N} & \boldsymbol{0} \end{bmatrix}^{2} = \begin{bmatrix} -\frac{1}{m_{e}}\boldsymbol{\kappa} & \boldsymbol{0} \\ \boldsymbol{0} & -\frac{1}{m_{e}}\boldsymbol{\kappa} \end{bmatrix}$$
(33)

and

$$\boldsymbol{B}^{2k} = \begin{bmatrix} \left(-\frac{1}{m_e}\right)^k \boldsymbol{\kappa}^k & \boldsymbol{0} \\ \boldsymbol{0} & \left(-\frac{1}{m_e}\right)^k \boldsymbol{\kappa}^k \end{bmatrix},$$
(34)

which implies

$$\sum_{k=0}^{\infty} \frac{t^{2k}}{(2k)!} \boldsymbol{B}^{2k} = \begin{bmatrix} \cos\left(\frac{t}{\sqrt{m_e}} \boldsymbol{\kappa}^{\frac{1}{2}}\right) & \boldsymbol{0} \\ \boldsymbol{0} & \cos\left(\frac{t}{\sqrt{m_e}} \boldsymbol{\kappa}^{\frac{1}{2}}\right) \end{bmatrix}$$
(35)

and

$$\sum_{k=0}^{\infty} \frac{t^{2k+1}}{(2k+1)!} \boldsymbol{B}^{2k+1} = \begin{bmatrix} \boldsymbol{0} & -\sqrt{m_e} \boldsymbol{\kappa}^{\frac{1}{2}} \sin\left(\frac{t}{\sqrt{m_e}} \boldsymbol{\kappa}^{\frac{1}{2}}\right) \\ -tS\left(\frac{t}{\sqrt{m_e}} \boldsymbol{\kappa}^{\frac{1}{2}}\right) & \boldsymbol{0} \end{bmatrix}.$$
 (36)

The newly appearing entity S(x) is a univariate function whose explicit structure is given below

$$S(x) \equiv \frac{\sin(x)}{x}.$$
(37)

This function is used with a matrix argument in (36). It has a removable singularity at x = 0and used in (36) to avoid a respresentation giving singularity impression. As remembered κ is a diagonal matrix, some of whose diagonal elements may be vanishing and the remaining ones are positive. If at least one of its diagonal elements vanishes then its inverse does not exist. Its square root is defined and is a diagonal matrix whose elements are square root of its original form. However $\kappa^{-\frac{1}{2}}$ is not defined when a zero diagonal element exists. S(x) definition enables us to avoid this undefined matrix square root.

We can get the following equation by joining (35) and (36)

$$e^{t\boldsymbol{B}} = \begin{bmatrix} \cos\left(\frac{t}{\sqrt{m_e}}\kappa^{\frac{1}{2}}\right) & -\sqrt{m_e}\kappa^{\frac{1}{2}}\sin\left(\frac{t}{\sqrt{m_e}}\kappa^{\frac{1}{2}}\right) \\ -tS\left(\frac{t}{\sqrt{m_e}}\kappa^{\frac{1}{2}}\right) & \cos\left(\frac{t}{\sqrt{m_e}}\kappa^{\frac{1}{2}}\right) \end{bmatrix}.$$
(38)

The blocks appearing in the last equation are diagonal because of the matrix κ 's diagonality.

Now, by having the information obtained above we can set k = 1 in (30) and write the following definition for $\mathbf{Z}_1(t)$ by making an analogy to the structure of $\mathbf{Z}_0(t)$

$$\mathbf{Z}_{1}(t) \equiv e^{t \mathbf{A}^{(1)}} \overline{\mathbf{Z}}_{1}(t), \qquad (39)$$

where $\overline{Z}_1(t)$ is a new unknown. This transformation enables us to write

$$\frac{d\mathbf{Z}_{1}(t)}{dt} = e^{-t\mathbf{A}^{(1)}}\mathbf{A}^{(2)}(t)\mathbf{Z}_{0}(t), \qquad \mathbf{Z}_{1}(0) = \mathbf{0},$$
(40)

which means

$$\frac{d\overline{\boldsymbol{Z}}_{1}(t)}{dt} = e^{-t\boldsymbol{A}^{(1)}}\boldsymbol{A}^{(2)}(t)e^{t\boldsymbol{A}^{(1)}}, \qquad \overline{\boldsymbol{Z}}_{1}(0) = \boldsymbol{0}.$$
(41)

If t is replaced by τ in this equation and the resulting equation's both sides are integrated over τ from 0 to t and the accompanying initial condition is taken into consideration then

$$\overline{\boldsymbol{Z}}_{1}(t) = \int_{0}^{t} d\tau \mathrm{e}^{-\tau \boldsymbol{A}^{(1)}} \boldsymbol{A}^{(2)}(\tau) \mathrm{e}^{\tau \boldsymbol{A}^{(1)}}$$
(42)

and from this with the aid of (39)

$$\boldsymbol{Z}_{1}(t) = \int_{0}^{t} d\tau e^{(t-\tau)} \boldsymbol{A}^{(1)} \boldsymbol{A}^{(2)}(\tau) e^{\tau \boldsymbol{A}^{(1)}}$$
(43)

is obtained. This result reveals the fact that the integration is needed to get the second contribution (first order) from perturbation expansion. If this integration can be performed analytically the result become analytic at this level. Otherwise, numerical integration is required for computation.

The way we have traced in the evaluation of last term can be followed for the general term of the recursion above. If this is done for general k values in (30) we obtain the following equation instead of (43)

$$\boldsymbol{Z}_{k}(t) = \int_{0}^{t} d\tau e^{(t-\tau)} \boldsymbol{A}^{(1)} \boldsymbol{A}^{(2)}(\tau) \boldsymbol{Z}_{k-1}(\tau), \qquad k \ge 1.$$
(44)

This recursive relation suffices to determine the terms of perturbation expansion. Its consecutive use for increasing k values starting from k = 1 allows us to obtain perturbative terms as much as we want and to construct approximants to the evolution matrix.

