Nondegenerate perturbation theory: A solution to the eigenvalue problem by the adiabatic-theorem formalism

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A nondegenerate perturbation theory is studied using the adiabatic-theorem formalism. A new factorization of the adiabatic time-evolution operator analogous to that obtained by Morita in his derivation of the linked-cluster theorem is given by carrying out a series of intermediate representations. This leads to a new expansion for the perturbed eigenvectors $|\alpha\rangle$ and energies E_α which are analytically expressed with respect to two matrix series $\mathscr{V}^{(n)}$ and $E^{(n)}$. The nth term of each series ($\mathscr{V}^{(n)}$ and $E^{(n)}$) includes perturbational contributions of power p, $p \geq n$. An explicit recurrence, $\mathscr{V}^{(n)}=V(\mathscr{V}^{n-1)}, E^{(n-1)})$, $E^{(n)}=E(\mathscr{V}^{(n-1)}, E^{(n-1)})$ is derived and allows us to calculate these matrices to an arbitrary iteration order (n). This is a central feature of our approach since the eigenvectors and eigenvalues can be generated, to this arbitrary order, by a simple numerical program.

I. INTRODUCTION

Perturbation theory is a theoretical physicist's most powerful tool.[1–5] However, the derivation of an explicit expansion of the wave function and eigenvalues in the perturbation is a complicated problem. This difficulty is apparent when the well-known Rayleigh-Schrödinger series for the nondegenerate case is considered.[2]

An explicit expression for the wave function and for the energy in a power series of the perturbation was first obtained by Goldstone.[6] An elegant derivation of the Goldstone formula from the Brillouin-Wigner expansion[7] can be found in the work of Brandow.[4] The Goldstone solution which concerns a system of interacting fermions uses time-dependent perturbation theory in the interaction representation. Introducing the time-dependent perturbation $\widetilde{V}(t)=\exp(iH_0t)V\exp(-iH_0t)\exp(\gamma t)$, the corresponding evolution operator

$$U_\gamma = \sum_n (-i)^n \times \int_{0<t_1<\cdots<t_n} V(t_1)V(t_2)\cdots V(t_n)dt_1\cdots dt_n ,$$

and analyzing the products of time-ordered operators which comprise the operator U_γ by the same algebra as is used in proving Wick's theorem,[8] Goldstone showed that the limit

$$\lim_{\gamma\to 0}\frac{U_\gamma\Phi_0}{\langle\Phi_0|U_\gamma|\Phi_0\rangle} \tag{1}$$

exists and has an explicit expression with respect to the linked Feynman graphs (with Φ_0 being the nondegenerate ground state of H_0). Finally, it was proved that the perturbed eigenfunction can be written

$$\Psi_0=\sum_L\left[\frac{1}{E_0-H_0}V\right]^n\Phi_0 , \tag{2}$$

where L represents the sum over the linked graphs and the energy shift

$$\Delta E=\sum_L\left\langle\Phi_0\left|V\left[\frac{1}{E_0-H_0}V\right]^n\right|\Phi_0\right\rangle , \tag{3}$$

where $\sum_L$ means summation over all connected graphs leading from Φ_0 to Φ_0, that is, with no external lines.

Following Brueckner's suggestions,[9] Huby[10] obtained tractable calculation rules for the two series (2) and (3). Nevertheless, these rules include nontrivial linear combinations of matrix elements whose complexity increases dramatically with the perturbation order. Thus it is impossible to use a numerical treatment to generate these series up to a high order of perturbation.

This method of investigation, through a power expansion of the U operator and the linked-cluster theorem is not the only possible solution. In the one-dimensional (and thus nondegenerate) case, a high-order perturbation treatment of the Rayleigh-Schrödinger (RS) series making use of general hypervirial theorems has been proposed by Killinbeck.[11] This formulation has the great advantage that it does not involve the calculation of perturbed wave functions and some applications concerning the hydrogen atom and the perturbed harmonic oscillator have been successful.[12] Unfortunately it seems very difficult to generalize this approach to the degenerate case.

In this work a third method is presented. A new factorization of the term $U_\gamma|\Phi_0\rangle$ in the adiabatic limit $\gamma\to 0+$ is derived, namely [cf. Eq. (1)]

$$U_\gamma|\Phi_0\rangle=U_R\langle\Phi_0|U_\gamma|\Phi_0\rangle ,$$

by using a series of intermediate representations of the adiabatic evolution operator. The basic hypotheses are presented in Sec. II in a formulation which does not use second quantization and thus not the exclusion principle. (As in the works of Klein[13] and Shavitt and Redmon[14] attention is focused on the formal aspects and the many-body application is ignored.) The explicit calculation of the factorization is presented in Sec. III. This leads to a new, nonperturbative expansion of the two factors U_R

and $\langle \Phi_0 | U_\gamma | \Phi_0 \rangle$. Iterative rules for the building up of these two terms from the matrices H_0 and V are given. Section IV puts forward a simple application concerning the case of a linearly perturbed harmonic oscillator.

II. ADIABATIC-THEOREM FORMALISM FOR NONDEGENERATE PROBLEMS

Let H be the Hermitian Hamiltonian of an isolated system. H is split into a zeroth-order Hamiltonian and a perturbation:

$$H = H_0 + V \,. \tag{4}$$

It is supposed that H_0 and H both have a discrete spectrum. In adiabatic perturbation theory the interaction V is switched on by adding a factor $\exp(\gamma t)$:

$$V(t) = \lim_{\gamma \to 0+} V \exp(\gamma t), \quad -\infty < t \le 0 \,. \tag{5}$$

The nondegenerate unperturbed state is denoted by $|\alpha_0\rangle$ and the corresponding unperturbed energy by E_α^0. According to the adiabatic theorem the zeroth-order eigenket evolves adiabatically into the perturbed eigenket of energy E_α, namely[15-17] (the convention $\hbar = 1$ is adopted)

$$\lim_{\gamma \to 0+} \exp\left[-i \int_{-\infty}^{0} H_0 dt\right] U(0,-\infty) P_0 = P \lim_{\gamma \to 0+} \exp\left[-i \int_{-\infty}^{0} H_0 dt\right] U(0,-\infty) \,, \tag{6a}$$

where

$$P_0 = |\alpha_0\rangle\langle\alpha_0| \,, \quad P = |\alpha\rangle\langle\alpha| \tag{6b}$$

and where $U(0,-\infty)$ represents the time-evolution operator defined by the equation

$$i \frac{\partial U(t,-\infty)}{\partial t} = \widetilde{V}(t) U(t,-\infty) \tag{6c}$$

with

$$\widetilde{V}(t) = \exp\left[i \int_{-\infty}^{t} H_0 dt'\right] V(t) \exp\left[-i \int_{-\infty}^{t} H_0 dt'\right] \,. \tag{6d}$$

The resolution of the stationary eigenvalue problem in the adiabatic-theorem formalism is then focused on the expression

$$U(0,-\infty)|\alpha_0\rangle = \exp\left[\int_{-\infty}^{0} [E_\alpha(t) - E_\alpha^0(t)] dt\right] |\alpha\rangle \,, \tag{7}$$

where $E_\alpha(t=0)$ and $|\alpha\rangle$ are the required eigenvalue and eigenvector.

