Theor Chim Acta (1993) 86: 511–524

Theoretica Chimica Acta

On the development of exponential ansatze for quantum dynamics in finite dimensional vector spaces

G. Sree Latha and M. Durga Prasad
School of Chemistry, University of Hyderabad, Hyderabad 500 134, India

Received October 23, 1992/Accepted April 27, 1993

Summary. A Wei–Norman type of exponential ansatz is constructed for the time evolution operator in finite dimensional vector spaces. Based on an analysis of the structure of the concerned operator algebra, it is shown that a reduction principle exists even for simple algebras that goes beyond the Wei–Norman result when a specific ordering of the operators is used such that the equations of motion for different generators belonging to different classes are decoupled. It is shown that the solution in this case is global. Some specific approximation schemes are considered and their strengths and weaknesses are analyzed. Model calculations are presented to bring out these features.

Key words: Perturbative molecular dynamics – Exponential ansatze – Algebraic approaches

1 Introduction

Time-dependent approaches for describing quantum dynamical processes have been growing exponentially due to the computational advantages these methods offer compared to the time independent methods. Obtaining the information of a molecular process in time domain requires integrating the Schroedinger equation in a large Hilbert space [1]. Exact methods require large basis sets which grow exponentially with the size of the system and hence it is desirable to develop and test approximate methods. Several such methods have been discussed in literature [2–21]. Among the available approximate methods perturbation theory enjoys a pre-eminent position.

In the conventional approach to the perturbation theory, the time evolution operator is expanded as power series in terms of the interaction hamiltonian. In such an expansion, the approximate evolution operator obtained by truncating the series is not always unitary. For developing unitary approximations to the evolution operator the exponential ansatz is very appropriate. In this approach, the evolution operator U_F is parameterized as:

$$U_F(t) = \exp[A(t)]. \tag{1.1}$$

Here A is evaluated perturbatively instead of U_F. In addition, A is constrained to remain antihermitian at each order, thus guaranteeing an unitary evolution operator irrespective of the order of truncation in A. A perturbation theory based on the above prescription is termed as the Magnus expansion [6–9].

Several authors discussed the validity of Magnus expansion [10–17]. The Magnus expansion is subject to two major limitations. First, the theory is essentially perturbative, since the governing equation for the generator A obtained from the Schroedinger's equation:

$$iU_F^{-1}\dot{U}_F = U_F^{-1}HU_F, \tag{1.2}$$

contains infinite-order polynomials in A. Consequently no non-perturbative solution is possible in practice. Second, under certain exotic conditions, even the existence of the solution to that equation is questionable [7, 16]. In addition to these formal problems, the Magnus expansion breaks down when some of the unperturbed wave functions are degenerate. To avoid this, degenerate perturbation theories have been developed [24–28]. In these approaches a set of quasi-degenerate states is defined as a model space and an effective hamiltonian is posited that generates the dynamics of the projection of the exact wave function in the model space. A wave operator is invoked to map the model space component to the exact wave function. Perturbation theory is now applied to the wave operator, while the couplings within the model space are treated exactly. It is possible to modify the Magnus expansion to provide such an effective hamiltonian. The resulting theory has an appearance very similar to canonical van Vleck perturbation theory [25]. However, it is also subject to the same deficiencies of the original formulation.

In view of the above considerations it is desirable to develop alternate perturbation theories for model space effective hamiltonians based on exponential ansatze. Such a theory should be free from any questions regarding the existence of solution. In addition, the equations for the generators of such a theory should be finite-order polynomials, so that a non-perturbative solution is possible. The earliest attempts to eliminate the existence problems are due to Wei and Norman [18, 19]. These authors parametrized the time evolution operator in a non-canonical product of exponential operators.

