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Harmonic perturbation

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## Determinantal solution of density matrix equations in time-dependent quantum mechanics: II. Harmonic perturbation

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**Abstract.** The determinantal formalism for solving the time-dependent Schrödinger equation of the density matrix has been developed, in I, assuming a constant external perturbation. The important case of an harmonic perturbation is presented here. Detailed expressions are derived for diagonal and non-diagonal elements of the response, and the trace conservation theorem is shown to be satisfied at all time and any order. The results are again applied to the simple illustrative quantum system considered in I.

### 1. Introduction

A lot of effort has been devoted, in the last two decades, to the resolution of the time-dependent Schrödinger equation in the case of harmonically excited quantum systems. The relevance of these investigations is easily understood because of their direct connection with the fundamental problem of the interaction of matter and radiation in the semi-classical scheme.

Since the early work of Shirley (1965) which related the periodic Hamiltonian to a static one represented by an infinite matrix, and several more recent extensions (see Milfeld and Wyatt 1983 for a review and references) various solutions have been proposed to improve the perturbation theory (Sambe 1973, Sen Gupta 1973), the propagator (Salzmann 1974), or the mean energy calculation (Kelemen 1979), in the case of oscillatory Hamiltonians. None of them, however, lead directly to tractable expressions of the density matrix. In the previous paper (Fortini 1983, hereafter referred to as I) the determinantal formalism initially built up in the evolution operator problem (Fortini 1981), has been extended to the time-dependent Schrödinger equation of the density matrix when the external perturbation is constant. The purpose of this second part is to work out a further extension to the case of an harmonic external perturbation.

Following Shirley (1965), by means of the introduction of the so-called 'Floquet states', the determinantal expressions of the evolution operator pertaining to an oscillating excitation was already shown to reduce to the case of constant one (Fortini 1981). The same is true in the density matrix formalism, although the relevant Schrödinger equation, the related kernel, etc. . . . are rather more involved. Thanks to this property the harmonic case will not cause any extra formal difficulty, so that we shall only have to choose a suitable set of notations (§ 2) and to proceed to an appropriate extension of the most important equations (§ 3). Our illustrative simple system will be, once more, reconsidered as an application in § 4.

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## 2. Integral equation of the density matrix and Fredholm–Laplace solution

The Hamiltonian of the physical system will be written as

$$H = H_0 + V + (A e^{-i\omega t} + A^+ e^{i\omega t}) Y(t) \quad (2.1)$$

where  $H_0$  denotes an unperturbed Hamiltonian of eigenkets  $|k\rangle$  and eigenvalues  $\varepsilon_k = \hbar\omega_k$ .  $V$  is a collision potential and the constant operator  $A$  represents the amplitude of the external harmonic perturbation, of circular frequency  $\omega$ , assumed to be applied from  $t=0$ , as stated by the Heaviside step function  $Y(t)$ .

Let us rewrite, for clarity, the equation defining the density matrix  $\rho(t)$  from the evolution operator  $U(t)$

$$\rho(t) = U(t)\rho(0)U^+(t), \quad (2.2)$$

and the time-dependent Schrödinger equation

$$d\rho/dt = (i\hbar)^{-1}[H, \rho(t)]. \quad (2.3)$$

The Laplace transform of  $\rho(t)$

$$R(\nu) = \int_0^\infty e^{-\nu t} \rho(t) dt \quad \text{or} \quad R(\nu) \subset \rho(t),$$

will again be introduced, making use of the elementary properties

$$\begin{aligned} d\rho/dt \supset \nu R(\nu) - \rho(0) \\ \exp(\pm i\omega t)\rho(t) \supset R(\nu \mp i\omega). \end{aligned} \quad (2.4)$$

Equation (2.3) thus becomes

$$\begin{aligned} \nu R(\nu) = \rho(0) + (i\hbar)^{-1}[H_0 + V, R(\nu)] + (i\hbar)^{-1}[A, R(\nu + i\omega)] \\ + (i\hbar)^{-1}[A^+, R(\nu - i\omega)]. \end{aligned} \quad (2.5)$$