An explicit calculation of $|\alpha\rangle$ and E_α can then be made from Eq. (7) by using Wick's theorem to express the Feynman[18] diagrams and Dyson's expansion[19] of U,

$$U(0,-\infty) = \sum_{n=0}^{\infty} U_{(0,-\infty)}^{(n)} = \frac{(-i)^n}{n!} \int_{-\infty}^{0} dt_1 \cdots dt_n T[\widetilde{V}(t_1) \cdots \widetilde{V}(t_n)] \,. \tag{8}$$

This application of Wick's theorem to the eigenvalue and eigenvector problem is known as the linked-cluster theorem. A simple derivation of this theorem and its extension to the degenerate case has been proposed by Morita.[20] In Ref. 20 it is shown that $U(t,-\infty)|\alpha_0\rangle$ has the following form:

$$U(t,-\infty)|\alpha_0\rangle = U(t,-\infty)|\alpha_0\rangle_c \langle\alpha_0| U(t,-\infty)|\alpha_0\rangle \,. \tag{9}$$

The first factor on the right-hand side (rhs) represents the sum of all possible products of connected diagrams which are open to the future at time t. This term is regular in the adiabatic limit (i.e., its power expansion does not introduce any singularity proportional to $1/\gamma^n$) and verifies the condition

$$\lim_{\gamma \to 0+} \left[\frac{\partial}{\partial t} U(t,-\infty)|\alpha_0\rangle_c\right] = 0 \,. \tag{10}$$

Moreover, Morita showed that

$$U(0,-\infty)|\alpha\rangle_c \tag{11a}$$

and

$$E_\alpha = \langle\alpha_0| H U(0,-\infty)|\alpha_0\rangle_c \tag{11b}$$

are the eigenvector (normalized so that $\langle\alpha_0|\alpha\rangle = 1$) and the corresponding eigenvalue. These results which were derived by Morita for H_0 quadratic in the electron operator have been generalized by Bulaevski[21] for arbitrary H_0 and V.

The main feature of Feynman-diagram expansions relevant to Eqs. (11) is that the expansions are power series in the perturbation $\widetilde{V}$. Unfortunately, there is not an explicit recurrence between two successive perturbation orders and it seems difficult to generate these series with a recursive numerical program.

The aim of this work is to show that it is nevertheless possible to derive a factorization similar to Eq. (9) in which each factor, calculated in an iterative procedure, is expressible with respect to two matrix series $\mathscr{V}$ and E; the nth-order terms $\mathscr{V}^{(n)}$ and $E^{(n)}$ include contributions of power p ($n \le p < \infty$) in the perturbation and are generated by a recursive program from $\mathscr{V}(n-1)$ and $E^{(n-1)}$.

To do this no particular form is given to the Hamiltonian H. It is only assumed that H is Hermitian and that it can be expanded on the basis of H_0 eigenkets:

$$H(t) = \sum_i E_i^0 |i_0\rangle\langle i_0| + \sum_i \sum_j \langle i_0| V(t) |j_0\rangle |i_0\rangle\langle j_0| \,. \tag{12}$$

The operator $|i_0\rangle\langle j_0|$ which includes the transition $i_0 \to j_0$ obeys the following commutation rule:

$$[\,|i_0\rangle\langle j_0|, |k_0\rangle\langle l_0|\,] = |i_0\rangle\langle l_0| \delta_{j,k} - |k_0\rangle\langle j_0| \delta_{l,i} \,. \tag{13}$$

As previously noted this expression does not guarantee the preservation of antisymmetry and many-body applications would require further expansions.

III. THEORY

The aim of the calculations presented in this section is to factor the term $U(0,-\infty)\,|\alpha_0\rangle$ as in Eq. (9). This factorization is obtained by making a series of intermediate representations.[22] The two following basic points concern the choice of this series.

(i) In Eq. (9), the term $U(0,-\infty)\,|\alpha\rangle_c$ is relevant to the diagrams open to the future at time 0 and which connect $|\alpha_0\rangle$ to $|j_0\rangle, j\neq\alpha$. The operators $|j_0\rangle\langle\alpha_0|$ which produce these transitions can be extracted in the intermediate-representation formalism by making representations with respect to

$$\sum_{\substack{j\\(j\neq\alpha)}} (\langle j_0|V|\alpha_0\rangle)\,|j_0\rangle\langle\alpha_0|\ .$$

(ii) The complexity of the expansion of the eigenvalue and eigenvector in the RS series comes partly from the fact that this series involves the unperturbed eigenvalues exclusively. This can be overcome in the representation formalism by making representations with respect to the projection operators $|j_0\rangle\langle j_0|$.

The series of representation will now be set out in detail. Its choice is based partly on the suggestions given previously in points (i) and (ii) and partly on intuition.

A. First interaction representation

A first interaction representation is made with respect to the diagonal part. To the partitioning

$$H(t)=V_{\rm d}^{0}+V_{\rm od}^{(0)}$$

corresponds the expansion

$$U(H)P_0=U(V_{\rm d}^{(0)})U(V_{\rm od}^{(1)})P_0 \tag{14a}$$

with

$$P_0=|\alpha_0\rangle\langle\alpha_0| \tag{14b}$$

and with

$$V_{\rm od}^{(1)}(t)=\sum_i\sum_{\substack{j\\(j\neq i)}} V_{ij}(t)\exp\left[\int_{-\infty}^{t}(E_i^0+V_{ii}-E_j^0-V_{jj})dt'\right]\times|i_0\rangle\langle j_0|\ . \tag{14c}$$

(The subscripts d and od, respectively, indicate the diagonal and the off-diagonal parts. It must be noted that $V_{\rm d}^{(0)}$ incorporates H_0. For the sake of clarity the two limits $t=-\infty$ and $t=0$ do not need to be written in each U factor.) This first operation is the standard intermediate representation often introduced in the adiabatic approach.

B. Continuation with series of similar transformations

The calculation is then continued using an infinite series of similar transformations. Each one is composed of two successive operations. For instance, the pth transformation is composed as follows. First an interaction representation with respect to the non-Hermitian term is made:

$$V_{\rm od}^{(p)}P_0=\sum_{\substack{j\\(j\neq\alpha)}}(V_{\rm od}^{(p)})_{j\alpha}\,|j_0\rangle\langle\alpha_0|$$

[where $V_{\rm od}^{(p)}$ represents the residual Hamiltonian after the $(p-1)$th transformation] and then an interaction representation with respect to the diagonal terms

$$V_{\rm d}^{(p)}=\sum_j(V_{\rm d}^{(p)})_{jj}\,|j_0\rangle\langle j_0|$$

which have been generated by the first operation of the pth transformation. This leads finally to the expansion

$$U(H)P_0=\lim_{p\to\infty}U(V_{\rm d}^{(0)})U(V_{\rm od}^{(1)}P_0)U(V_{\rm d}^{(1)})\times\cdots\times U(V_{\rm od}^{(p-1)}P_0)U(V_{\rm d}^{(p-1)})U(V_{\rm od}^{(p)})P_0\ . \tag{15}$$

It must be noted that each factor $U(V_{\rm od}^{(p)}P_0)$ is nonunitary since the operator $V_{\rm od}^{(p)}P_0$ is non-Hermitian. Nevertheless, the whole product, equal to $U(H(t))$, is unitary. The detailed calculations are set out in Appendixes A and B. They lead to an explicit expression for each factor in Eq. (15) and give a recurrent relationship between the matrix series $V_{\rm d}^{(l)}$ and $V_{\rm od}^{(l)}$. These results can be summarized as follows.

The operator $V_{\rm od}^{(n+1)}$ can be expressed in the form

$$V_{\rm od}^{(n+1)}(t)=\sum_{\substack{j\\(j\neq k)}}\sum_k V_{jk}^{(n+1)}(t)\,|j_0\rangle\langle k_0| \tag{16a}$$

with

$$\lim_{\gamma\to0+}V_{jk}^{(n+1)}(t)=\mathscr{V}_{jk}^{(n+1)}(t)\exp\left[\int_{-\infty}^{t}(\phi_j^{(n+1)}-\phi_k^{(n+1)})dt'+i\int_{-\infty}^{t}(E_j^{(n+1)}-E_k^{(n+1)})dt'\right]. \tag{16b}$$

The matrices $\mathscr{V}$, E, and ϕ appearing in Eq. (16b) will be given later in Eqs. (21) and (22).

