

A general non-Markovian master equation for time-dependent Hamiltonians with coupling that is weak, strong, or anything in between

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I. THE HAMILTONIAN

We start with a time-dependent Hamiltonian of the form:

$$H(t) = H_S(t) + H_I + H_B, \quad (1)$$

$$H_S(t) = \varepsilon_0(t) |0\rangle \langle 0| + \varepsilon_1(t) |1\rangle \langle 1| + V_{10}(t) |1\rangle \langle 0| + V_{01}(t) |0\rangle \langle 1|, \quad (2)$$

$$H_I = s \sum_{\mathbf{k}} g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}), \quad (3)$$

$$H_B = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}}. \quad (4)$$

We will start with a system-bath coupling operator of the form $s = |1\rangle \langle 1|$.

A. Variational Transformation

We consider the following canonical transformation

$$\overline{H} \equiv e^V H e^{-V}, \quad (5)$$

$$e^{\pm V} \equiv |0\rangle \langle 0| + |1\rangle \langle 1| B_{\pm}, \quad (6)$$

$$B_{\pm} \equiv \prod_{\mathbf{k}} D(\pm \alpha_{\mathbf{k}}), \quad (7)$$

$$D(\pm \alpha_{\mathbf{k}}) \equiv \exp\left(\pm \alpha_{\mathbf{k}} (b_{\mathbf{k}}^\dagger - b_{\mathbf{k}})\right), \quad (8)$$

$$\alpha_{\mathbf{k}} \equiv f_{\mathbf{k}}/\omega_{\mathbf{k}}, \quad (9)$$

Here $\{f_{\mathbf{k}}\}$, which we assume to be real, represent a set of variational parameters that we will optimize to make our master equation as accurate as possible.

We use the following identities:

$$\overline{|0\rangle \langle 0|} = e^V |0\rangle \langle 0| e^{-V} \quad (10)$$

$$= (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |0\rangle \langle 0| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \quad (11)$$

$$= |0\rangle \langle 0| |0\rangle \langle 0| |0\rangle \langle 0| \quad (12)$$

$$= |0\rangle \langle 0| \quad (13)$$

$$\overline{|1\rangle \langle 1|} = e^V |1\rangle \langle 1| e^{-V} \quad (14)$$

$$= (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |1\rangle \langle 1| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \quad (15)$$

$$= B_+ |1\rangle \langle 1| B_- \quad (16)$$

$$= |1\rangle \langle 1| \quad (17)$$

$$\overline{|0\rangle\langle 1|} = e^V |0\rangle\langle 1| e^{-V} \quad (18)$$

$$= (|0\rangle\langle 0| + |1\rangle\langle 1| B_+) |0\rangle\langle 1| (|0\rangle\langle 0| + |1\rangle\langle 1| B_-) \quad (19)$$

$$= |0\rangle\langle 0| |0\rangle\langle 1| |1\rangle\langle 1| B_- \quad (20)$$

$$= |0\rangle\langle 1| B_- \quad (21)$$

$$\overline{|1\rangle\langle 0|} = e^V |1\rangle\langle 0| e^{-V} \quad (22)$$

$$= (|0\rangle\langle 0| + |1\rangle\langle 1| B_+) |1\rangle\langle 0| (|0\rangle\langle 0| + |1\rangle\langle 1| B_-) \quad (23)$$

$$= |1\rangle\langle 1| B_+ |1\rangle\langle 0| |0\rangle\langle 0| \quad (24)$$

$$= |1\rangle\langle 0| B_+ \quad (25)$$

$$\overline{b_{\mathbf{k}}} = e^V b_{\mathbf{k}} e^{-V} \quad (26)$$

$$= (|0\rangle\langle 0| + |1\rangle\langle 1| B_+) b_{\mathbf{k}} (|0\rangle\langle 0| + |1\rangle\langle 1| B_-) \quad (27)$$

$$= |0\rangle\langle 0| b_{\mathbf{k}} |0\rangle\langle 0| + |1\rangle\langle 1| B_+ b_{\mathbf{k}} |1\rangle\langle 1| B_- \quad (28)$$

$$= |0\rangle\langle 0| b_{\mathbf{k}} + |1\rangle\langle 1| (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \quad (29)$$

$$\overline{b_{\mathbf{k}}^\dagger} = e^V b_{\mathbf{k}}^\dagger e^{-V} \quad (30)$$

$$= (|0\rangle\langle 0| + |1\rangle\langle 1| B_+) b_{\mathbf{k}}^\dagger (|0\rangle\langle 0| + |1\rangle\langle 1| B_-) \quad (31)$$

$$= |0\rangle\langle 0| b_{\mathbf{k}}^\dagger |0\rangle\langle 0| + |1\rangle\langle 1| B_+ b_{\mathbf{k}}^\dagger |1\rangle\langle 1| B_- \quad (32)$$

$$= |0\rangle\langle 0| b_{\mathbf{k}}^\dagger + |1\rangle\langle 1| (b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}}) \quad (33)$$

We have used the following:

$$B_+ b_{\mathbf{k}} B_- = b_{\mathbf{k}} - \alpha_{\mathbf{k}} \quad (34)$$

$$B_+ b_{\mathbf{k}}^\dagger B_- = b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}} \quad (35)$$

We therefore have the following relationships:

$$\overline{\varepsilon_0(t) |0\rangle\langle 0|} = \varepsilon_0(t) |0\rangle\langle 0| \quad (36)$$

$$\overline{\varepsilon_1(t) |1\rangle\langle 1|} = \varepsilon_1(t) |1\rangle\langle 1| \quad (37)$$

$$\overline{V_{10}(t) |1\rangle\langle 0|} = V_{10}(t) |1\rangle\langle 0| B_+ \quad (38)$$

$$\overline{V_{01}(t) |0\rangle\langle 1|} = V_{01}(t) |0\rangle\langle 1| B_- \quad (39)$$

$$\overline{g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}) |1\rangle\langle 1|} = g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}}) |1\rangle\langle 1| \quad (40)$$

$$\overline{\omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}}} = \overline{\omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} |0\rangle\langle 0| + \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} |1\rangle\langle 1|} \quad (41)$$

$$= \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} |0\rangle\langle 0| + \omega_{\mathbf{k}} D(\alpha_{\mathbf{k}}) b_{\mathbf{k}}^\dagger b_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) |1\rangle\langle 1| \quad (42)$$

Let's focus our attention in the following calculation, as we can see it appears as consequence of the application of the transformation (5) on the bath (4), here we define $D(\pm\alpha_{\mathbf{k}})$ as shown in the equation (8) as a displacement operator associated with the frequency $\omega_{\mathbf{k}}$. We will calculate $\omega_{\mathbf{k}} D(\alpha_{\mathbf{k}}) b_{\mathbf{k}}^\dagger b_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) |1\rangle\langle 1|$ inserting the identity operator between $b_{\mathbf{k}}^\dagger$ and $b_{\mathbf{k}}$ written like $D(-\alpha_{\mathbf{k}}) D(\alpha_{\mathbf{k}}) = 1$ in order to facilitate the calculations:

$$\omega_{\mathbf{k}} D(\alpha_{\mathbf{k}}) b_{\mathbf{k}}^{\dagger} D(-\alpha_{\mathbf{k}}) D(\alpha_{\mathbf{k}}) b_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) |1\rangle \langle 1| = \omega_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} - \alpha_{\mathbf{k}} \right) (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) |1\rangle \langle 1| \quad (43)$$

$$= \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} |1\rangle \langle 1| + \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 |1\rangle \langle 1| - \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) |1\rangle \langle 1| \quad (44)$$

