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Convergence of the Magnus expansion

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The convergence of the Magnus expansion in the Schrödinger representation is investigated with the aid of two exactly solvable models. A baffling observation regarding the dynamics of a spin-$\frac{1}{2}$ system driven by a superposition of a constant and a rotating magnetic field is elucidated. Perturbation theory is applied to the exponential solution to the Schrödinger equation for a time-dependent harmonic oscillator. The first terms of the perturbation expansion for the singularity of the exponent are exactly calculated. The scope and limitations of perturbation theory in obtaining the range of validity of the Magnus expansion are discussed.

I. INTRODUCTION

The Magnus expansion[1] (ME) has been widely applied to many physical problems. Among them we mention line broadening,[2] nuclear magnetic resonance[3] (NMR), multiphoton absorption,[4] energy transfer in molecular collisions,[5,6] differential equations in classical and quantum mechanics,[7] Born-Oppenheimer separation,[8] and scattering theory.[9] This formulation is appealing for at least two reasons. First, one can truncate the expansion at any order and obtain a time-evolution operator that is still unitary. Second, a representation can usually be found in which results are free from divergences on resonance. However, incorrect long-time behavior of spectroscopic properties obtained from the average Hamiltonian theory has been attributed to the failure of the ME (see Ref. 10 and those cited therein).

In order to solve the time-evolution equation (units are chosen so that $\hbar=1$)

$$\frac{d}{dt}U(t)=-iH(t)U(t)\ , \tag{1}$$

where $U(0)=I$ is the identity operator, Magnus[1] proposed to write $U(t)$ as

$$U(t)=e^{-iA(t)}\ , \tag{2}$$

where $A(0)=0$. If H is Hermitian, then A is also Hermitian and U is unitary.

An approximation to A is obtained as follows. First, ξH, ξ being a dummy expansion parameter, is substituted for H in Eq. (1). Second, $A(\xi,t)$ is expanded in a power series of ξ,

$$A(\xi,t)=A_1(t)\xi+A_2(t)\xi^2+\cdots\ . \tag{3}$$

Finally, ξ is set equal to unity at the end of the calculation. Several terms of this series have been obtained[11-14] and a recursive generation of higher-order terms has recently been proposed.[15]

In order to obtain A, the ME (3) has to converge for $\xi=1$. It has been shown that there is always a neighborhood of $t=0$ in which the exponential form (2) exists and the ME converges.[1] Equation (2) is no longer valid when the difference between two eigenvalues of A is a multiple of 2π.[1] If t_0 is the smallest time value for which this condition occurs, then the ME diverges for all $t\geq t_0$. The above-mentioned convergence criterion is not of much practical utility because it is based on the eigenvalues of the unknown operator A. Since a general statement that only takes into account the properties of H has not been given, except for some particular cases,[16] many authors have resorted to simple models to obtain information about the range of validity of the ME. Among them there are spin systems driven by magnetic fields and other forces,[16-20] harmonically and nonharmonically driven harmonic oscillators,[18,20,21] and damped harmonic oscillators.[22,23]

Examples of the form $H=H_0+\beta V(t)$, where H_0 is the Hamiltonian of the isolated system and V represents the time-dependent driving forces, are amenable to perturbation theory treatment, β being the perturbation parameter.[18-20] Perturbation theory appears to be an acceptable way of estimating the range of validity of the ME and has led to the striking result that the ME is divergent when the driving frequency is smaller than or equal to the frequency of the isolated system.[18-20] For this reason the ME would be useless in studying resonances. This conclusion has also been drawn for the general case by Maricq[24] by means of a simple, approximate argument which has recently been derived in a more rigorous way.[25] Theoretical results suggest that perturbation theory[18-20] overestimates the range of validity of the ME; in other words, $t_0(TP)>t_0$.[25] This conclusion is confirmed in Sec. II.

The exact treatment of the linearly driven harmonic oscillator $[H_0=\omega_0(p^2+q^2)/2,\ V(t)=f(t)q]$ (Ref. 21) reveals the actual cause for divergence of the ME predicted by perturbation theory.[18,20] For instance, when $f(t)=\cos(\omega t)$, it follows from the results of Ref. 21 that the ME converges provided that $t<2\pi/\omega_0$ and $\omega>\omega_0$ are simultaneously obeyed. (A slightly more general model is discussed in Ref. 25.) However, the linearly driven harmonic oscillator poses a trivial example in the sense that the eigenvalues of A are independent of β and the level spacing can be exactly obtained from the first term of the ME.[21,25] Besides, since first-order perturbation theory yields the operator A exactly,[21] this model is not the most suitable one to test such an approach.