Taking this equation between the bra  $\langle a|$  and the ket  $|b\rangle$  gives

$$\begin{aligned} \nu R_b^c(\nu) = \rho_b^c(0) - i\omega_{cb} R_b^c(\nu) - i\hbar^{-1}[V_k^c R_b^k(\nu) - R_k^c(\nu) V_b^k] \\ - i\hbar^{-1}[A_k^c R_b^k(\nu + i\omega) - R_k^c(\nu + i\omega) A_b^k] \\ - i\hbar^{-1}[A_k^{+c} R_b^k(\nu - i\omega) - R_k^c(\nu - i\omega) A_b^{+k}], \end{aligned} \quad (2.6)$$

where  $\omega_{cb} = \omega_c - \omega_b$ ,  $R_b^c(\nu) = \langle c|R(\nu)|b\rangle$ . In the following,  $\rho_b^c$  will stand for  $\rho_b^c(0)$  as long as no confusion may occur with  $\rho_b^c(t)$ , and summations over repeated indices will be implicit, as usual.

In Fortini (1981) the linear system was constructed whose complete solution is represented by the ‘column vector’ defined by the following  $bn$  components

$$F_a^b(\nu + in\omega) = F_a^{bn}(\nu), \quad (2.7)$$

where  $n$  is an integer and  $a$  the starting state of the physical system.  $F^{bn}$  was defined as a vector belonging to the Hilbert space  $\mathcal{E}_{H_0}$  sustained by the eigenstates of  $H_0$ , and extended so as to include all the vectors deduced from any  $\nu$ -dependent vector by the  $in\omega$  translations.  $R(\nu)$  will now be regarded as a vector belonging to the tensorial product of this extended representative space by its own dual, i.e.

$$\mathcal{E}_{H_0 H_0} = \mathcal{E}_{H_0} \otimes \mathcal{E}_{H_0}^*$$

and, as such, it will be noted  $\mathbf{R}(\nu)$ . Its *cnbm* components will be denoted  $\mathbf{R}^{cn}_{bm}(\nu)$ . Let us assume, for simplicity, that  $\rho(t)$  is diagonal at  $t=0$ . From equation (2.2), the *cnbm* component of  $\mathbf{R}(\nu)$  is defined by the following convolution integration

$$\mathbf{R}^{cn}_{bm}(\nu) = [F_a^c(\nu + i n \omega) * F_b^{+a}(\nu - i m \omega)] \rho_a^a(0),$$

or

$$\mathbf{R}^{cn}_{bm}(\nu) = [F_a^{cn}(\nu) * F_b^{+a}(\nu)] \rho_a^a(0). \tag{2.8}$$

$F_b^{+a}$  is a row-vector component such that

$$F_b^{+a}(\nu) = F_b^{+a}(\nu - i m \omega) = [F_a^{bm}(\nu^*)]^*$$

in accordance with both the definition of the complex conjugate of the Laplace transform of a given function and the conjugate of a given matrix. As defined by equation (2.8),  $\mathbf{R}^{cn}_{bm}$  is only dependent on the difference  $n - m$ . On returning to the original, and using elementary properties of the Laplace transform, we have

$$U_a^{cn}(t) = e^{-i n \omega t} U_a^{co}(t), \quad U_{bm}^{+a}(t) = e^{i m \omega t} U_{bo}^{+a}(t),$$

whence

$$\begin{aligned} \rho_{bm}^{cn}(t) &= U_a^{cn}(t) U_{bm}^{+a}(t) \rho_a^a(0) \\ &= e^{-i(n-m)\omega t} U_a^{co}(t) U_{bo}^{+a}(t) \rho_a^a(0), \end{aligned} \tag{2.9}$$

and the subsequent Laplace transform

$$\mathbf{R}^{cn}_{bm}(\nu) = \mathbf{R}^{co}_{bo}(\nu + i \omega_{nm}) \tag{2.10}$$

with  $\omega_{nm} = (n - m)\omega$ . Thus we may write

$$\mathbf{R}^{cn}_{bm}(\nu) = \mathbf{R}^{cn-m}_{bo}(\nu) = \mathbf{R}^{co}_{bm-n}(\nu), \tag{2.11}$$

so that the  $\mathbf{R}$ -components are labelled by the set of quantum numbers *cb* and one integer only.