Respect to H_S , H_I , H_B as shown in (2), (3) and (4) the transformed hamiltonian of each one of these terms using the equations (36) to (42) is:

$$\overline{H_S} = \overline{\varepsilon_0(t) |0\rangle \langle 0|} + \overline{\varepsilon_1(t) |1\rangle \langle 1|} + \overline{V_{10}(t) |1\rangle \langle 0|} + \overline{V_{01}(t) |0\rangle \langle 1|} \quad (45)$$

$$= \varepsilon_0(t) |0\rangle \langle 0| + \varepsilon_1(t) |1\rangle \langle 1| + V_{10}(t) |1\rangle \langle 0| B_+ + V_{01}(t) |0\rangle \langle 1| B_- \quad (46)$$

$$\overline{H_I} = \sum_{\mathbf{k}} g_{\mathbf{k}} \left(\bar{b}_{\mathbf{k}}^{\dagger} + \bar{b}_{\mathbf{k}} \right) |1\rangle \langle 1| \quad (47)$$

$$= \sum_{\mathbf{k}} g_{\mathbf{k}} \left(\left(b_{\mathbf{k}}^{\dagger} - \alpha_{\mathbf{k}} \right) + (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \right) |1\rangle \langle 1| \quad (48)$$

$$= \sum_{\mathbf{k}} g_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}} \right) |1\rangle \langle 1| \quad (49)$$

$$\overline{H_B} = \sum_{\mathbf{k}} \overline{\omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}} \quad (50)$$

$$= \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(|0\rangle \langle 0| b_{\mathbf{k}}^{\dagger} + |1\rangle \langle 1| \left(b_{\mathbf{k}}^{\dagger} - \alpha_{\mathbf{k}} \right) \right) \left(|0\rangle \langle 0| b_{\mathbf{k}} + |1\rangle \langle 1| (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \right) \quad (51)$$

$$= |0\rangle \langle 0| \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} - \alpha_{\mathbf{k}} \right) (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \quad (52)$$

$$= (|0\rangle \langle 0| + |1\rangle \langle 1|) \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \alpha_{\mathbf{k}} \quad (53)$$

$$= \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 \quad (54)$$

Finally merging these expressions gives the transformed Hamiltonian:

$$\overline{H} = \varepsilon_1(t) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) |1\rangle \langle 0| B_+ + V_{01}(t) |0\rangle \langle 1| B_- + \sum_{\mathbf{k}} g_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}} \right) |1\rangle \langle 1| \quad (55)$$

$$+ \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) |1\rangle \langle 1| + \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 |1\rangle \langle 1| \quad (56)$$

Also we may write this transformed Hamiltonian as a sum of the form:

$$\overline{H} = \overline{H_S} + \overline{H_B} + \overline{H_I} \quad (57)$$

Let's define:

$$R_1 \equiv \sum_{\mathbf{k}} (\omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 - 2\alpha_{\mathbf{k}} g_{\mathbf{k}}) \quad (58)$$

$$B_z \equiv \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) \quad (59)$$

We assume that the bath is at equilibrium with inverse temperature $\beta = 1/k_B T$:

$$\rho_B = \frac{e^{-\beta H_B}}{\text{Tr}(e^{-\beta H_B})} \quad (60)$$

We can show that:

$$\langle B_z \rangle_{H_B} = \text{Tr}(\rho_B B_z) \quad (61)$$

$$= \text{Tr} \frac{(e^{-\beta H_B} B_z)}{\text{Tr}(e^{-\beta H_B})} \quad (62)$$

$$= 0, \quad (63)$$

and:

$$\langle B_{\pm} \rangle_{H_B} = \text{Tr}(\rho_B B_{\pm}) \quad (64)$$

$$= \exp \left(- (1/2) \sum_{\mathbf{k}} (\alpha_{\mathbf{k}})^2 \coth(\beta \omega_{\mathbf{k}}/2) \right) \quad (65)$$

$$\equiv B \quad (66)$$

In order to (i) ensure that $\langle \overline{H_I} \rangle_{H_B} = 0$ which simplifies the form of the master equation to be derived; (ii) introduce the bath renormalizing driving in $\overline{H_S}$ to treat it non-perturbatively in the subsequent formalism, we associate the terms related with $B_+ \sigma_+$ and $B_- \sigma_-$ to the interaction part of the Hamiltonian $\overline{H_I}$ and we subtract their expected value in order to satisfy $\langle \overline{H_I} \rangle_{H_B} = 0$. Furthermore we add the subtracted terms to the $\overline{H_S}$. The final form of the terms of the splitted Hamiltonian \overline{H} is:

$$\overline{H_S} = (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \sigma_+ + V_{01}(t) B \sigma_- \quad (67)$$

$$\overline{H_I} = \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) (b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}}) |1\rangle \langle 1| + V_{10}(t) (\sigma_+ B_+ - \sigma_+ B) + V_{01}(t) (\sigma_- B_- - \sigma_- B) \quad (68)$$

$$H_B = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} \quad (69)$$

Let's consider the following Hermitian combinations:

$$B_x = \frac{B_+ + B_- - 2B}{2} \quad (70)$$

$$B_y = \frac{B_- - B_+}{2i} \quad (71)$$

$$B_z = \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) (b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}}) \quad (72)$$

Writing the equations (67) and (68) using the previous combinations we obtain that:

$$\overline{H_S} = (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \sigma_+ + V_{01}(t) B \sigma_- \quad (73)$$

$$= (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \frac{\sigma_x + i\sigma_y}{2} + V_{01}(t) B \frac{\sigma_x - i\sigma_y}{2} \quad (74)$$

$$= (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + \frac{B\sigma_x}{2} (V_{10}(t) + V_{01}(t)) + \frac{iB\sigma_y}{2} (V_{10}(t) - V_{01}(t)) \quad (75)$$

$$\overline{H_I} = B_z |1\rangle \langle 1| + V_{10}(t) (\sigma_+ B_+ - \sigma_+ B) + V_{01}(t) (\sigma_- B_- - \sigma_- B) \quad (76)$$

$$= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) \left(\frac{\sigma_x + i\sigma_y}{2} B_+ - \frac{\sigma_x + i\sigma_y}{2} B + \frac{\sigma_x - i\sigma_y}{2} B_- - \frac{\sigma_x - i\sigma_y}{2} B \right) \quad (77)$$

$$+ i\Im(V_{10}(t)) \left(\frac{\sigma_x + i\sigma_y}{2} B_+ - \frac{\sigma_x + i\sigma_y}{2} B - \frac{\sigma_x - i\sigma_y}{2} B_- + \frac{\sigma_x - i\sigma_y}{2} B \right) \quad (78)$$

$$= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) (B_x \sigma_x + B_y \sigma_y) + i\Im(V_{10}(t)) (iB_x \sigma_y - iB_y \sigma_x) \quad (79)$$

$$= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) (B_x \sigma_x + B_y \sigma_y) - \Im(V_{10}(t)) (B_x \sigma_y - B_y \sigma_x). \quad (80)$$