$$U_F = \prod_K \exp(g_K A_K) \tag{1.3}$$

The time-dependent complex functions g_K are defined by a set of nonlinear differential equations obtained by substituting ansatz (1.3) into Eq. (1.2). The operators A_K are the generators of the Lie-algebra to which the hamiltonian belongs. Wei and Norman have shown that when the Lie-algebra to which the hamiltonian belongs is a solvable algebra, it is possible to choose the sequence of operators in Eq. (1.3) such that a global solution to U_F can be obtained [18, 19, 21]. In addition, they have developed a reduction principle to disentangle the equations of motion of different sets of coefficients when the Lie-algebra in question is not simple [19, 21]. The projection operator algebra that we use is a simple algebra and hence cannot be subjected to their analysis. Not withstanding this, it is possible to parametrize the evolution operator in a product form such that a limited version of reduction principle is available in that the equation of motion of different groups of coefficients are decoupled from each other and contain finite order polynomials only. In addition, the existence of the solution to these equations can be proved rigorously. This part of the theory is presented in Sect. 2.2 in the context of

quasi-degenerate perturbation theory after a review of the elements of the requisite effective hamiltonian theory for model spaces in Sect. 2.1. We then consider three different methods to generate approximations to these equations in Sect. 2.3 and assess their relative merits. We have applied our methodology to a laser driven Morse oscillator with a view to understand the characteristic convergence pattern of these methods and the results obtained are discussed in Sect. 3. We have presented our conclusions in Sect. 4.

2 Methodology

2.1 Model space dynamics

We assume that the state of the system whose dynamics are to be described is an element of a finite dimensional vector space spanned by the basis functions $\{|n\rangle, 1 \leqslant n \leqslant N\}$. The dynamics of a system are often confined to a small subspace and the states involved in this space require exact treatment while the rest of the states mix only weakly with the manifold of the strongly interacting states. Accordingly we partition the Hilbert space into two orthogonal subspaces: Model space M, consisting of the strongly interacting states characterized by the projection operator P and the remaining states spanning the virtual space V, (projection operator Q).

We now focus our attention on a nonstationary state ψ that was constructed as a superposition of the model space functions alone at some initial time:

$$\psi(0) = \sum_{n \in M} c_n |n\rangle, \tag{2.1a}$$

and evolves in time according to the Schroedinger equation:

$$i\dot{\psi} = H\psi. \tag{2.1b}$$

Our desire is to define a model space effective hamiltonian H_M such that the evolution of the model space component $\phi(t)$ of $\psi(t)$ is described via an equation of motion of the form:

$$i\dot{\phi} = H_M \phi, \tag{2.2a}$$

$$\phi(t) = P\psi(t) = \sum_{n \in M} c_n(t)|n\rangle. \tag{2.2b}$$

With this goal in mind, we define a common wave operator [24] U for all the states in M. Thus:

$$\psi(t) = U(t)\phi(t). \tag{2.2c}$$

From Eq. (2.1b) and (2.2c) it follows that ϕ satisfies:

$$i\dot{\phi} = \bar{H}\phi, \tag{2.3a}$$

$$\bar{H} = U^{-1}HU - iU^{-1}\dot{U}. \tag{2.3b}$$

Comparing Eq. (2.2a) and (2.3a) we find:

$$H_M = P\bar{H}P. \tag{2.3c}$$

The function ϕ will evolve within the model space as long as:

$$Q\bar{H}P = 0 \tag{2.3d}$$

is satisfied and the initial conditions are specified by Eq. (2.1a). Consequently, the governing equation for U is given by:

$$Q(U^{-1}HU - iU^{-1}\dot{U})P = 0. \tag{2.3e}$$

Note that Eq. (2.3d) serves to define the coefficients of only such operators in U which induce transitions from the model space to virtual space. The coefficients of the rest of the operators are indeterminate, and as it turns out, irrelevant. Additional conditions can be imposed on $\bar{H}$ to determine $\dot{U}$ more fully such as, for example:

$$P\bar{H}Q = 0, \tag{2.3f}$$

which then allows the determination of coefficients of operators which induce transitions from the virtual space to the model space. However, as long as Eq. (2.3e) is exactly satisfied, these additional conditions have no influence on H_M as we shall demonstrate in Sect. 2.3.

