Brief Reports

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New criterion for convergence of exponential perturbation theory in the Schrödinger representation

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It is shown by calculations on several model systems of physical importance that exponential time-dependent perturbation theory (the Magnus expansion and similar expansions) does not converge in the Schrödinger representation for time intervals larger than the natural period of the system. This result has serious implications for the application of exponential perturbation theory in the Schrödinger representation to scattering and adiabatic turn-on problems. It is argued that this result also applies generally to more realistic systems.

The Magnus expansion,[1–4] and some of its more accessible modern counterparts,[5] have been applied to a wide range of problems in time-dependent quantum mechanics in recent years.[6] The expansions provide a prescription for calculating the time-development operator, $U(\tau)\equiv U(\tau,0)$, for a mechanical system as the exponential of an anti-Hermitian operator. In other words, given the Schrödinger equation,

$$i\hbar\frac{\partial}{\partial t}U(t)=H(t)U(t)\ , \tag{1}$$

then

$$U(\tau)=e^{A(\tau)}\ , \tag{2}$$

where A is expanded as a sum of terms $A_1+A_2+A_3+\cdots$, each of which is anti-Hermitian, and A_1 is first order in $H(t)$, A_2 is second order, and so on. These expansions are generally regarded as a particularly elegant way to do time-dependent perturbation theory, in part because they can be truncated at any order to give an approximate time-development operator which is still unitary. Also, the approximate time-development operator so obtained, with its finite-order exponent, will contain contributions from all orders. These features give the method the potential of providing many new and interesting approximations.

A more extensive utilization of exponential perturbation theory has been hampered by two problems. The first and, until recently, the most severe problem has been the extreme complexity of the expansion terms above second order. Traditional derivations are very difficult and the different derivations lead to forms for the various terms, as time-ordered multiple integrals of nested commutators, which are not at all transparent and which differ from each other. Furthermore, the different *appearing* terms are not obviously equivalent. (For example, Milfeld and Wyatt[7] have suggested that the form of the third term presented by Pechukas and Light[3] is incorrect and have given an alternate form for that term.)

The complexity problem has recently been solved by the discovery of a new and much simplified derivation, much in the spirit of a cumulant expansion.[5] The form of the terms is simple enough that they can be written by inspection to arbitrary order. In addition, the relationship of the new expansion to previous versions is straightforward. (For example, it was shown that the Milfeld and Wyatt form for the third term is equivalent to that of Pechukas and Light.)

The other problem, which is now being addressed more actively,[8,9] in part because of the new ability to generate expansion terms to arbitrary order, is the question of convergence.[10–12] Until recently very little was known about the convergence of exponential perturbation theory. Magnus's convergence criterion for his original version of the expansion is stated in terms of the eigenvalues of the exponent itself. Namely, if $ia_j(t)$ are the eigenvalues of $A(t)$ at some time t, then the expansion which calculates A at some time $t+\delta$ in the neighborhood of t converges, provided none of the $|a_j-a_k|$

equals an integral multiple of 2π. This convergence criterion, in terms of the eigenvalues of the operator A itself, does not lend itself readily to consideration of actual calculational procedures since convergence must be tested after each infinitesimal transit along the interval $[0,\tau]$.

Recent progress has been made in understanding the convergence of exponential perturbation theory by performing actual calculations, to hitherto unheard of orders, on physically interesting model systems.[13,14] The model systems treated to date in the attempt to understand the convergence properties of exponential perturbation theory have been simple models with a periodic Hamiltonian. These include several versions of the harmonically driven two-level system (including one model which has a known closed-form exponential time-development operator in the rotating-frame representation), the harmonically driven harmonic oscillator (which has a known closed-form solution in the interaction representation[3]), and an NMR multiple-pulse model. All of these models were treated in the Schrödinger representation and other representations, as appropriate. They all have a characteristic natural frequency ω_0. The conclusions based on calculations on these models were, first, that the convergence properties are highly representation dependent. Models which have closed-form exponential time-development operators in some other representation give expansions in the Schrödinger representation which diverge for the interesting ranges of the parameters. Second, the models all give the convergence criterion in the Schrödinger representation as

$$\omega > \omega_0 \ , \tag{3}$$

where ω is the driving frequency. (This, of course, severely limits the utility of the method in the Schrödinger representation, particularly for discussions of resonance or multiphoton processes.)