II. FREE-ENERGY MINIMIZATION

The true free energy A is bounded by the Bogoliubov inequality:

$$A \leq A_B \equiv -\frac{1}{\beta} \ln \left(\text{Tr} \left(e^{-\beta(\overline{H}_S + H_B)} \right) \right) + \langle \overline{H}_I \rangle_{\overline{H}_S + H_B} - \frac{1}{\beta} (??) + O^? \left(\langle \overline{H}_I \rangle_{\overline{H}_S + H_B} \right). \quad (81)$$

We will optimize the set of variational parameters $\{f_{\mathbf{k}}\}$ in order to minimize A_B (i.e. to make it as close to the true free energy A as possible). Neglecting the higher order terms and using $\langle \overline{H}_I \rangle_{\overline{H}_S + H_B} = 0$ we can obtain the following condition to obtain the set $\{f_{\mathbf{k}}\}$:

$$\frac{\partial A_B}{\partial f_{\mathbf{k}}} = 0. \quad (82)$$

Using this condition and given that $[\overline{H}_S, H_B] = 0$ then $e^{-\beta(\overline{H}_S + H_B)} = e^{-\beta\overline{H}_S} e^{-\beta H_B}$, furthermore $\text{Tr} \left(e^{-\beta\overline{H}_S} e^{-\beta H_B} \right) = \text{Tr} \left(e^{-\beta\overline{H}_S} \right) \text{Tr} \left(e^{-\beta H_B} \right)$ from the fact that \overline{H}_S and H_B relate to different Hilbert spaces. Given that $\frac{\partial \text{Tr}(e^{-\beta H_B})}{\partial f_{\mathbf{k}}} = 0$ then it's possible to write the equation (82) in the following way:

$$\frac{\partial \text{Tr} \left(e^{-\beta\overline{H}_S} \right)}{\partial f_{\mathbf{k}}} = 0 \quad (83)$$

The variational parameters are:

$$f_{\mathbf{k}} = \frac{g_{\mathbf{k}} \left(1 - \frac{\tanh(\frac{\beta\eta}{2})}{\eta} (\varepsilon_1(t) + R_1 - \varepsilon_0(t)) \right)}{1 - \frac{\tanh(\frac{\beta\eta}{2})}{\eta} \left(\varepsilon_1(t) + R_1 - \varepsilon_0(t) - \frac{2|V_{10}(t)|^2 B^2}{\omega_{\mathbf{k}}} \coth \left(\frac{\beta\omega_{\mathbf{k}}}{2} \right) \right)} \quad (84)$$

where $\eta = \sqrt{(\text{Tr}(\overline{H}_S))^2 - 4\text{Det}(\overline{H}_S)}$ and $f_{\mathbf{k}} = \alpha_{\mathbf{k}}\omega_{\mathbf{k}}$.

III. MASTER EQUATION

In order to describe the dynamics of the QD under the influence of the phonon environment, we use the time-convolutionless projection operator technique. We consider the QD in its ground state. The initial density operator $\rho(0) = |0\rangle\langle 0| \otimes \rho_B$, the transformed density operator is equal to:

$$e^V \rho(0) e^{-V} = \left(|0\rangle\langle 0| + |1\rangle\langle 1| \prod_{\mathbf{k}} D(\alpha_{\mathbf{k}}) \right) (|0\rangle\langle 0| \otimes \rho_B) \left(|0\rangle\langle 0| + |1\rangle\langle 1| \prod_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) \right) \quad (85)$$

$$0 = |0\rangle\langle 0| \otimes \rho_B \quad (86)$$

$$0 = \rho(0) \quad (87)$$

We transform any operator O into the interaction picture in the following way:

$$\tilde{O} \equiv U^\dagger(t) O U(t) \quad (88)$$

$$U(t) \equiv \mathcal{T} \exp \left(-i \int_0^t dv \overline{H}_S(v) \right). \quad (89)$$

Therefore:

$$\widetilde{\overline{\rho_S}}(t) = U^\dagger(t) \overline{\rho_S}(t) U(t), \text{ where} \quad (90)$$

$$\overline{\rho_S}(t) = \text{Tr}_B(\bar{\rho}(t)) \quad (91)$$

We define $A_1 = \sigma_x$, $A_2 = \sigma_y$, $A_3 = \frac{I + \sigma_z}{2}$, $A_4 = \sigma_x$ and $A_5 = -\sigma_y$. Furthermore we label $B_1(t) = B_x = -B_5(t)$, $B_2(t) = B_y = B_4(t)$ and $B_3(t) = B_z$, also $C_1(t) = \Re(V_{10}(t)) = C_2(t)$, $C_3(t) = 1$ and $C_4(t) = \Im(V_{10}(t)) = -C_5(t)$. The precedent notation allows us to write the interaction Hamiltonian in $H_I(t)$ as:

$$H_I(t) = \sum_i C_i(t) (A_i \otimes B_i(t)) \quad (92)$$

Taking as reference state ρ_B and truncating at second order in $H_I(t)$, we obtain our master equation in the interaction picture:

$$\frac{d\widetilde{\overline{\rho_S}}(t)}{dt} = - \int_0^t \text{Tr}_B \left[\tilde{H}_I(t), \left[\tilde{H}_I(s), \widetilde{\overline{\rho_S}}(s) \rho_B \right] \right] ds \quad (93)$$

From the interaction picture applied on H_I we find:

$$\tilde{H}_I(t) = U^\dagger(t) H_I(t) U(t) \quad (94)$$

$t = 0$, we use the time-ordering operator \mathcal{T} because in general $\overline{H_S}(t)$ doesn't commute with itself at two different times. We write the interaction Hamiltonian as:

$$\tilde{H}_I(t) = \sum_i C_i(t) (\tilde{A}_i \otimes \tilde{B}_i(t)) \quad (95)$$

$$\tilde{A}_i(t) = U_S^\dagger(t) A_i U_S(t) \quad (96)$$

$$\tilde{B}_i(t) = e^{iH_B t} B_i(t) e^{-iH_B t} \quad (97)$$

Using the expression (95) we have

$$\frac{d\overline{\rho_S}(t)}{dt} = -i[H_S(t), \overline{\rho_S}(t)] - \sum_{ij} \int_0^t d\tau \left(C_i(t) C_j(t-\tau) \Lambda_{ij}(\tau) [A_i, \tilde{A}_j(t-\tau, t) \overline{\rho_S}(t)] \right. \quad (98)$$

$$\left. + C_j(t) C_i(t-\tau) \Lambda_{ji}(-\tau) [\overline{\rho_S}(t) \tilde{A}_j(t-\tau, t), A_i] \right) \quad (99)$$

where $i, j \in \{1, 2, 3, 4, 5\}$.