In most practical calculations one is more interested in the full wave function or expectation values of the dynamical variables rather than the projection of the wave function onto the model space. The full wave function is obtained by operating on ϕ by U. We now turn to the calculation of physically relevent expectation values. The expectation value of a dynamical variable O is given by:

$$\langle O\rangle = \langle\psi|O|\psi\rangle = \langle\phi|U^{+}OU|\phi\rangle \tag{2.4a}$$

With ϕ and U obtained from Eq. (2.2a) and (2.3e) respectively, this provides a straight forward procedure. An alternative procedure, more in the spirit of the effective operators in the model space, is the following. We define an auxiliary bra function $\langle\bar{\phi}|$ by:

$$\langle\bar{\phi}| = \langle\psi|U. \tag{2.4b}$$

From the Schroedinger equation for $\langle\psi|$:

$$-i\langle\dot{\psi}| = \langle\psi|H, \tag{2.4c}$$

we get the working equation for $\langle\bar{\phi}|$ as:

$$-i\langle\dot{\bar{\phi}}| = \langle\bar{\phi}|\bar{H}, \tag{2.4d}$$

where $\bar{H}$ is defined as before. The expectation value of O is now given by:

$$\langle O\rangle = \langle\psi|O|\psi\rangle = \langle\bar{\phi}|\bar{O}|\phi\rangle, \tag{2.4e}$$

where

$$\bar{O} = U^{-1}OU. \tag{2.4f}$$

Here $\bar{O}$ is the model space effective operator corresponding to O in the full Hilbert space. This result holds irrespective of whether U is unitary or not. One noteworthy point here is that $\langle\bar{\phi}|$ is not the complex conjugate of $|\phi\rangle$, but must be evolved independently according to Eq. (2.4d). In general, $\langle\bar{\phi}|$ is not confined to the model space. However, if Eq. (2.3f) is imposed in addition to Eq. (2.3d) while determining U, $\langle\bar{\phi}|$ would also be confined to the model space.

2.2 Operator algebra and the evolution operator

We now turn to the construction of the model space wave operator U. The set of operators that act on the vector space of functions is a complete set and is spanned by the basis set of the generators of the unitary group, $L = \{|i\rangle\langle j|, 1 \leqslant i, j \leqslant N\}$. This operator set is closed under commutation, thus forming a Lie algebra. The hamiltonian, given by:

$$H = \sum_{i,j} h_{ij} |i\rangle\langle j| \tag{2.5}$$

is an element of this Lie algebra. Given that the hamiltonian is an element of the algebra, the full evolution operator U_F can be parametrized, atleast locally, as:

$$U_F = \exp\left[\sum_{i,j} f_{ij}(t) |i\rangle\langle j|\right], \tag{2.6a}$$

and satisfies the Schroedinger equation:

$$i\dot{U}_F = HU_F. \tag{2.6b}$$

Eq. (2.6a) is the starting point for the Magnus expansion approach [6–9]. An alternative form is to write U as a product of exponentials (the so called Wei–Norman form) rather than a single exponential [18, 21]:

$$U_F = U_1 U_2 U_3 \cdots, \tag{2.7}$$

where each U_i is an exponential operator. Wei and Norman [18] and more recently Wolf and Korsch [21] have discussed a reduction principle to obtain a convenient sequence of U_i when the algebra is semisimple. In addition, Wei and Norman have shown that for solvable Lie-algebras there exists a basis and an ordering of the basis for which the product form is global. Since the only invariant sub-algebras of the projection operator algebra are the null set and itself, the algebra is simple and hence this procedure cannot be used here. However, it turns out that a limited form of a reduction principle is possible in this case also due to the special structure of the algebra. To exhibit this structure we classify the operator set as follows:

set of excitation operators: $E = \{X_{vm} = |v\rangle\langle m|;\ v \in V,\ m \in M\}$

set of deexcitation operators: $D = \{Y_{mv} = |m\rangle\langle v|;\ v \in V,\ m \in M\}$

set of shift operators: $S = \{Z_{mn} = |m\rangle\langle n|,\ W_{uv} = |u\rangle\langle v|;\ m, n \in M;\ u, v \in V\}$