The operator $V_{\rm d}^{(n+1)}$ has the expansion

$$V_{\rm d}^{(n+1)}(t)=\sum_j|j_0\rangle\langle j_0|\,V_{jj}^{(n+1)}(t) \tag{17a}$$

with

$$V_{jj}^{(n+1)}(t)=E_j^{(n+2)}-E_j^{(n+1)}-i(\phi_j^{(n+2)}-\phi_j^{(n+1)})\ . \tag{17b}$$

The various matrix elements $\mathscr{V}_{jk}$, E_j, and ϕ_j are all real functions of time. They can be separated into two groups according to their behavior in the adiabatic limit $\gamma\to0+$ A first group is constituted by the functions $\mathscr{V}(t)$ and $E(t)$ which become bound, adiabatically varying functions in the limit $\gamma\to0+$. A second group is constituted by the functions $\delta\mathscr{V}(t)$ and $\phi(t)$ which converge to zero at this limit. Moreover, they converge to zero sufficiently rapidly so that for any arbitrary complex function $\beta(t)$ whose modulus is bounded on the real axis $-\infty<t\leq0$ the following integrals converge:

$$\lim_{\gamma\to0+}\int_{-\infty}^{t}\chi(t')\beta(t')dt',\ \ \chi=\delta\mathscr{V}\text{ or }\phi\ . \tag{18}$$

(It must be noted that the functions $d\mathscr{V}/dt$ and dE/dt belong to this second group of functions.) Each group of equations is generated by recurrence equations. The introduction of the notations

$$Z_{j\alpha}^{(n)}=\frac{-\mathscr{V}_{j\alpha}^{(n)}}{E_j^{(n)}-E_\alpha^{(n)}}\ , \tag{19}$$

$$\delta Z_{j\alpha}^{(n)}=\frac{-\delta\mathscr{V}_{j\alpha}^{(n)}}{E_j^{(n)}-E_\alpha^{(n)}}-\frac{\mathscr{V}_{j\alpha}^{(n)}(\phi_j^{(n)}-\phi_\alpha^{(n)})}{E_j^{(n)}-E_\alpha^{(n)}} -\frac{d}{dt}\left[\frac{\mathscr{V}_{j\alpha}^{(n)}}{E_j^{(n)}-E_\alpha^{(n)}}\right]\frac{1}{E_j^{(n)}-E_\alpha^{(n)}} \tag{20}$$

leads to, for the first group,

$$E_j^{(1)}(t)=E_j^0+\langle j_0\mid V(t)\mid j_0\rangle,\quad \mathscr{V}_{jk}^{(1)}(t)=\langle j_0\mid V(t)\mid k_0\rangle\ , \tag{21a}$$

$$E_j^{(n+1)}=E_j^{(n)}-Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha j}^{(n)}\quad\text{for } j\neq\alpha\ , \tag{21b}$$

$$E_\alpha^{(n+1)}=E_\alpha^{(n)}+\sum_j \mathscr{V}_{\alpha j}^{(n)}Z_{j\alpha}^{(n)}\quad\text{for } j\neq\alpha\ , \tag{21c}$$

$$\mathscr{V}_{jk}^{(n+1)}=\mathscr{V}_{jk}^{(n)}-Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha k}^{(n)}\quad\text{for } j\neq k,\ k\neq\alpha\ , \tag{21d}$$

$$\mathscr{V}_{j\alpha}^{(n+1)}=\sum_{\substack{l\\(l\neq\alpha)}}\mathscr{V}_{jl}^{(n)}Z_{l\alpha}^{(n)}-Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha l}^{(n)}Z_{l\alpha}^{(n)}\quad\text{for } j\neq\alpha\ . \tag{21e}$$

Equations (21) constitute for matrix series $E^{(n)}$ and $\mathscr{V}^{(n)}$ a closed group of recurrence equations. The second group is

$$\phi_l^{(1)}=0,\quad \delta\mathscr{V}_l^{(1)}=0\ , \tag{22a}$$

$$\phi_l^{(n+1)}=\phi_l^{(n)}-\delta Z_{l\alpha}^{(n)}\mathscr{V}_{\alpha l}^{(n)}-Z_{l\alpha}^{(n)}\delta\mathscr{V}_{\alpha l}^{(n)}\quad\text{for } j\neq\alpha\ , \tag{22b}$$

$$\phi_\alpha^{(n+1)}=\phi_\alpha^{(n)}+\sum_j\delta\mathscr{V}_{\alpha j}^{(n)}Z_{j\alpha}^{(n)}+\mathscr{V}_{\alpha j}^{(n)}\delta Z_{j\alpha}^{(n)}\quad\text{for } j\neq\alpha\ , \tag{22c}$$

$$\delta\mathscr{V}_{jk}^{(n+1)}=\delta\mathscr{V}_{jk}^{(n)}-\delta Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha k}^{(n)}-Z_{j\alpha}^{(n)}\delta\mathscr{V}_{\alpha k}^{(n)}\quad\text{for } j\neq k,\ k\neq\alpha\ , \tag{22d}$$

$$\delta\mathscr{V}_{j\alpha}^{(n+1)}=\sum_{\substack{l\\(l\neq\alpha)}}\delta\mathscr{V}_{jl}^{(n)}Z_{l\alpha}^{(n)}+\mathscr{V}_{jl}^{(n)}\delta Z_{l\alpha}^{(n)}-\delta Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha l}^{(n)}Z_{l\alpha}^{(n)}-Z_{j\alpha}^{(n)}\delta\mathscr{V}_{\alpha l}^{(n)}Z_{l\alpha}^{(n)}-Z_{j\alpha}^{(n)}\mathscr{V}_{\alpha l}^{(n)}\delta Z_{l\alpha}^{(n)}\ . \tag{22e}$$

This second group of recurrence equations includes as an unknown the solutions $E^{(n)}$ and $\mathscr{V}^{(n)}$ from the first group of equations.

C. Propagation of operators $U(V_{\rm d}^{(n)})$

A last operation consists of propagating in Eq. (15) all the operators $U(V_{\rm d}^{(n)})$ from the left to the right of the series of operators. Equation (16) reveals that all the operators which compose $V_{\rm od}^{(n)}P_0$ commute. This is also the case for the operators constituting $V_{\rm d}^{(n)}$ [cf. Eq. (17)]. Consequently, the two corresponding evolution operators have the simple expressions[23]

$$U(0,-\infty;V_{\rm od}^{(n)}P_0)=\exp\left[\sum_{\substack{j\\(j\neq\alpha)}}\mid j_0\rangle\langle\alpha_0\mid\frac{1}{i}\int_{-\infty}^{0}V_{j\alpha}^{(n)}dt\right]\ ,$$

$$U(0,-\infty;V_{\rm d}^{(n)})=\exp\left[\sum_j\mid j_0\rangle\langle j_0\mid\frac{1}{i}\int_{-\infty}^{0}V_{jj}^{(n)}dt\right]\ .$$

The propagating of the operators $U(V_{\rm d}^{(n)})$ can be done gradually as explained in Appendix C. This leads to the final formula

$$\lim_{\gamma\to 0+}U(0,-\infty;H(t))P_0=\lim_{n\to\infty}\exp\left[\sum_{\substack{k\\(k\neq\alpha)}}\mid k_0\rangle\langle\alpha_0\mid\left[\sum_{l=1}^{n-1}\frac{-\mathscr{V}_{k\alpha}^{(l)}(t=0)}{E_k^{(l)}(t=0)-E_\alpha^{(l)}(t=0)}\right]\right]\times\exp\left[\sum_k\mid k_0\rangle\langle k_0\mid\int_{-\infty}^{0}\left[-iE_k^{(n)}-\phi_k^{(n)}\right]dt\right]U(V_{\rm od}^{(n+1)})P_0\ . \tag{23}$$