Equation (2.14*b*) in I again holds

$$(\mathbf{I} + \mathbf{d}^{-1} \mathbf{K}) |\mathbf{R}\rangle = \mathbf{d}^{-1} |\rho\rangle \tag{2.12}$$

and is equivalent to the system

$$\mathbf{R}^{cn}_{bm} + \frac{\mathbf{K}_{c_1 n_1 b_1 m_1}^{cn \ b_1 m_1}}{\mathbf{d}_{cnbm}} \mathbf{R}^{c_1 n_1 b_1 m_1} = \frac{\rho_{b_1 m_1}^{c_1 n}}{\mathbf{d}_{cnbm}} \tag{2.13}$$

$1_m^n$  stands for a column vector, all components of which are equal to 1 in the relevant *nm* subspace. Matrix elements of the commutation kernel  $\mathbf{K}$  are derived from those of the kernel  $K$  of the  $F$ -equation as

$$\langle c_2 n_2 b_2 m_2 | \mathbf{K} | c_1 n_1 b_1 m_1 \rangle = \mathbf{K}_{c_1 n_1 b_2 m_2}^{c_2 n_2 b_1 m_1} = \mathbf{K}_{c_1 n_1}^{c_2 n_2} \delta_{b_2 m_2}^{b_1 m_1} - \delta_{c_1 n_1}^{c_2 n_2} \mathbf{K}_{b_2 m_2}^{b_1 m_1}, \tag{2.14}$$

with

$$\mathbf{K}_{c_1 n_1}^{c_2 n_2} = i \hbar^{-1} (V_{c_1}^{c_2} \delta_{n_1}^{n_2} + A_{c_1}^{c_2} \delta_{n_1}^{n_2+1} + A_{c_1}^{+c_2} \delta_{n_1}^{n_2-1}). \tag{2.15}$$

Since  $\rho(0)$  does not depend on  $n, m$ , the components of the vector  $|\rho(0)\rangle$  in the *nm* subspace are all equal to 1. This is recalled in equation (2.13) by the  $1_m^n$ , for notational consistency.  $\mathbf{d}_{cnbm}$  denotes a current eigenvalue of the following diagonal operator in  $\mathcal{E}_{H_0 H_0}$

$$\mathbf{d} = \nu \mathbf{1} + i \hbar^{-1} \mathbf{H}_0 + i \omega \left[ \left( \sum_n n P_n \right) \delta - \delta \left( \sum_m m P_m \right) \right] \tag{2.16}$$

where  $\mathbf{1}$  and  $H_0$  have the same meaning as in I, and  $P_n, P_m$ , denote the projectors on  $n, m$  states. The system (2.13) represents the set of difference equations derived from equation (2.6) in all substitutions  $\nu \rightarrow \nu + i n \omega$ .

Since the kernel matrix elements (2.14) are obviously dependent on  $n_1 - n_2, m_1 - m_2$  only, it can be easily verified that if  $R^{cn}_{bm}$  is a solution of the system (2.13), the same is true for  $R^{cn+p}_{bm+p}$  in accordance with the property (2.10). The point is that, in applications,  $n, m$  refer to integer numbers of quanta in the field, whose variations we are concerned with in the transitions under consideration. We thus can take  $m = 0$  in equation (2.13). Furthermore, as in I, the hermiticity of the kernel  $iK$  which writes

$$(K^{c_2 n_2}_{c_1 n_1})^* = -K^{c_1 n_1}_{c_2 n_2}$$

entails the hermiticity of the kernel  $iK$  in the form

$$(K^{c_2 n_2}_{c_1 n_1} b_1 m_1)^* = -K^{c_1 n_1}_{c_2 n_2} b_2 m_2 = K^{b_2 m_2}_{b_1 m_1} c_1 n_1 \tag{2.17}$$

whereas the trace property is simply expressed as

$$\text{Tr}[K, 0] = \sum_b \langle b n b m | K | 0 \rangle = 0 \tag{2.18}$$

for any operator 0.

The Fredholm solution of the system (2.13) can then be written as

$$R^{cn}_{bo} = (D^{cn}_{c_1 n_1, b_1 m_1} / d_{c_1 n_1, b_1 m_1} D) \rho^{c_1 n_1}_{b_1 m_1} \tag{2.19}$$

where  $D$  is the determinant of the system and  $D^{cn}_{c_1 n_1, b_1 m_1}$  the algebraic minor at the crossing of the  $c_1 n_1, b_1 m_1$ -row and the  $cnbo$ -column. In fact it is sufficient to calculate the solution  $R^{co}_{bo}$ , since on account of (2.10),

$$R^{cn}_{bo} = R^{co}_{bo}(\nu + i n \omega).$$

As resulting from the property (2.17), the conjugate of  $R^{cn}_{bo}$  is

$$(R^{cn}_{bo})^* = R^{bo}_{cn}$$

in accordance with the hermiticity of the  $R$  matrix in  $\mathcal{E}_{H_0}$ .

