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Time-dependent perturbation treatment of independent Raman schemes

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Abstract
The problem of a trapped ion subjected to the action of two or more independent Raman schemes is analysed through a suitable time-dependent perturbative approach based on the factorization of the evolution operator in terms of other unitary operators. We show that the dynamics of the system may be traced back to an effective Hamiltonian up to a suitable dressing. Moreover, we give the method to write the master equation corresponding to the case wherein spontaneous decays occur.

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1. Introduction

Over the last few decades, trapped ions have been exploited to realize interesting applications in the field of quantum computing [1] and teleportation [2], and to test fundamental aspects of quantum mechanics through the generation of nonclassical states [3].

The confinement of an atomic ion in a Paul trap is achieved by the action of a quadrupolar time-dependent electric field that gives rise to a complicate motion which, in correspondence to the stability regimes of the trap, is separable into a secular component and a micromotion. The latter is a small and fast oscillation, while the former perfectly matches the motion of a harmonic oscillator [4–6]. In several applications, a trapped ion can be treated as a finite-level system, associated with the 'relevant electronic levels' of the ion, coupled with a bosonic system describing the degrees of freedom related to the centre-of-mass motion (considered as the motion of a harmonic oscillator) [7, 8]. In the simplest case, the finite-level system is a *two-level* system. For instance, in experiments using a $^9Be^+$ ion such two levels are hyperfine sub-levels of the ion ground state [7].

Driving a trapped ion by means of suitable laser beams, it is possible to induce ‘vibronic’ couplings—involving both vibrational (bosonic) and internal (fermionic) degrees of freedom—giving rise to interesting dynamical effects and to the possibility of applications in quantum technology [9–12]. In many relevant applications, the vibronic transitions involving a couple of electronic levels—that, keeping in mind applications to quantum computing, will be called ‘qubit levels’ in the following—are induced via Raman couplings, i.e. the two $^9Be^+$ hyperfine sub-levels, for instance, are not directly coupled by a single resonant laser field; instead, a third (‘auxiliary’) level, non-resonantly coupled by two laser fields with the qubit levels, is exploited. The two associated detunings are fixed in such a way that, even if transitions from/to the qubit levels to/from the auxiliary one ‘violate energy conservation’, second-order processes involving the two qubit levels take place. Such a coupling scheme is usually called ‘Λ Raman coupling scheme’.

The analysis of a trapped ion *single* Raman scheme dynamics has been carried out by means of the method called ‘adiabatic elimination’ [13–15], widely used in different physical situations [16–20] and, in some sense, overcome by techniques based on unitary transformations which decouple the auxiliary level [21, 22]. Recently, a more systematic and transparent approach relying on a time-independent perturbative method [23–25] has been used. Indeed, by switching to a suitable *rotating frame*, one is able to eliminate the time dependence from the Hamiltonian. On the other hand, in the case where two Λ Raman couplings are simultaneously active, there is no elementary way for removing the time dependence from the Hamiltonian. Such a problem should not be regarded as an academical one. In fact, in contrast, it is of crucial importance in the implementation of some models aimed to realize suitable quantum state manipulations [13, 26].

Scope of the present contribution is to extend our previous work on (single) Raman schemes for investigating the dynamics of a trapped ion double Λ Raman scheme (the further extension to the case of $n \geqslant 3$ Λ-couplings is straightforward). In this case, it will be necessary to introduce a *time-dependent* perturbative approach based on a suitable decomposition of the time evolution operator of the system [27]. This new approach can be regarded as a non-trivial generalization of the time-independent approach previously introduced and allows us to study in a unique framework closely related models—such as the single and multiple Raman couplings—whose Hamiltonians, depending on the particular case, may or may not be treated by means of time-independent perturbative methods.

Our previous study of trapped ion single Raman schemes (see [23]) has put in evidence that, in addition to the well-known Rabi oscillations involving the qubit levels, also fast transitions between the qubit levels and the auxiliary one take place. Such transitions—that have been called *anomalous transitions*—although too fast to be revealed directly in ordinary experiments, in the case where the auxiliary level has a non-vanishing decay rate towards the other two levels, have the effect of gradually injecting decoherence in the coherent Rabi dynamics involving the qubit levels. This phenomenon has actually been observed experimentally [12]. In the present paper, we will show that an analogous situation occurs in the case of a double Λ-coupling.

We will also show that the behaviour of a trapped ion Raman scheme can be described in a way somewhat similar to the ordinary interaction picture. Precisely, one can assume that the outcome of an experimentally observed quantity $\mathcal{O}$—to which is associated an abstract quantum observable O (i.e. a self-adjoint operator)—at a certain time t is given by

$$\mathcal{O}(t) = \mathrm{tr}(\bar{O}\rho_{\mathrm{eff}}(t)),$$

where $\rho_{\mathrm{eff}}(t) = T_{\mathrm{eff}}(t)\rho(0)T_{\mathrm{eff}}(t)^{\dagger}$ is the density matrix of the system subject to an effective evolution determined by $T_{\mathrm{eff}}(t)$ (corresponding to the evolution obtainable by the procedure of ‘adiabatic elimination’) and $\bar{O}$ is a self-adjoint operator—a ‘coarse-grained observable’—which takes into account suitable corrections to the effective evolution and the additional effect of temporal coarse-graining of a real experimental setup, effect that can be reproduced by means of a time-averaging procedure. We will further show that the coarse-grained observable $\bar{O}$ can be obtained from the abstract observable O via a ‘coarse-graining superoperator’,

$$\widehat{\mathsf{CG}} : O \mapsto \bar{O}.$$

The paper is organized as follows. In the next section, we recall the result of the time-independent perturbation theory applied on the physical situation corresponding to a single trapped ion subjected to the action of a *single Raman scheme*. In section 3, we sketch a new approach on time-dependent perturbation theory based on the idea of factorizing the evolution operator in terms of *simpler* unitary operators. Since such an expansion is not unique, specific solutions are given up to a *gauge* condition. Therefore, we discuss such a point and bring to the light the connection between a specific gauge condition and the possibility of extracting effective Hamiltonians. Subsequently, in section 4, we analyse the physical problem of two Raman schemes simultaneously active applying the reported perturbation theory. The meaning and validity of the coarse-graining approach are discussed in section 4.2. Finally, in section 5, the case wherein the auxiliary level is a metastable one is considered and the relevant master equation is given.

2. Single Λ Raman scheme

The physical system that we will study in this section is a three-level harmonically trapped ion subjected to a single Λ Raman coupling scheme. Although this system can be obviously considered as a special case of the double Λ Raman scheme which will be studied in section 4, it is worth considering it here separately since, in this particular case, one can use a time-independent perturbative technique. This will also allow us to better illustrate the time-dependent perturbative method introduced in section 3.

The relevant Schrödinger picture Hamiltonian of the single Λ Raman coupling scheme is given by

$$\hat{H}_\Lambda(t) = \hat{H}_0 + \hat{H}_B + [\hbar g_{13}\, \mathrm{e}^{-\mathrm{i}(\vec{k}_{13}\cdot\vec{r}-\omega_{13}t)}\hat{\sigma}_{13} + \mathrm{h.c.}] + [\hbar g_{23}\, \mathrm{e}^{-\mathrm{i}(\vec{k}_{23}\cdot\vec{r}-\omega_{23}t)}\hat{\sigma}_{23} + \mathrm{h.c.}], \tag{1}$$

where

$$\hat{H}_0 = \sum_{l=1,2,3} \hbar\omega_l \hat{\sigma}_{ll}, \qquad \hat{H}_B = \hbar\nu \sum_{j=x,y,z} \hat{a}_j^\dagger \hat{a}_j^\dagger, \tag{2}$$

with $\hat{\sigma}_{lm} \equiv |l\rangle\,\langle m|$ (with $l, m = 1, 2, 3$), $\{|l\rangle\}$ being the considered three atomic levels and $\{\hbar\omega_l\}$ the corresponding energies; $\hat{a}_\alpha^\dagger$ ($\alpha = x, y, z$) is the annihilation operator related to the centre-of-mass harmonic motion along the direction α (we will denote the associated Fock basis by $\{\psi_n^\alpha\}$):