Here $\tilde{A}_j(s, t) = U_S(t) U_S^\dagger(s) A_j U_S(s) U_S^\dagger(t)$. The equation obtained is a non-Markovian master equation which describes the QD exciton dynamics in the variational frame with a general time-dependent Hamiltonian, and valid at second order in $H_I(t)$. The environmental correlation functions are given by:

$$\Lambda_{ij}(\tau) = \text{Tr}_B \left(\tilde{B}_i(t) \tilde{B}_j(s) \rho_B \right) \quad (100)$$

$$= \text{Tr}_B \left(\tilde{B}_i(\tau) \tilde{B}_j(0) \rho_B \right) \quad (101)$$

Using the coherent-state representation of the bath density operator we find that the correlation functions are equal to:

$$\Lambda_{11}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) \quad (102)$$

$$= \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \quad (103)$$

$$\Lambda_{22}(\tau) = \text{Tr}_B \left(\tilde{B}_2(\tau) \tilde{B}_2(0) \rho_B \right) \quad (104)$$

$$= \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \quad (105)$$

$$\Lambda_{33}(\tau) = \int_0^\infty d\omega J(\omega) (1 - F(\omega))^2 G_+(\tau) \quad (106)$$

$$\Lambda_{32}(\tau) = B(\tau) \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega) (1 - F(\omega)) iG_-(\tau) \quad (107)$$

$$\Lambda_{23}(\tau) = -B(0) \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega, \tau) (1 - F(\omega, \tau)) iG_-(\tau) \quad (108)$$

$$\Lambda_{12}(\tau) = \Lambda_{21}(\tau) = \Lambda_{13}(\tau) = \Lambda_{31}(\tau) = 0 \quad (109)$$

With the phonon propagator given by:

$$\phi(\tau) = \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} F(\omega)^2 G_+(\tau) \quad (110)$$

defined in terms of $G_\pm(\tau) = (n(\omega) + 1) e^{-i\tau\omega} \pm n(\omega) e^{i\tau\omega}$ with $n(\omega) = (e^{\beta\omega} - 1)^{-1}$ the occupation number. The matrix $\Lambda(\tau)$ called correlation matrix defined in terms of the equation (100) allows us to write all the correlations functions as:

$$\Lambda(\tau) = \begin{pmatrix} \Lambda_{11}(\tau) & 0 & 0 & 0 & -\Lambda_{11}(\tau) \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ 0 & \Lambda_{32}(\tau) & \Lambda_{33}(\tau) & \Lambda_{32}(\tau) & 0 \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ -\Lambda_{11}(\tau) & 0 & 0 & 0 & \Lambda_{11}(\tau) \end{pmatrix} \quad (111)$$

The eigenvalues of the Hamiltonian $\overline{H_S}$ are given by the solution of the following algebraic equation:

$$\lambda^2 - \text{Tr}(\overline{H_S}) \lambda + \text{Det}(\overline{H_S}) = 0 \quad (112)$$

The solutions of this equation written in terms of η and ξ as defined in the precedent section are given by $\lambda_\pm = \frac{\xi \pm \eta}{2}$ and they satisfy $H_S |\pm\rangle = \lambda_\pm |\pm\rangle$. Using this notation is possible to write $H_S = \lambda_+ |+\rangle \langle +| + \lambda_- |-\rangle \langle -|$.

The time-dependence of the system operators $\tilde{A}_i(t)$ may be made explicit using the Fourier decomposition:

$$\tilde{A}_i(\tau) = e^{i\overline{H_S}\tau} A_i e^{-i\overline{H_S}\tau} = \sum_{\zeta} e^{-i\zeta\tau} A_i(\zeta) \quad (113)$$

Where the sum is defined on the set of all the differences between the eigenvalues of the system, in our case $\zeta \in \{0, \pm\eta\}$.

Extending the Fourier decomposition to the matrix $\tilde{A}_j(t - \tau, t)$ we will obtain

$$\tilde{A}_j(t - \tau, t) = U_S(t - \tau) U_S^\dagger(t) A_j U_S(t) U_S^\dagger(t - \tau) \quad (114)$$

$$= \sum_{\zeta, \zeta'} e^{-i\zeta\tau + i\tau(\zeta - \zeta')} A_i(\zeta, \zeta') \quad (115)$$

where ζ' and ζ belongs to the set of the differences of the eigenvalues of the Hamiltonian $H_S(t - \tau)$ and $H_S(t)$ respectively.

In order to show the explicit form of the matrices present in the RHS of the equation (113) for a general 2×2 matrix let's write the matrix A_i in the base $V = \{|+\rangle, |-\rangle\}$ in the following way:

$$A_i = \sum_{\alpha, \beta \in V} \langle \alpha | A_i | \beta \rangle | \alpha \rangle \langle \beta | \quad (116)$$

Given that $[|+\rangle \langle +|, |-\rangle \langle -|] = 0$, then using the Zassenhaus formula we obtain:

$$e^{i\overline{H_S}\tau} = e^{i(\lambda_+|+\rangle \langle +| + \lambda_-|-\rangle \langle -|)\tau} \quad (117)$$

$$= e^{i\lambda_+|+\rangle \langle +|\tau} e^{i\lambda_-|-\rangle \langle -|\tau} \quad (118)$$

$$= (|-\rangle \langle -| + e^{i\lambda_+\tau} |+\rangle \langle +|) (|+\rangle \langle +| + e^{i\lambda_-\tau} |-\rangle \langle -|) \quad (119)$$

$$= e^{i\lambda_+\tau} |+\rangle \langle +| + e^{i\lambda_-\tau} |-\rangle \langle -| \quad (120)$$

Calculating the transformation (113) directly using the precedent relationship we find that:

$$\tilde{A}_i(\tau) = (e^{i\lambda_+\tau} |+\rangle \langle +| + e^{i\lambda_-\tau} |-\rangle \langle -|) \left(\sum_{\alpha, \beta \in V} \langle \alpha | A_i | \beta \rangle | \alpha \rangle \langle \beta | \right) (e^{-i\lambda_+\tau} |+\rangle \langle +| + e^{-i\lambda_-\tau} |-\rangle \langle -|) \quad (121)$$

$$= \langle + | A_i | + \rangle | + \rangle \langle + | + e^{i\eta\tau} \langle + | A_i | - \rangle | + \rangle \langle - | + e^{-i\eta\tau} \langle - | A_i | + \rangle | - \rangle \langle + | + \langle - | A_i | - \rangle | - \rangle \langle - | \quad (122)$$

Here $\eta = \lambda_+ - \lambda_-$. Comparing the RHS of the equations (113) and the explicit expression for $\tilde{A}_i(\tau)$ and we obtain the form of the expansion matrices of the Fourier decomposition for a general 2×2 matrix:

$$A_i(0) = \langle + | A_i | + \rangle | + \rangle \langle + | + \langle - | A_i | - \rangle | - \rangle \langle - | \quad (123)$$

$$A_i(\zeta) = \langle + | A_i | - \rangle | + \rangle \langle - | \quad (124)$$

$$A_i(-\zeta) = \langle - | A_i | + \rangle | - \rangle \langle + | \quad (125)$$