It will be supposed that the process converges in the sense that the matrix elements $\langle k_0\mid V_{\rm od}^{(n+1)}\mid\alpha_0\rangle$, $k\neq\alpha$ converge to zero as $n\to\infty$ so that $\lim_{n\to\infty}U(V_{\rm od}^{(n+1)})P_0=1$. This implies that the series $E_k^{(l)}$ and $\sum_l -\mathscr{V}_{k\alpha}^{(l)}/(E_k^{(l)}-E_\alpha^{(l)})$ which are power series in the perturbation converge.[24] Without developing this point any further it will be simply assumed that the perturbation is sufficiently weak to be in the convergence interval of these series noting, nevertheless, that in some special cases the convergence radius can be equal to zero.[25]

It is now possible to compare Eq. (23) with the factorization of Morita [Eq. (9)]. A rapid calculation made on Eq. (23) leads to

$$\langle\alpha_0\mid U(0,-\infty)\mid\alpha_0\rangle =\exp\left[\mid\alpha_0\rangle\langle\alpha_0\mid\lim_{n\to\infty}\int_{-\infty}^{0}(-iE_\alpha^{(n)}-\phi_\alpha^{(n)})dt\right]\ . \tag{24}$$

Moreover, the first term on the rhs of Eq. (23), equal to

$$\mathscr{V}_R=\left[1+\sum_{\substack{k\\(k\neq\alpha)}}\mid k_0\rangle\langle\alpha_0\mid\left[\sum_l -\mathscr{V}_{k\alpha}^{(l)}/(E_k^{(l)}-E_\alpha^{(l)})\right]\right]\ ,$$

verifies the condition $(d/dt)\mathscr{V}_R=0$ and can be expanded, through the expansion of the series $\mathscr{V}$ and E, as a power series in the perturbation (this is clearly not the case for the second factor because the integrals $\int_{-\infty}^{0} E_k^{(n)}dt$ diverge). Thus the factorization written in (23) can be assimilated to the factorization in (9); both have the same definitions. Moreover, using Eq. (7) the following is obtained for the eigenket normalized to unity:

$$|\alpha\rangle=\exp\left[\int_{-\infty}^{0}-\phi_\alpha^{(\infty)}dt\right]\times\left[|\alpha_0\rangle+\sum_{\substack{k\\(k\neq\alpha)}}|k_0\rangle\left[\sum_{l=1}^{\infty}\frac{-\mathscr{V}_{k\alpha}^{(l)}(0)}{E_k^{(l)}(0)-E_\alpha^{(l)}(0)}\right]\right]. \tag{25}$$

It is evident from Eqs. (25), (21), and (22) that the greatest difficulty comes from the series $\phi_\alpha^{(l)}$ because this series gives the functions δZ which introduce time derivatives [Eq. (20)]. However, this series is not needed since all the components of vector $|\alpha\rangle$ are proportional to the factor $\exp\{\int_{-\infty}^{0}-\phi_\alpha^{(\infty)}dt\}$. Finally, Eq. (25) can be rewritten

$$|\alpha\rangle=\lim_{N\to\infty}\sum_j A_{j\alpha}^{(N)}|j_0\rangle\ , \tag{26a}$$

with

$$A_{j\alpha}^{(N)}=\left[\sum_{n=1}^{N}-\mathscr{V}_{j\alpha}^{(n)}/(E_j^{(n)}-E_\alpha^{(n)})\right]D^{-1} \quad \text{for } j\neq\alpha \tag{26b}$$

$$A_{\alpha\alpha}^{(N)}=D^{-1}\ ,$$

where

$$D=\left[1+\sum_{\substack{j\\(j\neq\alpha)}}\left[\sum_{n=1}^{N}[-\mathscr{V}_{j\alpha}^{(n)}/(E_j^{(n)}-E_\alpha^{(n)})]\right]^2\right]^{1/2} \tag{26c}$$

so that the group of equations (22) does not need to be integrated.

Equations (21) and (26) constitute the central result of this work.

D. Directions for use

The eigenvector to which we shall pay attention is [cf. Eqs. (26)]

$$|\alpha\rangle=\left[|\alpha_0\rangle+\sum_{\substack{k\\(k\neq\alpha)}}|k_0\rangle\left[\sum_{l=1}^{\infty}-\frac{\mathscr{V}_{k\alpha}^{(l)}(0)}{E_k^{(l)}(0)-E_\alpha^{(l)}(0)}\right]\right]\left\{1+\sum_{\substack{j\\(j\neq\alpha)}}\left[\sum_{l=1}^{\infty}\left[-\frac{\mathscr{V}_{j\alpha}^{(l)}(0)}{E_j^{(l)}(0)-E_\alpha^{(l)}(0)}\right]\right]^2\right\}^{-1/2}. \tag{27a}$$

The corresponding eigenvalue is

$$E_\alpha=\lim_{l\to\infty}E_\alpha^{(l)}(0)\ . \tag{27b}$$

Thus the resolution of this perturbation problem simply implies the calculation of the two matrix series $\mathscr{V}^{(n)}$ and $E^{(n)}$.

Equation (21a) give the first-order terms, $E^{(1)}$ and $\mathscr{V}^{(1)}$, with respect to the Hamiltonian H_0 and to the perturbation V. Equations (21b) and (21c), and (21d) and (21e), respectively, constitute the recurrence equations:

$$E^{(n+1)}=E(E^{(n)},\mathscr{V}^{(n)})\ ,$$

$$\mathscr{V}^{(n+1)}=V(E^{(n)},\mathscr{V}^{(n)})\ .$$

This closed group of equations has been expressed in a numerical program to resolve the simple application presented in Sec. IV.

IV. A SIMPLE APPLICATION: THE LINEARLY FORCED HARMONIC OSCILLATOR

A simple application is the case of a linearly forced harmonic oscillator. The Hamiltonian of this system is

$$H=H_0+V\ , \tag{28}$$

with

$$H_0=-\frac{\partial^2}{\partial y^2}+\tfrac{1}{2}ky^2 \quad\text{and}\quad V=Ay\ .$$

The introduction of the adiabatically translated vibrational coordinate $z=y+A/k$ leads to

$$H=-\frac{\partial^2}{\partial z^2}+\tfrac{1}{2}kz^2-b,\quad b=A^2/2k \tag{29}$$

so that the nonperturbed $\phi_n^0(y)$ and the corresponding perturbed eigenvectors $\phi_n(y)$ are related by the equations

$$\phi_n(y)=\phi_n^0(z)\ . \tag{30}$$

The perturbed eigenvalues are

$$E_n=(n+\tfrac{1}{2})(2k)^{1/2}-b\ . \tag{31}$$

The functions $\phi_n^0(y)$ have well-known expressions on the Hermite polynomial basis so that the coefficients $A_{n,k}$ of the expansion,

$$\phi_n=\sum_k A_{n,k}\phi_k^0,\quad n=0,1,2,\ldots \tag{32}$$

can be calculated without difficulty using the relationship $A_{n,k}=\langle\phi_k^0|\phi_n\rangle$. This leads to a recurrence relation:

$$
\begin{aligned}
A_{n,0} &= \left[\frac{2^n}{n!}\right]^{1/2}\left[\frac{c}{2}\right]^n \exp\left[-\frac{c^2}{4}\right], \\
A_{n,1} &= \frac{1}{n^{1/2}}\left[n-\frac{c^2}{4}\right]A_{n-1,0}, \\
A_{n,p+1} &= \frac{1}{[n(p+1)]^{1/2}}\left[\left[p+n-\frac{c^2}{4}\right]A_{n-1,p} - [p(n-1)]^{1/2}A_{n-2,p-1}\right], \\
A_{p,n} &= (-1)^{n-p}A_{n,p},
\end{aligned}
\tag{33}
$$

with

$$c=(2A^2)^{1/2}.$$

The perturbational series $A_{j\alpha}^{(N)}$ [Eq. (26b)] and $E_\alpha^{(N)}$ [Eq. (21c)] were calculated using a truncated basis constituted by the 40 first unperturbed states and compared in Figs. 1–4 with the exact results given in Eqs. (31) and (33). The case of the ground state ($n=0$) for the coupling strengths $A=0.2$, 0.5, and 1 [cf. Eq. (28)] and that of the first excited state for $A=0.2$ and 0.5 (it was assumed that $k=0.5$) were analyzed. In every case the series converges to the exact values.