$$\hat{a}_x^\dagger = \left(\frac{\mu\nu}{2\hbar}\right)^{1/2}\left(\hat{x} + \frac{\mathrm{i}}{\mu\nu}\hat{p}_x^\dagger\right), \ldots, \hat{a}_z^\dagger = \left(\frac{\mu\nu}{2\hbar}\right)^{1/2}\left(\hat{z} + \frac{\mathrm{i}}{\mu\nu}\hat{p}_z^\dagger\right),$$

with μ denoting the mass of the ion. For simplicity, we have assumed to deal with a 3D degenerate parabolic trap with single frequency ν. The two laser fields responsible for the coupling terms are characterized by complex strengths (proportional to the laser amplitude and to the atomic dipole operator, and including the laser phases), wave vectors and frequencies $g_{13}, \vec{k}_{13}, \omega_{13}$ and $g_{23}, \vec{k}_{23}, \omega_{23}$, respectively. The *auxiliary level* $|3\rangle$ is assumed to be dipole-coupled to both the levels $|1\rangle$ and $|2\rangle$ via two far detuned lasers. Precisely, the two laser frequencies are chosen in such a way that

$$\Delta \equiv \omega_3 - \omega_1 - \omega_{13} = \omega_3 - \omega_2 - \omega_{23}, \tag{3}$$

where the detuning Δ satisfies the condition

$$|\Delta| \gg |g_{13}|, |g_{23}|, \nu. \tag{4}$$

A detailed analysis of such a model has been already performed in [23] by means of a suitable (time-independent) operator perturbative approach. In that paper, it is shown that, in addition to the standard Rabi oscillations between levels $|1\rangle$ and $|2\rangle$, also fast transitions coupling levels $|1\rangle$ and $|2\rangle$ with the auxiliary level $|3\rangle$—the so-called *anomalous transitions*—take place. Then, if the auxiliary level is an excited level with non-negligible decay rates towards levels $|1\rangle$ and $|2\rangle$, the fast transitions to the auxiliary level, composed with decays, gradually inject decoherence into the effective coherent cycle involving levels $|1\rangle$ and $|2\rangle$. This result is actually in agreement with experimental observations [12].

In the following, we resume the main results obtained in [23] and we put them in a form which is compatible with the time-dependent perturbative method which is used in the present paper. The first step of our treatment consists in passing to the interaction picture (usually named 'rotating frame') associated with the transformation

$$\hat{R}(t) = \mathrm{e}^{-\mathrm{i}\hat{A}t}, \tag{5}$$

where

$$\begin{aligned}\hat{A} &= (\omega_3 - \Delta)\,(\hat{\sigma}_{11} + \hat{\sigma}_{22} + \hat{\sigma}_{33}) - \omega_{13}\hat{\sigma}_{11} - \omega_{23}\hat{\sigma}_{22} \\ &= \omega_1\hat{\sigma}_{11} + \omega_2\hat{\sigma}_{22} + (\omega_3 - \Delta)\,\hat{\sigma}_{33},\end{aligned} \tag{6}$$

in such a way that the time-dependent Hamiltonian $\hat{H}_\Lambda$ is transformed into the following time-independent Hamiltonian:

$$\hat{\mathfrak{H}} := \hat{R}(t)^\dagger(\hat{H}_\Lambda(t) - \hat{A})\hat{R}(t) = \hbar\Delta\hat{H} = \hbar\Delta\big(\hat{\sigma}_{33} + \hat{H}_B^{(\Delta)} + \hat{H}_\updownarrow\big), \tag{7}$$

where $\hat{H}$ is a *dimensionless Hamiltonian* which is the sum of the three Hermitian operators, $\hat{H}_B^{(\Delta)}$ and $\hat{H}_\updownarrow$ being defined as

$$\begin{cases} \hat{H}_B^{(\Delta)} := \dfrac{\nu}{\Delta} \displaystyle\sum_{\alpha=x,y,z} \hat{a}_\alpha^\dagger \hat{a}_\alpha^\dagger, \\ \hat{H}_\updownarrow := \left[\dfrac{g_{13}}{\Delta}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{13}\cdot\vec{r}}\hat{\sigma}_{13} + \mathrm{h.c.}\right] + \left[\dfrac{g_{23}}{\Delta}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{23}\cdot\vec{r}}\hat{\sigma}_{23} + \mathrm{h.c.}\right]. \end{cases} \tag{8}$$

Considering the assumption given by the inequality (4), both $\hat{H}_B^{(\Delta)}$ and $\hat{H}_\updownarrow$ may be thought of as perturbations with respect to $\hat{\sigma}_{33}$. In fact, introducing the dimensionless perturbative parameter

$$\lambda := \frac{g}{\Delta}, \qquad g \equiv \max\{\nu, |g_{13}|, |g_{23}|\}, \tag{9}$$

both $\hat{H}_B^{(\Delta)}$ and $\hat{H}_\updownarrow$ are first-order perturbations in λ:

$$\hat{H} = \hat{H}(\lambda) = \hat{\sigma}_{33} + \lambda\varkappa \sum_{\alpha=x,y,z} \hat{a}_\alpha^\dagger\hat{a}_\alpha^\dagger + \lambda \sum_{j=1,2} [\varkappa_{j3}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{j3}\cdot\vec{r}}\hat{\sigma}_{j3} + \mathrm{h.c.}], \tag{10}$$

where $\varkappa \equiv \nu/g \leqslant 1$, $\varkappa_{j,3} \equiv g_{j,3}/g$, $|\varkappa_{j,3}| \leqslant 1$, and we note that, due to condition (4), $\lambda \ll 1$.

Our time-independent perturbative approach relies on a suitable canonical transformation $\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)}$ of the interaction picture Hamiltonian such that

$$\hbar\Delta\,\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)}\,\hat{H}(\lambda)\,\mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda)} = \hbar\Delta(\hat{\sigma}_{33} + \hat{C}(\lambda)), \tag{11}$$

where $\hat{C}(\lambda)$ and $\hat{Z}(\lambda)$ depend analytically on the perturbative parameter λ and $\hat{C}(\lambda)$ is a *constant of the motion* with respect to the unperturbed dynamics, i.e. $[\hat{\sigma}_{33}, \hat{C}(\lambda)] = 0$. This transformation allows us to give a very convenient decomposition of the evolution operator associated with the rotating frame Hamiltonian:

$$\exp\left(-\frac{\mathrm{i}}{\hbar}\hat{\mathfrak{H}}t\right) = \mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda)} \exp(\mathrm{i}\Delta\hat{\sigma}_{33}t) \exp(\mathrm{i}\Delta\hat{C}(\lambda)t)\, \mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)}. \tag{12}$$

At this point, truncating the power expansions

$$\hat{C}(\lambda) = \lambda\hat{C}_1 + \lambda^2\hat{C}_2 + \cdots + \lambda^n\hat{C}_n + \cdots, \qquad \hat{Z}(\lambda) = \lambda\hat{Z}_1 + \lambda^2\hat{Z}_2 + \cdots + \lambda^n\hat{Z}_n + \cdots$$

at a given perturbative order, one obtains by formula (12) useful expressions of the evolution operator. The operators $\hat{C}_1, \hat{C}_2, \ldots, \hat{Z}_1, \hat{Z}_2, \ldots$ are not uniquely determined and can be computed by a suitable iterative algebraic procedure. Fixing a certain *gauge* (namely, a specific solution corresponding to a certain condition discussed in [23]), one obtains the following results:

$$\lambda\hat{C}_1 = \hat{H}_B^{(\Delta)}, \tag{13}$$

$$\lambda^2\hat{C}_2 = -\frac{|g_{13}|^2}{\Delta^2}\hat{\sigma}_{11} - \frac{|g_{23}|^2}{\Delta^2}\hat{\sigma}_{22} + \frac{|g_{13}|^2+|g_{23}|^2}{\Delta^2}\hat{\sigma}_{33} - \left(\frac{g_{13}g_{32}}{\Delta^2}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{13}\cdot\vec{r}}\,\mathrm{e}^{\mathrm{i}\vec{k}_{23}\cdot\vec{r}}\hat{\sigma}_{12} + \mathrm{h.c.}\right), \tag{14}$$