This latter result has recently been reaffirmed and generalized for the harmonically driven two-level system by Maricq[9] using an elegant treatment reminiscent of the Feynman-Vernon-Hellwarth formulation.[15] Maricq's criterion reduces to Eq. (3) in appropriate limits.

It is not immediately clear how a convergence criterion derived for harmonically driven systems should be extended to systems with a nonharmonic time dependence (for example, some scattering models, and models with an adiabatic turn on). Taking the reciprocal of both sides of Eq. (3) to arrive at

$$\tau < \tau_0 \ , \tag{4}$$

where $[0,\tau]$ is the time interval over which the system is to evolve and τ_0 is the natural period of the system, is suggestive but needs independent justification. In order to pursue this question we have applied the techniques of Ref. 14 to a series of models of the form

$$H = H_0 + \beta f(t) V \ , \tag{5}$$

where H_0 and V are time-independent operators in the Schrödinger representation, β is a coupling constant, and $f(t)$ is a simple, nonharmonic function of time which provides the time dependence of the model. The models included were the nonharmonically driven two-level system, with

$$H_0 = (\hbar\omega_0/2)\sigma_z \tag{6}$$

and

$$V = \sigma_x \ , \tag{7}$$

where σ_x and σ_z are the usual Pauli spin matrices; and the nonharmonically driven harmonic oscillator with

$$H_0 = \hbar\omega_0 a^\dagger a \tag{6$'$}$$

and

$$V = a + a^\dagger \ , \tag{8}$$

where $a^\dagger$ and a are the usual harmonic-oscillator raising and lowering operators. We have used the simplest possible forms for the driving function, namely, $f(t)=t$, $f(t)=t^2$, and $f(t)=t^3$, and linear combinations of these on the time interval $[0,\tau]$ to calculate the exponent in exponential perturbation theory to first order in β and infinite order in H. (It is well known that, in the Schrödinger representation, the expansion must be taken to infinite order in H to get all of the first-order contribution of β. If the first order in β terms diverges, then the expansion is not useful at higher orders of β. Also, it was shown in Ref. 14, through high-order calculations on harmonically driven systems, that the β^2 terms lead to the same convergence criteria as the β terms.)

The results are particularly simple but nevertheless enlightening. For the linear driving function, $f(t)=t$, we find that, to first order in β, the expansion terms have the following form.

For the two-level system,

$$A_{\text{even } n} = (-i\beta/\hbar)(-1)^{n/2}\tau^2(\omega_0\tau)^{n-1}B_n\sigma_y \ , \tag{9a}$$

$$A_{\text{odd } n} = 0 \quad (n>1) \ , \tag{9b}$$

and for the harmonic oscillator,

$$A_{\text{even } n} = (\beta/\hbar)(-1)^{n/2}\tau^2(\omega_0\tau)^{n-1}B_n(a - a^\dagger) \ , \tag{10a}$$

$$A_{\text{odd } n} = 0 \quad (n>1) \ . \tag{110b}$$

The B_n, the same for both models, are members of a sequence of numbers related to Bernoulli numbers the first few of which are given in Table I. They have the property that the ratio B_{n+2}/B_n converges to $-1/(2\pi)^2$ in the limit of large n. Thus the ratio A_{n+2}/a_n converges to $(\omega_0\tau/2\pi)^2$ in the limit of large n, and the expansion will diverge unless $\omega_0\tau/2\pi < 1$, or unless $\tau < \tau_0$ *independent of the value of the coupling constant* β.

The results for t^2, and t^3, and linear combinations of all three are different in detail but lead to the same convergence criterion. For example, for a t^2 driving term both even and odd terms occur as follows.