The purpose of this paper is to investigate other exactly solvable models in order to obtain a better understanding of the limitations of the ME. Two examples are considered in Sec. II, namely, a spin-$\frac{1}{2}$ system driven by a superposition of a constant and a rotating magnetic field, and a harmonic oscillator with a time-dependent quadratic perturbation. The former is used to investigate what may be called Salzman's paradox,[18] and the latter proves to be valuable in checking perturbation theory results. Further comments and conclusions are found in Sec. III.

II. EXAMPLES

It is considered here the case that H can be written

$$H=\sum_{j=1}^{n} f_j(t)X_j \ , \tag{4}$$

where $\{X_1,X_2,\ldots,X_n\}$ are time-independent operators spanning an n-dimensional Lie algebra[26,27] and $f_j(t)$, $j=1,2,\ldots,n$, are continuous functions of time. Under such conditions A is known to be of the form[1,26,27]

$$A=\sum_{j=1}^{n} a_j(t)X_j \ , \tag{5}$$

where $a_j(0)=0$, $j=1,2,\ldots,n$. There are remarkable papers containing summaries of the most useful properties of finite-dimensional Lie algebras[26,27] and for this reason we do not discuss them here. In spite of the fact that the global validity of the exponential form (2) for several finite-dimensional Lie algebras has been extensively discussed,[28] present results are believed to be a valuable contribution. Besides, a matrix representation has recently been proposed[25] which might facilitate the treatment of the problem.

The first example is given by a spin-$\frac{1}{2}$ system driven by a superposition of a constant and a rotating magnetic field

$$H=\omega_0 s_3+\beta[s_1\cos(\omega t)+s_2\sin(\omega t)] \ , \tag{6}$$

where

$$s_1=\frac{1}{2}\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix} , \quad s_2=\frac{1}{2}\begin{bmatrix}0 & -i\\ i & 0\end{bmatrix} , \quad s_3=\frac{1}{2}\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix} \tag{7}$$

are proportional to the well-known Pauli spin matrices.

Since H is a periodic function of time with period $\tau=2\pi/\omega$, it follows from the Floquet theorem that $U(t)$ can be written as $P(t)e^{-itS}$, where S is time independent and $P(t+\tau)=P(t)$.[10] Therefore $P(\tau)=P(0)=I$ is the identity operator and

$$U(\tau)=e^{-i\tau S} \ . \tag{8}$$

At times $t=N\tau$ the propagator is exactly given by $U(N\tau)=U(\tau)^N$ and the system, when observed stroboscopically, appears to evolve under a time-independent Hamiltonian. This is the basis of the average Hamiltonian theory which has become a powerful method of analysis of high-resolution NMR.[10] The ME has been widely used in obtaining approximations to S (see Ref. 10 and those cited therein).

On using well-known methods of the linear algebra,[29] it is not difficult to verify that for any operator of the form (5) with $X_j=s_j$, $j=1,2,3$, one has

$$e^{-iA}=\begin{bmatrix}u & v\\ -v^* & u^*\end{bmatrix} , \tag{9}$$

where

$$u=\cos a - ia_3(\sin a)/(2a) \ , \tag{10a}$$

$$v=-(a_2+ia_1)(\sin a)/(2a) \ , \tag{10b}$$

$$a=(a_1^2+a_2^2+a_3^2)^{1/2}/2 \ , \tag{10c}$$

and the asterisk stands for complex conjugation.

The solution of the Schrödinger equation (1) with the Hamiltonian (6) can be written

$$U(t)=\exp(-i\omega t s_3)\exp[-i\beta t s_1-i(\omega_0-\omega)t s_3] \ . \tag{11}$$

When $t=\tau=2\pi/\omega$ it becomes[19]

$$\begin{aligned}U(\tau)&=-\exp[-i\beta\tau s_1-i(\omega_0-\omega)\tau s_3]\\ &=\exp[-i\beta\tau s_1-i(\omega_0-\omega)\tau s_3+i\pi I] \ ,\end{aligned} \tag{12}$$

where I is the 2×2 identity matrix. As noticed by Salzman[19] there is no apparent explanation for the perturbation-theory results that predict the divergence of the ME because the exponent in Eq. (12) has no singularity. The reason for this inconsistency is that while the exponent in Eq. (12) is not an element of the algebra spanned by the Pauli matrices due to the occurrence of the identity matrix, every term of the ME is known to belong to that algebra.[1] Therefore, in order to understand the cause for the divergence, one has to look for an operator A belonging to the algebra.