Equation (44) defines a matrix recursion. Although the order of recursion is just one it requires matrix algebraic operations and hence it is not a desired algorithm in numerical evaluations or in computer programming. Therefore, it is reasonable to put this recursion into a form where recursion completely or partially occurs between scalar entities. If the (k - 1)-th indexed term in the recursion above is expressed in terms of (k - 2)-th one by using recursion's itself the following double integration containing recursion is obtained

$$\boldsymbol{Z}_{k}(t) = \int_{0}^{t} d\tau e^{(t-\tau)} \boldsymbol{A}^{(1)} \boldsymbol{A}^{(2)}(\tau) \int_{0}^{\tau} d\tau_{1} e^{(\tau-\tau_{1})} \boldsymbol{A}^{(1)} \boldsymbol{A}^{(2)}(\tau_{1}) \boldsymbol{Z}_{k-2}(\tau_{1}), \qquad k \ge 2.$$
(45)

This is a second order recursion, that is, it does not relate consecutive terms but connects even and odd index terms separately. This twofold integral can be reduced to a univariate one after some manipulations. We can start by defining

$$\mathbf{A}^{(3)}(t,\tau,\tau_{1}) \equiv e^{(t-\tau)\mathbf{A}^{(1)}}\mathbf{A}^{(2)}(\tau)e^{(\tau-\tau_{1})\mathbf{A}^{(1)}}\mathbf{A}^{(2)}(\tau_{1}) = \\
 = \begin{bmatrix} -\frac{W_{p}(\tau_{1})}{W_{E}(\tau)}e^{(t-\tau)\mathbf{B}}\boldsymbol{u}_{1}\boldsymbol{u}_{2}^{T}e^{(\tau-\tau_{1})\mathbf{B}}\boldsymbol{u}_{3}\boldsymbol{u}_{4}^{T} & \mathbf{0} \\
 & \mathbf{0} & -\frac{W_{p}(\tau)}{W_{E}(\tau_{1})}e^{(t-\tau)\mathbf{B}}\boldsymbol{u}_{3}\boldsymbol{u}_{4}^{T}e^{(\tau-\tau_{1})\mathbf{B}}\boldsymbol{u}_{1}\boldsymbol{u}_{2}^{T} \end{bmatrix}. \quad (46)$$

A careful glance at the explicit matrix structure above reveals the fact that $\boldsymbol{u}_2^T e^{(\tau-\tau_1)\boldsymbol{B}} \boldsymbol{u}_3$ and $\boldsymbol{u}_4^T e^{(\tau-\tau_1)\boldsymbol{B}} \boldsymbol{u}_1$ are scalar entities. Therefore we can define

$$\varphi_{1}(\tau,\tau_{1}) \equiv -\frac{W_{p}(\tau_{1})}{W_{E}(\tau)}\boldsymbol{u}_{2}^{T}e^{(\tau-\tau_{1})}\boldsymbol{B}\boldsymbol{u}_{3},$$

$$\varphi_{2}(\tau,\tau_{1}) \equiv -\frac{W_{p}(\tau)}{W_{E}(\tau_{1})}\boldsymbol{u}_{4}^{T}e^{(\tau-\tau_{1})}\boldsymbol{B}\boldsymbol{u}_{1}$$
(47)

and rewrite (46) as follows

$$\boldsymbol{A}^{(3)}(t,\tau,\tau_1) = \begin{bmatrix} \varphi_1(\tau,\tau_1) \mathrm{e}^{(t-\tau)} \boldsymbol{B}_{\boldsymbol{u}_1} \boldsymbol{u}_4^T & \boldsymbol{0} \\ \boldsymbol{0} & \varphi_2(\tau,\tau_1) \mathrm{e}^{(t-\tau)} \boldsymbol{B}_{\boldsymbol{u}_3} \boldsymbol{u}_2^T \end{bmatrix},$$
(48)

which leads us to rewrite (45) as

$$\boldsymbol{Z}_{k}(t) = \int_{0}^{t} d\tau \int_{0}^{\tau} d\tau_{1} \boldsymbol{A}^{(3)}(t,\tau,\tau_{1}) \boldsymbol{Z}_{k-2}(\tau_{1}), \qquad k \ge 2.$$
(49)

We can use the integration's triangular identity which can be expressed as follows

$$\int_{0}^{t} d\tau \int_{0}^{\tau} d\tau_{1} f(\tau, \tau_{1}) \equiv \int_{0}^{t} d\tau_{1} \int_{\tau_{1}}^{t} d\tau f(\tau, \tau_{1}),$$
(50)

where $f(\tau, \tau_1)$ is an integrable bivariate function. Therefore, as an intermediate conclusion, we can get the following recursion

$$\boldsymbol{Z}_{k}(t) = \int_{0}^{t} d\tau_{1} \int_{\tau_{1}}^{t} d\tau \boldsymbol{A}^{(3)}(t,\tau,\tau_{1}) \boldsymbol{Z}_{k-2}(\tau_{1}), \qquad k \ge 2.$$
(51)

If we define

$$\mathbf{A}^{(4)}(t,\tau_1) \equiv \int_{\tau_1}^t d\tau \mathbf{A}^{(3)}(t,\tau,\tau_1),$$
(52)

then (51) becomes

$$\boldsymbol{Z}_{k}(t) = \int_{0}^{t} d\tau \boldsymbol{A}^{(4)}(t,\tau) \boldsymbol{Z}_{k-2}(\tau), \qquad k \ge 2.$$
(53)

The most important aspect of this recursion is the fact that the integration kernel $A^{(4)}(t,\tau)$ is block diagonal.

