ARTICLES

Driven harmonic oscillators in the adiabatic Magnus approximation

S. Klarsfeld
Division de Physique Théorique, Institut de Physique Nucléaire, 91406 Orsay CEDEX, France

J. A. Oteo
Departament de Física Teòrica and Instituto de Física Corpuscular, Universitat de València, 46100 Burjassot, Spain

(Received 10 July 1992)

The time evolution of driven harmonic oscillators is determined by applying the Magnus expansion in the basis set of instantaneous eigenstates of the total Hamiltonian. It is shown that the first-order approximation already provides transition probabilities close to the exact values even in the intermediate regime.

PACS number(s): 03.65.−w, 34.50.Ez

I. INTRODUCTION

Recently we proposed a simple approximate nonperturbative approach for treating time-dependent Hamiltonians in the intermediate regime, i.e., situations that are far from both the sudden and the adiabatic limits [1]. The method is based on applying the Magnus expansion [2] in a time-dependent basis which we called *adiabatic* because it consists of eigenvectors of the instantaneous Hamiltonian which the system would follow in the limit of infinitely slow motion. This basis set is currently used in adiabatic perturbation theory [3]. By analogy we shall refer to the present approach as the *adiabatic Magnus expansion* although its validity extends far into the intermediate regime.

Comparison with exact calculations for various two-state problems [1, 4] definitely shows that the method is very effective indeed. While the generalization to systems with more than two levels is straightforward it was not clear how the accuracy of the approximation will be affected by augmenting their number.

In order to examine this point we consider here a class of driven harmonic oscillators, thus passing from two to an infinite number of levels. Such systems have been extensively investigated in the literature because they serve as semiclassical models for vibrational excitation in molecular collisions [5, 6]. To be more specific, we study the collinear collision of a point atom A with a diatomic molecule BC. The latter is represented by a quantum-mechanical harmonic oscillator while the relative motion of the two systems is treated classically. The corresponding Hamiltonian reads

$$H(t) = H_0 + V_1(t) + V_2(t)\,, \tag{1}$$

where

$$H_0 = p^2/(2m) + m\omega_0^2 q^2/2\,, \tag{2}$$

and

$$V_1(t) = f(t)q\,, \quad V_2(t) = g(t)q^2\,. \tag{3}$$

Here p, q are the momentum and position operators, $m = m_B m_C/(m_B + m_C)$ and ω_0 are the mass and frequency of the harmonic oscillator, and f, g are scalar functions to be defined later. For the time being we assume only that both f and g vanish for $t \to \pm\infty$.

In Sec. II we discuss the simpler problem in which the quadratic term $V_2(t)$ is missing. This offers a good opportunity to compare the usual Magnus approach with the adiabatic variant. The latter is then extended in Sec. III to the complete Hamiltonian defined above and an analytic expression is derived for the corresponding time-evolution operator in the first-order Magnus approximation. Explicit formulas are also given for several transition probabilities. Finally, Sec. IV presents a comparison of numerical results obtained from these formulas with exact values computed by a recent operator method. This allows us to emphasize the remarkable qualities of the adiabatic Magnus approximation as a reliable computational tool in molecular collision theory.

II. LINEARLY DRIVEN HARMONIC OSCILLATOR

Before proceeding with the full Hamiltonian in Eq. (1) it is instructive indeed to assume $g = 0$. This case corresponds to the well-known problem of an ordinary linearly driven harmonic oscillator which is exactly solvable by various methods, including the Magnus expansion [7]. The latter has been applied in the Dirac interaction picture, i.e., the Hilbert space was referred to the set $\{\exp(-iE_n t/\hbar)|n\rangle\}$, where $|n\rangle$ and E_n are the eigenstates and eigenenergies of the unperturbed Hamiltonian,

$$H_0|n\rangle = E_n|n\rangle\,, \quad E_n = \hbar\omega_0(n + \tfrac{1}{2})\,. \tag{4}$$