For a decomposition of the interaction Hamiltonian in terms of Hermitian operators, i.e. $\tilde{A}_i(\tau) = \tilde{A}_i^\dagger(\tau)$ and $\tilde{B}_i(\tau) = \tilde{B}_i^\dagger(\tau)$ we can use the equation (113) to write the master equation in the following neater form:

$$\frac{d\bar{\rho}_S}{dt} = -i[H_S(t), \bar{\rho}_S(t)] - \frac{1}{2} \sum_{ij} \sum_{\zeta, \zeta'} \gamma_{ij}(\zeta, \zeta', t) [A_i, A_j(\zeta, \zeta') \bar{\rho}_S(t) - \bar{\rho}_S(t) A_j^\dagger(\zeta, \zeta')] - \sum_{ij} \sum_{\zeta} S_{ij}(\zeta, \zeta', t) [A_i, A_j(\zeta, \zeta') \bar{\rho}_S(t) + \bar{\rho}_S(t) A_j^\dagger(\zeta, \zeta')] \quad (126)$$

where $A_j^\dagger(\zeta) = A(-\zeta)$ as expected from the equations (124) and (125). As we can see the equation shown contains the rates and energy shifts $\gamma_{ij}(\zeta, \zeta', t) = 2\Re(K_{ij}(\zeta, \zeta', t))$ and $S_{ij}(\zeta, \zeta', t) = \Im(K_{ij}(\zeta, \zeta', t))$, respectively, defined in terms of the response functions

$$K_{ij}(\zeta, \zeta', t) = \int_0^t C_i(t) C_j(t - \tau) \Lambda_{ij}(\tau) e^{i\zeta\tau} e^{-it(\zeta - \zeta')} d\tau \quad (127)$$

If we extend the upper limit of integration to ∞ in the equation (127) then the system will be independent of any preparation at $t = 0$, so the evolution of the system will depend only on its present state as expected in the Markovian approximation.

IV. LIMIT CASES

In order to show the plausibility of the master equation (??) for a time-dependent Hamiltonian we will show that this equation reproduces the following cases under certain limits conditions that will be pointed in each subsection.

A. Time-independent variational quantum master equation

At first let's show that the master equation (??) reproduces the results of the reference [1], for the latter case we have that $i, j \in \{1, 2, 3\}$ and $\omega \in (0, \pm\eta)$. The Hamiltonian of the system considered in this reference written in the same basis than the Hamiltonian (1) is given by:

$$H = \left(\delta + \sum_j g_k (b_k^\dagger + b_k) \right) |1\rangle \langle 1| + \frac{\Omega}{2} \sigma_x + \sum_k \omega_k b_k^\dagger b_k \quad (128)$$

After performing the transformation (5) on the Hamiltonian (128) it's possible to split that result in the following set of Hamiltonians:

$$\overline{H_S} = (\delta + R) |1\rangle \langle 1| + \frac{\Omega_r}{2} \sigma_x \quad (129)$$

$$\overline{H_I} = B_z |1\rangle \langle 1| + \frac{\Omega}{2} (B_x \sigma_x + B_y \sigma_y) \quad (130)$$

$$H_B = \sum_k \omega_k b_k^\dagger b_k \quad (131)$$

The Hamiltonian (129) differs from the transformed Hamiltonian H_S of the reference written like $H_S = \frac{R}{2} \mathbb{I} + \frac{\epsilon}{2} \sigma_z + \frac{\Omega_r}{2} \sigma_x$ by a term proportional to the identity, this can be seen in the following way taking $\epsilon = \delta + R$

$$(\delta + R) |1\rangle \langle 1| - \frac{\delta}{2} \mathbb{I} = \left(\frac{\delta}{2} + R \right) |1\rangle \langle 1| - \frac{\delta}{2} |0\rangle \langle 0| \quad (132)$$

$$= \frac{R}{2} \mathbb{I} + \frac{\delta + R}{2} \sigma_z \quad (133)$$

$$= \frac{R}{2} \mathbb{I} + \frac{\epsilon}{2} \sigma_z \quad (134)$$

In this Hamiltonian we can write $A_1 = \sigma_x$, $A_2 = \sigma_y$ and $A_3 = \frac{I + \sigma_z}{2}$. In order to find the decomposition matrices of the Fourier decomposition let's obtain the eigenvalues and eigenvectors of the matrix $\overline{H_S}$.

$$\lambda_+ = \frac{\epsilon + \eta}{2} \quad (135)$$

$$\lambda_- = \frac{\epsilon - \eta}{2} \quad (136)$$

$$|+\rangle = \frac{1}{\sqrt{(\epsilon + \eta)^2 + \Omega_r^2}} \begin{pmatrix} \epsilon + \eta \\ \Omega_r \end{pmatrix} \quad (137)$$

$$|-\rangle = \frac{1}{\sqrt{(\epsilon + \eta)^2 + \Omega_r^2}} \begin{pmatrix} -\Omega_r \\ \epsilon + \eta \end{pmatrix} \quad (138)$$

Using this basis we can find the decomposition matrices using the equations (124)-(125) and the fact that $|+\rangle = \cos(\theta) |1\rangle + \sin(\theta) |0\rangle$ and $|-\rangle = -\sin(\theta) |1\rangle + \cos(\theta) |0\rangle$ with $\sin(\theta) = \frac{\Omega_r}{\sqrt{(\epsilon + \eta)^2 + \Omega_r^2}}$ and $\cos(\theta) = \frac{\epsilon + \eta}{\sqrt{(\epsilon + \eta)^2 + \Omega_r^2}}$:

$$\langle + | \sigma_x | + \rangle = (\cos(\theta) \ \sin(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (139)$$

$$= 2 \sin(\theta) \cos(\theta) \quad (140)$$

$$= \sin(2\theta) \quad (141)$$

$$\langle - | \sigma_x | - \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} \quad (142)$$

$$= -2 \sin(\theta) \cos(\theta) \quad (143)$$

$$= -\sin(2\theta) \quad (144)$$

$$\langle - | \sigma_x | + \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (145)$$

$$= \cos^2(\theta) - \sin^2(\theta) \quad (146)$$

$$= \cos(2\theta) \quad (147)$$

$$\langle + | \sigma_y | + \rangle = (\cos(\theta) \ \sin(\theta)) \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (148)$$

$$= i \sin(\theta) \cos(\theta) - i \sin(\theta) \cos(\theta) \quad (149)$$

$$= 0 \quad (150)$$

$$\langle - | \sigma_y | - \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} \quad (151)$$

$$= -i \sin(\theta) \cos(\theta) + i \sin(\theta) \cos(\theta) \quad (152)$$

$$= 0 \quad (153)$$

$$\langle - | \sigma_y | + \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (154)$$