Figures 1 and 2 illustrate the fact that the expansion is built on nonpower series since the shift of the energy is, in this case, a quadratic function of the perturbation. Figure 3 shows that more than ten iterations are needed to produce the convergence in this strong-coupling case. This figure also reveals the efficiency of the formulation since it would be very difficult to continue the RS series at this high order using Huby's rules. The convergence interval of the series has not been studied in detail. However, it can be said that in the case of the ground state the radius of convergence is between $A=1.0$ and 2.0 (the series diverges in this last case) and that in the case of the first

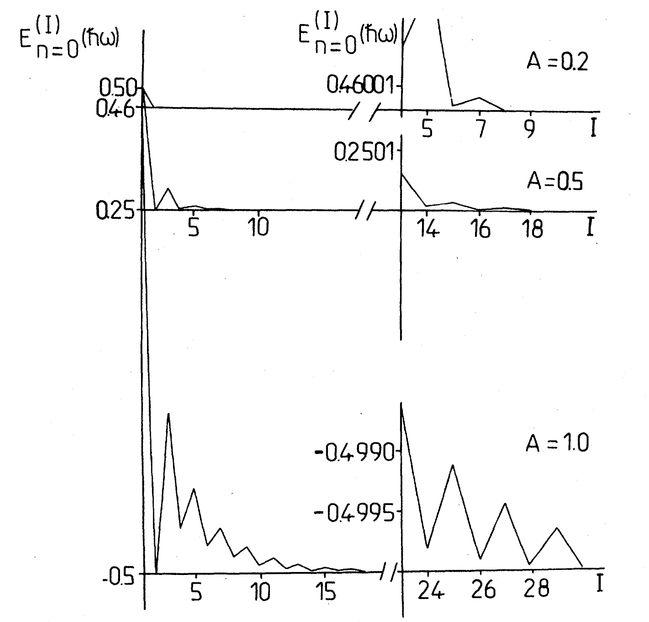

FIG. 1. Convergence of the perturbed eigenvalue $E_{n=0}^{(I)}$ (in units of $\hbar\omega$) for the ground state of the linearly forced harmonic oscillator vs the recurrence index I. Three values of the perturbation magnitude ($A=0.2$, 0.5, and 1.0) are investigated [cf. Eq. (29)]. The three values $E=0.46$, 0.25, and -0.5 are the exact values given by Eq. (31).

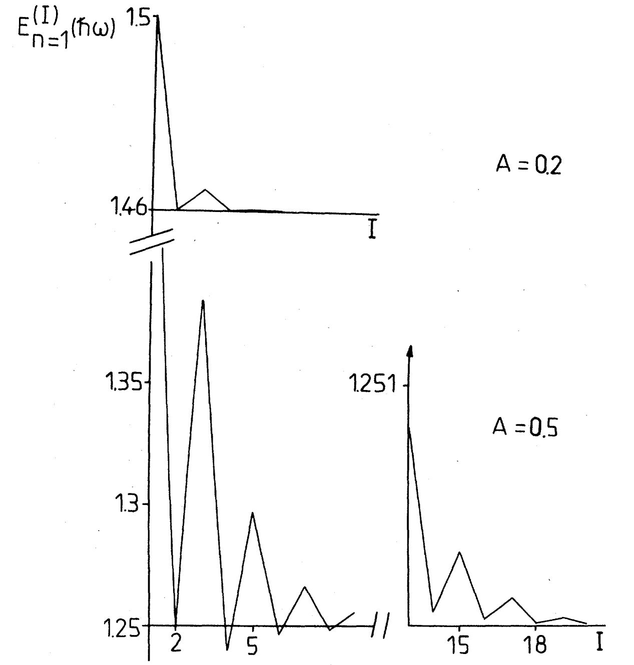

FIG. 2. Same as Fig. 1 for the first excited state. Two values of the perturbation magnitude are investigated ($A=0.2$ and 0.5).

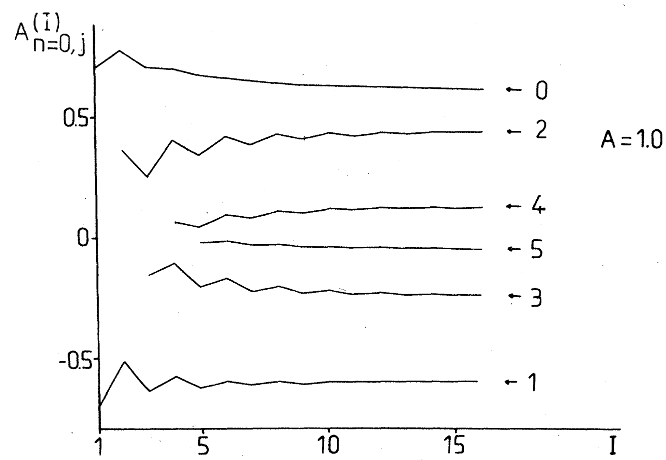

FIG. 3. Convergence of the coefficients $A_{n=0,j}^{(I)}$ [Eqs. (33)] of the expansion of the perturbed eigenvector of the ground state of the linearly forced oscillator on the unperturbed harmonic basis vs the recurrence index I. The perturbation magnitude $A=1.0$ is studied. The arrows associated with a number j ($0\le j\le 5$) give the exact values of the coefficients $A_{n=0,j}$ given by Eqs. (33).

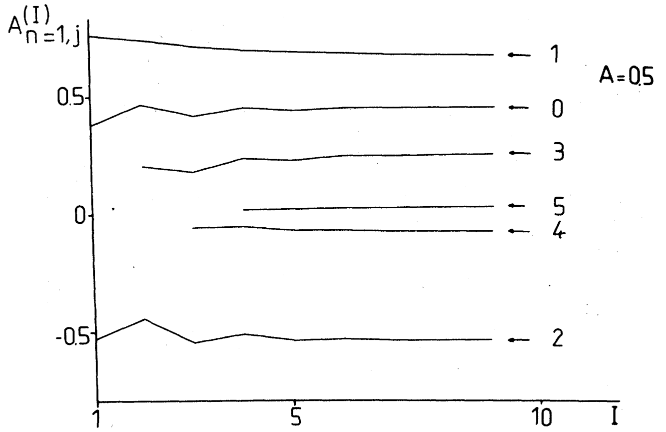

FIG. 4. Same as Fig. 3 for the first excited state and for a perturbation magnitude $A=0.5$.

excited state the radius is between $A=0.5$ and 1.

V. CONCLUSION

The simple application made in Sec. IV reveals that the explicit expansions in the perturbation derived in this work converge to the exact values for the wave function and for energy under conditions of sufficiently weak perturbation amplitudes. As expected the convergence is rapid for the weakest values of the perturbation and becomes slower and slower when approaching the convergence radius of the series. This finite radius of convergence comes from the natural finite interval of convergence of the series $E^{(N)}$ [Eq. (21c)] and $A^{(N)}$ [Eq. (26b)] and from the assumption that at each order of the recurrence the perturbed eigenvalue $E_\alpha^{(n)}$ does not cross any of the other eigenvalues $E_j^{(n)}$.

A large range of applications are currently under study concerning various vibrational and rotational systems. The first results confirm the adequecy of the theory and the existence in each case of a finite interval of convergence which is more or less large according to the nature of the perturbation matrix.

This formulation which uses a recursive numerical calculation is particularly efficient in the strong-coupling situations when a high perturbation order is required. In relation to Killinbeck's treatment the construction of the perturbation matrix on the zeroth-order representation is required, but it can be generalized to the degenerate case without introducing any fundamental difficulties.[26]

ACKNOWLEDGMENT

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APPENDIX A

The three appendixes set out the calculations for Eqs. (14)–(23). Appendix A shows more specifically the first operation in the pth nonunitary transformation, i.e., the interaction representation with respect to $V_{\rm od}^{(p)}P_0$; Appendix B the second operation, i.e., the interaction representation with respect to $V_{\rm d}^{(p)}$; and, finally, Appendix C presents the permutations of the operators which propagate in Eq. (15) the operators $U(V_{\rm d}^{(n)})$ to the right of this operator series.