By using last recursion above it is possible to express perturbation terms with even integer indices in terms of $\mathbf{Z}_0(t)$. If k is set equal to 2 in last recursion then we can write

$$\boldsymbol{Z}_{2}(t) = \int_{0}^{t} d\tau \boldsymbol{A}^{(4)}(t,\tau) \boldsymbol{Z}_{0}(\tau), \qquad (54)$$

which enables us to write the following equation by taking k = 4 in (51) and using last equation

$$\boldsymbol{Z}_{4}(t) = \int_{0}^{t} d\tau \boldsymbol{A}^{(4)}(t,\tau) \int_{0}^{\tau} d\tau_{1} \boldsymbol{A}^{(4)}(\tau,\tau_{1}) \boldsymbol{Z}_{0}(\tau_{1}).$$
(55)

If the order of integrations over τ and τ_1 is exchanged and the triangular identity of integration is used then

$$\boldsymbol{Z}_{4}(t) = \int_{0}^{t} d\tau_{1} \int_{\tau_{1}}^{t} d\tau \boldsymbol{A}^{(4)}(t,\tau) \boldsymbol{A}^{(4)}(\tau,\tau_{1}) \boldsymbol{Z}_{0}(\tau_{1})$$
(56)

is obtained. By defining

$$\boldsymbol{A}^{(5)}(t,\tau) \equiv \int_{\tau}^{t} d\tau_1 \boldsymbol{A}^{(4)}(t,\tau_1) \boldsymbol{A}^{(4)}(\tau_1,\tau)$$
(57)

one can arrive at the following equation

$$\boldsymbol{Z}_{4}(t) = \int_{0}^{t} d\tau \boldsymbol{A}^{(5)}(t,\tau) \boldsymbol{Z}_{0}(\tau)$$
(58)

(54) and (58) urges us to write the following generalization

$$\boldsymbol{Z}_{2k}(t) = \int_0^t d\tau \boldsymbol{A}^{(k+3)}(t,\tau) \boldsymbol{Z}_0(\tau), \qquad k \ge 1.$$
(59)

If this structure is used in the equation obtained from (53) after replacement of k by 2k then

$$\int_{0}^{t} d\tau \boldsymbol{A}^{(k+3)}(t,\tau) \boldsymbol{Z}_{0}(\tau) = \int_{0}^{t} d\tau \boldsymbol{A}^{(4)}(t,\tau) \int_{0}^{\tau} d\tau_{1} \boldsymbol{A}^{(k+2)}(\tau,\tau_{1}) \boldsymbol{Z}_{0}(\tau_{1}), \qquad k \ge 2$$
(60)

and the equation obtained from this by exchanging τ with τ_1 in the structure appearing after using triangular identity of integration

$$\int_{0}^{t} d\tau \mathbf{A}^{(k+3)}(t,\tau) \mathbf{Z}_{0}(\tau) = \int_{0}^{t} d\tau \int_{\tau}^{t} d\tau_{1} \mathbf{A}^{(4)}(t,\tau_{1}) \mathbf{A}^{(k+2)}(\tau_{1},\tau) \mathbf{Z}_{0}(\tau), \qquad k \ge 2, \tag{61}$$

can be written. This equation enables us to establish the following recursion

$$\mathbf{A}^{(k+3)}(t,\tau) = \int_{\tau}^{t} d\tau_1 \mathbf{A}^{(4)}(t,\tau_1) \mathbf{A}^{(k+2)}(\tau_1,\tau), \qquad k \ge 2.$$
(62)

If we take k = 2 in this recursion the matrix valued function $A^{(5)}(t,\tau)$ is obtained in a block diagonal structure with two blocks in its diagonal. The upper diagonal block of this matrix can be written as follows

$$\left[\boldsymbol{A}^{(5)}(t,\tau)\right]_{11} = \int_{\tau}^{t} d\tau_{1} \int_{\tau_{1}}^{t} d\tau_{2} \varphi_{1}(\tau_{2},\tau_{1}) \mathrm{e}^{(t-\tau_{2})} \boldsymbol{B} \boldsymbol{u}_{1} \int_{\tau}^{\tau_{1}} d\tau_{3} \varphi_{1}(\tau_{3},\tau) \boldsymbol{u}_{4}^{T} \mathrm{e}^{(\tau_{1}-\tau_{3})} \boldsymbol{B} \boldsymbol{u}_{1} \boldsymbol{u}_{4}^{T}.$$
 (63)

If we consider the fact that $\boldsymbol{u}_4^T e^{(\tau_1 - \tau_3)} \boldsymbol{B} \boldsymbol{u}_1$ is scalar in this expression and use the triangular identity of integration together with the following definition

$$\varphi_1^{(5)}(\tau_1,\tau) \equiv \int_{\tau}^{\tau_1} d\tau_2 \varphi_1(\tau_1,\tau_2) \int_{\tau}^{\tau_2} d\tau_3 \varphi_1(\tau_3,\tau) \boldsymbol{u}_4^T \mathrm{e}^{(\tau_2-\tau_3)} \boldsymbol{B}_{\boldsymbol{u}_1},\tag{64}$$

then we can write

$$\left[\boldsymbol{A}^{(5)}(t,\tau)\right]_{11} = \int_{\tau}^{t} d\tau_{1} \varphi_{1}^{(5)}(\tau_{1},\tau) \mathrm{e}^{(t-\tau_{1})\boldsymbol{B}} \boldsymbol{u}_{1} \boldsymbol{u}_{4}^{T}.$$
(65)

The lower diagonal block of the matrix valued function $A^{(5)}(t,\tau)$ can be written as follows by tracing exactly same ways above

$$\left[\boldsymbol{A}^{(5)}(t,\tau)\right]_{22} = \int_{\tau}^{t} d\tau_{1} \varphi_{2}^{(5)}(\tau_{1},\tau) \mathrm{e}^{(t-\tau_{1})\boldsymbol{B}} \boldsymbol{u}_{3} \boldsymbol{u}_{2}^{T}.$$
(66)

The explicit definition of $\varphi_2^{(5)}(\tau_1, \tau)$ here is given below

$$\varphi_2^{(5)}(\tau_1,\tau) \equiv \int_{\tau}^{\tau_1} d\tau_2 \varphi_2(\tau_1,\tau_2) \int_{\tau}^{\tau_2} d\tau_3 \varphi_2(\tau_3,\tau) \boldsymbol{u}_2^T \mathrm{e}^{(\tau_2-\tau_3)} \boldsymbol{B} \boldsymbol{u}_3.$$
(67)