In terms of raising and lowering operators, the Hamiltonian then reads

$$H^{(I)}(t) = \sigma_0 f(t)(e^{i\omega_0 t}a^\dagger + e^{-i\omega_0 t}a), \tag{5}$$

where σ_0 denotes the rms radius of the molecule in the ground state, i.e.,

$$\sigma_0^2 = \langle 0|q^2|0\rangle = \hbar/(2m\omega_0)\,. \tag{6}$$

Clearly the commutator $[H^{(I)}(t_1), H^{(I)}(t_2)]$ is a multiple of unity and higher-order commutators vanish. The Magnus expansion therefore reduces to just two terms and the time-evolution operator is given exactly by

$$U^{(I)}(t,-\infty) = \exp[\Omega_1(t) + \Omega_2(t)]\,, \tag{7}$$

where

$$\begin{aligned}\Omega_1(t) &= -(i/\hbar)\int_{-\infty}^{t} dt_1\; H_1^{(I)} \\ &= -i\xi(t)a^\dagger - i\xi^*(t)a\,,\end{aligned} \tag{8}$$

and

$$\begin{aligned}\Omega_2(t) &= -(1/2\hbar^2)\int_{-\infty}^{t} dt_2 \int_{-\infty}^{t_2} dt_1 [H_2^{(I)}, H_1^{(I)}] \\ &= -i\eta(t)\,.\end{aligned} \tag{9}$$

Here we used the shorthand notation $H(t_k) = H_k$ and the functions $\xi(t), \eta(t)$ are defined by

$$\xi = (\sigma_0/\hbar)\int_{-\infty}^{t} dt_1 f(t_1) e^{i\omega_0 t_1}\,, \tag{10}$$

$$\begin{aligned}\eta = (\sigma_0^2/2\hbar^2)\int_{-\infty}^{t} dt_2 f(t_2) \\ \times \int_{-\infty}^{t_2} dt_1 f(t_1)\sin\omega_0(t_1 - t_2)\,.\end{aligned} \tag{11}$$

From the above equations, assuming that the oscillator was initially in its ground state, we get in particular the familiar Poisson distribution for the transition probabilities,

$$\begin{aligned}P_{0\to n}(t) &= |\langle n|U^{(I)}(t,-\infty)|0\rangle|^2 \\ &= |\xi|^{2n} e^{-|\xi|^2}/n!\,.\end{aligned} \tag{12}$$

We now solve the same problem in the adiabatic interaction picture. To do this we rewrite the total Hamiltonian $H(t) = H_0 + V_1(t)$ as

$$H(t) = \frac{p^2}{2m} + \frac{m\omega_0^2}{2}[q - q_0(t)]^2 - \frac{m\omega_0^2}{2}q_0^2(t)\,, \tag{13}$$

where

$$q_0(t) = -f(t)/m\omega_0^2 \tag{14}$$

represents the instantaneous equilibrium position of the oscillator. The *adiabatic* basis set is obtained by solving the Schrödinger equation

$$H(t)|n(t)\rangle = E_n(t)|n(t)\rangle\,. \tag{15}$$

In the q representation this amounts simply to replacing q by $q - q_0(t)$ in the usual harmonic-oscillator wave functions and the corresponding eigenenergies are given by

$$E_n(t) = \hbar\omega_0(n + \tfrac{1}{2}) - m\omega_0^2 q_0^2(t)/2\,. \tag{16}$$

Alternatively we shall refer the system to the fixed set of unperturbed states $\{|n(-\infty)\rangle = |n\rangle\}$. This is related to the adiabatic set by a unitary transformation $G(t)$ such that $|n(t)\rangle = G(t)|n\rangle$. Hence

$$\langle k(t)|\dot{n}(t)\rangle = \langle k|G^\dagger\dot{G}|n\rangle\,. \tag{17}$$