Consider Eq. (15) and more particularly the term $U(V_{\rm od}^{(p)})$ which is the last term on the rhs. A particular form is assumed for the Hamiltonian $V_{\rm od}^{(p)}$ and the calculations of Appendixes A and B confirm this hypothesis by showing that $A_{\rm od}^{(p+1)}$, obtained from $V_{\rm od}^{(p)}$ using the pth transformation, has a similar expression.

Thus it is assumed that $V_{\rm od}^{(p)}(t)$ can be written

$$V_{\rm od}^{(p)}(t)=\sum_{\substack{j\\(j\neq k)}}\sum_k |j_0\rangle\langle k_0|(V_{\rm od}^{(p)})_{jk}$$
$$=\sum_{\substack{j\\(j\neq k)}}\sum_k |j_0\rangle\langle k_0|\left[(\mathscr{V}_{jk}^{(p)}+i\delta\mathscr{V}_{jk}^{(p)})\exp\left[\int_{-\infty}^t(\phi_j^{(p)}-\phi_k^{(p)}+iE_j^{(p)}-iE_k^{(p)})dt'\right]\right], \quad \text{(A1)}$$

where $\mathscr{V}_{jk}$, $\delta\mathscr{V}_{jk}$, ϕ_j, and E_j are finite real-time functions. It is simultaneously supposed that these functions obey the following conditions.

(1) In the adiabatic limit $\gamma\to 0+$, the functions $\mathscr{V}_{jk}^{(p)}$ and $E_j^{(p)}$ become bound, adiabatically varying functions.

(2) In the adiabatic limit the derivatives $d\mathscr{V}_{jk}^{(p)}/dt$ and $dE_j^{(p)}/dt$ and the functions $\delta\mathscr{V}_{jk}^{(p)}$ and $\phi_j^{(p)}$ converge to zero sufficiently rapidly that for any arbitrary complex function $\beta(t)$ whose modulus is bound on the real axis $-\infty<t\leq 0$ the integrals

$$\lim_{\gamma\to 0+}\int_{-\infty}^t \chi(t')\beta(t')dt', \quad \chi(t)=d\mathscr{V}/dt, dE/dt, \delta\mathscr{V}, \phi$$

converge.

(3) The frequencies $\omega_{j\alpha}^{(p)}(t)=E_j^{(p)}(t)-E_\alpha^{(p)}(t)$, $j\neq\alpha$ do not vanish at the limit $\gamma\to 0+$ for any value of t between $-\infty$ and 0.

It must be noted first that these conditions are consistent with the first-order result [Eq. (14b)] with the correspondence

$$\mathscr{V}^{(1)}_{jk}(t)=\langle j_0\,|\,V\,|\,k_0\rangle\exp(\gamma t),\quad (\delta\mathscr{V}^{(1)}_{(t)})_{jk}=0$$

$$E_j^{(1)}(t)=E_j^0+\langle j_0\,|\,V\,|\,j_0\rangle\exp(\gamma t),\quad \phi_{jk}^{(1)}(t)=0 \tag{A2}$$

where ϕ_{jk} designates $\phi_j-\phi_k$.

The first operation in the pth nonunitary transformation is the interaction representation with respect to

$$V_{\rm od}^{(p)}P_0=\sum_{\substack{j\\(j\neq\alpha)}}(V_{\rm od}^{(p)})_{j\alpha}\,|\,j_0\rangle\langle\alpha_0\,| \ . \tag{A3}$$

The corresponding evolution operator has the rigorous expression

$$U_{\rm int}(t)=\exp\left[\frac{1}{i}\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle\alpha_0\,|\left[\int_{-\infty}^{t}(V_{\rm od}^{(p)})_{j\alpha}dt'\right]\right] . \tag{A4a}$$

By noting

$$W_{j\alpha}^{(p)}(t)=\int_{-\infty}^{t}(V_{\rm od}^{(p)})_{j\alpha}dt' \tag{A4b}$$

and

$$A(t)=\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle\alpha_0\,|\,[iW_{j\alpha}^{(p)}(t)] ,$$

$$B(t)=\sum_{\substack{j\\(j\neq k)}}\sum_{\substack{k\\(k\neq\alpha)}}|\,j_0\rangle\langle k_0\,|\,[V_{\rm od}^{(p)}(t)]_{jk}$$

the calculations lead to

$$V_{\rm int}(t)=\exp[A(t)]B(t)\exp[-A(t)] . \tag{A5}$$

Using the well-known expansion

$$\exp(A)B\exp(-A)=B+[A,B]+\frac{1}{2!}[A,[A,B]]+\cdots \tag{A6}$$

and the commutation rules expressed by Eq. (13), the following is obtained:

$$[A,B]=\sum_{j}|\,j_0\rangle\langle\alpha_0\,|\left[-i\sum_{l}(V_{\rm od}^{(p)})_{jl}(W^{(p)})_{l\alpha}\right]+\sum_{\substack{j\\(j\neq\alpha)}}\sum_{\substack{k\\(k\neq\alpha)}}|\,j_0\rangle\langle k_0\,|\,[i(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha k}] ,$$

$$[A,[A,B]]=2\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle\alpha_0\,|\left[(W^{(p)})_{j\alpha}\sum_{\substack{l\\(l\neq\alpha)}}(V_{\rm od}^{(p)})_{\alpha l}(W^{(p)})_{l\alpha}\right] ,$$

$$[A,[A,[A,B]]]=0 .$$

Thus only the three terms contribute to the expansion (A6) so that the final result is

$$U(0,-\infty;V_{\rm od}^{(p)})=\exp\left[-i\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle\alpha_0\,|\,[W(t=0)]_{j\alpha}\right]\times U(0,-\infty;V_{\rm int})$$

with

$$V_{\rm int}=\sum_{j}\sum_{\substack{k\\(k\neq\alpha)}}|\,j_0\rangle\langle k_0\,|\,[(V_{\rm od}^{(p)})_{jk}+i(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha k}]+\sum_{j}|\,j_0\rangle\langle\alpha_0\,|\left[-i\sum_{l}(V_{\rm od}^{(p)})_{jl}(W^{(p)})_{l\alpha}+(W^{(p)})_{j\alpha}\sum_{l}(V_{\rm od}^{(p)})_{\alpha l}(W^{(p)})_{l\alpha}\right] . \tag{A7}$$

This last expression gives a diagonal part, denoted $V_{\rm d}^{(p)}$ in Eq. (15),

$$V_{\rm d}^{(p)}=\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle j_0\,|\,]i(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha j}]+|\,\alpha_0\rangle\langle\alpha_0\,|\left[-i\sum_{\substack{l\\(l\neq\alpha)}}(V_{\rm od}^{(p)})_{\alpha l}(W^{(p)})_{l\alpha}\right] \tag{A8}$$

which is used to build up the second operation of the pth transformation:

$$U(0,-\infty;V_{\rm int})=U(0,-\infty;V_{\rm d}^{(p)})U(0,-\infty;V_{\rm od}^{(p+1)}) . \tag{A9}$$

APPENDIX B

The interaction representation with respect to $V_{\rm d}^{(p)}$ [cf. Eq. (A9)] is presented. The evolution operator associated with $V_{\rm d}^{(p)}$ has the rigorous expression

$$U(t,-\infty;V_{\rm d}^{(p)})=\exp\left[-i\int_{-\infty}^{t}V_{\rm d}^{(p)}dt'\right] , \tag{B1}$$

where [Eq. (A8)]

$$V_{\rm d}^{(p)}=\sum_{\substack{j\\(j\neq\alpha)}}|\,j_0\rangle\langle j_0\,|\,[i(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha j}]+|\,\alpha_0\rangle\langle\alpha_0\,|\left[-i\sum_{\substack{l\\(l\neq\alpha)}}(V_{\rm od}^{(p)})_{\alpha l}(W^{(p)})_{l\alpha}\right] .$$

One difficulty is due to the presence of the divergent integral $\int_{-\infty}^{t} V_d^{(p)} dt'$. Thus it is necessary to retain in the expansion of $V_d^{(p)}$ the terms which are finite at the adiabatic limit and those which converge to zero and whose time integral is finite. The two factors $(V_{\text{od}}^{(p)})_{\alpha j}$ and $(W^{(p)})_{j\alpha}$ which appear in the expression of $V_d^{(p)}$ are studied.