$$= i \cos^2(\theta) + i \sin^2(\theta) \quad (155)$$

$$= i \quad (156)$$

$$\langle + | \frac{1 + \sigma_z}{2} | + \rangle = (\cos(\theta) \ \sin(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (157)$$

$$= \cos(\theta) \cos(\theta) \quad (158)$$

$$= \cos^2(\theta) \quad (159)$$

$$\langle - | \frac{1 + \sigma_z}{2} | - \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} \quad (160)$$

$$= \sin(\theta) \sin(\theta) \quad (161)$$

$$= \sin^2(\theta) \quad (162)$$

$$\langle - | \frac{1 + \sigma_z}{2} | + \rangle = (-\sin(\theta) \ \cos(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} \quad (163)$$

$$= -\sin(\theta) \cos(\theta) \quad (164)$$

$$= -\sin(\theta) \cos(\theta) \quad (165)$$

Composing the parts shown give us the Fourier decomposition matrices for this case:

$$A_1(0) = \sin(2\theta) (|+\rangle \langle +| - |-\rangle \langle -|) \quad (166)$$

$$A_1(\eta) = \cos(2\theta) |-\rangle \langle +| \quad (167)$$

$$A_2(0) = 0 \quad (168)$$

$$A_2(\eta) = i |-\rangle \langle +| \quad (169)$$

$$A_3(0) = \cos^2(\theta) |+\rangle \langle +| + \sin^2(\theta) |-\rangle \langle -| \quad (170)$$

$$A_3(\eta) = -\sin(\theta) \cos(\theta) |-\rangle \langle +| \quad (171)$$

Now to make comparisons between the model obtained and the model of the system under discussion we will define that the correlation functions of the reference [1] denoted by $\Lambda'_{ij}(\tau)$ relate with the correlation functions defined in the equation (101) in the following way:

$$\Lambda'_{ij}(\tau) = C_i(t) C_j(t - \tau) \Lambda_{ij}(\tau) \quad (172)$$

Using the notation of the master equation (??), we can say that $C_1(t) = \frac{\Omega}{2} = C_2(t)$ and $C_3(t) = 1$, being Ω a constant. Furthermore given that $\overline{H_S}$ is time-independent then $B(t) = B$. Taking the equations(102)-(109) we find that the correlation functions of the reference [1] written in terms of the RHS of the equation (101) are equal to:

$$\Lambda'_{11}(\tau) = \left(\frac{\Omega}{2}\right)^2 \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) \quad (173)$$

$$= \frac{\Omega_r^2}{8} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \quad (174)$$

$$\Lambda'_{22}(\tau) = \left(\frac{\Omega}{2}\right)^2 \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) \quad (175)$$

$$= \frac{\Omega_r^2}{8} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \quad (176)$$

$$\Lambda'_{33}(\tau) = \int_0^\infty d\omega J(\omega) (1 - F(\omega))^2 G_+(\tau) \quad (177)$$

$$\Lambda'_{32}(\tau) = \frac{\Omega_r}{2} \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega) (1 - F(\omega)) iG_-(\tau) \quad (178)$$

$$\Lambda'_{32}(\tau) = -\Lambda'_{23}(\tau) \quad (179)$$

$$\Lambda'_{12}(\tau) = \Lambda'_{21}(\tau) = \Lambda'_{13}(\tau) = \Lambda'_{31}(\tau) = 0 \quad (180)$$

Finally taking the Hamiltonian (128) and given that to reproduce this Hamiltonian we need to impose in (5) that $V_{10}(t) = \frac{\Omega}{2}$, $\varepsilon_0(t) = 0$ and $\varepsilon_1(t) = \delta$, then we obtain using the equation (67) that $\text{Det}(\overline{H_S}) = -\frac{\Omega_r^2}{4}$, $\text{Tr}(\overline{H_S}) = \epsilon$. Now $\eta = \sqrt{\epsilon^2 + \Omega_r^2}$ and using the equation (84) we have that:

$$f_k = \frac{g_k \left(1 - \frac{\epsilon \tanh\left(\frac{\beta\eta}{2}\right)}{\eta} \right)}{1 - \frac{\tanh\left(\frac{\beta\eta}{2}\right)}{\eta} \left(\epsilon - \frac{\Omega_r^2 \coth\left(\frac{\beta\omega_k}{2}\right)}{2\omega_k} \right)} \quad (181)$$

$$= \frac{g_k \left(1 - \frac{\epsilon \tanh\left(\frac{\beta\eta}{2}\right)}{\eta} \right)}{1 - \frac{\epsilon \tanh\left(\frac{\beta\eta}{2}\right)}{\eta} \left(1 - \frac{\Omega_r^2 \coth\left(\frac{\beta\omega_k}{2}\right)}{2\epsilon\omega_k} \right)} \quad (182)$$

This shows that the expression obtained reproduces the variational parameters of the time-independent model of the reference. In general we can see that the time-independent model studied can be reproduced using the master equation (99) under a time-independent approach providing similar results.

Given that the Hamiltonian of this system is time-independent, then $U_S(t) U_S^\dagger(t - \tau) = U_S(\tau)$. From the equation (??) and using the fact that

$$\tilde{A}_j(t - \tau, t) = U_S(\tau) A_j U_S(-\tau) \quad (183)$$

$$= \sum_{\zeta} e^{i\zeta\tau} A_i(-\zeta) \quad (184)$$

$$= \sum_{\zeta} e^{-i\zeta\tau} A_i(\zeta) \quad (185)$$

because the matrices $U_S(t)$ and $U_S(t - \tau)$ commute from the fact that $H_S(t)$ and $H_S(t - \tau)$ commute as well for time independent Hamiltonians. The master equation is equal to:

$$\frac{d\bar{\rho}_S(t)}{dt} = -i[H_S(t), \bar{\rho}_S(t)] - \frac{1}{2} \sum_{ij} \sum_{\zeta} \gamma_{ij}(\zeta, t) \left[A_i, A_j(\zeta) \bar{\rho}_S(t) - \bar{\rho}_S(t) A_j^\dagger(\zeta) \right] \quad (186)$$

$$- \sum_{ij} \sum_{\zeta} S_{ij}(\zeta, t) \left[A_i, A_j(\zeta) \bar{\rho}_S(t) + \bar{\rho}_S(t) A_j^\dagger(\zeta) \right] \quad (187)$$

where $A_j^\dagger(\zeta) = A(-\zeta)$, as we can see the equation (187) contains the rates and energy shifts $\gamma_{ij}(\zeta, t) = 2\Re(K_{ij}(\zeta, t))$ and $S_{ij}(\zeta, t) = \Im(K_{ij}(\zeta, t))$, respectively, defined in terms of the response functions

$$K_{ij}(\zeta, t) = \int_0^t \Lambda'_{ij}(\tau) e^{i\zeta\tau} d\tau \quad (188)$$