Since the states $|n(t)\rangle$ depend on time only through $q_0(t)$ the same scalar product can be expressed as

$$\langle k(t)|\dot{n}(t)\rangle = -(i/\hbar)\,\dot{q}_0(t)\,\langle k(t)|p|n(t)\rangle \tag{18}$$

and it is easily seen that the matrix element in the right-hand side is actually time independent. From this we infer

$$G^\dagger\dot{G} = -(i/\hbar)\,\dot{q}_0(t)\,p\,. \tag{19}$$

According to Ref. [1] the Hamiltonian in the adiabatic picture reads

$$H_G = E - i\hbar G^\dagger\dot{G}\,, \tag{20}$$

where $E = E(t)$ is the diagonal matrix with elements given in Eq. (16). At this point it will be useful to introduce again raising and lowering operators. Explicitly, recalling Eq.(14), one then obtains

$$H_G = \hbar\omega_0(a^\dagger a + \tfrac{1}{2}) - f^2/(2m\omega_0^2) + i(\sigma_0/\omega_0)\dot{f}(a^\dagger - a). \tag{21}$$

In H_G the diagonal and off-diagonal parts are clearly separated, and the passage to the adiabatic interaction picture proceeds very much as in the Dirac case. This results in a new Hamiltonian

$$H_G^{(I)}(t) = i(\sigma_0/\omega_0)\dot{f}(t)(e^{i\omega_0 t}a^\dagger - e^{-i\omega_0 t}a), \tag{22}$$

which represents the counterpart of Eq. (5) and eventually leads to a Poisson law similar to Eq. (12),

$$\begin{aligned}P_{0\to n}(t) &= |\langle n(t)|U_G^{(I)}(t,-\infty)|0\rangle|^2 \\ &= |\bar{\xi}|^{2n} e^{-|\bar{\xi}|^2}/n!\,,\end{aligned} \tag{23}$$

with

$$\bar{\xi} = i(\sigma_0/\hbar\omega_0)\int_{-\infty}^{t} dt_1 \dot{f}(t_1) e^{i\omega_0 t_1}\,. \tag{24}$$

The two transition probabilities are not equal for finite t because the final state is not quite the same, but they become so in the limit $t \to \infty$ as expected.

III. TRANSLATION-VIBRATION ENERGY TRANSFER IN THE GAZDY-MICHA MODEL

Let us now go back to the complete Hamiltonian in Eq. (1) thus assuming $g \neq 0$. As shown in Ref. [5] inclusion of the quadratic term is indeed essential for achieving approximate equivalence with the two-dimensional scattering model of Secrest and Johnson [8]. We first note that $H(t)$ may be rewritten as

$$H(t) = \frac{p^2}{2m} + \frac{m\omega^2(t)}{2}[q - q_0(t)]^2 - \frac{m\omega^2(t)}{2}q_0^2(t)\,, \tag{25}$$

where

$$\omega^2(t) = \omega_0^2 + 2g(t)/m \tag{26}$$

and

$$q_0(t) = -f(t)/m\omega^2(t)\,. \tag{27}$$

The main change with respect to the previous case is that here both the frequency and the equilibrium position depend on time (displaced parametric harmonic oscillator). In spite of this difference the instantaneous Hamiltonian is still easily diagonalized and the associated eigenenergies are found to be

$$E_n(t) = \hbar\omega(t)(n+\tfrac{1}{2}) - m\omega^2(t)q_0^2(t)/2\,. \tag{28}$$

Using explicit analytical expressions for the normalized harmonic-oscillator wave functions one can show that

$$\langle k(t)|\dot{n}(t)\rangle = -(i/\hbar)\langle k(t)|\dot{q}_0 p - (\dot{\omega}/4\omega)(pq+qp)|n(t)\rangle\,. \tag{29}$$