It follows from Eqs. (9)–(11) that

$$\begin{aligned}\cos a=&\cos(\omega t/2)\cos(\Delta t/2)\\ &-\epsilon\sin(\omega t/2)\sin(\Delta t/2)/\Delta \ ,\end{aligned} \tag{13a}$$

$$a_1(\sin a)/a=2\beta\cos(\omega t/2)\sin(\Delta t/2)/\Delta \ , \tag{13b}$$

$$a_2(\sin a)/a=-2\beta\sin(\omega t/2)\sin(\Delta t/2)/\Delta \ , \tag{13c}$$

$$\begin{aligned}a_3(\sin a)/a=&2[\sin(\omega t/2)\cos(\Delta t/2)\\ &+\epsilon\cos(\omega t/2)\sin(\Delta t/2)]/\Delta \ ,\end{aligned} \tag{13d}$$

where $\epsilon=\omega_0-\omega$ and $\Delta=(\epsilon^2+\beta^2)^{1/2}$. A unitary exponential propagator $U(t)$ only exists for those t values that admit real solutions a_1, a_2, and a_3.

When $t=\tau$, Eqs. (13) yield the desired exponent $A(\tau)$ that belongs to the algebra

$$A(\tau)=2\epsilon a(\tau)(\beta s_1+\epsilon s_3)/(|\epsilon|\Delta) , \quad (14)$$

where $a(\tau)=\pi+\epsilon\Delta\tau/(2|\epsilon|)$. Introducing the expansion parameter ξ is equivalent to substituting $\xi\omega_0$ and $\xi\beta$ for ω_0 and β, respectively. Therefore a pair of complex-conjugate singularities are obtained from the roots of $\Delta(\xi)=[(\xi\omega_0-\omega)^2+\xi^2\beta^2]^{1/2}=0$, namely, $\xi_\pm=\omega(\omega_0\pm i\beta)/(\omega_0^2+\beta^2)$. Consequently, the ME converges when $|\xi_\pm|>1$, which can be rewritten

$$\omega>(\omega_0^2+\beta^2)^{1/2} . \quad (15)$$

This result has been obtained by Fel'dman[17] in a different way and shows that the ME does not hold near resonance.

The fist two perturbation terms given in Ref. 18 are obtained by expanding Eq. (14) in a Taylor series around $\beta=0$. Every term of this series is singular at $\omega=\omega_0$, which is a mere consequence of the fact that the perturbation expansion converges for $|\beta|<|\omega_0-\omega|$. Besides, it has been inferred from perturbation theory that the ME converges provided $\omega>\omega_0$ (Ref. 18), which is in fairly good agreement with (15) when $\beta\ll\omega_0$. Clearly these results completely solve Salzman's paradox.[18] In addition to this, it is found that the exponent $A(\tau)$ is not singular at $\omega=\omega_0$ and exhibits a factor $1/\Delta$ that reminds one of the Lorentzian factor conjectured by Salzman[19] in the case of the harmonically driven two-level system $[V=2(\cos(\omega t)s_1]$.

It is worth noticing that although $A(\tau)$ exists for all real values of ω, ω_0, and β, the ME only converges when the condition (15) is satisfied because of the complex poles of $1/\Delta(\xi)$. This fact is closely related to the elegant and more general argument developed by Maricq.[16] According to the formula derived in Refs. 16 and 25 for an arbitrary time-dependent two-level system, the ME for the example above converges for all $t<2\pi/(\omega_0^2+\beta^2)^{1/2}$, which reduces to Eq. (15) when $t=\tau$. Although there is loss of generality in the treatment of particular examples, they allow one to investigate the cause for the divergence of the ME in more detail. For instance, when $\omega=\omega_0$, Eq. (13a) becomes $\cos a=\cos(\omega_0 t/2)\cos(|\beta|t/2)$ and a cannot, in general, equal π except for particular values of the ratio $|\beta|/\omega_0$. However, even when this ratio allows A to exist for all t values, the ME is not globally valid as shown before.