Hence we obtain the following result

$$\boldsymbol{A}^{(5)}(t,\tau) = \begin{bmatrix} \int_{\tau}^{t} d\tau_{1} \varphi_{1}^{(5)}(\tau_{1},\tau) \mathrm{e}^{(t-\tau_{1})} \boldsymbol{B} \boldsymbol{u}_{1} \boldsymbol{u}_{4}^{T} & \boldsymbol{0} \\ \boldsymbol{0} & \int_{\tau}^{t} d\tau_{1} \varphi_{2}^{(5)}(\tau_{1},\tau) \mathrm{e}^{(t-\tau_{1})} \boldsymbol{B} \boldsymbol{u}_{3} \boldsymbol{u}_{2}^{T} \end{bmatrix}.$$
(68)

If we define

$$\varphi_j^{(4)}(\tau_1, \tau) \equiv \varphi_j(\tau_1, \tau) \qquad j = 1, 2,$$
(69)

we get the following result from (48) and (52)

$$\boldsymbol{A}^{(4)}(t,\tau) = \begin{bmatrix} \int_{\tau}^{t} d\tau_{1} \varphi_{1}^{(4)}(\tau_{1},\tau) e^{(t-\tau_{1})} \boldsymbol{B}_{\boldsymbol{u}_{1}} \boldsymbol{u}_{4}^{T} & \boldsymbol{0} \\ \boldsymbol{0} & \int_{\tau}^{t} d\tau_{1} \varphi_{2}^{(4)}(\tau_{1},\tau) e^{(t-\tau_{1})} \boldsymbol{B}_{\boldsymbol{u}_{3}} \boldsymbol{u}_{2}^{T} \end{bmatrix},$$
(70)

which can be generalized as follows

$$\boldsymbol{A}^{(k+4)}(t,\tau) = \begin{bmatrix} \int_{\tau}^{t} d\tau_{1} \varphi_{1}^{(k+4)}(\tau_{1},\tau) e^{(t-\tau_{1})\boldsymbol{B}} \boldsymbol{u}_{1} \boldsymbol{u}_{4}^{T} \quad \boldsymbol{0} \\ \boldsymbol{0} \quad \int_{\tau}^{t} d\tau_{1} \varphi_{2}^{(k+4)}(\tau_{1},\tau) e^{(t-\tau_{1})\boldsymbol{B}} \boldsymbol{u}_{3} \boldsymbol{u}_{2}^{T} \end{bmatrix}, \\ 0 \leq k < \infty. \tag{71}$$

The validity of this equation enforces its expression to satisfy (62). This is possible if and only if the following scalar recursions are satisfied

$$\varphi_1^{(k+5)}(\tau_1,\tau) \equiv \int_{\tau}^{\tau_1} d\tau_2 \varphi_1(\tau_1,\tau_2) \int_{\tau}^{\tau_2} d\tau_3 \varphi_1^{(k+4)}(\tau_3,\tau) \boldsymbol{u}_4^T \mathrm{e}^{(\tau_2-\tau_3)\boldsymbol{B}} \boldsymbol{u}_1, \quad 0 \le k < \infty, \quad (72)$$

$$\varphi_2^{(k+5)}(\tau_1,\tau) \equiv \int_{\tau}^{\tau_1} d\tau_2 \varphi_2(\tau_1,\tau_2) \int_{\tau}^{\tau_2} d\tau_3 \varphi_2^{(k+4)}(\tau_3,\tau) \boldsymbol{u}_2^T \mathrm{e}^{(\tau_2-\tau_3)} \boldsymbol{B}_{\boldsymbol{u}_3}, \quad 0 \le k < \infty.$$
(73)

The uniqueness of the solution of these recursions necessitates initial values which have been given by (69). Last two equations together with (71) and (59) enable us to determine the $\mathbf{Z}(t)$ matrix valued functions with even integer indices uniquely. A similar and detailed investigation permits us to write the following equation for the determination of matrix valued functions $\mathbf{Z}(t)$ with odd integer indices.

$$\boldsymbol{Z}_{2k+1}(t) = \int_0^t d\tau \boldsymbol{A}^{(k+3)}(t,\tau) \boldsymbol{Z}_1(\tau), \qquad k \ge 1.$$
(74)

3. Convergence of the perturbative expansion

Now it is time to deal with the convergence of the perturbation expansion developed above. To this end we may start with the norm of the scalar constructed by sandwiching a square matrix between the transpose of a vector and another vector. Since the entities had physical units until now it is better to scale them with an appropriately chosen diagonal matrix. We can start by considering the following norm inequality

$$\left\|\boldsymbol{u}_{1}^{T} e^{(\tau_{2}-\tau_{3})\boldsymbol{B}} \boldsymbol{u}_{4}\right\| \leq \left\|\boldsymbol{u}_{1} \boldsymbol{M}\right\| \left\|\boldsymbol{M}^{-1} e^{(\tau_{2}-\tau_{3})\boldsymbol{B}} \boldsymbol{M}\right\| \left\|\boldsymbol{M}^{-1} \boldsymbol{u}_{4}\right\|,$$
(75)

where

$$\boldsymbol{M} \equiv \begin{bmatrix} \boldsymbol{I}_N & \boldsymbol{0} \\ \boldsymbol{0} & \frac{1}{\sqrt{m_e \kappa_{max}}} \boldsymbol{I}_N \end{bmatrix}$$
(76)

and I_N stands for $N \times N$ unit matrix. κ_{max} symbolizes the greatest element of κ diagonal matrix. The reason why this matrix is entered above formula lies beneath the desire to get rid of physical units. Indeed p(t) and q(t) are given in [Mass]×[Length]/[Time] and [Length] units because they correspond to momentum and position respectively. By choosing the second

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block of M diagonal matrix in [Mass]/[Time] unit the elements of the vector obtained from the original ones via the transformation characterized by this matrix are brought to same physical units. Same thing is also valid for $\mathbf{r}(t)$ and $\mathbf{s}(t)$ vectors.