The expression (2.19) generalises the result (2.17) in I, by simply substituting the  $cnbm$  Floquet-states of the properly extended Hilbert space for the initial  $cb$  states. The harmonic perturbation case is thus reduced to the static one, and all the subsequent results of I can be easily rewritten, in the same way as in (2.19). This will be done for the most important of them in the next section.

### 3. Reduced determinantal expressions

#### 3.1. First reduced form

The system will be conveniently assumed in diagonal states, at  $t = 0$ . This assumption, however, is not essential. The foregoing results could be written for any set of initial states as well. The starting equation (3.3) in I is first extended for any  $anam$  initial state as follows

$$(I + d^{-1} K Q_{anam})^{-1} (I + d^{-1} K) |R\rangle = d^{-1} |anam\rangle \tag{3.1}$$

and leads to the following expressions of the  $R$ -components. For the non-diagonal

cobo-component

$$R^{co}_{bo} = -\frac{\langle cobo | d^{-1} K S_{anam}^{-1} | anam \rangle}{\nu + i\omega_{nm} + \langle anam | K S_{anam}^{-1} | anam \rangle} \rho_a^{a1n} \tag{3.2}$$

and for the diagonal bobo-component

$$R^{bo}_{bo} = \frac{\rho_b^{b1o}}{\nu + \langle bobo | K S_{bobo}^{-1} | bobo \rangle} - \frac{\langle bobo | d^{-1} K S_{bnbm}^{-1} | bnbm \rangle \rho_b^{b1n}}{\nu + i\omega_{nm} + \langle bnbm | K S_{bnbm}^{-1} | bnbm \rangle} - \frac{\langle bobo | d^{-1} K S_{anam}^{-1} | anam \rangle \rho_a^{a1n}}{\nu + i\omega_{nm} + \langle anam | K S_{anam}^{-1} | anam \rangle} \tag{3.3}$$

(anam ≠ bobo), with

$$S_{anam} = I + Q_{anam} d^{-1} K. \tag{3.4}$$

In (3.3), the contributions in  $\rho_b^{b1o}$  and  $\rho_b^{b1n}$  ( $nm \neq 00$ ) are separated out because of their particular mathematical form.

Again, these expressions can be obtained by means of a systematic ‘reduction’ procedure of the related Fredholm expressions derived from (2.19)

$$R^{co}_{bo} = \frac{D_{anbo}^{co\ am} (D_{anam}^{an\ am})^{-1}}{(\nu + i\omega_{nm}) D (D_{anam}^{an\ am})^{-1}} \rho_a^{a1n} \tag{3.5}$$

and

$$R^{bo}_{bo} = \frac{\rho_b^{b1o}}{\nu D (D_{bobo}^{bo\ bo})^{-1}} + \frac{D_{bnbo}^{bo\ bm} (D_{bnbm}^{bn\ bm})^{-1}}{(\nu + i\omega_{nm}) D (D_{bnbm}^{bn\ bm})^{-1}} \rho_b^{b1n} \tag{3.6}$$

$$+ \frac{D_{anbo}^{bo\ am} (D_{anam}^{an\ am})^{-1}}{(\nu + i\omega_{nm}) D (D_{anam}^{an\ am})^{-1}} \rho_a^{a1n} \tag{3.6}$$

(a ≠ b).

### 3.2. Second reduced form

We now proceed to work out the second reduction stage by solving X systems of the type

$$S_{anam} | X \rangle = d^{-1} K | anam \rangle = d_{k pam}^{-1} K_{an}^{kp} | k pam \rangle - d_{ankp}^{-1} K_{kp}^{am} | ank p \rangle \tag{3.7}$$

whence

$$X^{co}_{bo} = \frac{D_{kpbo}^{co\ am}}{d_{k pam} D} K_{an}^{kp} - \frac{D_{anbo}^{co\ kp}}{d_{ankp} D} K_{kp}^{am}, \tag{3.8}$$