The time dependence of the matrix elements involved is a little bit more complicated than for the linearly driven harmonic oscillator. Eventually the analysis results in the following expression for the transformed Hamiltonian:

$$\begin{aligned} H_G = {} & \hbar\omega(a^\dagger a + \tfrac{1}{2}) - f^2/(2m\omega^2) \\ & + i(\sigma/\omega)(\dot{f} - 2f\dot{\omega}/\omega)(a^\dagger - a) \\ & + i\hbar(\dot{\omega}/4\omega)(a^{\dagger 2} - a^2) \end{aligned} \tag{30}$$

with $\sigma = \sigma(t)$ defined by

$$\sigma^2 = \hbar/[2m\omega(t)]\,. \tag{31}$$

Obviously, the above Hamiltonian reduces to that of Eq. (21) when $g = 0$.

The next step is to introduce the adiabatic interaction picture via a unitary transformation generated by the diagonal part of H_G [1]. As a result we get the new Hamiltonian

$$\begin{aligned} H_G^{(I)}(t) = {} & i(\sigma/\omega)(\dot{f} - 2f\dot{\omega}/\omega)(e^{i\phi}a^\dagger - e^{-i\phi}a) \\ & + i\hbar(\dot{\omega}/4\omega)(e^{2i\phi}a^{\dagger 2} - e^{-2i\phi}a^2), \end{aligned} \tag{32}$$

where

$$\phi = \phi(t) = \int_0^t dt'\,\omega(t')\,. \tag{33}$$

Because of the presence of quadratic terms the Magnus expansion in this case is infinite. Retaining only the first order yields the approximate time-evolution operator

$$U_G^{(I)}(t,-\infty) \simeq \exp\left[-(i/\hbar)\int_{-\infty}^t dt'\,H_G^{(I)}(t')\right]\,, \tag{34}$$

from which we shall derive transition probabilities for $t \to \infty$ to be compared with the exact results of Gazdy and Micha [5]. We henceforth specialize by using their expressions for f, g, viz.,

$$f(t) = (\gamma/a)E/\cosh^2(t/T)\,, \quad g(t) = (\gamma/2a)f(t)\,. \tag{35}$$

Here $\gamma = m_C/(m_B+m_C)$, a is a range parameter, and E is the relative kinetic energy of the system. In order to ensure microscopic reversibility for the inelastic transitions in the semiclassical approach one defines the latter by the average value

$$E = (\mu/2)[(v_i+v_f)/2]^2\,, \tag{36}$$

where $\mu = m_A(m_B+m_C)/(m_A+m_B+m_C)$ is the global reduced mass and v_i, v_f are the initial and final relative velocities, respectively. Finally $T = (2\mu/E)^{1/2}a$ is a measure of the collision time.

In view of the similar functional form of f and g one has $\dot{f} - 2f\dot{\omega}/\omega = (\omega_0/\omega)^2\dot{f}$. Taking into account the parity of each function involved we arrive at

$$U_G^{(I)}(\infty,-\infty) \simeq \exp[-i\xi_2(a^{\dagger 2}+a^2) - i\xi_1(a^\dagger + a)] \tag{37}$$

where

$$\xi_1 = (2\omega_0^2/\hbar)\int_0^\infty dt\,(\sigma/\omega^3)\dot{f}\sin\phi\,, \tag{38}$$

$$\xi_2 = \tfrac{1}{2}\int_0^\infty dt\,(\dot{\omega}/\omega)\sin 2\phi\,. \tag{39}$$

Moreover, the phase ϕ can now be calculated analytically [cf. Eq. (18) in Ref. [4]]. The result reads

$$\phi = (\omega_0 T/2)\left\{\ln[(1+\rho)/(1-\rho)] + 2\lambda\arctan\lambda\rho\right\}\,, \tag{40}$$

where $(s = t/T)$

$$\begin{aligned} \rho &= \sinh s/(\lambda^2+\cosh^2 s)^{1/2}\,, \\ \lambda &= (\gamma/\omega_0 a)(E/m)^{1/2}. \end{aligned} \tag{41}$$