(i) The matrix element $(V_{\text{od}}^{(p)})_{\alpha j}$ is given by Eq. (A1). A rapid analysis of Eq. (A7) reveals that the successive intermediate representations have not affected the functions $\mathscr{V}_{\alpha j}^{(p)} + i(\delta\mathscr{V}^{(p)})_{\alpha j}$, so that

$$\mathscr{V}_{\alpha j}^{(p)} + i(\delta\mathscr{V}^{(p)})_{\alpha j} = \mathscr{V}_{\alpha j}^{(1)} + i(\delta\mathscr{V}^{(1)})_{\alpha j} = \langle \alpha_0 | V | j_0 \rangle \exp(\gamma t) , \tag{B2}$$

thus

$$(V_{\text{od}}^{(p)})_{\alpha j} = \langle \alpha_0 | V | j_0 \rangle \exp\left[\gamma t + \int_{-\infty}^{t} (\phi_\alpha^{(p)} - \phi_j^{(p)} + iE_\alpha^{(p)} - iE_j^{(p)}) dt'\right] . \tag{B3}$$

(ii) The equations (A1) and (A4b) lead to

$$[W^{(p)}(t)]_{j\alpha} = \int_{-\infty}^{t} (\mathscr{V}_{j\alpha}^{(p)} + i\delta\mathscr{V}_{j\alpha}^{(p)}) \exp\left[\int_{-\infty}^{t'} (\phi_j^{(p)} - \phi_\alpha^{(p)} + iE_j^{(p)} - iE_\alpha^{(p)}) dt''\right] dt' . \tag{B4}$$

This integral can be calculated using the two functions

$$U(t) = (\mathscr{V}_{j\alpha}^{(p)} + i\delta\mathscr{V}_{j\alpha}^{(p)}) \exp\left[\int_{-\infty}^{t} (\phi_j^{(p)} - \phi_\alpha^{(p)}) dt'\right] / i(E_j^{(p)} - E_\alpha^{(p)})$$

and

$$\frac{dV(t)}{dt} = i(E_j^{(p)} - E_\alpha^{(p)}) \exp\left[i\int_{-\infty}^{t} (E_j^{(p)} - E_\alpha^{(p)}) dt'\right] .$$

The recurrent procedure of integration by parts leads to

$$(W^{(p)})_{j\alpha} = (I_0 + I_1 + \cdots + I_N) \exp\left[i\int_{-\infty}^{t} (E_j^{(p)} - E_\alpha^{(p)}) dt'\right] + R_{n+1} \tag{B5}$$

with

$$I_0 = \chi_{j\alpha}^{(p)}(t) ,$$

$$I_1 = -\frac{d}{dt}[\chi_{j\alpha}^{(p)}(t)] / i(E_j^{(p)} - E_\alpha^{(p)}) ,$$

$$\vdots$$

$$I_N = (-1)^N \frac{d}{dt}\left[\frac{d}{dt}\left[\cdots \frac{d}{dt}(\chi_{j\alpha}^{(p)}) \frac{1}{i(E_j^{(p)} - E_\alpha^{(p)})} \cdots\right] \frac{1}{i(E_j^{(p)} - E_\alpha^{(p)})}\right] \frac{1}{i(E_j^{(p)} - E_\alpha^{(p)})} ,$$

$$R_{N+1} = \int_{-\infty}^{t} I_{N+1}(t_1) \exp\left[i\int_{-\infty}^{t_1} (E_j^{(p)} - E_\alpha^{(p)}) dt_2\right] dt_1$$

with

$$\chi_{j\alpha}^{(p)}(t) = (\mathscr{V}_{j\alpha}^{(p)} + i\delta\mathscr{V}_{j\alpha}^{(p)}) \exp\left[\int_{-\infty}^{t} (\phi_j^{(p)} - \phi_\alpha^{(p)}) dt'\right] / i(E_j^{(p)} - E_\alpha^{(p)}) .$$

In light of the hypothesis made in Appendix A relative to the matrices $\mathscr{V}^{(p)}$, $\delta\mathscr{V}^{(p)}$, $E^{(p)}$, and $\phi^{(p)}$ it is possible to select the terms of W which are finite at the adiabatic limit and those which vanish and whose time integral is finite. This leads to

$$(W^{(p)})_{j\alpha} = (iZ_{j\alpha}^{(p)} - \delta Z_{j\alpha}^{(p)}) \times \exp\left[\int_{-\infty}^{t} (\phi_j^{(p)} - \phi_\alpha^{(p)} + iE_j^{(p)} - iE_\alpha^{(p)}) dt'\right] + O(\gamma^2) , \tag{B6a}$$

where

$$Z_{j\alpha}^{(p)} = \frac{-\mathscr{V}_{j\alpha}^{(p)}}{E_j^{(p)} - E_\alpha^{(p)}} , \tag{B6b}$$

$$(\delta Z^{(p)})_{j\alpha} = \frac{-(\delta\mathscr{V}^{(p)})_{j\alpha}}{E_j^{(p)} - E_\alpha^{(p)}} - \frac{\mathscr{V}_{j\alpha}^{(p)}(\phi_j^{(p)} - \phi_\alpha^{(p)})}{E_j^{(p)} - E_\alpha^{(p)}} - \frac{d}{dt}\left[\frac{\mathscr{V}_{j\alpha}^{(p)}}{E_j^{(p)} - E_\alpha^{(p)}}\right] \frac{1}{E_j^{(p)} - E_\alpha^{(p)}} . \tag{B6c}$$

In this expansion $\lim_{\gamma\to 0^+} Z_{j\alpha}^{(p)}$ is finite, $\lim_{\gamma\to 0}(\delta Z^{(p)})_{j\alpha} = \lim_{\gamma\to 0^+}[O(\gamma^2)] = 0$. Moreover, for any arbitrary complex function $\beta(t)$ whose modulus is bound in the real axis $(-\infty, 0]$, the integral $\int_{-\infty}^{(t)} [(\delta Z^{(p)})_{j\alpha}\beta] dt'$

converges to a finite complex value and the integral $\int_{-\infty}^{t}[O(\gamma^2)\beta]dt'$ converges to zero.