and similar expressions for the elements  $X^{an}_{am}$ ,  $X^{bo}_{bo}$  arising in (3.2) and (3.3). The D and the relevant minors now pertain to the matrix (3.4).  $X^{co}_{bo}$  is next reduced as follows

$$X^{co}_{bo} = \frac{D_{kpbo}^{co\ am} (D_{k pam}^{kp\ am})^{-1}}{d_{k pam} D (D_{k pam}^{kp\ am})^{-1}} K_{an}^{kp} - \frac{D_{anbo}^{co\ kp} (D_{ankp}^{an\ kp})^{-1}}{d_{ankp} D (D_{ankp}^{an\ kp})^{-1}} K_{kp}^{am}. \tag{3.9}$$

In a more explicit form

$$X^{co}_{bo} = -\frac{\langle cobo | d^{-1} K S_{anamk pam}^{-1} | k pam \rangle K_{an}^{kp}}{d_{k pam} + \langle k pam | K S_{anamk pam}^{-1} | k pam \rangle} + \frac{\langle cobo | d^{-1} K S_{anamk p}^{-1} | ank p \rangle K_{kp}^{am}}{d_{ankp} + \langle ank p | K S_{anamk p}^{-1} | ank p \rangle} \tag{3.10}$$

with

$$S_{anamankp} = I + Q_{anam}Q_{ankp}d^{-1}K, \text{ etc. . . . .} \tag{3.11}$$

The second reduced form will be finally obtained by using the above expressions of the  $X$  matrix elements in (3.2) and (3.3). The initial state of the relevant equation (3.7) is recalled in a bracket

$$R^{co}_{bo} = -\frac{X^{co}_{bo}(anam)\rho_a^a 1_m^n}{(\nu + i\omega_{nm})[1 + X^{an}_{am}(anam)]}, \tag{3.12}$$

$$R^{bo}_{bo} = \frac{\rho_b^b 1_o^o}{\nu[1 + X^{bo}_{bo}(bobo)]} - \frac{X^{bo}_{bo}(bnbm)\rho_b^b 1_m^n}{(\nu + i\omega_{nm})[1 + X^{bn}_{bm}(bnbm)]} \quad (nm \neq 00) \\ - \frac{X^{bo}_{bo}(anam)\rho_a^a 1_m^n}{(\nu + i\omega_{nm})[1 + X^{an}_{am}(anam)]} \quad (b \neq a). \tag{3.13}$$

### 3.3. Trace conservation

In spite of the similarity with the results of I, it is worthwhile reconsidering the trace conservation theorem with regard to the complications arising from the extra  $n, m$  quantum numbers in the representative space. Confining ourselves to the two times reduced expressions, which are of the most interest in applications, we start with equation (3.13)

$$\text{Tr } R = \sum_b R^{bo}_{bo} \\ = \sum_b \frac{\rho_b^b 1_o^o}{\nu[1 + X^{bo}_{bo}(bobo)]} - \sum_{\substack{b \\ nm \neq 00}} \frac{X^{bo}_{bo}(bnbm)\rho_b^b 1_m^n}{(\nu + i\omega_{nm})[1 + X^{bn}_{bm}(bnbm)]} \\ - \sum_{b \neq a} \frac{X^{bo}_{bo}(anam)\rho_a^a 1_m^n}{(\nu + i\omega_{nm})[1 + X^{an}_{am}(anam)]}. \tag{3.14}$$

Consider the third term on the right-hand side.  $X^{bo}_{bo}(anam)$  is deduced from (3.9) and (3.10) by taking  $c = b$ . Summing over all states, except  $a$ , first gives, in the numerators

$$\sum_{b \neq a} D^{bo}_{kp\ bo} (D^{kp\ am}_{kp\ am})^{-1} \\ = - \sum_{b \neq a} \langle bobo | d^{-1} K S^{-1}_{anamk\ pam} | k\ pam \rangle \\ = \frac{1}{\nu} \langle aoao | d^{-1} K S^{-1}_{anamk\ pam} | k\ pam \rangle, \tag{3.15}$$

on account of the property (2.18). Using this result in the third term on the right-hand side of (3.14), leads to

$$\sum_b R^{bo}_{bo} = \sum_b \frac{\rho_b^b 1_o^o}{\nu[1 + X^{bo}_{bo}(bobo)]} - \sum_{\substack{b \\ nm \neq 00}} \frac{X^{bo}_{bo}(bnbm)\rho_b^b 1_m^n}{(\nu + i\omega_{nm})[1 + X^{bn}_{bm}(bnbm)]} \\ + \sum_a \frac{X^{ao}_{ao}(anam)\rho_a^a 1_m^n}{(\nu + i\omega_{nm})[1 + X^{an}_{am}(anam)]}.$$