Alternatively the time-evolution operator may be written in a simpler form,

$$U_G^{(I)}(\infty,-\infty) \simeq \exp[-i\xi_2(b^{\dagger 2}+b^2) + i\xi_0]\,, \tag{42}$$

where $b = a+\beta$, with $\beta = \xi_1/(2\xi_2)$ and $\xi_0 = \beta\xi_1$. Notice that the operators $b, b^\dagger$ obey the same commutation relation as $a, a^\dagger$. For the exponential operator in Eq. (42) we use the disentangling formula given in Ref. [9] to obtain

$$\begin{aligned} \exp[-i\xi_2(b^{\dagger 2}+b^2)] = {} & (1-\sigma_2)^{1/2}\exp(-i\tau_2 b^{\dagger 2}) \\ & \times \sum_n \frac{(-\sigma_2)^n}{n!} b^{\dagger n} b^n \exp(-i\tau_2 b^2), \end{aligned} \tag{43}$$

where $\sigma_2 = 1 - 1/\cosh 2\xi_2$, $\tau_2 = (\tanh 2\xi_2)/2$. A little more algebra allows us to derive closed-form expressions for the transition probabilities generated by Eq. (42). For instance,

$$P_{0\to 0} = (1-\sigma_2)\exp(-2\beta^2\sigma_2)\,, \tag{44}$$

$$P_{0\to 1} = 2\beta^2\sigma_2(1-\sigma_2)\exp(-2\beta^2\sigma_2)\,, \tag{45}$$

$$P_{0\to 2} = 2(\tau_2^2 - 4\beta^2\sigma_2\tau_2^2 + \beta^4\sigma_2^2)(1-\sigma_2)\exp(-2\beta^2\sigma_2)\,, \tag{46}$$

$$\begin{aligned} P_{0\to 3} = {} & 4\beta^2\sigma_2(3\tau_2^2 - 4\beta^2\sigma_2\tau_2^2 + \beta^4\sigma_2^2/3)(1-\sigma_2) \\ & \times \exp(-2\beta^2\sigma_2)\,. \end{aligned} \tag{47}$$

When the quadratic term V_2 in Eq. (1) is missing, $\omega = \omega_0$

is a constant, therefore $\phi = \omega_0 t$ and ξ_1 reduces to the integral called $\bar{\xi}$ in Eq. (24) while $\xi_2 = 0$. Thus from Eq. (37) it is apparent that the transition probabilities $P_{0\to n}$ must reduce to the Poisson form in Eq. (23). This can be checked also in the explicit formulas above by noticing that for $\xi_2 \to 0$ both σ_2 and τ_2 vanish while $2\beta^2\sigma_2 \to \xi_1^2$.

IV. DISCUSSION OF RESULTS

We have performed extensive numerical calculations for the three systems considered in Ref. [5]. Since no results were available in that paper for the transitions $0 \to 0$ and $0 \to 3$ we decided to redo the entire exact calculation independently by using the operator approach of Ma and Rhodes [10]. Where possible we checked our exact values against those in Refs. [5, 6] and found complete agreement. The parameters defining the colliding systems are the masses (in amu), the range a (in Å), and the dimensionless quantity $\alpha = \sqrt{2}\gamma(\sigma_0/a)$. Specifically the values used are as follows.

System 1 : $m_A = 4$, $m_B = m_C = 1$; $a = 0.2$; $\alpha = 0.3$ (this mimics He-H_2 scattering).

System 2 : $m_A = 2$, $m_B = m_C = 12$; $a = 0.2214$; $\alpha = 0.1287$.

System 3 : same as system 2, but with $\alpha = 0.3$.

For an inelastic process in which the molecule BC undergoes a vibrational transition $0 \to n$ conservation of energy gives

$$E_t = \mu v_i^2/2 + \hbar\omega_0/2 = \mu v_f^2/2 + (2n+1)\hbar\omega_0/2\,, \qquad (48)$$

from which one readily determines the average collision energy E in terms of the total energy E_t according to Eq. (36) .