These operators satisfy the following commutation relations,

$$[X, X] = [Y, Y] = 0, \tag{2.8a}$$

$$[X, Y] = Z + W, \tag{2.8b}$$

$$[X, Z] = X = [X, W], \tag{2.8c}$$

$$[Y, Z] = Y = [Y, W]. \tag{2.8d}$$

Note that each of these sets defines a subalgebra of L. In addition $S \cup E$ and $S \cup D$ are also closed under commutation. We now note that (a) the excitation operators

in H can be eliminated by a similarity transformation generated by X alone: Consider the parametrization:

$$U_F = U_X U_R, \tag{2.9a}$$

$$U_X = \exp(X). \tag{2.9b}$$

The equation of motion for U_R is given by:

$$i\dot{U}_R = H_R U_R, \tag{2.9c}$$

$$H_R = U_X^{-1} H U_X - i U_X^{-1} \dot{U}_X. \tag{2.9d}$$

In other words, U_R is generated by the effective hamiltonian operator H_R. It is possible to eliminate all X operators in H_R by requiring:

$$Q(U_X^{-1} H U_X - i U_X^{-1} \dot{U}_X) P = 0, \tag{2.9e}$$

which provides the working equation for $\dot{U}_X$. (b) If the effective hamiltonian operator H_R does not contain X operators then U_R can be parametrized as:

$$U_R = \exp[Y + Z + W], \tag{2.9f}$$

since $S \cup D$ is a Lie algebra and H_R belongs to it. Combining these two statements, and invoking similar arguments with respect to the deexcitation operators, we arrive at:

$$U_F = \exp(X)\exp(Y)\exp(Z + W). \tag{2.10}$$

Thus the model space wave operator U is given by:

$$U = \exp(X)\exp(Y), \tag{2.11}$$

since the effect of Z can be absorbed into ϕ and W acting on the model space gives zero. The generators X and Y satisfy:

$$i\dot{X} = Q \exp(-X) H \exp(X) P, \tag{2.12a}$$

$$i\dot{Y} = P \exp(-Y) [\exp(-X) H \exp(X) - i\dot{X}] \exp(Y) Q. \tag{2.12b}$$

These equations are decoupled since X does not depend upon Y. In this sense we have obtained a reduction of the operator set. Let us note that any other ordering of operators (e.g. $\exp(Z)\exp(X)\exp(Y)\exp(W)$) does not lead to decoupled sets of equations.

2.3 Perturbation theory

We now turn to the explicit solution of Eq. (2.12). From Eq. (2.12a) we obtain by Hausdorff expansion:

$$\begin{aligned} i\dot{X} &= Q(H + [H, X] + 1/2! [[H, X], X]) P, \\ &= H_{QP} + H_{QQ} X - X H_{PP} - X H_{PQ} X. \end{aligned} \tag{2.13a}$$

This equation is a matrix Ricatti equation and is the result of a multistate generalization of the nonlinear quotient approach discussed by Dion and Hirschfelder [29]. The existence theorem proved in that context (theorem 9 of

Ref. [29]) holds here also. Since X is a $v \times m$ matrix operator where v and m are the dimensions of the V and M respectively, parametrizing X formally as:

$$X = FG^{-1}, \tag{2.13b}$$

where F is a $v \times m$ matrix and G is a $m \times m$ matrix and substituting it into Eq. (2.13a) we obtain:

$$i\dot{F}G^{-1} - iFG^{-1}\dot{G}G^{-1} = H_{QP} + H_{QQ}FG^{-1} - FG^{-1}H_{PP} - FG^{-1}H_{PQ}FG^{-1}. \tag{2.13c}$$

This equation can now be decoupled into two sets by writing:

$$i\dot{G} = H_{PP}G + H_{PQ}F, \tag{2.13d}$$

$$i\dot{F} = H_{QP}G + H_{QQ}F. \tag{2.13e}$$

Note that Eqs. (2.13d, 2.13e) recover the original Schroedinger equation. Since the solution to this always exists, X also has a solution. The initial condition:

$$X(0) = 0 \tag{2.13f}$$

is satisfied by requiring:

$$F(0) = 0, \tag{2.13g}$$

$$G(0) = 1 \tag{2.13h}$$

without loss of generality. In a similar fashion, it can be shown that Eq. (2.12b) for Y also has a global solution.

Since the multicommutator expansion in Eq. (2.13a) is finite, a nonperturbative solution for X is possible in contrast to Magnus expansion. In addition, a perturbative expansion for X by this approach has far fewer terms at any order compared to its counterpart by Magnus expansion.