Finally, Eqs. (A7), (A9), (B1), (B3), and (B6) lead to

$$U(0,-\infty;V_{\rm od}^{(p)}) = \exp\left[-i\sum_{\substack{j\\(j\neq\alpha)}}|j_0\rangle\langle\alpha_0|[W_{j\alpha}^{(p)}(t=0)]\right] \times U(0,-\infty;V_{\rm d}^{(p)})U(0,-\infty;V_{\rm od}^{(p+1)}) \tag{B7a}$$

with

$$\lim_{\gamma\to 0+} W_{j\alpha}^{(p)}(0) = \frac{-i\mathscr{V}_{j\alpha}^{(p)}(0)}{E_j^{(p)}(0)-E_\alpha^{(p)}(0)} \times\exp\left[\int_{-\infty}^{0}(\phi_j^{(p)}-\phi_\alpha^{(p)}+iE_j^{(p)}-iE_\alpha^{(p)})dt\right] \tag{B7b}$$

and with

$$\lim_{\gamma\to 0+} U(0,-\infty;V_{\rm d}^{(p)}) = \exp\left[\sum_j |j_0\rangle\langle j_0|\int_{-\infty}^{0}(-\phi_j^{(p+1)}+\phi_j^{(p)} -iE_j^{(p+1)}+iE_j^{(p)})dt\right], \tag{B7c}$$

where

$$E_j^{(p+1)}-E_j^{(p)} = -Z_{j\alpha}^{(p)}\mathscr{V}_{\alpha j}^{(p)} \quad \text{for } j\neq\alpha ,$$

$$E_\alpha^{(p+1)}-E_\alpha^{(p)} = -\sum_{\substack{j\\(j\neq\alpha)}} E_j^{(p+1)}-E_j^{(p)} ,$$

$$\phi_j^{(p+1)}-\phi_j^{(p)} = (\delta Z^{(p)})_{j\alpha}\mathscr{V}_{\alpha j}^{(p)} \quad \text{for } j\neq\alpha ,$$

$$\phi_\alpha^{(p+1)}-\phi_\alpha^{(p)} = -\sum_{\substack{j\\(j\neq\alpha)}}(\phi_j^{(p+1)}-\phi_j^{(p)}) .$$

The residual Hamiltonian $V_{\rm od}^{(p+1)}$ is expressed as

$$\begin{aligned} V_{\rm od}^{(p+1)} = &\sum_{\substack{j\\(j\neq k)}}\sum_{\substack{k\\(k\neq\alpha)}}|j_0\rangle\langle k_0|[(V_{\rm od}^{(p)})_{jk}+i(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha k}] \\ &\times\exp\left[i\int(E_j^{(p+1)}-E_j^{(p)}-E_k^{(p+1)}+E_k^{(p)})dt+\int(\phi_j^{(p+1)}-\phi_j^{(p)}-\phi_k^{(p+1)}+\phi_k^{(p)})dt\right] \\ &+\sum_{\substack{j\\(j\neq\alpha)}}|j_0\rangle\langle\alpha_0|\left[\sum_l -i(V_{\rm od}^{(p)})_{jl}(W^{(p)})_{l\alpha}+(W^{(p)})_{j\alpha}(V_{\rm od}^{(p)})_{\alpha l}(W^{(p)})_{l\alpha}\right] \\ &\times\exp\left[i\int(E_j^{(p+1)}-E_j^{(p)}-E_\alpha^{(p+1)}+E_\alpha^{(p)})dt+\int(\phi_j^{(p+1)}-\phi_j^{(p)}-\phi_\alpha^{(p+1)}+\phi_\alpha^{(p)})dt\right] . \end{aligned}$$

In this expression the adiabatic limit is taken and the terms which contribute to the next nonunitary transformation are retained, i.e., the finite terms and terms which converge to zero and whose time integral is finite. The final result of this operation is summarized in Eqs. (18)–(22). A detailed investigation of Eq. (B8) reveals that it complies with Eq. (A1).

APPENDIX C

Consider now Eq. (15). On the rhs of this equation the operators $U(V_{\rm d}^{(l)})$ can be propagated from the left to the right of this series of operators. This can be done for instance using the following particular commutation scheme:

$$U(H)=U(V_{\rm d}^{(0)})U(V_{\rm od}^{(1)}P_0)U(V_{\rm d}^{(1)})\cdots U(V_{\rm d}^{(p-1)})U(V_{\rm od}^{(p)}) ; \tag{C1a}$$

first commutation,

$$U(V_{\rm d}^{(0)})U(V_{\rm od}^{(1)}P_0)=U(Y^{(1)})U(V_{\rm d}^{(0)}) \tag{C1b}$$

with

$$Y_{(t)}^{(1)}=U^{-1}(t,-\infty;V_{\rm d}^{(0)})V_{\rm od}^{(1)}(t)P_0U(t,-\infty;V_{\rm d}^{(0)}) ; \tag{C1c}$$

second commutation,

$$U(V_{\rm d}^{(0)})U(V_{\rm d}^{(1)})U(V_{\rm od}^{(2)}P_0)=U(Y^{(2)})U(V_{\rm d}^{(0)})U(V_{\rm d}^{(1)}) \tag{C1d}$$

with

$$Y^{(2)}(t)=U^{-1}(t,-\infty;V_{\rm d}^{(1)})U^{-1}(t,-\infty;V_{\rm d}^{(0)}) \times V_{\rm od}^{(2)}(t)P_0U(t,-\infty;V_{\rm d}^{(0)})U(t,-\infty;V_{\rm d}^{(1)}) ; \tag{C1e}$$

etc. For the sake of clarity the two time limits, 0 and $-\infty$ in the U factors, have been eliminated in Eqs. (C1a), (C1b), and (C1d).

An explicit expression of the operator $U(V_{\rm d}^{(l)})$ is given by Eq. (B7c). All the operators $\{V_{\rm d}^{(l)},\ l=1,2,\ldots\}$ commute so that after the $(l-1)$th commutation the following sequence of operators is obtained:

$$U(V_{\rm d}^{(0)})U(V_{\rm d}^{(1)})\cdots U(V_{\rm d}^{(l-1)})=U\left[\sum_{p=1}^{l-1}V_{\rm d}^{(p)}\right] \tag{C2}$$

with

$$U\left[\sum_{p=1}^{l-1}V_{\rm d}^{(p)}\right]=\exp\left[\sum_{j}|j_0\rangle\langle j_0|\left[\int_{-\infty}^{0}-iE_j^{(l)}-\phi_j^{(l)}\right]\right].$$

The following sequence is obtained:

$$\mathscr{P}=U\left[\sum_{p=1}^{l-1}V_{\rm d}^{(p)}\right]U(V_{\rm od}^{(l)}P_0)U(V_{\rm d}^{(l)}) \tag{C3}$$

with [cf. Eq. (B7)]

$$U(V_{\rm od}^{(l)}P_0)=\exp\left[\sum_{\substack{j\\(j\neq\alpha)}}|j_0\rangle\langle\alpha_0|\left[\frac{\mathscr{V}_{j\alpha}^{(l)}(t=0)}{E_j^{(l)}(t=0)-E_\alpha^{(l)}(t=0)}\right]\exp\left[\int_{-\infty}^{0}\phi_j^{(l)}-\phi_\alpha^{(l)}+iE_j^{(l)}-iE_\alpha^{(l)}\right]\right].$$

This product is transformed after the lth commutation into

$$\mathscr{P}=\exp\left[\sum_{\substack{k\\(k\neq\alpha)}}|k_0\rangle\langle\alpha_0|\left[\frac{-\mathscr{V}_{k\alpha}^{(l)}(t=0)}{E_k^{(l)}(t=0)-E_\alpha^{(l)}(t=0)}\right]\right]U\left[\sum_{p=1}^{l}V_{\rm d}^{(p)}\right]. \tag{C4}$$

Finally, using the fact that all the operators

$$\left\{\sum_{\substack{k\\(k\neq\alpha)}}|k_0\rangle\langle\alpha_0|\left[\frac{-\mathscr{V}_{k\alpha}^{(l)}}{E_k^{(l)}-E_\alpha^{(l)}}\right],\ l=1,2,\ldots\right\}$$

commute, the following can be obtained from Eq. (C1):

$$\lim_{\gamma\to0+}U(0,-\infty;H(t))=\exp\left[\sum_{\substack{k\\(k\neq\alpha)}}|k_0\rangle\langle\alpha_0|\left[\sum_{l=1}^{p-1}\frac{-\mathscr{V}_{k\alpha}^{(l)}(t=0)}{E_k^{(l)}(t=0)-E_\alpha^{(l)}(t=0)}\right]\right]$$
$$\times\exp\left[\sum_{k}|k_0\rangle\langle k_0|\int_{-\infty}^{0}(-iE_k^{(p)}-\phi_k^{(p)}dt\right]U(V_{\rm od}^{(p)}). \tag{C5}$$

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