For the two-level system,

$$A_{\text{even } n} = (-i\beta/\hbar)(-1)^{n/2}\tau^3(\omega_0\tau)^{n-1}B_n\sigma_y \tag{11a}$$

$$A_{\text{odd } n} = (i\beta/\hbar)(-1)^{(n+1)/2}\tau^3 \times(\omega_0\tau)^{n-1}(-2B_{n+1})\sigma_x \quad (n>1) \tag{11b}$$

TABLE I. Numerical coefficients of the operators in the various terms of exponential perturbation theory given in Eqs. (9)–(12).

$B_2 = 1/12$
$B_4 = -1/720$
$B_6 = 1/30240$
$B_8 = -1/1209600$
$B_{10} = 1/47900160$
$B_{12} = -691/1307674368000$

and for the harmonic oscillators,

$$A_{\text{even } n} = (\beta/\hbar)(-1)^{n/2}\tau^3(\omega_0\tau)^{n-1}B_n(a-a^\dagger) , \quad (12a)$$

$$A_{\text{odd } n} = (i\beta/\hbar)(-1)^{(n+1)/2}\tau^3(\omega_0\tau)^{n-1} \times(-2B_{n+1})(a+a^\dagger), \quad (n>1) , \quad (12b)$$

where the B_n are the same as above.

Again we see that the ratios of successive coefficients of the operators in the A_n are the same as in the linear driving term case. Thus the convergence condition is still Eq. (4).

For t^3, numerical coefficients are not B_n and the coefficients of the odd and even terms do not have the same simple relationship as in the t^2 case. Nevertheless, the ratios of successive coefficients converge to the same limit $-1/(2\pi)^2$ giving the same convergence criterion. Similar procedures with a driving term consisting of a linear combination of t, t^2, and t^3 are more complicated but they lead to the same convergence criterion (the details will be reported elsewhere).

We repeat, for emphasis, that these results are independent of the magnitude of the coupling constant. This means that the problem is not a failure of perturbation theory due to an excessively strong perturbation, since the convergence condition holds for arbitrarily small β, but rather a fundamental problem with the exponential expansion itself in the Schrödinger representation. Here it is important to recall that the convergence properties of exponential perturbation theory are strongly representation dependent and that there are other representations where the exponent is known to converge for all values of the parameters of the problem. So far there is no evidence of divergence in the interaction representation, although the calculations are sufficiently difficult that the question is just beginning to be tested. The new convergence criterion makes it clear that exponential perturbation theory is unlikely to be useful, in the Schrödinger representation, for problems where it is necessary to consider times long with respect to a natural period. This unfortunately includes a number of models used in scattering and dynamics, and problems where a perturbation is turned on slowly over a long time (adiabatic approximation).

The models treated here are the simplest finite Hilbert-space model and probably the simplest model with an infinite Hilbert space. An important question is whether or not the convergence criterion derived also applies to not-so-simple systems. That question cannot be answered definitively yet, but the following argument is suggestive. Any system may be partitioned into two subspaces using projection operators P and Q, with $P+Q=1$ and $P^2=P$, $Q^2=Q$, and $PQ=QP=0$. The Hamiltonian may be divided into three parts, $H=PHP+QHQ+(PHQ+QHP)$. The projected expansions generated strictly within the P and Q defined subspaces will not interact with each other; however, the portions of the expansion generated from inclusion of the "off-diagonal" portions of H will mix the P and Q subspaces and have components on both. If we select the P subspace to be two levels which are coupled by the driving term, then the P space expansion will lead to the above convergence criterion. In order for the criterion not to be valid one would have to argue that the contributions to the expansion generated from the PHQ and QHP portions of the Hamiltonian *exactly* cancel those from the P space. Although this is conceivable, it seems exceedingly unlikely (further tests of this are in progress for more complicated models). Thus the criterion would be that the expansion converges only for times smaller than the smallest natural period of the system, as defined by direct coupling of states by the driving perturbation.

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