Although the above inequality does not explicitly depend on how the norm is defined we will use natural norm here. That is, the norm of a vector will be defined as the square root of the sum of the elements' squares. On the other hand the norm of a matrix is defined as the maximum value of the ratio whose denominator is defined as the norm of a vector arbitrarily chosen from the domain of matrix while the numerator is the norm of the image of same vector under this matrix over its domain. Therefore we can write the following equations from (9) and (76)

$$\|\boldsymbol{u}_1\boldsymbol{M}\| = \|\boldsymbol{\mu}\| = \sqrt{\boldsymbol{\mu}^T\boldsymbol{\mu}},\tag{77}$$

$$\left\|\boldsymbol{M}^{-1}\boldsymbol{u}_{4}\right\| = \sqrt{\left\|\boldsymbol{\alpha}'\right\|^{2} + m_{e}\kappa_{max}\left\|\boldsymbol{\beta}'\right\|^{2}} = \sqrt{\boldsymbol{\alpha}'^{T}\boldsymbol{\alpha}' + m_{e}\kappa_{max}\boldsymbol{\beta}'^{T}\boldsymbol{\beta}'}.$$
 (78)

Now we can consider the action of the exponential matrix in (75) on any arbitrary vector $\boldsymbol{\xi}$ to evaluate its norm. We can write

$$\boldsymbol{\zeta} \equiv \begin{bmatrix} \boldsymbol{\zeta}_1 \\ \boldsymbol{\zeta}_2 \end{bmatrix} \equiv \boldsymbol{M}^{-1} \mathrm{e}^{(\tau_2 - \tau_3)} \boldsymbol{B} \boldsymbol{M} \boldsymbol{\xi} = \boldsymbol{M}^{-1} \mathrm{e}^{(\tau_2 - \tau_3)} \boldsymbol{B} \boldsymbol{M} \begin{bmatrix} \boldsymbol{\xi}_1 \\ \boldsymbol{\xi}_2 \end{bmatrix},$$
(79)

where ζ_1 , ζ_2 , ξ_1 , and ξ_2 symbolize *N*-element vectors. The block nature of the exponential matrix above and last equation enables us to write

$$\boldsymbol{\zeta}_{1} = \cos\left(\frac{(\tau_{2}-\tau_{3})}{\sqrt{m_{e}}}\boldsymbol{\kappa}^{\frac{1}{2}}\right)\boldsymbol{\xi}_{1} - \frac{1}{\kappa_{max}}\boldsymbol{\kappa}\left((\tau_{2}-\tau_{3})\sqrt{\frac{\kappa_{max}}{m_{e}}}\right)S\left(\frac{(\tau_{2}-\tau_{3})}{\sqrt{m_{e}}}\boldsymbol{\kappa}^{\frac{1}{2}}\right)\boldsymbol{\xi}_{2},$$

$$\boldsymbol{\zeta}_{2} = \left(\sqrt{\frac{\kappa_{max}}{m_{e}}}\right)S\left(\frac{(\tau_{2}-\tau_{3})}{\sqrt{m_{e}}}\boldsymbol{\kappa}^{\frac{1}{2}}\right)\boldsymbol{\xi}_{1} + \cos\left(\frac{(\tau_{2}-\tau_{3})}{\sqrt{m_{e}}}\boldsymbol{\kappa}^{\frac{1}{2}}\right)\boldsymbol{\xi}_{2}.$$
 (80)

The norms of the functions S and cosine are bounded by 1 as long as their arguments are real. Same thing is also true for $\frac{1}{\kappa_{max}}\kappa$. By considering these bounds and the fact that the norm of a sum can not exceed the sum of the norms of its summands we can arrive at the following inequalities

$$\begin{aligned} \|\boldsymbol{\zeta}_{1}\|^{2} + \|\boldsymbol{\zeta}_{2}\|^{2} &= \|\boldsymbol{\zeta}\|^{2} \leq \left[1 + \left((\tau_{2} - \tau_{3})\sqrt{\frac{\kappa_{max}}{m_{e}}}\right)^{2}\right] \left(\|\boldsymbol{\xi}_{1}\|^{2} + \|\boldsymbol{\xi}_{2}\|^{2}\right) \\ &\leq \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}\left(\tau_{2} - \tau_{3}\right)\right)^{2} \|\boldsymbol{\xi}\|^{2}. \end{aligned}$$
(81)

These enable us to write

$$\left\|\boldsymbol{M}^{-1}\mathrm{e}^{(\tau_{2}-\tau_{3})}\boldsymbol{B}\boldsymbol{M}\boldsymbol{\xi}\right\|^{2} \leq \left(1+\sqrt{\frac{\kappa_{max}}{m_{e}}}(\tau_{2}-\tau_{3})\right)^{2}\|\boldsymbol{\xi}\|^{2}$$
(82)

and finally

$$\left\| \boldsymbol{M}^{-1} \mathrm{e}^{(\tau_2 - \tau_3)} \boldsymbol{B} \boldsymbol{M} \right\| \leq \left(1 + \sqrt{\frac{\kappa_{max}}{m_e}} (\tau_2 - \tau_3) \right).$$
(83)

In this formulae τ_2 is greater than or equal to τ_3 . If $(\tau_2 - \tau_3)$ term at the right hand side of this inequality is replaced by T then we can get rid of time dependence at the expense of obtaining a more pessimistic inequality.

Now, by using all knowledge we have obtained until this moment, we can produce the following inequalities

$$\begin{aligned} |\varphi_{1}(\tau,\tau_{1})| &\leq \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \|\boldsymbol{M}\boldsymbol{u}_{2}\| \|\boldsymbol{M}^{-1}\boldsymbol{u}_{3}\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}T\right), \\ |\varphi_{2}(\tau,\tau_{1})| &\leq \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \|\boldsymbol{M}\boldsymbol{u}_{4}\| \|\boldsymbol{M}^{-1}\boldsymbol{u}_{1}\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}T\right). \end{aligned}$$
(84)