In Figs. 1–3 we have plotted the exact inelastic transition probabilities $P_{0\to n}$, $n = 1$–3, together with those calculated from Eqs. (45)–(47) for the three systems defined above. The horizontal axis carries the total collision energy in units of the zero-point energy of the oscillator, i.e., $\epsilon_t = E_t/(\hbar\omega_0/2)$. We present plots in a linear scale rather than in the semilog scale used in Ref. [5] since otherwise one could hardly distinguish the curves from each other. Similar results hold for all the other systems investigated by Secrest and Johnson [8]. Needless to say, since unitarity is satisfied in Eq. (34) the agreement for elastic scattering $0 \to 0$ is equally good.

All that witnesses to the high quality of the first-order Magnus approximation in the adiabatic picture. From

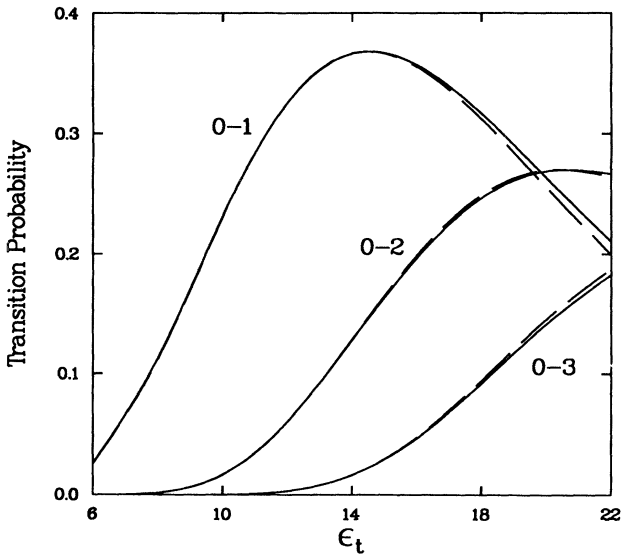

FIG. 1. Probabilities of inelastic transitions $0 \to 1$, $0 \to 2$, $0 \to 3$ for system 1 ($m_A = 4$, $m_B = m_C = 1$, $\alpha = 0.3$) as functions of the total energy in units of the oscillator zero-point energy [$\epsilon_t = E_t/(\hbar\omega_0/2)$]. Solid lines: exact results. Dashed lines: adiabatic Magnus approximation.

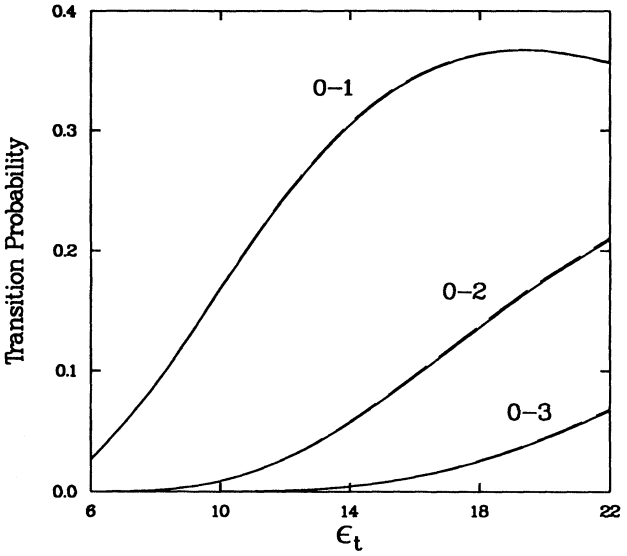

FIG. 2. Same as Fig. 1 for system 2 ($m_A = 2$, $m_B = m_C = 12$, $\alpha = 0.1287$).