where we have set $g_{3j} \equiv g_{j3}^*$, and

$$\lambda\hat{Z}_1 = \mathrm{i}\left(\frac{g_{13}}{\Delta}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{13}\cdot\vec{r}}\hat{\sigma}_{13} - \mathrm{h.c.}\right) + \mathrm{i}\left(\frac{g_{23}}{\Delta}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{23}\cdot\vec{r}}\hat{\sigma}_{23} - \mathrm{h.c.}\right), \tag{15}$$

$$\lambda^2\hat{Z}_2 = \frac{\nu}{\Delta}\left\{\left(\frac{g_{13}}{\Delta}\hat{X}_{13}\hat{\sigma}_{13} + \frac{g_{31}}{\Delta}\hat{X}_{31}\hat{\sigma}_{31}\right) + \left(\frac{g_{23}}{\Delta}\hat{X}_{23}\hat{\sigma}_{23} + \frac{g_{32}}{\Delta}\hat{X}_{32}\hat{\sigma}_{32}\right)\right\}, \tag{16}$$

where

$$\begin{cases} \hat{X}_{j3} := \mathrm{i}\left[\mathrm{e}^{-\mathrm{i}\vec{k}_{j3}\cdot\vec{r}}, \displaystyle\sum_{\alpha=x,y,z}\hat{a}_\alpha^\dagger\hat{a}_\alpha^\dagger\right], \\ \hat{X}_{3j} := \mathrm{i}\left[\mathrm{e}^{\mathrm{i}\vec{k}_{j3}\cdot\vec{r}}, \displaystyle\sum_{\alpha=x,y,z}\hat{a}_\alpha^\dagger\hat{a}_\alpha^\dagger\right] = \hat{X}_{j3}^\dagger, \end{cases}$$

with $j = 1, 2$.

The interpretation of this result leads to a very interesting fact. Indeed, once the unitary transformation $\mathrm{e}^{\mathrm{i}Z(\lambda)}$ has been applied to the interaction picture Hamiltonian $\hat{\mathfrak{H}}$ (recall equation (11)), the time evolution of the system is described, at the second order in the parameter λ, by the Hamiltonian

$$\hbar\Delta(\hat{\sigma}_{33} + \lambda\hat{C}_1 + \lambda^2\hat{C}_2) = \hat{\mathfrak{H}}_{12} + \hat{\mathfrak{H}}_3 := \hbar\Delta(\hat{H}_{12} + \hat{H}_3), \tag{17}$$

where, in order to display a more transparent formula, we set

$$\begin{aligned} \hat{\mathfrak{H}}_{12} \equiv \hbar\Delta\hat{H}_{12} &:= \hbar\nu\sum_{\alpha=x,y,z}\left(\hat{a}_\alpha^\dagger\hat{a}_\alpha^\dagger\right)\otimes(\hat{\sigma}_{11}+\hat{\sigma}_{22}) + \hbar\breve{\omega}_1\hat{\sigma}_{11} + \hbar\breve{\omega}_2\hat{\sigma}_{22} \\ &\quad + [\hbar g_{12}\,\mathrm{e}^{-\mathrm{i}\vec{k}_{12}\cdot\vec{r}}\hat{\sigma}_{12} + \mathrm{h.c.}], \end{aligned} \tag{18}$$

$$\hat{\mathfrak{H}}_3 \equiv \hbar\Delta\hat{H}_3 := \hbar\nu\sum_{\alpha=x,y,z}\left(\hat{a}_\alpha^\dagger\hat{a}_\alpha^\dagger\right)\otimes\hat{\sigma}_{33} + \hbar(\Delta+\breve{\omega}_3)\hat{\sigma}_{33}, \tag{19}$$

with

$$\breve{\omega}_j = -\frac{|g_{j3}|^2}{\Delta}, \qquad j = 1, 2, \qquad \breve{\omega}_3 = \frac{|g_{13}|^2+|g_{23}|^2}{\Delta},$$

$$g_{12} = \frac{g_{13}g_{32}}{\Delta}, \qquad \vec{k}_{12} = \vec{k}_{13} - \vec{k}_{23}.$$

Thus, the transformed Hamiltonian is the sum of two decoupled Hamiltonians $\tilde{H}_{12}$ and $\tilde{H}_3$, $[\tilde{H}_{12}, \tilde{H}_3] = 0$, 'living' respectively in the ranges of the orthogonal projectors $\hat{P}_g = \hat{\sigma}_{11} + \hat{\sigma}_{22}$, $\hat{P}_e = \hat{\sigma}_{33}$. It is worth noting that the Hamiltonian $\tilde{H}_{12}$ can be regarded as the rotating frame Hamiltonian of a trapped two-level ion in interaction with a laser field characterized by the following parameters:

$$\begin{cases} \omega_{12} \equiv \omega_{13} - \omega_{23} = \omega_2 - \omega_1, \\ \vec{k}_{12} \equiv \vec{k}_{13} - \vec{k}_{23}. \end{cases} \tag{20}$$

This effective coupling can be compared with the result found performing the adiabatic elimination of level $|3\rangle$ (see [13]). The question of what the *complete* dynamics of the system is now arises. First, it will be convenient to adopt the following notation. Given a couple of functions f and h of the perturbative parameter λ, if $f(\lambda) = h(\lambda) + O(\lambda^3)$, we will simply write

$$f(\lambda) \overset{\lambda^2}{\approx} h(\lambda).$$

Next, let us denote by $\hat{U}_\Lambda$ the evolution operator associated with the Raman scheme:

$$\mathrm{i}\hbar \left(\frac{\mathrm{d}}{\mathrm{d}t} \hat{U}_\Lambda \right)(t) = \hat{H}_\Lambda(t) \hat{U}_\Lambda(t), \; \hat{U}_\Lambda(0) = \mathrm{Id}. \tag{21}$$

Expressing $\hat{U}_\Lambda$ in terms of the evolution operator associated with the rotating frame Hamiltonian yields

$$\hat{U}_\Lambda(t) = \hat{R}(t) \exp\left(-\frac{\mathrm{i}}{\hbar} \hat{\mathfrak{H}} t \right). \tag{22}$$

Now, according to what we have previously shown, we have

$$\begin{aligned} \exp\left(-\frac{\mathrm{i}}{\hbar} \hat{\mathfrak{H}} t \right) &= \mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda)} \exp\left(-\mathrm{i}\Delta\, \mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)} \hat{H}(\lambda)\, \mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda)} t \right) \mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)} \\ &\overset{\lambda^2}{\approx} \mathrm{e}^{-\mathrm{i}(\lambda \hat{Z}_1 + \lambda^2 \hat{Z}_2)}\, \mathrm{e}^{-\mathrm{i}\Delta(\hat{\sigma}_{33} + \lambda \hat{C}_1 + \lambda^2 \hat{C}_2)t}\, \mathrm{e}^{\mathrm{i}(\lambda \hat{Z}_1 + \lambda^2 \hat{Z}_2)}, \end{aligned} \tag{23}$$

where we have truncated the power expansions of $\hat{Z}(\lambda)$ and $\hat{C}(\lambda)$ at the second order in λ; hence,

$$\hat{U}_\Lambda(t) \overset{\lambda^2}{\approx} \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}(\lambda \hat{Z}_1(t) + \lambda^2 \hat{Z}_2(t))}\, \mathrm{e}^{-\mathrm{i}\Delta(\lambda \hat{C}_1 + \lambda^2 \hat{C}_2)t}\, \mathrm{e}^{\mathrm{i}(\lambda \hat{Z}_1 + \lambda^2 \hat{Z}_2)}, \tag{24}$$

with $\hat{H}_0$ given by equation (2) and

$$Z_k(t) := \mathrm{e}^{\mathrm{i}\Delta\hat{\sigma}_{33} t} Z_k\, \mathrm{e}^{-\mathrm{i}\Delta\hat{\sigma}_{33} t}. \tag{25}$$

Observe that formula (24) suggests that one can consider a general perturbative decomposition of the evolution operator associated with a quantum system, decomposition which, as it will be shown in the next section, holds also in the case where the Hamiltonian of the system depends on time.