All contributions  $n$  or  $m \neq 0$  cancel with each other in the second and the third term

on the right. Detailing the remaining terms finally gives

$$\sum_b R^{bo}_{bo} = \sum_b \rho_b^b 1_o^o / \nu - \frac{\langle bobo | \mathbf{KS}_{bobolpbo}^{-1} | lpbo \rangle K_{bo}^{lp}}{d_{lpbo} + \langle lpbo | \mathbf{KS}_{bobolpbo}^{-1} | lpbo \rangle} + \text{CC}$$

$$- \sum_{a \neq b} \frac{\langle aoao | \mathbf{KS}_{aoakpao}^{-1} | kpao \rangle K_{ao}^{kp} \rho_a^a 1_o^o}{d_{kpao} + \langle kpao | \mathbf{KS}_{aoakpao}^{-1} | kpao \rangle} + \text{CC}$$

$$\nu \left( \nu - \frac{\langle aoao | \mathbf{KS}_{aoaolqao}^{-1} | lqao \rangle K_{ao}^{lq}}{d_{lqao} + \langle lqao | \mathbf{KS}_{aoaolqao}^{-1} | lqao \rangle} + \text{CC} \right). \tag{3.16}$$

It is now quite visible that, by simply associating these two summations, the trace conservation theorem is recovered as

$$\text{Tr } R = \sum_b \frac{\rho_b^b 1_o^o}{\nu} = \frac{1}{\nu}, \tag{3.17}$$

and remains true at any order of the kernel  $\mathbf{K}$ .

#### 4. Application to a simple model

We will, once more, consider the simplified physical system defined in previous publications (Fortini 1981, 1983). The external potential  $A(t)$  is now taken of the form

$$A(t) = A \exp(-i\omega t) + A^+ \exp(i\omega t), \tag{4.1}$$

and induces transitions from an initial state  $a$  toward a continuum of states  $b, c, \dots$  lying around energies  $\hbar\omega_a \pm \hbar\omega$ . Through direct application of formulae (3.13) and (3.10), we first found that in the occupation probability of the  $a$ -state, only the first term on the right-hand side of (3.13) remains. Using equation (A3) the relevant matrix element  $X^{bo}_{bo}(bob_o)$  is shown to involve two types of contributions, depending on whether the transition under consideration relates to an absorption or an emission process, i.e. on whether the continuum energies  $\omega_b, \omega_c, \dots, \omega_k, \dots$  are larger or smaller than  $\omega_a$ . Assuming an absorption process for definiteness, we finally obtain the  $aa$ -matrix element of  $\mathbf{R}$  as follows

$$R^{ao}_{ao} = \left\{ \nu + \frac{|A_a^k|^2}{\hbar^2} \left[ \frac{1}{\nu + i(\omega_{ka} - \omega) + |A_a^l|^2/\hbar^2(\nu + i\omega_{kl})} + \frac{1}{\nu + i(\omega_{ak} + \omega) + |A_a^l|^2/\hbar^2(\nu + i\omega_{lk})} \right] \right\}^{-1}. \tag{4.2}$$

The terms in the square brackets are conjugate to each other.

The current matrix element between excited states  $b, c$  is next calculated, starting with (3.12). Making use of (A2) and (A3), we readily obtain, in the absorption case

$$R^{co}_{bo} = \frac{A_a^c A_b^{+a}}{\hbar^2(\nu + i\omega_{cb})}$$

$$\times \frac{[\nu + i(\omega_{ca} - \omega) + \hbar^{-2}|A_a^l|^2/(\nu + i\omega_{cl})]^{-1} + [\nu + i(\omega_{ab} + \omega) + \hbar^{-2}|A_a^l|^2/(\nu + i\omega_{lb})]^{-1}}{\nu + \frac{|A_a^l|^2}{\hbar^2} \left[ \frac{1}{\nu + i(\omega_{ka} - \omega) + \hbar^{-2}|A_a^l|^2/(\nu + i\omega_{kl})} + \frac{1}{\nu + i(\omega_{ak} + \omega) + \hbar^{-2}|A_a^l|^2/(\nu + i\omega_{lk})} \right]}. \tag{4.3}$$