Expanding X perturbatively we obtain:

$$X = \sum_n \lambda^n X_n, \tag{2.14a}$$

$$i\dot{X}_n = H_{QP}\delta_{n1} + H_{QQ}X_{n-1} - X_{n-1}H_{PP} - \sum_{r=1}^{n-2} X_r H_{PQ} X_{n-r-1}, \tag{2.14b}$$

where n is the order of perturbation. The model space effective hamiltonian (Eq. (2.2b)) becomes:

$$H_M = H_{PP} + H_{PQ}X - Y(H_{QP} + H_{QQ}X - XH_{PP} - XH_{PQ}X - i\dot{X}). \tag{2.15a}$$

If X satisfies Eq. (2.13a) exactly, this reduces to:

$$H_M = H_{PP} + H_{PQ}X. \tag{2.15b}$$

This is expected since the action of Y on the model space gives zero. Consequently these operators should have no influence on the model space dynamics. Thus a simple minded perturbation theory consists of two steps: (*1*) solve Eq. (2.14b) to some finite order. (*2*) Construct H_M according to Eq. (2.15b) and integrate Eq. (2.2a) for ϕ. We shall refer to this approach as the similarity transformation based perturbation theory (STP), since in effect, it postulates $U = \exp(X)$.

One possible problem with STP is that, in the strong coupling regime the perturbative dynamics may not conserve the norm of the wave function. This is the well-known problem of intruder states [24]. When Eq. (2.3d) is satisfied, these states have no influence on the model space dynamics because, any vector from the model space would evolve within the model space as long as $\bar{H}_{QP} = 0$. If X is obtained from perturbation theory, Eq. (2.3d) is violated. In such a case, invocation of Eq. (2.15b) is equivalent to replacing $\bar{H}$ in Eq. (2.3b) with H_A such that:

$$H_A = \bar{H} - R, \tag{2.16a}$$

where

$$QRP = Q\bar{H}P. \tag{2.16b}$$

The hamiltonian corresponding to H_A in the full Hilbert space is

$$\tilde{H} = H - URU^{-1} \tag{2.17}$$

If the $\tilde{H}$ operator is non-hermitian, it could, in the course of its evolution, develop complex eigenvalues. The eigenvectors associated with these eigenvalues are the intruder states. When the intruder states develop a large component in the model space at some stage, the model space states would grow exponentially. Such a situation can be expected when the states in the model space interact strongly with the virtual space.

There are two ways to eliminate the influence of the intruder states. The first approach is to expand the model space to incorporate all the strongly interacting states as advocated by Jolicard and Grosjean [24]. This forms the basis of the intermediate hamiltonian approach discussed in the context of the usage of incomplete model spaces along with the coupled cluster method for stationary states [28]. A second approach is to ensure that $\tilde{H}$ is hermitian through out the course of the evolution. Note that only the QP block of the R operator is specified by Eq. (2.16b). It is possible to use this flexibility to ensure a hermitian $\tilde{H}$. Essentially $\tilde{H}$ is hermitian if $\bar{R} = URU^{-1}$ is hermitian. Thus, in terms of the sub-blocks of the R operator the following equations must be satisfied to guarantee the hermiticity of $\tilde{H}$.

$$\bar{R}_{QQ} = R_{QQ} + XR_{PQ} = \bar{R}^{+}_{QQ}, \tag{2.18a}$$

$$\bar{R}_{PP} = R_{PP} - R_{PP}X = \bar{R}^{+}_{PP}, \tag{2.18b}$$

$$\bar{R}_{PQ} = R_{PQ} = \bar{R}^{+}_{QP}. \tag{2.18c}$$

Since R_{PP}, R_{PQ} and R_{QQ} are not defined by Eq. (2.16b) additional conditions can be imposed. For example setting:

$$\bar{R}_{PQ} = R_{PQ} = R_{PP} = 0, \tag{2.19}$$

we arrive at:

$$\bar{R}_{PP} = 0, \tag{2.20a}$$

$$\bar{R}_{QP} = (R_{QP} - R_{QQ}X) = 0. \tag{2.20b}$$

Thus choosing an R_{QQ} such that $R_{QQ} = R^{+}_{QQ}$ and requiring $R_{QP} = R_{QQ}X$ would guarantee that $\tilde{H}$ is hermitian. In such a case X must satisfy:

$$i\dot{X} = H_{QP} + (H_{QQ} - R_{QQ})X - XH_{PP} - XH_{PQ}X. \tag{2.21}$$

In this case X cannot be obtained as a power series. Instead approximations to X are obtained by defining different R_{QQ} matrices. We term approaches based on Eq. (2.21) as the hermitised similarity transformation-based theories (HST).