3. Time-dependent perturbative expansion of the evolution operator of a quantum system

Let us consider a time-dependent perturbed Hamiltonian $\hat{H}(\lambda; t)$, namely a self-adjoint linear operator of the form

$$\hat{H}(\lambda; t) = \hat{H}_0 + \hat{H}_\diamond(\lambda; t), \tag{26}$$

where $\hat{H}_0$ is a self-adjoint operator—the 'unperturbed component'—and $\hat{H}_\diamond(\lambda; t)$ is a time-dependent perturbation; precisely, we will assume that $\lambda \mapsto \hat{H}_\diamond(\lambda; t)$ is (for the perturbative parameter λ in a certain neighbourhood of zero and for any t) a real analytic, self-adjoint operator-valued function, with $\hat{H}_\diamond(0;t) = 0$. A real analytic function can be extended to a domain in the complex plane. Keeping this fact in mind, we will specify that a given property *holds for* λ *real*. For instance, the analytic function $\lambda \mapsto \hat{H}_\diamond(\lambda;t)$ will take values in the self-adjoint operators for λ real only.

Let $\hat{U}(\lambda;t)$ be the evolution operator associated with $\hat{H}(\lambda;t)$, with initial time $t_0 = 0$:

$$\mathrm{i}\dot{\hat{U}}(\lambda;t) = \hat{H}(\lambda;t)\hat{U}(\lambda;t), \qquad \hat{U}(\lambda;0) = \mathrm{Id}, \tag{27}$$

where the dot denotes the time derivative and we have set $\hbar = 1$. Then, we have that

$$\hat{U}(\lambda;t) = \hat{U}_0(t)T(\lambda;t), \tag{28}$$

where $\hat{U}_0(t)$ and $T(\lambda;t)$ are, respectively, the evolution operator associated with the unperturbed component $\hat{H}_0$ (evolution operator which is obviously given by $\mathrm{e}^{-\mathrm{i}\hat{H}_0 t}$) and the evolution operator associated with the interaction picture Hamiltonian

$$\tilde{H}(\lambda;t) := \hat{U}_0(t)^\dagger \hat{H}_\diamond(\lambda;t)\hat{U}_0(t). \tag{29}$$

Let us explicitly note that, since $\tilde{H}(0;t) = 0$, we have

$$T(0;t) = \mathrm{Id}. \tag{30}$$

We will suppose that the unperturbed evolution $\hat{U}_0(t)$ is explicitly known. Then the problem is to determine perturbative expressions of $T(\lambda;t)$. To this aim, the central point of the paper is to assume for $T(\lambda;t)$ the following general decomposition:

$$T(\lambda;t) = \exp(-\mathrm{i}Z(\lambda;t)) \exp\left(-\mathrm{i}\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right) \exp(\mathrm{i}Z(\lambda)), \tag{31}$$

where $(\lambda;t) \mapsto Z(\lambda;t), (\lambda;t) \mapsto C(\lambda;t)$ are operator-valued functions which depend analytically on the perturbative parameter λ and $Z(\lambda) \equiv Z(\lambda;0)$; in agreement with condition (30), we set

$$\hat{Z}(0;t) = 0, \qquad \hat{C}(0;t) = 0, \quad \forall t. \tag{32}$$

It will be seen that decomposition (31) has a wide range of solutions and that a possible choice for fixing a certain class of solutions is given by imposing the condition $\hat{C}(\lambda;t) = \hat{C}(\lambda)$, i.e. assuming that the function $(\lambda;t) \mapsto \hat{C}(\lambda;t)$ does not depend on time. This decomposition includes, as particular cases, two decompositions of the evolution operator that have been considered in the literature:

- the decomposition that is obtained setting $\hat{Z}(\lambda;t) = 0, \forall t$, in formula (31), decomposition which is at the root of the *Magnus expansion* of the evolution operator [28];
- the classical *Floquet decomposition* that holds in the case where the interaction picture Hamiltonian depends periodically on time (say with period T)—decomposition which is obtained setting $\hat{C}(\lambda;t) \equiv \hat{C}(\lambda)$, $\hat{Z}(\lambda;0) = 0$ and assuming that $(\lambda,t) \mapsto \hat{Z}(\lambda;t)$ is periodic with respect to time with period T—and that is at the root of the *Floquet–Magnus expansion* of the evolution operator [28].

We are now ready to obtain a perturbative expansion of $T(\lambda;t)$. In fact, if we require the interaction picture evolution operator to satisfy the Schrödinger equation, we get

$$\begin{aligned}\tilde{H}(\lambda;t)T(\lambda;t) &= \mathrm{i}\dot{T}(\lambda;t)\\ &= \mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda;t)}\int_0^1 (\mathrm{e}^{\mathrm{i}s\hat{Z}(\lambda;t)}\dot{Z}(\lambda;t)\,\mathrm{e}^{-\mathrm{i}s\hat{Z}(\lambda;t)})\,\mathrm{d}s \exp\left(-\mathrm{i}\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right)\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)}\\ &\quad + \mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda;t)}\int_0^1\left(\exp\left(-\mathrm{i}s\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right)\hat{C}(\lambda;t)\exp\left(\mathrm{i}s\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right)\right)\mathrm{d}s\\ &\quad \times \exp\left(-\mathrm{i}\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right)\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda)},\end{aligned} \tag{33}$$

where we have used the remarkable formula (see, for instance, [29])

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\mathrm{e}^{F} = \mathrm{e}^{F}\int_0^1 (\mathrm{e}^{-sF}\,\dot{F}\,\mathrm{e}^{sF})\,\mathrm{d}s = \int_0^1 (\mathrm{e}^{sF}\,\dot{F}\,\mathrm{e}^{-sF})\,\mathrm{d}s\,\mathrm{e}^{F}, \tag{34}$$

which extends to an operator-valued function $t \mapsto F(t)$ the formula for the derivative of the exponential of an ordinary function. Next, let us apply to each member of equation (33) the operator $\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda;t)}$ on the left and the operator $\mathrm{e}^{-\mathrm{i}\hat{Z}(\lambda)}\exp\left(\mathrm{i}\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t}\right)$ on the right:

$$\mathrm{Ad}_{\exp(\mathrm{i}\hat{Z}(\lambda;t))}\tilde{H}(\lambda;t) = \int_0^1 \left(\mathrm{Ad}_{\exp(\mathrm{i}s\hat{Z}(\lambda;t))}\dot{Z}(\lambda;t) + \mathrm{Ad}_{\exp(-\mathrm{i}s\int_0^t C(\lambda;\mathfrak{t})\mathrm{d}\mathfrak{t})}\hat{C}(\lambda;t)\right)\mathrm{d}s, \tag{35}$$

where we recall that, given linear operators $\mathfrak{X}$ and Y, with $\mathfrak{X}$ invertible, $\mathrm{Ad}_{\mathfrak{X}}Y := \mathfrak{X}Y\mathfrak{X}^{-1}$. Then, since $\mathfrak{X}$ is of the form e^{X}, we can use the well-known relation

$$\mathrm{Ad}_{\exp(X)}Y = \exp([X,\cdot])Y = \sum_{k=0}^{\infty}\frac{1}{k!}\,[X,\cdot]^k\,Y, \tag{36}$$

with $[X,\cdot]^k$ denoting the kth power ($[X,\cdot]^0 \equiv \mathrm{Id}$) of the superoperator $[X,\cdot]$ defined by $[X,\cdot]\,Y := [X,Y]$. Eventually, applying formula (36) to equation (35) and performing the integrals, we obtain

$$\sum_{k=0}^{\infty}\frac{\mathrm{i}^k}{k!}[\hat{Z}(\lambda;t),\cdot]^k\tilde{H}(\lambda;t) = \sum_{k=0}^{\infty}\frac{\mathrm{i}^k}{(k+1)!}[\hat{Z}(\lambda;t),\cdot]^k\dot{Z}(\lambda;t)$$
$$+\sum_{k=0}^{\infty}\frac{(-\mathrm{i})^k}{(k+1)!}\left[\int_0^t C(\lambda;\mathfrak{t})\,\mathrm{d}\mathfrak{t},\cdot\right]^k\hat{C}(\lambda;t). \tag{37}$$

Expanding the operators $\tilde{H}(\lambda;t)$, $\hat{Z}(\lambda;t)$ and $\hat{C}(\lambda;t)$ in series of the perturbative parameter λ as follows

$$\tilde{H}(\lambda;t) = \sum_{n=1}^{\infty}\lambda^n\tilde{H}_n(t) \tag{38}$$

$$\hat{Z}(\lambda;t) = \sum_{n=1}^{\infty}\lambda^n Z_n(t), \qquad \hat{C}(\lambda;t) = \sum_{n=1}^{\infty}\lambda^n C_n(t), \tag{39}$$

formula (37) provides the possibility of determining the operators $\{Z_k(t)\}$ and $\{C_k(t)\}$ at each order of λ.