B. Time-dependent polaron quantum master equation

Following the reference [1], when $\Omega_k \ll \omega_k$ then $f_k \approx g_k$ so we recover the full polaron transformation. It means from the equation (72) that $B_z = 0$. The Hamiltonian studied is given by:

$$H = \left(\delta + \sum_{\mathbf{k}} \left(g_{\mathbf{k}} b_{\mathbf{k}}^\dagger + g_{\mathbf{k}}^* b_{\mathbf{k}} \right) \right) |1\rangle \langle 1| + \frac{\Omega(t)}{2} \sigma_x + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \quad (189)$$

If $f_{\mathbf{k}} \approx g_{\mathbf{k}}$ then $B(\tau) = B$, so B is independent of the time. In order to reproduce the Hamiltonian of the equation (189) using the Hamiltonian of the equation (1) we can say that $\delta = \varepsilon_1(t)$, $\varepsilon_0(t) = 0$, $V_{10}(t) = \frac{\Omega(t)}{2}$. Now given that $f_{\mathbf{k}} \approx g_{\mathbf{k}}$ then, in this case and using the equation (75) and (80) we obtain the following transformed Hamiltonians:

$$\overline{H}_S = (\delta + R_1) |1\rangle \langle 1| + \frac{B\sigma_x}{2} \Omega(t) \quad (190)$$

$$\overline{H}_I = \frac{\Omega(t)}{2} (B_x \sigma_x + B_y \sigma_y) \quad (191)$$

In this case $R_1 = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 - 2\alpha_{\mathbf{k}} g_{\mathbf{k}})$ from (27) and given that $f_{\mathbf{k}} \approx g_{\mathbf{k}}$ and $\alpha_{\mathbf{k}} = g_{\mathbf{k}}/\omega_{\mathbf{k}}$ then $R_1 = \sum_{\mathbf{k}} (-\omega_{\mathbf{k}}^{-1} |g_{\mathbf{k}}|^2) = \sum_{\mathbf{k}} (-\omega_{\mathbf{k}} |\alpha_{\mathbf{k}}|^2)$ as expected, take $\delta + R_1 = \delta'$. If $F(\omega_{\mathbf{k}}) = 1$ and using the equations (102)-(109) we can deduce that the only terms that survive are $\Lambda_{11}(\tau)$ and $\Lambda_{22}(\tau)$. The phonon propagator for this case is:

$$\phi(\tau) = \int_0^\infty \frac{J(\omega)}{\omega^2} G_+(\tau) d\omega \quad (192)$$

Writing $G_+(\tau) = \coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\tau) - i \sin(\omega\tau)$ so (192) can be written as:

$$\phi(\tau) = \int_0^\infty \frac{J(\omega)}{\omega^2} \left(\coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\tau) - i \sin(\omega\tau) \right) d\omega \quad (193)$$

Writing the interaction Hamiltonian (191) in the similar way to the equation (80) allow us to write $A_1 = \sigma_x$, $A_2 = \sigma_y$, $B_1(t) = B_x$, $B_2(t) = B_y$ and $C_1(t) = \frac{\Omega(t)}{2} = C_2(t)$. Now taking the equation (75) with $\delta'|1\rangle\langle 1| = \frac{\delta'}{2}\sigma_z + \frac{\delta'}{2}\mathbb{I}$ help us to reproduce the hamiltonian of the reference [2]. Then $\overline{H_S}$ is equal to:

$$\overline{H_S} = \frac{\delta'}{2}\sigma_z + \frac{B\sigma_x}{2}\Omega(t) \quad (194)$$

As we can see the function B is a time-independent function because we consider that g_k doesn't depend of the time. In this case the relevant correlation functions are given by:

$$\Lambda_{11}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) \quad (195)$$

$$= \frac{B^2}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \quad (196)$$

$$\Lambda_{22}(\tau) = \text{Tr}_B \left(\tilde{B}_2(\tau) \tilde{B}_2(0) \rho_B \right) \quad (197)$$

$$= \frac{B^2}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \quad (198)$$

These functions match with the equations $\Lambda_x(\tau)$ and $\Lambda_y(\tau)$ of the reference [2] and $\Lambda_i(\tau) = \Lambda_i(-\tau)$ for $i \in \{x, y\}$ respectively. The master equation for this section based on the equation(99) is:

$$\frac{d\overline{\rho_S}(t)}{dt} = -i \left[\frac{\delta'}{2}\sigma_z + \frac{\Omega_r(t)\sigma_x}{2}, \rho_S(t) \right] - \sum_{i=1}^2 \int_0^t d\tau \left(C_i(t) C_i(t-\tau) \Lambda_{ii}(\tau) \left[A_i, \tilde{A}_i(t-\tau, t) \rho_S(t) \right] \right. \quad (199)$$

$$\left. + C_i(t) C_i(t-\tau) \Lambda_{ii}(-\tau) \left[\rho_S(t) \tilde{A}_i(t-\tau, t), A_i \right] \right) \quad (200)$$

Replacing $C_i(t) = \frac{\Omega(t)}{2}$ and $\tilde{A}_i(t-\tau, t) = \tilde{\sigma}_i(t-\tau, t)$, also using the equations (196) and (198) on the equation (200) we obtain that:

$$\frac{d\overline{\rho_S}(t)}{dt} = -\frac{i}{2} [\delta'\sigma_z + \Omega_r(t)\sigma_x, \rho_S(t)] - \frac{\Omega(t)}{4} \int_0^t d\tau \Omega(t-\tau) ([\sigma_x, \tilde{\sigma}_x(t-\tau, t) \rho_S(t)] \Lambda_x(\tau) \quad (201)$$

$$+ [\sigma_y, \tilde{\sigma}_y(t-\tau, t) \rho_S(t)] \Lambda_y(\tau) + [\rho_S(t) \tilde{\sigma}_x(t-\tau, t), \sigma_x] \Lambda_x(\tau) + [\rho_S(t) \tilde{\sigma}_y(t-\tau, t), \sigma_y] \Lambda_y(\tau)) \quad (202)$$

As we can see $[A_j, \tilde{A}_i(t-\tau, t) \rho_S(t)]^\dagger = [\rho_S(t) \tilde{A}_i(t-\tau, t), A_j]$, $\Lambda_x(\tau) = \Lambda_x(-\tau)$ and $\Lambda_y(\tau) = \Lambda_y(-\tau)$, so the result obtained is the same master equation (21) of the reference [2] extended in the hermitian conjugate.