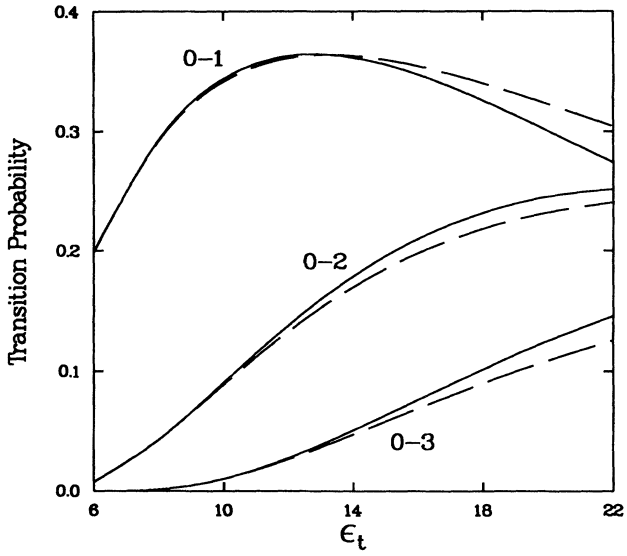

FIG. 3. Same as Fig. 1 for system 3 ($m_A = 2$, $m_B = m_C = 12$, $\alpha = 0.3$).

the magnitude of the transition probabilities it is quite obvious that in the energy range considered one is already very far from the adiabatic regime. A rapid estimate confirms this conclusion. The relevant quantity here is the ratio χ between the collision time T and the internal characteristic time $2\pi/\omega_0$. Thus $\chi \gg 1$ or $\chi \ll 1$ according to whether the perturbation is adiabatic or sudden. In our case

$$\chi = \omega_0 T/2\pi = (\gamma/\pi\alpha)(\mu/m)^{1/2}\epsilon^{-1/2}, \tag{49}$$

where $\epsilon = E/(\hbar\omega_0/2)$. For the $0 \rightarrow 1$ transition, for instance, χ is less than unity for the three systems even at the lowest energy shown in the figures. The process is therefore strongly nonadiabatic indeed, and the higher the energy the closer to the sudden limit. Increasing α should also result in a lower χ, which explains the change from Figs. 2 to 3. Nevertheless the adiabatic Magnus approximation proves remarkably accurate also under such extreme circumstances.

Judging from the examples treated here and in our previous papers [1,4] we expect quite generally the adiabatic Magnus approximation to be effective in the intermediate regime whenever first-order adiabatic perturbation theory adequately describes the situation near the adiabatic limit. This covers a very wide class of problems indeed. Special phenomena occurring only at infinite order in the adiabatic expansion (e.g., in field theory) lie outside the scope of our method.

ACKNOWLEDGMENTS

Continuing support by the IN2P3-CICYT international exchange program and by DGICYT (Spain) under Grant No. PB-88-0064 is gratefully acknowledged. The Division de Physique Théorique is "Unité de Recherche des Universités Paris 6 et Paris 11 associée au CNRS."

[1] S. Klarsfeld and J.A. Oteo, Phys. Rev. A **45**, 3329 (1992).
[2] W. Magnus, Commun. Pure Appl. Math. **7**, 649 (1956).
[3] L.I. Schiff, *Quantum Mechanics*, 3rd ed. (McGraw-Hill, New York, 1968), Chap. 8.
[4] S. Klarsfeld and J.A. Oteo, Phys. Lett. A **164**, 38 (1992).
[5] B. Gazdy and D.A. Micha, J. Chem. Phys. **82**, 4926 (1985).
[6] F.M. Fernández, D.A. Micha, and J. Echave, Phys. Rev. A **40**, 74 (1989).
[7] P. Pechukas and J.C. Light, J. Chem. Phys. **44**, 3897 (1966).
[8] D. Secrest and B.R. Johnson, J. Chem. Phys. **45**, 4556 (1966).
[9] R.A. Fisher, M.M. Nieto, and V.D. Sandberg, Phys. Rev. D **29**, 1107 (1984).
[10] X. Ma and W. Rhodes, Phys. Rev. A **39**, 1941 (1989).