It is worth noting that, for each couple of operators $Z_k(t)$ and $C_k(t)$ of a generic order k, there is just one equation. Hence, to fix a solution it is necessary to impose some gauge and moreover to determine the initial condition $Z_k(0)$.

Effective Hamiltonian as result of gauge conditions. A particularly interesting solution of equation (37) is that corresponding to the condition that all the operators $\{C_k(t)\}$ are constant in time. In such a case, that is when $C_k(t) = C_k(0) := C_k$, by integrating formula (37) one obtains the following order by order equations:

$$\begin{aligned}
&\text{first order:} && \lambda Z_1(t) &&= \int_0^t \lambda\tilde{H}_1(s)\,\mathrm{d}s - \lambda C_1 t + \lambda Z_1(0);\\
&\text{second order:} && \lambda^2 Z_2(t) &&= \tfrac{\mathrm{i}}{2}\int_0^t [\lambda Z_1(s),\ \lambda\tilde{H}_1(s) + \lambda C_1]\,\mathrm{d}s\\
&&& &&\quad + \int_0^t \lambda^2\tilde{H}_2(s)\,\mathrm{d}s - \lambda^2 C_2 t + \lambda^2 Z_2(0);\\
&\text{third order:} && &&\quad \ldots
\end{aligned} \tag{40}$$

Imposing that all $C_k(t)$'s are time independent is not enough to determine them at all. A further constraint that can be imposed is that they are such constant operators which 'eliminate' the terms linearly dependent on time from $Z_k(t)$'s (for details, see the appendix).

Depending on the structure of the Hamiltonian, the 'removal' of terms linear on time from $Z_k(t)$ (with $k = 1, \ldots, N$) coincides with the removal of all terms which indefinitely 'magnify' with time. In such cases, the mentioned gauge condition provides (as better clarified in the following) the possibility of describing the time evolution substantially as a time-independent Hamiltonian dynamics (governed by the evolution operator $\mathrm{e}^{-\mathrm{i}C(\lambda)t}$) up to unitary transformations (essentially $\mathrm{e}^{-\mathrm{i}Z(\lambda;t)}$ and $\mathrm{e}^{-\mathrm{i}Z(\lambda;0)}$) that turn out to be near to the identity at any instant of time, because their generators are bounded (i.e., not indefinitely magnifying with time) and of the order of λ or higher.

The mentioned gauge condition can be better formalized through the following tern of constraints:

(1) $C(\lambda; t) = C(\lambda; 0) \equiv C(\lambda), \forall t$;

(2) the function $t \mapsto Z(\lambda; t)$ satisfies

$$\lim_{t\to\infty} t^{-1} Z(\lambda; t) = 0; \tag{41}$$

(3) the *mean value* of the function $t \mapsto Z(\lambda; t)$ is zero, namely

$$\lim_{\tau\to\infty} \frac{1}{\tau} \int_0^\tau Z(\lambda; t)\, \mathrm{d}t = 0. \tag{42}$$

In the following we shall concentrate to such cases. Therefore, from equation (31) it turns out that

$$T(\lambda; t) = \exp(-\mathrm{i}Z(\lambda; t)) \exp(-\mathrm{i}C(\lambda)t) \exp(\mathrm{i}Z(\lambda)), \tag{43}$$

with $C(\lambda) = \sum_{n=1}^{\infty} \lambda^n C_n$ and $\exp(-\mathrm{i}Z(\lambda; t))$ a unitary operator. We here stress again that, since $Z(\lambda; t)$ is a bounded operator obtained as the sum of terms of the first order in λ or higher, it turns out that the unitary operator $\exp(-\mathrm{i}Z(\lambda; t))$ approaches the identity operator at any instant of time up to terms of the first or higher order in λ.

The complete time evolution of the system, according with equations (28) and (43), may then be written as

$$\begin{aligned} \hat{U}(\lambda, t) &= \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}Z(\lambda;t)}\, \mathrm{e}^{-\mathrm{i}C(\lambda)t}\, \mathrm{e}^{\mathrm{i}Z(\lambda;0)} \\ &= \mathrm{e}^{-\mathrm{i}\tilde{Z}(\lambda;t)}\, \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}C(\lambda)t}\, \mathrm{e}^{\mathrm{i}Z(\lambda;0)} \end{aligned} \tag{44}$$

with

$$\tilde{Z}(\lambda; t) := \mathrm{e}^{-\mathrm{i}\hat{H}_0 t} Z(\lambda; t)\, \mathrm{e}^{\mathrm{i}\hat{H}_0 t} \tag{45}$$

an operator whose time dependence is characterized by only oscillatory terms, due to the choice of the suitable gauge condition as previously discussed.

Inserting the identity operator $\mathrm{Id} = \mathrm{e}^{\mathrm{i}C(\lambda)t}\, \mathrm{e}^{\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}C(\lambda)t}$ on the right, it turns out

$$\hat{U}(\lambda, t) = \mathrm{e}^{-\mathrm{i}\tilde{Z}(\lambda;t)}\, \mathrm{e}^{-\mathrm{i}\breve{Z}(\lambda;t)}\, \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}C(\lambda)t} \tag{46}$$

with

$$\breve{Z}(\lambda; t) = \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\, \mathrm{e}^{-\mathrm{i}C(\lambda)t} Z(\lambda; 0)\, \mathrm{e}^{\mathrm{i}C(\lambda)t}\, \mathrm{e}^{\mathrm{i}\hat{H}_0 t}.$$

This operator, as well as $\tilde{Z}(\lambda, t)$, is constituted by constant in time and oscillatory terms only and, moreover, such terms are of the first or higher order in λ.

As anticipated, the total evolution operator may be expressed as the product of two unitary operators:

$$\hat{U}(\lambda, t) = T_d(\lambda, t) T_{\text{eff}}(\lambda, t) \tag{47}$$

with

$$T_d(\lambda, t) := \mathrm{e}^{-\mathrm{i}\tilde{Z}(\lambda;t)}\,\mathrm{e}^{-\mathrm{i}\breve{Z}(\lambda;t)} \tag{48}$$

$$T_{\mathrm{eff}}(\lambda, t) := \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\,\mathrm{e}^{-\mathrm{i}C(\lambda)t}. \tag{49}$$

We emphasize again that, since both $\tilde{Z}(\lambda; t)$'s and $\breve{Z}(\lambda; t)$'s contain only stationary and oscillating terms which are also terms of the first and higher orders in the perturbative parameter, then we are legitimated to assume that these operators are small at any instant of time. This circumstance leads to the fact that the dressing operator is near to the unity operator at any instant of time.

Concerning the unitary operator $\mathrm{e}^{-\mathrm{i}\hat{H}_0 t}\,\mathrm{e}^{-\mathrm{i}C(\lambda)t}$, it may be thought of as a Schrödinger picture evolutor factorized in terms of the unperturbed evolution $\mathrm{e}^{-\mathrm{i}\hat{H}_0 t}$ and the interaction picture evolution $\mathrm{e}^{-\mathrm{i}C(\lambda)t}$ generated by the interaction picture Hamiltonian $C(\lambda)$. The corresponding Schrödinger picture effective Hamiltonian is given by $\hat{H}_0 + \mathrm{e}^{\mathrm{i}\hat{H}_0 t} C(\lambda)\,\mathrm{e}^{-\mathrm{i}\hat{H}_0 t}$.