An alternative approach to avoid norm violations is to insist that the full evolution operator of Eq. (2.10) underlying the model space evolution operator to be unitary. It can be shown by direct substitution that U_F is unitary if Y, Z and W satisfy the following equations:

$$Y = -(1 + X^+X)^{-1} X^+, \tag{2.22a}$$

$$Z = -1/2 \ln(1 + X^+X), \tag{2.22b}$$

$$W = -1/2 \ln(1 + XY). \tag{2.22c}$$

Thus, usage of Eq. (2.15a) with Eq. (2.22a) for Y ensures that there is no norm violation. This approach will be called as the unitary transformation based perturbation theory (UTP) in the following.

In the next section we study the numerical performances of these three methods.

3 Model applications

To examine the relative performances of the three approaches described in the previous section and also to gain an understanding of their convergence properties we have used the three approaches to follow the dynamics of a harmonically driven Morse oscillator. The hamiltonian of the system is given by:

$$H = H_0 + V \tag{3.1}$$

$$H_0 = p^2/2m + D(1 - e^{-\alpha x})^2 \tag{3.2a}$$

$$V = A_0 x \cos(\omega t) \tag{3.2b}$$

The parameters D, α, and m were chosen to correspond to that of HF molecule [22]. A_0 was chosen such that the intensity of the external field was in the range of 1 to 2 TW/cm^2 [23].

In all the calculations presented below the frequency of the radiation field was set to be in 1:1 resonance between ground and the first excited states. Consequently, these two states are degenerate and interact strongly, while the coupling to the rest of the states is taken to be weak enough to be subjected to a perturbative treatment. Thus all the perturbative calculations were carried out in the interaction picture with a model space spanning the ground and first excited states and the lowest ten eigenstates of the Morse oscillator were used to define the complete vector space.

In Fig. 1 we plot the expectation value of the unperturbed hamiltonian of the Morse oscillator calculated by STP as a function of time when the intensity of the driving field is 2 TW/cm^2. Upto about 15 optical cycles (o.c.) the perturbation theory provides an improvement over the zeroth order description with the accuracy of the perturbation solution improving with increasing order. Beyond that the energy calculated by all the orders blows up indicating the divergent nature of the perturbation series. This conclusion is reinforced by Fig. 2 in which norm of the perturbed wave function defined by:

$$N = \langle \phi | \exp(X^+) \exp(X) | \phi \rangle^{1/2} \tag{3.3}$$

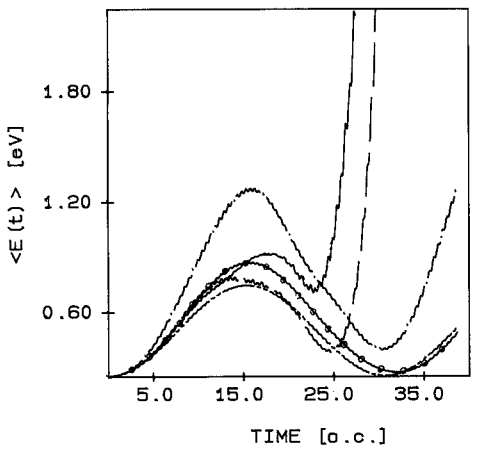

Fig. 1. Energy of the Morse oscillator as a function of time with the field intensity $I = 2.0\ \mathrm{TW/cm^2}$ by STP. *Continuous line with circles*: Converged basis set calculation. *Dash* and *double dotted line*: Basis set restricted to two functions. *Dash* and *dotted line*: First order. *Dashed line*: third order. *Continuous line*: Fifth order