Setting  $c = b$ , then summing over the final continuum, it is straightforward to see that

$$\mathbf{R}^{ao} + \sum_{b \neq a} \mathbf{R}^{bo} = \frac{1}{\nu},$$

which simply expresses the trace conservation theorem in the present model. We notice, in addition, that there exists a long-term distribution in the excited states, which is defined by

$$\rho_b^b(t \rightarrow \infty) = \lim_{\nu \rightarrow +0} [\nu \mathbf{R}^{bo}(\nu)]$$

whereas the initial state becomes empty in that limit

$$\rho_a^a(t \rightarrow \infty) = \lim_{\nu \rightarrow +0} [\nu \mathbf{R}^{ao}(\nu)] = 0.$$

The physical meaning of these results is evident.

The broadening terms in expressions (4.2) and (4.3) involve summations over dummy indices, along the cut defined by the spectrum of the system, as explained in I. The actual range of integration, however, is more restricted, in the vicinity of  $\gamma \sim 0$ , consistent with energy conservation to within the uncertainty relation. Through integration over  $k$  (or  $l$ ), resonant  $1/d$  terms lead to complex roots in the denominators, confined close to the origin. Non-resonant  $1/d$  terms (which have been deleted) should yield negligible additional contributions to these roots. Going on to calculations in the same simplifying assumptions as in I, we have typically for the broadening terms

$$\sum_l \frac{\hbar^{-2} |A_a^l|^2}{\nu + i\omega_{kl}} \cong \frac{|A|^2 \theta_k}{\hbar^2} \ln \frac{\nu + i\omega_{k_2}}{\nu + i\omega_{k_1}} = i\beta_k \pm \gamma_k,$$

then

$$\begin{aligned} \sum_k \frac{\hbar^{-2} |A_a^k|^2}{\nu + i(\omega_{ka} - \omega + \beta_k) \pm \gamma_k} &\cong \frac{|A|^2 \theta_a}{\hbar^2} \ln \frac{\nu + i(\omega_{2a} - \omega + \beta_k) \pm \gamma_k}{\nu + i(\omega_{2a} - \omega + \beta_k) \pm \gamma_k} \\ &\cong -i\beta_a + \gamma_a. \end{aligned}$$

All the broadening parameters have nearly the same value. Using these results, equations (4.2) and (4.3) simplify as follows

$$R_a^a(i\eta) = \frac{1}{i\eta \pm 2\gamma_a}$$

$$R_b^c(i\eta) = \frac{\hbar^{-2} A_a^c A_b^{+a}}{i(\eta + \omega_{cb})(\eta + 2\gamma_a)} \left( \frac{1}{i(\eta + \omega_{ca} - \omega + \beta_a) \pm \gamma_a} + \frac{1}{i(\eta + \omega_{ab} + \omega - \beta_a) \pm \gamma_a} \right)$$

valid on each side of the cut. Except for the  $\omega$  translation, these formulae are the same as in I. The resulting original functions give

$$\rho_a^a(t) = e^{-2\gamma_a t}$$

$$\rho_b^c(t) = \frac{A_a^c A_b^{+a}}{\hbar^2} e^{-i\omega_{cb} t} \frac{[e^{i(\omega_{ca} - \omega + \beta_a)t - \gamma_a t} - 1][e^{i(\omega_{ab} + \omega - \beta_a)t - \gamma_a t} - 1]}{[i(\omega_{ca} - \omega + \beta_a) - \gamma_a][i(\omega_{ab} + \omega - \beta_a) - \gamma_a]}$$

in agreement with the trace conservation theorem, as it could be easily verified.