The complete time evolution may be considered in its exact form or truncated to some order N. In this case, taking into account equation (39), equation (43) may be substituted by

$$T(\lambda; t) \approx \exp(-\mathrm{i}Z^{(N)}(\lambda; t))\exp(-\mathrm{i}C^{(N)}(\lambda)t)\exp(\mathrm{i}Z^{(N)}(\lambda)), \tag{50}$$

where

$$C^{(N)}(\lambda) = \sum_{n=1}^{N} \lambda^n C_n, \qquad Z^{(N)}(\lambda; t) = \sum_{n=1}^{N} \lambda^n Z_n(t) \tag{51}$$

are the up to Nth order 'truncated' $C(\lambda)$ and $Z(\lambda; t)$ operators, respectively.

4. Double Λ Raman schemes

Let us now apply such a perturbative approach to the specific problem of two simultaneously active Raman schemes in trapped ion, and let us carry on our analysis up to the second order, in the same spirit of [30].

The Schrödinger picture Hamiltonian describing a laser-driven trapped ion is given by

$$\hat{H}_{\mathbb{A}}(t) = \hat{H}_0 + \hat{H}_B + \hat{H}_R(t) \tag{52}$$

with $\hat{H}_0$ and $\hat{H}_B$ given by equation (2), and

$$\hat{H}_R(t) = [\hbar\Omega_{13}(t)\hat{\sigma}_{13} + \mathrm{h.c.}] + [\hbar\Omega_{23}(t)\hat{\sigma}_{23} + \mathrm{h.c.}], \tag{53}$$

being

$$\Omega_{j3}(t) = g'_{j3}\,\mathrm{e}^{-\mathrm{i}(\vec{k}'_{j3}\cdot\vec{r} - \omega'_{j3}t)} + g''_{j3}\,\mathrm{e}^{-\mathrm{i}(\vec{k}''_{j3}\cdot\vec{r} - \omega''_{j3}t)}, \tag{54}$$

where g'_{j3}, ω'_{j3}, $\vec{k}'_{j3}$ ($j = 1, 2$) are coupling constant, frequency and wave vector related to the first couple of Raman lasers, and g''_{j3}, ω''_{j3}, $\vec{k}''_{j3}$ the analogous for the second Raman scheme.

Passing to the interaction picture related to $\hat{H}_0$, one obtains

$$\hat{\mathfrak{H}}_{\mathbb{A}}(t) = \hat{H}_B + \tilde{H}_R(t) \tag{55}$$

with

$$\tilde{H}_R(t) = [\hbar\tilde{\Omega}_{13}(t)\hat{\sigma}_{13} + \mathrm{h.c.}] + [\hbar\tilde{\Omega}_{23}(t)\hat{\sigma}_{23} + \mathrm{h.c.}], \tag{56}$$

$$\tilde{\Omega}_{j3}(t) = g'_{j3}\,\mathrm{e}^{-\mathrm{i}(\vec{k}'_{j3}\cdot\vec{r} - \tilde{\omega}'_{j3}t)} + g''_{j3}\,\mathrm{e}^{-\mathrm{i}(\vec{k}''_{j3}\cdot\vec{r} - \tilde{\omega}''_{j3}t)}, \tag{57}$$

where we have introduced the interaction picture laser frequencies

$$\begin{aligned} \tilde{\omega}'_{13} &= \omega'_{13} - (\omega_3 - \omega_1), & \tilde{\omega}'_{23} &= \omega'_{23} - (\omega_3 - \omega_2); \\ \tilde{\omega}''_{13} &= \omega''_{13} - (\omega_3 - \omega_1), & \tilde{\omega}''_{23} &= \omega''_{23} - (\omega_3 - \omega_2). \end{aligned} \tag{58}$$

It is worth mentioning that in this situation we do not use a rotating frame like in the case of single Raman scheme, because no rotating frame able to eliminate the whole time dependence in the Hamiltonian exists. Therefore, we make the most natural choice of considering $\hat{H}_0$ as the generator of the transformation.

Introducing the following resonance conditions

$$\tilde{\omega}'_{13} = \tilde{\omega}'_{23} =: \Delta'; \tilde{\omega}''_{13} = \tilde{\omega}''_{23} =: \Delta'',$$

it turns out

$$\tilde{\Omega}_{j3}(t) = g'_{j3}\, \mathrm{e}^{-\mathrm{i}(\vec{k}'_{j3}\cdot\vec{r} - \Delta' t)} + g''_{j3}\, \mathrm{e}^{-\mathrm{i}(\vec{k}''_{j3}\cdot\vec{r} - \Delta'' t)}. \tag{59}$$

In accordance with notation in equation (38) we take

$$\hat{\mathfrak{H}}_{\mathbb{A}} = \lambda \hat{\mathfrak{H}}_{\mathbb{A},1}, \qquad \hat{\mathfrak{H}}_{\mathbb{A},n} = 0, \qquad n \geqslant 2, \tag{60}$$

that is, the ‘perturbation’ ($\hat{H}_B + \tilde{H}_R(t)$) is assumed to be a first-order perturbation.

Considering equations (55) and (60), putting them into equation (40) and taking into account the gauge condition discussed in the previous section, one easily obtains

$$\lambda C_1 = \hat{H}_B \tag{61}$$

$$\begin{aligned} \lambda^2 C_2 = {} & \frac{1}{2}\left(\frac{\alpha'^{\dagger}_{13}\alpha'^{\dagger}_{13}}{\Delta'} + \frac{\alpha''^{\dagger}_{13}\alpha''^{\dagger}_{13}}{\Delta''}\right)\hat{\sigma}_{11} + \frac{1}{2}\left(\frac{\alpha'^{\dagger}_{23}\alpha'^{\dagger}_{23}}{\Delta'} + \frac{\alpha''^{\dagger}_{23}\alpha''^{\dagger}_{23}}{\Delta''}\right)\hat{\sigma}_{22} \\ & - \frac{1}{2}\left(\frac{\alpha'^{\dagger}_{13}\alpha'^{\dagger}_{13}}{\Delta'} + \frac{\alpha''^{\dagger}_{13}\alpha''^{\dagger}_{13}}{\Delta''} + \frac{\alpha'^{\dagger}_{23}\alpha'^{\dagger}_{23}}{\Delta'} + \frac{\alpha''^{\dagger}_{23}\alpha''^{\dagger}_{23}}{\Delta''}\right)\hat{\sigma}_{33} \\ & + \frac{1}{2}\left[\left(\frac{\alpha'^{\dagger}_{13}\alpha'^{\dagger}_{23}}{\Delta'} + \frac{\alpha''^{\dagger}_{13}\alpha''^{\dagger}_{23}}{\Delta''}\right)\hat{\sigma}_{12} + \text{h.c.}\right] \end{aligned} \tag{62}$$

with $\alpha'_{j3} = g'_{j3}\, \mathrm{e}^{-\mathrm{i}\vec{k}'_{j3}\cdot\vec{r}}$, $\alpha''_{j3} = g''_{j3}\, \mathrm{e}^{-\mathrm{i}\vec{k}''_{j3}\cdot\vec{r}}$ and $j = 1, 2$.