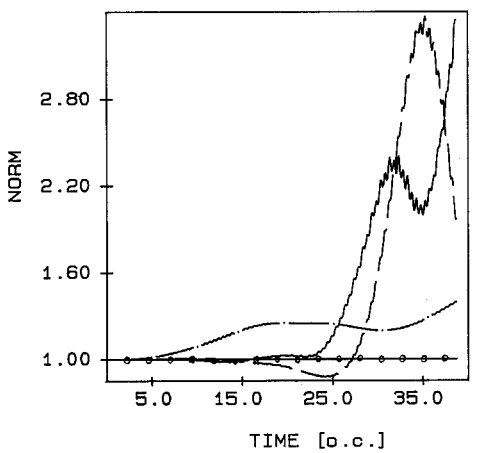

Fig. 2. Norm of the perturbed wave function at $I = 2\ \mathrm{TW/cm^2}$. Figure conventions are the same as before

is plotted as a function of time. It appears from this and other unpublished data that the higher the order of the perturbation theory the later is the occurrence of the norm violation. But once the norm violation begins it appears to grow faster in higher order approximations.

In Fig. 3 we present a similar calculation to Fig. 1 but based on HST in which we have set $R_{QQ} = H_{QQ}$. This is equivalent to solving the equation:

$$i\dot{X} = H_{QP} - XH_{PP} - XH_{PQ}X \tag{3.4}$$

for X. This is a nonperturbative approximation and as may be expected performs quite satisfactorily.

In Fig. 4 the energy expectation value calculated by the UTP approximations is plotted as a function of time at the same intensity as Fig. 1. In the early phase of the time development (upto about 20 o.c.) the approximation based on the unitary transformation is far worse than the corresponding STP version. However, since there is no norm violation in this approach its validity improves at longer times

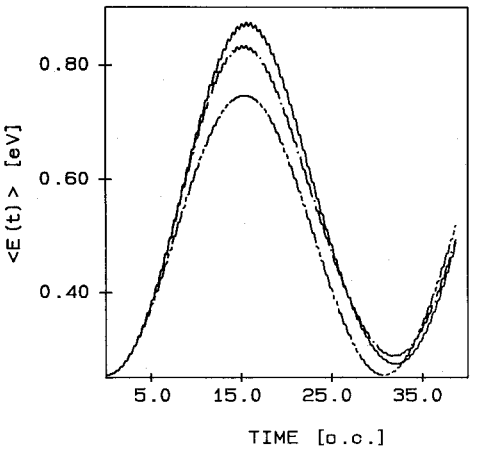

Fig. 3. Energy of the Morse oscillator as a function of time by HST. *Continuous line*: Converged basis set calculation. *Dash* and *double dotted line*: Basis set restricted to two basis functions. *Dash* and *dotted line*: HST calculation

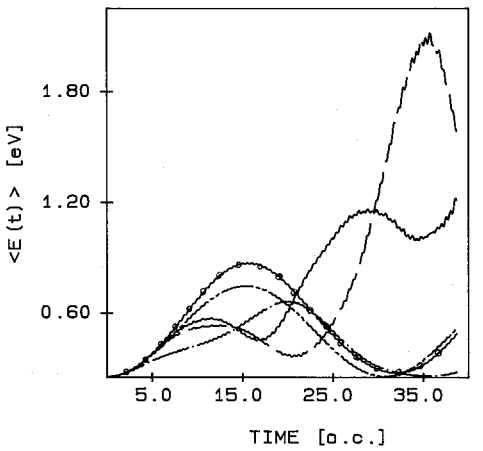

Fig. 4. Energy of the Morse oscillator as a function of time with the field intensity $I = 2.0\,\mathrm{TW/cm^2}$ by UTP. Figure conventions are the same as in Fig. 1

when the approximations based on the STP breakdown due to norm violations. However these approximations seem to be worse than the hermitised version.

Comparing the energy expectation values obtained from STP and UTP approaches (Figs. 1 and 4), it appears that the result of STP version is close to exact result upto about 20 o.c. Beyond that it diverges. The UTP version underestimates the energy atleast in the initial phase. The explanation for this can be given as follows: Substituting for Y in Eq. (2.15a) results in the equation:

$$H_M = (1 + X^+X)^{-1}(H_{PP} + H_{PQ}X + X^+H_{QP} + X^+H_{QQ}X - iX^+X) \tag{3.5}$$

It can be seen from this equation that when a particular X value turns out to be large, the corresponding value of the factor $(1 + X^+X)^{-1}$ will be very small, consequently the matrix elements in the corresponding row of H_M become negligible. This means, the model space state which is strongly interacting with the virtual space is being eliminated from the model space for all practical purposes. In effect then the model space is reduced to one function and the wave packet is unable to gain energy.