We can write the following inequalities from (72)

$$\left| \varphi_1^{(k+5)}(\tau_1, \tau) \right| \le \int_{\tau}^{\tau_1} d\tau_2 \left| \varphi_1(\tau_1, \tau_2) \right| \int_{\tau}^{\tau_2} d\tau_3 \left| \varphi_1^{(k+4)}(\tau_3, \tau) \right| \left| \boldsymbol{u}_4^T e^{(\tau_2 - \tau_3)} \boldsymbol{B} \boldsymbol{u}_1 \right|,$$

$$0 \le k < \infty,$$
 (85)

$$\begin{aligned} |\varphi_{1}(\tau_{1},\tau_{2})| \left| \boldsymbol{u}_{4}^{T} \mathrm{e}^{(\tau_{2}-\tau_{3})} \boldsymbol{B}_{\boldsymbol{u}_{1}} \right| &\leq \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \left\| \boldsymbol{M}\boldsymbol{u}_{2} \right\| \left\| \boldsymbol{M}^{-1}\boldsymbol{u}_{3} \right\| \left\| \boldsymbol{M}\boldsymbol{u}_{4} \right\| \left\| \boldsymbol{M}^{-1}\boldsymbol{u}_{1} \right\| \times \\ &\times \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}} T \right)^{2}. \end{aligned}$$

$$(86)$$

If we define

$$C(T) \equiv \frac{W_p^{(max)}}{W_E^{(min)}} \|\boldsymbol{M}\boldsymbol{u}_2\| \|\boldsymbol{M}^{-1}\boldsymbol{u}_3\| \|\boldsymbol{M}\boldsymbol{u}_4\| \|\boldsymbol{M}^{-1}\boldsymbol{u}_1\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_e}}T\right)^2,$$
(87)

(85) takes the following form

$$\left|\varphi_{1}^{(k+5)}(\tau_{1},\tau)\right| \leq C(T) \int_{\tau}^{\tau_{1}} d\tau_{2} \int_{\tau}^{\tau_{2}} d\tau_{3} \left|\varphi_{1}^{(k+4)}(\tau_{3},\tau)\right|, \qquad 0 \leq k < \infty.$$
(88)

If we set k = 0 in this inequality and use (69) together with (84) then the following inequality can be written

$$\left|\varphi_{1}^{(5)}(\tau_{1},\tau)\right| \leq \frac{1}{2} \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \|\boldsymbol{M}\boldsymbol{u}_{2}\| \|\boldsymbol{M}^{-1}\boldsymbol{u}_{3}\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}T\right) C(T)(\tau_{1}-\tau)^{2}.$$
(89)

By generalizing this inequality, that is, using consecutively, we can obtain the following inequality

$$\left|\varphi_{1}^{(k+4)}(\tau_{1},\tau)\right| \leq \frac{1}{(2k)!} \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \left\|\boldsymbol{M}\boldsymbol{u}_{2}\right\| \left\|\boldsymbol{M}^{-1}\boldsymbol{u}_{3}\right\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}T\right) C(T)^{k} (\tau_{1}-\tau)^{2k}. (90)$$

A similar investigation shows that almost same result can be obtained for also $\varphi_2^{(k+4)}(\tau_1,\tau)$

$$\left|\varphi_{2}^{(k+4)}(\tau_{1},\tau)\right| \leq \frac{1}{(2k)!} \frac{W_{p}^{(max)}}{W_{E}^{(min)}} \left\|\boldsymbol{M}\boldsymbol{u}_{4}\right\| \left\|\boldsymbol{M}^{-1}\boldsymbol{u}_{1}\right\| \left(1 + \sqrt{\frac{\kappa_{max}}{m_{e}}}T\right) C(T)^{k}(\tau_{1}-\tau)^{2k}.$$
(91)

We need transformations using M matrix to get rid of physical units, for passing to $Z_k(t)$ matrices. In this connection one can construct bounds to the norms of the matrices appearing in (71) and write the following inequalities

$$\begin{aligned} \left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{u}_1 \boldsymbol{u}_4^T \boldsymbol{M} \right\| &= \left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{M} \boldsymbol{M}^{-1} \boldsymbol{u}_1 \boldsymbol{u}_4^T \boldsymbol{M} \right\| \leq \\ &\leq \left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{M} \right\| \left\| \boldsymbol{M}^{-1} \boldsymbol{u}_1 \right\| \left\| \boldsymbol{u}_4^T \boldsymbol{M} \right\|, \end{aligned}$$
(92)

$$\left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{u}_2 \boldsymbol{u}_3^T \boldsymbol{M} \right\| = \left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{M} \boldsymbol{M}^{-1} \boldsymbol{u}_2 \boldsymbol{u}_3^T \boldsymbol{M} \right\| \leq \\ \leq \left\| \boldsymbol{M}^{-1} \mathbf{e}^{(t-\tau_1)\boldsymbol{B}} \boldsymbol{M} \right\| \left\| \boldsymbol{M}^{-1} \boldsymbol{u}_2 \right\| \left\| \boldsymbol{u}_3^T \boldsymbol{M} \right\|.$$
(93)

If we define the following $4N \times 4N$ type matrix

$$\overline{M} \equiv \begin{bmatrix} M & \mathbf{0} \\ \mathbf{0} & M \end{bmatrix},\tag{94}$$

then the following inequality related to, in one sense, the physical unitless form of $\mathbf{A}^{(k+4)}(t,\tau)$ matrix in (71), can be written as follows

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{A}^{(k+4)}(t,\tau)\overline{\boldsymbol{M}}\right\| \leq \frac{2}{(2k+1)!}C(T)^{k+1}(t-\tau)^{2k+1}, \qquad t \geq \tau.$$
(95)

The right hand side of this inequality has been simplified by using the above definition of C(T)and the fact that the norm of a block diagonal matrix is less than or equal to the sum of the individual norms of its blocks.

Now a careful analysis shows that the initial values of the recursions for odd and even indexed perturbation terms obey the following inequalities

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{Z}_{0}(t)\overline{\boldsymbol{M}}\right\| \leq 2\left(1+\sqrt{\frac{\kappa_{max}}{m_{e}}}T\right),\tag{96}$$

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{Z}_{0}(t)\overline{\boldsymbol{M}}\right\| \leq 8\left(1+\sqrt{\frac{\kappa_{max}}{m_{e}}}T\right)C(T).$$
(97)

If the inequality in (95) is used in (59) and (75) together with last two equalities then

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{Z}_{2k}(t)(t,\tau)\boldsymbol{M}\right\| \leq \frac{4}{(2k+2)!} \left(1 + \sqrt{\frac{\kappa_{max}}{m_e}}T\right) C(T)^{k+1}T^{2k+2}, \qquad 0 \leq k < \infty$$
(98)

and

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{Z}_{2k+1}(t)(t,\tau)\boldsymbol{M}\right\| \leq \frac{16}{(2k+2)!} \left(1 + \sqrt{\frac{\kappa_{max}}{m_e}}T\right) C(T)^{k+2} t^{2k+2}, \qquad 0 \leq k < \infty, \tag{99}$$

are obtained. We can arrive at the following inequality from last two formulae

$$\left\|\overline{\boldsymbol{M}}^{-1}\boldsymbol{Z}(t,\nu)(t,\tau)\boldsymbol{M}\right\| \le (4+16\,|\nu|)\left(1+\sqrt{\frac{\kappa_{max}}{m_e}}T\right)\sum_{k=0}^{\infty}\frac{\nu^{2k}C(T)^{k+1}T^{2k+2}}{(2k+2)!}.$$
(100)

This result shows that, the perturbation expansion proposed in this paper converges for all finite values of T although the convergence may slow down as T increases.