4.1. Factorization in terms of the effective dynamics and dressing

According with the decomposition in equation (50), the total evolution operator, up to the second order, may be expressed as the product of two unitary operators:

$$\hat{U}(\lambda, t) = T_d^{(2)}(\lambda, t) T_{\text{eff}}^{(2)}(\lambda, t),$$

with $T_d^{(2)}(\lambda, t) = \exp(-\mathrm{i}Z^{(2)}(\lambda; t))$ being the dressing operator and $T_{\text{eff}}^{(2)}(\lambda, t)$ the effective time evolution operator corrected up to the second order, hence associated with the effective Hamiltonian $\hat{H}_{\text{eff}} = \hat{H}_0 + \mathrm{e}^{\mathrm{i}\hat{H}_0 t} C(\lambda)\, \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}$,

$$\begin{aligned} \hat{H}_{\text{eff}} :={} & \hat{H}_0 + \mathrm{e}^{-\mathrm{i}\hat{H}_0 t}(\lambda C_1 + \lambda^2 C_2)\, \mathrm{e}^{\mathrm{i}\hat{H}_0 t} \\ ={} & \hat{H}_0 + \hat{H}_B + \frac{1}{2}\left(\frac{g'^{*}_{13}g'^{*}_{13}}{\Delta'} + \frac{g''^{*}_{13}g''^{*}_{13}}{\Delta''}\right)\hat{\sigma}_{11} + \frac{1}{2}\left(\frac{g'^{*}_{23}g'^{*}_{23}}{\Delta'} + \frac{g''^{*}_{23}g''^{*}_{23}}{\Delta''}\right)\hat{\sigma}_{22} \\ & - \frac{1}{2}\left(\frac{g'^{*}_{13}g'^{*}_{13}}{\Delta'} + \frac{g''^{*}_{13}g''^{*}_{13}}{\Delta''} + \frac{g'^{*}_{23}g'^{*}_{23}}{\Delta'} + \frac{g''^{*}_{23}g''^{*}_{23}}{\Delta''}\right)\hat{\sigma}_{33} \\ & + \frac{1}{2}\left[\left(\frac{g'^{*}_{13}g'^{*}_{23}}{\Delta'} + \frac{g''^{*}_{13}g''^{*}_{23}}{\Delta''}\right)\mathrm{e}^{\mathrm{i}\omega_{12}t}\hat{\sigma}_{12} + \text{h.c.}\right] \end{aligned} \tag{63}$$

with $\omega_{12} := \omega_2 - \omega_1$.

In the interaction picture associated with $\hat{H}_0$, the perturbation $C^{(2)}(\lambda) = \lambda C_1 + \lambda^2 C_2$ turns out to be exactly time independent (exactly, i.e. without rotating wave approximation).

It is worth noting that the effective Hamiltonian in this case is just the sum of the two effective Hamiltonians by which it is possible to describe the two dynamics induced by each one of the two Raman schemes when it is the only active one. Such a fact, which has been assumed as an obvious result, is now *formally* proved.

4.2. Validity of the adiabatic elimination

At this point some words are worthy in order to better understand the meaning and the limits of validity of the usual procedure consisting in evaluating the dynamics of the system as if induced by the effective Hamiltonian only. To this end, consider the evolution of the mean value of the generic operator:

$$\begin{aligned}\langle\hat{O}\rangle(t) &= \mathrm{tr}[\rho\,(t)\,\hat{O}] \\ &= \mathrm{tr}\big[\hat{T}_d\,(t)\,\hat{T}_{\mathrm{eff}}\,(t)\,\rho\,(0)\,\hat{T}_{\mathrm{eff}}^{\dagger}\,(t)\,\hat{T}_d^{\dagger}\,(t)\,\hat{O}\big]\end{aligned} \tag{64}$$

where $\rho(t)$ is the density matrix describing the state of the system at time t.

In many physical contexts, it is not possible to establish with a high at will precision the instant of time at which the observable $\hat{O}$ is measured. Therefore, in many realistic situations, it is meaningless to consider the rigorously 'instantaneous' mean value of the operator, whereas it is meaningful to average it over a time interval eventually considering a suitable probability density distribution of the measurement time instant.

Denoting by $\varphi(s)$ the probability density distribution of the instant of time at which the measurement is performed, the time average of the expectation value of the operator $\hat{O}$ in the time interval $[t - T/2, t + T/2]$ is given by

$$\begin{aligned}\int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\langle\hat{O}\rangle\,(t+s)\;\mathrm{d}s &= \int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\,\mathrm{tr}[\rho\,(t+s)\,\hat{O}] \\ &= \int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\,\mathrm{tr}\big[\hat{T}_d\,(t+s)\,\hat{T}_{\mathrm{eff}}\,(t+s)\,\rho\,(0)\,\hat{T}_{\mathrm{eff}}^{\dagger}\,(t+s)\,\hat{T}_d^{\dagger}\,(t+s)\,\hat{O}\big] \\ &= \int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\,\mathrm{tr}\big[\hat{T}_{\mathrm{eff}}\,(t+s)\,\rho\,(0)\,\hat{T}_{\mathrm{eff}}^{\dagger}\,(t+s)\,\hat{T}_d^{\dagger}\,(t+s)\,\hat{O}\hat{T}_d\,(t+s)\,\big]\end{aligned} \tag{65}$$

which may be evaluated for all $t \geqslant \frac{T}{2}$. Due to the presence of many random processes that can render uncertain the instant of measurement, a Gaussian distribution could be appropriate to describe the situation: $\varphi(s) = \frac{1}{\sigma\sqrt{2\pi}}\,\mathrm{e}^{-\frac{s^2}{2\sigma^2}}$.

Assuming that the effective dynamics evolution operator does not change meaningfully in the time interval $\left[-\frac{T}{2}, \frac{T}{2}\right]$, the average of the mean value of the operator $\hat{O}$ may be cast in the form

$$\int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\langle\hat{O}\rangle\,(t+s)\;\mathrm{d}s \simeq \mathrm{tr}\big[\hat{T}_{\mathrm{eff}}\,(t+s)\,\rho(0)\,\hat{T}_{\mathrm{eff}}^{\dagger}\,(t+s)\,\overline{O}(t)\,\big] \tag{66}$$

with

$$\overline{O} = \int_{-\frac{T}{2}}^{\frac{T}{2}} \mathrm{d}s\,\varphi(s)\hat{T}_d^{\dagger}\,(t+s)\,\hat{O}\hat{T}_d\,(t+s)\,. \tag{67}$$

Technically speaking, at this point wherein the effective dynamics has been taken out from the integral, we can consider the limit $T \longrightarrow \infty$ taking into account that the weight function $\varphi(s)$ quickly approaches zero for large values of s. Therefore, finally, we reach the definition

$$\widehat{\mathsf{CG}} : O \mapsto \overline{O} = \int_{-\infty}^{+\infty} \mathrm{d}s\, \varphi(s) \hat{T}_d^\dagger\,(t+s)\, \hat{O} \hat{T}_d\,(t+s)\,. \tag{68}$$

The assumption that the effective dynamics is a good approximation of the complete dynamics consists in the fact that the dressed and averaged (coarse-grained) operator $\overline{O}$ well approximates the bare operator $\hat{O}$.

The dressing operator $\hat{T}_d$ may be expressed as follows:

$$\hat{T}_d\,(t) \simeq \hat{1} + \sum_{ij} f_{ij}\big(\{\hat{a}_k, \hat{a}_k^\dagger\}, \lambda, t\big)\hat{\sigma}_{ij}, \tag{69}$$

where f_{ij}'s vanish for $\lambda \to 0$, that is, they are of first or higher order in the perturbative parameter. Approximation in equation (69) is valid when the operator $\hat{T}_d$ is truncated at any order in the perturbative parameter. In the following, for the sake of notational brevity, we omit the dependence of the f_{ij} operators: $f_{ij}\big(\{\hat{a}_k, \hat{a}_k^\dagger\}, \lambda, t\big) =: f_{ij}$.

Let us consider how the generic operator $\hat{O}$ does transform under the action of the dressing:

$$\hat{T}_d^\dagger\,(t)\, \hat{O} \hat{T}_d\,(t) = \hat{O} + \sum_{ij} f_{ij}^\dagger \hat{\sigma}_{ji} \hat{O} + \sum_{ij} f_{ij} \hat{O} \hat{\sigma}_{ij} + \sum_{ijkl} f_{ij}^\dagger f_{ij} \hat{\sigma}_{ji} \hat{O} \hat{\sigma}_{kl}. \tag{70}$$

Thanks to this formula, it is straightforward to find that deviations of the *dressed* operator $\hat{O}$ from the *bare* one are of the first or higher order in the perturbative parameter. Moreover, averaging the dressed operator as in equation (67) makes even smaller the corrections.

This circumstance enforces the reasonableness of exploiting the effective Hamiltonian to describe the behaviour of the system, evaluating the expectation values of *bare* operators. Nevertheless, has to be kept in mind that in general no warranty is given that the discrepancy between the operators, its dressed counterpart and the coarse-grained operator is small enough to be considered as negligible. Therefore, in principle, a case-by-case check would be necessary.