4 Concluding remarks

Degenerate perturbation theory is a convenient tool for the description of dynamical systems with a large number of strongly interacting states. Any approach to such a perturbation theory defines a common wave operator for all the states in the model space [24–28]. In this work we have studied the development of three such approaches based on exponential ansatze. We have opted for Wei–Norman type of product form for the time evolution operator rather than a single exponential type normally invoked in the Magnus expansion. Based on the structural properties of the Lie algebra under consideration we have shown that the evolution operator is partially reducible in the sense that the computation of the time dependent coefficients associated with the excitation operators is decoupled from the computation of the rest of the operators. More generally, if the algebra L underlying the dynamics can be decomposed into a sequence of subalgebras L_K such that:

$$L = L_0 \supseteq L_1 \supseteq L_2 \supseteq \cdots, \tag{4.1}$$

then, parametrization of U as:

$$U = \exp(X_0)\exp(X_1)\exp(X_2)\cdots; \quad X_K \in L_K - L_{K-1} \tag{4.2}$$

leads to decoupling of the equations of motion for different X_K. A perturbation theory based on such decouplings has the advantage that it requires less computational effort than the Magnus expansion. First, the equations for different generators are decoupled, and consequently the original problem is broken down into several subproblems which can be solved sequentially. Second, since the equations for the generators contain finite order polynomials, the number of terms at each order of perturbation theory are fewer than in the corresponding Magnus expansion. Lastly, the resulting equations provide a global solution to the wave operator. We note in passing that algebraic methods have been used in the past to provide approximate solutions to the Schroedinger equation [4, 5]. In all these approaches the hamiltonian is not in general an element of the algebra used. Consequently, the algebraic approach does not provide the complete solution. Instead it is used to construct a convenient time-dependent coordinate system in which a basis set is defined to carry out the dynamical calculations. The most commonly used algebras in this context are the harmonic oscillator algebra and its multidimensional extensions. Because of the nonlinearities in the equations of motion for the generators, these approaches are also subject to the question of the existence of the solution depending on the order of the operators used. However, the harmonic oscillator algebra is a semisimple algebra and can be treated on the lines discussed by Wolfe and Korsch [21]. A particularly well-developed approximation method based on this approach is the time-dependent Rotated Hartree method discussed by Kucar et al. [5], who also prove the existence of a global solution when a particular ordering of the operators is chosen. However, all these approaches differ from ours because we work with the algebra that contains the hamiltonian. The work of Mukherjee [28] is closest in spirit to ours, in that he uses an exponential ansatz for the full evolution operator which he then factorises into the model space wave operator and the closed part containing the norm and the phase corrections. However, his ansatz is motivated by the requirements of the asymptotic separability. In our case the factorisation of the excitation and shift operators (open and closed operators in the terminology of Mukherjee) is a consequence of the structure present in the algebra.

While the decoupling of equations of motion provide significant formal advantages as discussed above, in an approximate calculation they could lead to norm violations because the effective hamiltonian underlying specific approximations in the full Hilbert space could become nonhermitian. Analysing the source of such nonhermiticity we found criteria by which an approximation could be tested *a priori* as to whether it would lead to norm violation or not. Alternatively, the model space wave operator could also be designed such that the full evolution operator from which it is generated is unitary.

We have studied the performances of these three methods in a model system. It appears from these studies that the perturbation theory based on similarity transformation seems to be adequate atleast for weak coupling or short time dynamics while at longer times the norm violation effects seem to be predominant. We have also tested a nonpreturbative approximation which was guaranteed to conserve the norm. The performance of this approximation was quite good. Approximations based on unitary transformation do not suffer from norm violation; however, this seems to be achieved at the cost of practically eliminating some of the states from the model space. Its performance is in general worse than an unconstrained similarity transformation based approach. These conclusions are, of course, tentative and would have to be backed up by more extensive studies on these approaches.

Acknowledgements. Financial support from DST, New Delhi is gratefully acknowledged. G. Sree Latha acknowledges a sustaining fellowship from UGC, New Delhi.

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