(2k+2)! term appearing in the denominator of the summand in last inequality means that the general term of the series expansion in (100) rapidly diminishes as k grows unboundedly. Although the general term contains $C(T)^k$ in numerator and this power grows as k increases, the growth is overwhelmed by (2k+2)! in denominator because of faster-than-power growth of factorial. This absolutely means that the general term of the perturbation expansion decreases rapidly as k grows. Of course, the theoretical convergence may not give information about how rapid the numerical convergence is. That is, it does not say anything about how many terms are required to get a prescribed precision unless an error estimation formula constructed. Qualitatively we can say that C(T) has an important role about the speed of convergence. As a conclusion, now, we have an always convergent perturbation expansion as long as T remains finite and we can construct approximants from this expansion by truncations after certain order.

4. Concluding remarks

We have shown that a perturbation expansion which assumes weight function containing part of the coefficient matrix appearing in the ordinary differential equation of the evolution matrix with unit matrix initial condition as the parturbation can be developed and it converges for all finite values of control time. The theoretical convergence may not be so practical when the control time incerases. We have also developed a scalar recursion to evaluate the perturbation terms. This reduces the computational complexity enormously. The method is applicable to many cases of weight functions.

The perturbation term can be taken as a deviation from an appropriately defined nominal value of the coefficient matrix in the ordinary differential equation for the evolution matrix. If the time variations in weight functions are quite small this may work well since it will require a small number of first terms from the perturbation expansion.

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References

- Amstrup, J. D., Doll, R. A., Sauerbrey, G., Szab, G., Lorincz, A., (1993), Optimal control of quantum systems by chirped pulses, *Phys. Rev. A*,48, p. 3830.
- [2] Balint-Kurti, G., Manby, Q.F., Artamonov, Ho, T.-S., Rabitz, H., (2005), Quantum control of molecular motion including electronic polarization effects with a two-stage toolkit, J. Chem. Phys., (122), p. 084110.
- [3] Biteen, J.S., Geremia, J.M., Rabitz, H., (2001), Quantum Optimal Quantum Control Field Design Using Logarithmic Maps, *Chem. Phys. Lett.*, 348, p. 440-446.
- [4] Dahleh, M., Peirce, A., Rabitz, H., Ramakrishna, V., (1996), Control of Molecular Motion, Proceedings of the IEEE, 84, pp. 7-15.
- [5] Demiralp, M.,(1994), Determination of the Quantum Motion of the One Dimensional Harmonic Oscillator via Expectation Value Evolutions,), Bull. Tech. Univ. İstanbul, 47 (4), p. 357.
- [6] Demiralp, M., Rabitz, H., (1993), Optimally controlled quantum molecular dynamics: A perturbation formulation and the existence of multiple solutions, *Phys. Rev. A*, 47, p. 809.
- [7] Demiralp, M., Rabitz, H., (1993), Optimally controlled quantum molecular dynamics: The effect of nonlinearities on the magnitude and multiplicity of control-field solutions, *Phys. Rev. A*, 47, p. 831.
- [8] Demiralp, M., Rabitz, H., (1994), Optimal Control of Classical Molecular Dynamics: A Perturbation Formulation and the Existence of Multiple Solutions, J. Math. Chem., 16, p. 185.
- [9] Demiralp, M., Rabitz, H., (1996), Upper and Lower Bounds on the Control Field and the Quality of Achieved Optimally Controlled Quantum Molecular Motion, J. Math. Chem., 19, p. 337-352.
- [10] Demiralp, M., Rabitz, H., (1997), Dispersion-free Wave Packets and Feedback Solitonic Motion in Controlled Quantum Dynamics, Phys. Rev. A, 55, p. 673.
- [11] Demiralp, M., Rabitz, H., (1998), Assessing Optimality and Robustness of Control over Quantum Dynamics, *Phys. Rev. A*, 57, p. 2420-2425.
- [12] Demiralp,M., Rabitz,H., (2000), Assessing Optimality and Robustness for the Control of Dynamical Systems, *Phys. Rev. A*, 61, p. 2569.
- [13] Demiralp, M., Tunga, B., (2005), Instantaneous Stability and Robustness Investigations in Quantum Optimal Control: Harmonic Oscillator Under Linear Dipole and Quadratic Control Agents, ANACM, 2(1), p. 60-69.
- [14] Dey, B.K., Rabitz, H., Aşkar, A., (2000), Optimal control of molecular motion expressed through quantum fluid dynamics, *Phys. Rev. A*, 61, p. 043412-1-6.
- [15] Dey, B., Rabitz, H., Aşkar, A., (2003), Optimal Reduced Dimensional Representation of Classical Dynamics, *J. Chem. Phys.*, 119, p. 5379-5387.
- [16] Geremia, J.M., Rabitz, H., (2001), Global nonlinear algorithm for inverting quantum-mechanical observations, Phys. Rev. A, 64, p. 022710-1-13.
- [17] Judson, R.S., Rabitz, H., (1992), Teaching Lasers to Control Molecules, Phys. Rev. Lett., 68, p. 1500.
- [18] Kurşunlu, A., Yaman, I., Demiralp, M., (2003), Optimal Control of One Dimensional Quantum Harmonic Oscillator Under an External Field With Quadratic Dipole Function and Penalty on Momentum: Construction of the Linearised Field Amplitude Integral Equation, ANACM, 1, p. 277-286.
- [19] Li, B., Turinici, G., Ramakrishna, V., Rabitz, H., (2002), Optimal Dynamical Discrimination of Similar Molecules Through Quantum Learning Control, J. Phys. Chem. B, 106, p. 8125-8131.
- [20] de Lima, E., Ho, T.-S., Rabitz, H., (2008), Solution of the Schrödinger equation for the Morse potential with an infinite barrier at long range, J. Phys. A: Math. Gen, 41, p. 335303.
- [21] Luo, Z-P., Rabitz, H., (1987), Expected Value Analysis: Experimental Considerations and Convergence Properties, Appl. Math. and Comp., 23, p.25.
- [22] Meral, E., Demiralp, M.,(2009), Determination of the external field amplitude and deviation parameter through expectation value based quantum optimal control of multiharmonic oscillators under linear control agents, J. Math. Chem., 46, pp. 834-852.