According to Eq. (11) the ME in the interaction picture converges for all t values when $\omega=\omega_0$ since the series reduces to just the first term. Unfortunately, in general, the Hamiltonian in the interaction picture is no longer periodic and the stroboscopic evolution mentioned before does not apply. For this reason, alternative transformations have been proposed that preserve the period of the Hamiltonian in the Schrödinger representation.[16,24]

The next example is given by the following harmonic oscillator with a quadratic time-dependent perturbation:

$$H=\omega_0(a^\dagger a+\tfrac{1}{2})+\tfrac{1}{2}\beta[\exp(2i\omega_0 t)a^2+\exp(-2i\omega_0 t)(a^\dagger)^2] , \quad (16)$$

where $a^\dagger$ and a are the creation and annihilation operators, respectively, satisfying $[a,a^\dagger]=I$. The operators $X_1=a^\dagger a+\frac{1}{2}$, $X_2=\frac{1}{2}a^2$, and $X_3=\frac{1}{2}(a^\dagger)^2$ span a three-dimensional Lie algebra and can be represented by two-dimensional matrices that obey the same commutation relations. These matrices are easily obtained from the general method discussed in Ref. 25 and are given by

$$X_1\to M_1=\begin{bmatrix}-1 & 0\\ 0 & 1\end{bmatrix} ,$$

$$X_2\to M_2=\begin{bmatrix}0 & 1\\ 0 & 0\end{bmatrix} , \quad (17)$$

$$X_3\to M_3=\begin{bmatrix}0 & 0\\ -1 & 0\end{bmatrix} .$$

The time-evolution operator can be exactly written

$$U(t)=\exp(-i\omega_0 tX_1)\exp[-i\beta t(X_2+X_3)] . \quad (18)$$

On arguing as in the previous example one immediately finds

$$\cos\lambda=\cos(\omega_0 t)\cosh(\beta t) , \quad (19a)$$

$$a_1(\sin\lambda)/\lambda=\sin(\omega_0 t)\cosh(\beta t) , \quad (19b)$$

$$a_2(\sin\lambda)/\lambda=[i\cos(\omega_0 t)-\sin(\omega_0 t)]\sinh(\beta t) , \quad (19c)$$

$$a_3=a_2^* , \quad (19d)$$

where $\lambda=(a_1^2-|a_2|^2)^{1/2}$. These equations hold for all t values satisfying $-1\le f(t)\le 1$, where $f(t)=\cos(\omega_0 t)\cosh(\beta t)$. When $f(t)>1$, $\cosh\Omega$ and $(\sinh\Omega)/\Omega$ have to be substituted for $\cos\lambda$ and $(\sin\lambda)/\lambda$, respectively. Because of the quadratic perturbation both $A(t)$ and its eigenvalues depend on β in such a way that they cannot be exactly obtained from a finite number of terms of the perturbation or the Magnus expansion as in the trivial case of the linearly driven oscillator.[15,21]

It is clear that $A(t)$ does not exist when $f(t)\le -1$ because Eq. (19a) no longer holds. Since $f(\pi/(2\omega_0))=0$ and $f(\pi/\omega_0)=-\cosh(\beta\pi/\omega_0)<-1$, then there exists t_0, $\pi/(2\omega_0)<t_0<\pi/\omega_0$, so that

$$\cos(\omega_0 t_0)\cosh(\beta t_0)=-1 . \quad (20)$$

When $t\to t_0$, $\lambda\to\pi$, and $|a_2|=|a_3|\to\infty$ from which it is concluded that the ME diverges for all $t>t_0$. This result is not surprising. According to the argument in Ref. 25, if the form of H is properly taken into account, the level spacing to be considered in the Magnus condition[1] is in the present case equal to 2λ, which leads to the conclusion above. However, as pointed out in Ref. 25 the structure of H is usually overlooked.

The critical time value t_0 can be obtained in terms of β from the smallest root of Eq. (20). In order to check the perturbation theory predictions, this root is expanded in a power series of β. The first terms are easily found to be

$$t_0=\pi\omega_0^{-1}[1-\gamma+\gamma^2+(\pi^2/6-1)\gamma^3+\cdots] , \quad \gamma=|\beta/\omega_0| . \quad (21)$$

As conjectured in Ref. 25, the first term in this series, which is the only usually obtained,[18–20] overestimates the critical time value; i.e., $\pi/\omega_0>t_0$.