5. Master equation and conclusive remarks

So far, the dynamics of the systems has been considered as purely unitary being traceable back to coherent transitions involving the three levels, and induced by four external laser fields responsible for two Raman couplings. Each one of such two Raman couplings gives rise to an effective dynamics governed by an effective time-independent Hamiltonian up to a dressing. Both the single Raman scheme effective couplings, and hence their sum, involve the two lowest levels, while the highest one turns out to be decoupled. In other words, two dressed states are coherently coupled and perform Rabi oscillations, while the third dressed state is a stationary one.

Nevertheless, as mentioned in [31] and further considered in [23], the real dynamics of a trapped ion subjected to Raman schemes involves such non-unitary elements imputable to decays from the highest atomic level, the auxiliary one, to the lowest ones.

It can be considered that two independent *channels* are present, giving rise to a master equation that in the Schrödinger picture may be cast in the form

$$\dot{\varrho} = \mathbb{L}(\hbar^{-1} \hat{H}_{\mathbb{A}}(t); \sqrt{\gamma_{3\downarrow 1}}\hat{\sigma}_{31}, \sqrt{\gamma_{3\downarrow 2}}\hat{\sigma}_{32})\varrho, \tag{71}$$

where $\mathbb{L}(\cdot)[\cdot]$ is the *Lindblad superoperator* defined by

$$\mathbb{L}(\hat{G}; \hat{F}_1^\dagger, \hat{F}_2^\dagger)\varrho = -i[\hat{G}, \varrho] + \sum_{j=1,2} \left(\hat{F}_j^\dagger \varrho \hat{F}_j^\dagger - \frac{1}{2}\{\hat{F}_j^\dagger \hat{F}_j^\dagger, \varrho\} \right), \tag{72}$$

with $\{\cdot,\cdot\}$ denoting the anti-commutator. The first part, i.e. the commutator, with $\hat{G}$ describes the unitary evolution, while the second part is the *dissipator* describing the effects of the environment, that is the decays. Of course, for $\gamma_{3\downarrow 1} = \gamma_{3\downarrow 2} = 0$, equation (71) reduces to the standard Schrödinger equation of the Raman scheme, otherwise the Raman scheme with relaxation of the auxiliary level $|3\rangle$ is considered.

Assuming that the dissipator is a first-order superoperator, the *dressed* master equation correct up to the second order is given by

$$\dot{\varrho} = \mathbb{L}(\hbar^{-1}\hat{H}_{\rm eff}(t); \sqrt{\gamma_{3\downarrow 1}}\tilde{\sigma}_{31}, \sqrt{\gamma_{3\downarrow 2}}\tilde{\sigma}_{32})\varrho, \tag{73}$$

with

$$\begin{aligned}\tilde{\sigma}_{3j} &:= \mathrm{e}^{\mathrm{i}\tilde{Z}(\lambda;t)}\hat{\sigma}_{3j}\,\mathrm{e}^{-\mathrm{i}\tilde{Z}(\lambda;t)} \approx \hat{\sigma}_{3j} + \mathrm{i}[\lambda Z_1(\lambda;t), \hat{\sigma}_{3j}] \\ &= \hat{\sigma}_{3j} + \beta_{23}(t)\,(\hat{\sigma}_{22} - \hat{\sigma}_{33}) + \mathrm{i}\beta_{13}(t)\hat{\sigma}_{12}\end{aligned}$$

where

$$\beta_{j3}(t) = \frac{\alpha'_{j3}}{\omega'_{j3}}\,\mathrm{e}^{\mathrm{i}\omega'_{j3}t} + \frac{\alpha''_{j3}}{\omega''_{j3}}\,\mathrm{e}^{\mathrm{i}\omega''_{j3}t}.$$

As in the case of a single Raman scheme active on the system we obtain the very expressive result that, despite in the *reference frame* corresponding to the dressing $\mathrm{e}^{\mathrm{i}\hat{Z}(\lambda;t)}$ the coherent dynamics of the two lowest levels is decoupled from the coherent and trivial dynamics of the upper level, the incoherent (that is dissipative) dynamics maintains couplings between the auxiliary and the two effective levels.

In conclusion, in this paper we have investigated the dynamics of a single trapped ion subjected to the action of two simultaneously active Raman schemes founding two remarkable results. The first one is the rigorous proof of the intuitive idea that the effective Hamiltonian associated with such a physical scenario is given by the sum of the two effective Hamiltonians corresponding to the two single Raman schemes. We have traced back this 'additivity' of the effective Hamiltonians to the 'vicinity' between the bare operator and the corresponding averaged dressed ones. The second result we have achieved in this paper generalizes a result found in the case of single Raman coupling in the case wherein the auxiliary level is a metastable one. In such a situation, indeed, we succeed in writing the relevant master equation describing the 'effective evolution' of the system.

Appendix. Linear terms in $Z(\lambda; t)$

Consider the first of equation (40) with $\lambda\tilde{H}_1(t) = \hat{H}_B + \tilde{H}_R(t)$ according to equations (52), (55) and (60). The integral $\int_0^t \lambda\tilde{H}_1(s)\,\mathrm{d}s$ gives rise to a term linear in time, that is $\hat{H}_B t$, and to constant and oscillatory terms originated by quantities of the form $\int_0^t \mathrm{e}^{\mathrm{i}\omega t}\,\mathrm{d}s = \frac{\mathrm{e}^{\mathrm{i}\omega t}-1}{\mathrm{i}\omega}$. Therefore to eliminate linear terms in $Z_1(t)$ we need to choose $\lambda C_1 = \hat{H}_B$.

Consider now the second of equation (40) which may be cast in the convenient form

$$\begin{aligned}\lambda^2 Z_2(t) = \frac{\mathrm{i}}{2}\int_0^t \mathrm{d}s\int_0^s \mathrm{d}\eta[\hat{H}_R(\eta), \tilde{H}_R(s)] + \mathrm{i}\int_0^t \mathrm{d}s\int_0^s \mathrm{d}\eta[\tilde{H}_R(\eta), \hat{H}_B] \\ + \mathrm{i}\int_0^t \mathrm{d}s[\lambda Z_1(0), \hat{H}_B] + \frac{\mathrm{i}}{2}\int_0^t \mathrm{d}s[\lambda Z_1(0), \tilde{H}_R(s)] - \lambda^2 C_2 t + \lambda^2 Z_2(0).\end{aligned} \tag{A.1}$$

The most meaningful term in such an equation is given by

$$\int_0^t \mathrm{d}s\int_0^s \mathrm{d}\eta[\tilde{H}_R(\eta), \tilde{H}_R(s)] \tag{A.2}$$

involving terms of the form

$$D(\omega_a, \omega_b) := \int_0^t \mathrm{d}s \int_0^s \mathrm{d}\eta\, \mathrm{e}^{\mathrm{i}\omega_a s}\, \mathrm{e}^{\mathrm{i}\omega_b \eta} = \frac{\mathrm{e}^{\mathrm{i}\omega_a t} - 1}{\omega_a \omega_b} - \frac{\mathrm{e}^{\mathrm{i}(\omega_a+\omega_b)t} - 1}{\omega_b(\omega_a + \omega_b)}, \qquad \text{(A.3)}$$

giving rise to linear terms in time only in the two cases ($\omega_a = 0$; $\omega_b \neq 0$) or (ω_a; $\omega_b = -\omega_a$). The first condition does not match our case, because by hypothesis we deal with non-vanishing frequencies. The second condition corresponds to a resonance.

Observe in addition that other linear terms raising from

$$\int_0^t \mathrm{d}s \int_0^s \mathrm{d}\eta [\tilde{H}_R(\eta), \hat{H}_B] \qquad \text{(A.4)}$$

may be compensated (i.e. eliminated) by

$$\int_0^t \mathrm{d}s \int_0^s \mathrm{d}\eta [\lambda Z_1(0), \hat{H}_B] \qquad \text{(A.5)}$$

provided a suitable choice of the arbitrary constant $Z_1(0)$.

Since no other linear terms are present, from equation (A.3) it straightforwardly follows the result in equation (62).

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