- [23] Morris, D., Schwieters, C., Littman, M., Rabitz, H., (1994), Simulator of optimally controlled molecular motion, Am. J. Phys., 62, p. 817.
- [24] Pechen, A., Rabitz, H., (2006), Teaching the environment to control quantum systems, Phys. Rev. A, 73, p. 062102.
- [25] Peirce, A., Dahleh, M., Rabitz, H., (1988), Optimal Control of Quantum Mechanical Systems: Existence, Numerical Approximations, and Applications, *Phys. Rev. A*, 37, p. 4950.
- [26] Rabitz, H., (1992), Optimal Control of Molecular Motion, in *Coherence Phenomena in Atoms and Molecules in Laser Fields*, edited by A.D. Bandrauk and S.C. Wallace, Plenum Publishing Corporation, New York, 315 p.
- [27] Rabitz, H., (1994), Control of quantum dynamics: issues and alternatives, in Laser Techniques for State-Selected and State-to-State Chemistry II, SPIE Proceedings, 2124, p. 84.
- [28] Rabitz, H., (1995), Control of Quantum Dynamics: The Dream is Alive, AIP Conference Proceedings, 334, p. 160-163.
- [29] Rabitz, H., (2002), Controlling Molecular Motion: The Molecule Knows Best, in Laser Control and Manipulation of Molecules (ACS Symposium Series 821), A. Bandrauk, R.J. Gordon and Y. Fukimura, eds., American Chemical Society, Washington, DC, pp. 2-15.
- [30] Rabitz, H., Ho, T. -S., Hsieh, M., Kosut, R., Demiralp, M., (2006), Topology of optimally controlled quantum mechanical transition probability landscapes, *Phys. Rev. A*, 74, p. 012721.
- [31] Rabitz, H., Hsieh, M., Rosenthal, C., (2004), Quantum Optimally Controlled Transition Landscapes, Science, 303, p. 998.
- [32] Rabitz, H., Shi, S., (1991), Optimal Control of Molecular Motion: Making Molecules Dance, in Advances in Molecular Vibrations and Collision Dynamics, edited by Joel Bowman, 1, Part A, p. 187 - 214 (JAI Press, Inc.
- [33] Rabitz, H., Zhu, W., (2000), Optimal Control of Molecular Motion: Design, Implementation, and Inversion, Accts. Chem. Res., 33, p. 572-578.
- [34] Schwieters, C.D., Beumee, J.G.B., Rabitz, H., (1990), Optical Control of Molecular Motion with Robustness and Application to Vinylidene Fluoride, J. Opt. Soc. America B, 7, p. 1736.
- [35] Shankar, R., (1994), Principles of Quantum Mechanics (Hardcover), Springer Science.
- [36] Sharp, R., Rabitz, H., (2004), Mechanism analysis of controlled quantum dynamics in the coordinate representation, J. Chem. Phys., 121, p. 4516.
- [37] Shuang, F., Zhou, M., Pechen, A., Wu, R., Shir, O., Rabitz, H., (2008), Control of quantum dynamics by optimized measurements, *Phys. Rev. A*, 78, p. 063422.
- [38] Smith, L., Augustin, S., Rabitz, H., (1982), Numerical Methods for Solving Time-Dependent Quantum-Mechanical Problems with Applications, J. Comp. Phys., 45, p. 417.
- [39] Sugny, D., Kontz, C., (2008), Optimal control of a three-level quantum system by laser fields plus von Neumann measurements, *Phys. Rev. A*, 77, p. 063420.
- [40] Tunga, B., Demiralp, M., (2003), Optimally Controlled Dynamics of One Dimensional Harmonic Oscillator: Linear Dipole Function and Quadratic Penalty, ANACM, 1, pp. 245-253.
- [41] Turinici, G., Rabitz, H., (2001), Quantum Wave Function Controllability, Chem. Phys., 267, pp. 1-9.
- [42] Vugmeister, B.E., Rabitz, H., (1997), Optimal Control of Laser Induced Transient Birefringence in Liquid Crystals, *Ferroelectrics*, 202, pp. 105-114.
- [43] Walmsley, I., Rabitz, H., (2003), Quantum Physics Under Control, Physics Today, 56, pp. 43-49.
- [44] Wu, R., Sola, I., Rabitz, H., (2004), Optimal Quantum Control with Multi-Polarization Fields, Chem. Phys. Let, 400, pp. 469-475.
- [45] Wu, R., Rabitz, H., Turinici, G., Sola, I., (2004), Connectivity analysis of quantum control, Phys. Rev. A, 70, p. 052507.
- [46] Yao, K., Shi, S., Rabitz, H., (1991), Optimal Control of Molecular Motion: Nonlinear Field Effects, Chem. Phys., 150, p. 373.
- [47] Yip, F., Mazziotti, D., Rabitz, H., (2003), A Propagation Toolkit to Design Quantum Controls, J. Chem. Phys., 118, p. 8168-8172.
- [48] Zhu, W., Rabitz, H., (1999), Molecular Dipole Function Inversion from Time-dependent Probability Density and Electric Field Data, J. Phys. Chem. A, 103, p. 10187-10193.
- [49] Zhu, W., Rabitz, H., (2005), Perturbative and non-perturbative master equations for open quantum systems, NSF, DOD, J. Math. Phys., 46, p. 022105.



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