On the other hand, when $\beta\to\infty$, then $\omega_0 t_0\to\pi/2$ and t_0 can also be written as a series in powers of $1/\beta$. In this case we have

$$t_0=(\pi/(2\omega_0))[1+16\omega_0^2/(\pi^3\beta^2)+\cdots] . \quad (22)$$

A more general driven harmonic oscillator is given by

$$H=\omega_0(a^\dagger a+\tfrac{1}{2})+\tfrac{1}{2}\beta[\exp(2i\omega t)a^2+\exp(-2i\omega t)(a^\dagger)^2] , \quad (23)$$

which can be treated exactly as the previous examples. The main equations for this model can be obtained from those for the spin system by substituting 2ω, $2\omega_0$, and $2i\beta$ for ω, ω_0, and β, respectively. The factor $1/\Delta$ in $A(\tau)$, $\tau=\pi/\omega$, is no longer a Lorentzian because $\Delta=(\epsilon^2-\beta^2)^{1/2}$. For this reason it is found that the ME converges if

$$\omega>\omega_0+|\beta| . \quad (24)$$

Although this inequality is slightly different from (15), it also shows that the ME is not a useful tool in treating driving systems near resonance.

Finally, it is worth mentioning that the ME in the interaction picture converges for all t values because, as shown in Eq. (18), such a series reduces to just the first term.

III. COMMENTS AND CONCLUSIONS

Although the models considered in this paper are exactly solvable, they provide a good deal of information about the convergence properties of the ME. Particularly revealing is the conclusion that an exponential solution to the Schrödinger equation may in some cases exist that cannot be obtained by means of the ME [see Eq. (12) and the discussion below]. Needless to say, this can only occur outside the convergence interval of the ME. In addition to this, the examples in Sec. II reveal the scope and limitations of perturbation theory which yields a reasonable approximation to the range of validity of the ME provided the perturbation is weak enough.

As a by-product they also confirm that the Baker-Hausdorff theorem is not generally valid for Lie algebras that are not free.[28] As a result, whereas the time-evolution operator written as a product of two exponential operators exists for all t values, the single exponential is not globally valid.

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[1] W. Magnus, Commun. Pure Appl. Math. **7**, 649 (1954).
[2] W. A. Cady, J. Chem. Phys. **60**, 3318 (1974).
[3] J. S. Waugh, J. Magn. Reson. **50**, 30 (1982).
[4] I. Schek, J. Jortner, and M. L. Sage, Chem. Phys. **59**, 11 (1981).
[5] S.-K. Chen, J. C. Light, and J.-L. Lin, J. Chem. Phys. **49**, 86 (1968).
[6] J. D. Kelley, J. Chem. Phys. **56**, 6108 (1972).
[7] R. A. Marcus, J. Chem. Phys. **52**, 4803 (1970).
[8] J. C. Y. Chen and J. D. Kelley, J. Chem. Phys. **43**, 1429 (1965).
[9] R. J. Cross, J. Chem. Phys. **79**, 1272 (1983).
[10] M. M. Maricq, Phys. Rev. B **25**, 6622 (1982).
[11] P. Pechukas and J. C. Light, J. Chem. Phys. **44**, 3897 (1965).
[12] R. M. Wilcox, J. Math. Phys. **8**, 962 (1967).
[13] I. Bialynicki-Birula, B. Mielnik, and J. Plebański, Ann. Phys. (N.Y.) **51**, 187 (1969).
[14] K. F. Milfeld and R. E. Wyatt, Phys. Rev. A **27**, 72 (1983).
[15] S. Klarsfeld and J. A. Oteo, Phys. Rev. A **39**, 3270 (1989).
[16] M. M. Maricq, J. Chem. Phys. **86**, 5647 (1987).
[17] E. B. Fel'dman, Phys. Lett. **104A**, 479 (1984).
[18] W. R. Salzman, J. Chem. Phys. **85**, 4605 (1986).
[19] W. R. Salzman, Chem. Phys. Lett. **124**, 531 (1986).
[20] W. R. Salzman, Phys. Rev. A **36**, 5074 (1987).
[21] F. M. Fernández, J. Chem. Phys. **88**, 490 (1988).
[22] F. M. Fernández, J. Math. Phys. **28**, 2908 (1987).
[23] F. M. Fernández, J. Echave, and E. A. Castro, Chem. Phys. **117**, 101 (1987).
[24] M. M. Maricq, J. Chem. Phys. **85**, 5167 (1986).
[25] F. M. Fernández, J. Echave, and E. A. Castro, J. Math. Phys. (to be published).
[26] E. H. Wichmann, J. Math. Phys. **2**, 876 (1961).
[27] J. Wei and E. Norman, J. Math. Phys. **4**, 575 (1963).
[28] J. Wei, J. Math. Phys. **4**, 1337 (1963).
[29] F. R. Gantmacher, *The Theory of Matrices* (Chelsea, New York, 1959), Vol. I, Chap. V.