**Appendix. Calculation of X matrix elements in the simple model**

To second order in  $K$

$$X_{bo}^{co}(anam) = -\frac{d_{cobo}^{-1} K_{kp\ bo}^{co\ am} K_{an}^{kp}}{d_{kpam} - \langle kpam | K d^{-1} K | kpam \rangle} + \frac{d_{cobo}^{-1} K_{an\ bo}^{co\ kp} K_{kp}^{am}}{d_{ankp} - \langle ankp | K d^{-1} K | ankp \rangle}. \tag{A1}$$

Writing the first numerator on the right-hand side in detail gives

$$\begin{aligned} & -d_{cobo}^{-1} K_{kp\ bo}^{co\ am} K_{an}^{kp} \\ &= -d_{cobo}^{-1} (i\hbar^{-1})^2 [(A_k^c \delta_o^{o+1} + A_k^{+c} \delta_o^{o-1}) \delta_{bo}^{am} \\ &\quad - \delta_{kp}^{co} (A_b^a \delta_o^{m+1} + A_b^{+a} \delta_o^{m-1})] (A_a^k \delta_n^{p+1} + A_a^{+k} \delta_n^{p-1}) \\ &= \frac{\delta_{kp}^{co}}{\hbar^2 d_{cobo}} (A_b^a A_c^{+c} \delta_o^{m+1} \delta_n^{-1} + A_b^{+a} A_a^c \delta_o^{m-1} \delta_n^1), \end{aligned}$$

where terms which entail  $\omega_{nm} = 2\omega$ , giving negligible contributions near the resonance, have been dropped. Similarly

$$d_{cobo}^{-1} K_{an\ bo}^{co\ kp} K_{kp}^{am} = -\frac{\delta_{bo}^{kp}}{d_{cobo}} (A_a^c A_b^{+a} \delta_n^1 \delta_o^{m-1} + A_a^{+c} A_b^a \delta_n^{-1} \delta_o^{m+1}).$$

We thus obtain four contributions in which we have either  $kp = co$  or  $bo$  with  $m = n = \pm 1$ , and the denominators in (A1) are to be calculated accordingly. We will find for instance

$$\langle coa1 | K d^{-1} K | coa1 \rangle = -\frac{|A_c^1|^2}{\hbar^2} \left( \frac{1}{d_{l1a1}} + \frac{1}{d_{l-1a1}} \right) - \frac{|A_a^1|^2}{\hbar^2} \left( \frac{1}{d_{colo}} + \frac{1}{d_{coll}} \right).$$

On the right-hand side, the third term which is resonant will be retained only. Carrying out analogously the other similar calculations, we finally obtain

$$\begin{aligned} X_{bo}^{co}(anam) = & \frac{1}{\hbar^2 d_{cobo}} \left( \frac{A_a^{+c} A_b^a \delta_m^{-1} \delta_n^{-1}}{d_{coa-1} + |A_a^1|^2 / \hbar^2 d_{colo}} + \frac{A_a^c A_b^{+a} \delta_m^1 \delta_n^1}{d_{coa1} + |A_a^1|^2 / \hbar^2 d_{colo}} \right. \\ & \left. + \frac{A_a^c A_b^{+a} \delta_m^1 \delta_n^1}{d_{a1bo} + |A_a^1|^2 / \hbar^2 d_{lobo}} + \frac{A_2^{+c} A_b^2 \delta_m^{-1} \delta_n^{-1}}{d_{a-1bo} + |A_a^1|^2 / \hbar^2 d_{lobo}} \right). \tag{A2} \end{aligned}$$

It is readily recognised that either only the terms  $m = n = 1$  or  $m = n = -1$  remain, depending on whether the continuum  $b, c, \dots k, \dots$  lies above or below the initial state  $a$ . In the former case the transition relates to an absorption process, and in the latter case to an emission process.

We now turn to consider matrix elements of the form  $X_{am}^{an}(anam)$  which also arise in (3.12) and (3.13). Through quite similar calculations and deletion of non-resonant terms, we obtain

$$\begin{aligned} X_{am}^{an}(anam) = & \frac{|A_a^k|^2 \delta_m^n}{\nu \hbar^2} \left[ \left( d_{km+1am} + \frac{|A_a^l|^2}{\hbar^2 d_{km+1lm+1}} \right)^{-1} + \left( d_{km+1am} + \frac{|A_a^l|^2}{\hbar^2 d_{km+1lm+1}} \right)^{-1} \right. \\ & \left. + \left( d_{ankn-1} + \frac{|A_a^l|^2}{\hbar^2 d_{ln-1kn-1}} \right)^{-1} + \left( d_{ankn+1} + \frac{|A_a^l|^2}{\hbar^2 d_{ln+1kn+1}} \right)^{-1} \right] \tag{A3} \end{aligned}$$

where absorption and emission contributions will be separated again.

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