C. Time-Dependent Weak-Coupling Limit

In order to prove that the master equation deduced reproduces the equation (S17) of the reference [3] we will impose that $F(\omega) = 0$, so there is no transformation in this case. As we can see from the definition (101) the only term that survives is $\Lambda_{33}(\tau)$. Taking $\hbar = 1$ the Hamiltonian of the reference can be written in the form:

$$H = \Delta |1\rangle\langle 1| + \frac{\Omega(t)}{2} (|1\rangle\langle 0| + |0\rangle\langle 1|) + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + |1\rangle\langle 1| \sum_{\mathbf{k}} (g_{\mathbf{k}} b_{\mathbf{k}}^\dagger + g_{\mathbf{k}}^* b_{\mathbf{k}}) \quad (203)$$

Using the equation (??), from the fact that the Hamiltonian is time-independent in the evolution time allow us to write:

$$\frac{d\rho_S}{dt} = -i[H_S(t), \rho_S(t)] - \frac{1}{2} \sum_{\zeta} \gamma_{33}(\zeta, t) \left[A_3, A_3(\zeta) \rho_S(t) - \rho_S(t) A_3^\dagger(\zeta) \right] \quad (204)$$

$$- \sum_{\zeta} S_{33}(\zeta, t) \left[A_3, A_3(\zeta) \rho_S(t) + \rho_S(t) A_3^\dagger(\zeta) \right] \quad (205)$$

The correlation functions are relevant if $F(\omega) = 0$ for the weak-coupling approximation are:

$$\Lambda_{33}(\tau) = \int_0^\infty d\omega J(\omega) G_+(\tau) \quad (206)$$

$$\Lambda_{33}(-\tau) = \int_0^\infty d\omega J(\omega) G_+(-\tau) \quad (207)$$

In our case $A_3 = \frac{\mathbb{I} + \sigma_z}{2}$, the equation (205) can be transformed in

$$\frac{d\rho_S}{dt} = -i[H_S(t), \rho_S(t)] - \sum_{\zeta} (K_{33}(\zeta, t) [A_3, A_3(\zeta) \rho_S(t)] + K_{33}^*(\zeta, t) [\rho_S(t) A_3(\zeta), A_3]) \quad (208)$$

As the paper suggest we will consider that the quantum system is in resonance, so $\Delta = 0$ and furthermore, the relaxation time of the bath is less than the evolution time to be considered, so the frequency of the Rabi frequency of the laser can be taken as constant and equal to $\tilde{\Omega}$. To find the matrices $A_3(\zeta)$, we have to remember that $H_S = \frac{\Omega(t)}{2} (|1\rangle\langle 0| + |0\rangle\langle 1|)$, this Hamiltonian have the following eigenvalues and eigenvectors:

$$\lambda_+ = \frac{\tilde{\Omega}}{2} \quad (209)$$

$$|+\rangle = \frac{1}{\sqrt{2}} (|1\rangle + |0\rangle) \quad (210)$$

$$\lambda_- = -\frac{\tilde{\Omega}}{2} \quad (211)$$

$$|-\rangle = \frac{1}{\sqrt{2}} (-|1\rangle + |0\rangle) \quad (212)$$

The elements of the decomposition matrices are:

$$\langle + | \frac{1 + \sigma_z}{2} | + \rangle = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad (213)$$

$$= \frac{1}{2} \quad (214)$$

$$\langle - | \frac{1 + \sigma_z}{2} | - \rangle = \frac{1}{2} \begin{pmatrix} -1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} -1 \\ 1 \end{pmatrix} \quad (215)$$

$$= \frac{1}{2} \quad (216)$$

$$\langle - | \frac{1 + \sigma_z}{2} | + \rangle = \frac{1}{2} \begin{pmatrix} -1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad (217)$$

$$= -\frac{1}{2} \quad (218)$$

The decomposition matrices are

$$A_3(0) = \frac{1}{2} |+\rangle \langle +| + \frac{1}{2} |-\rangle \langle -| \quad (219)$$

$$= \frac{\mathbb{I}}{2} \quad (220)$$

$$A_3(\eta) = -\frac{1}{2} |-\rangle \langle +| \quad (221)$$

$$= \frac{1}{4} (\sigma_z + i\sigma_y) \quad (222)$$

$$A_3(-\eta) = -\frac{1}{2} |+\rangle \langle -| \quad (223)$$

$$= \frac{1}{4} (\sigma_z - i\sigma_y) \quad (224)$$

Neglecting the term proportional to the identity in the Hamiltonian we obtain that:

$$\frac{d\rho_S(t)}{dt} = -i\frac{\tilde{\Omega}}{2} [\sigma_x, \rho_S(t)] - K_{33}(\tilde{\Omega}, t) \left[\frac{\sigma_z}{2}, \frac{1}{4}(\sigma_z + i\sigma_y) \rho_S(t) \right] - K_{33}(-\tilde{\Omega}, t) \left[\frac{\sigma_z}{2}, \frac{1}{4}(\sigma_z - i\sigma_y) \rho_S(t) \right] \quad (225)$$

$$- K_{33}^*(\tilde{\Omega}, t) \left[\rho_S(t) \frac{1}{4}(\sigma_z + i\sigma_y), \frac{\sigma_z}{2} \right] - K_{33}^*(-\tilde{\Omega}, t) \left[\rho_S(t) \frac{1}{4}(\sigma_z - i\sigma_y), \frac{\sigma_z}{2} \right] \quad (226)$$

Calculating the response functions extending the upper limit of τ to ∞ , we obtain:

$$K_{33}(\tilde{\Omega}) = \int_0^\infty \int_0^\infty J(\omega) G_+(\tau) e^{i\tilde{\Omega}\tau} d\tau d\omega \quad (227)$$

$$= \int_0^\infty \int_0^\infty J(\omega) e^{i\tilde{\Omega}\tau} ((n(\omega) + 1) e^{-i\tau\omega} + n(\omega) e^{i\tau\omega}) d\tau d\omega \quad (228)$$

$$= \int_0^\infty \int_0^\infty J(\omega) e^{i\tilde{\Omega}\tau} (n(\omega) + 1) e^{-i\tau\omega} d\tau d\omega \quad (229)$$

$$= \int_0^\infty \int_0^\infty J(\omega) (n(\omega) + 1) e^{i\tilde{\Omega}\tau - i\tau\omega} d\tau d\omega \quad (230)$$

$$= \int_0^\infty J(\omega) (n(\omega) + 1) \pi \delta(\tilde{\Omega} - \omega) d\omega \quad (231)$$

$$= \pi J(\tilde{\Omega}) (n(\tilde{\Omega}) + 1) \quad (232)$$

$$K_{33}(-\tilde{\Omega}) = \int_0^\infty \int_0^\infty J(\omega) G_+(\tau) e^{-i\tilde{\Omega}\tau} d\tau d\omega \quad (233)$$

$$= \int_0^\infty \int_0^\infty J(\omega) e^{-i\tilde{\Omega}\tau} ((n(\omega) + 1) e^{-i\tau\omega} + n(\omega) e^{i\tau\omega}) d\tau d\omega \quad (234)$$

$$= \int_0^\infty \int_0^\infty J(\omega) e^{-i\tilde{\Omega}\tau} n(\omega) e^{i\tau\omega} d\tau d\omega \quad (235)$$

$$= \int_0^\infty \int_0^\infty J(\omega) n(\omega) e^{-i\tilde{\Omega}\tau + i\tau\omega} d\tau d\omega \quad (236)$$

$$= \int_0^\infty J(\omega) n(\omega) \pi \delta(-\tilde{\Omega} + \omega) d\omega \quad (237)$$

$$= \pi J(\tilde{\Omega}) n(\tilde{\Omega}) \quad (238)$$

Here we have used $\int_0^\infty ds e^{\pm i\varepsilon s} = \pi \delta(\varepsilon) \pm i \frac{V.P.}{\varepsilon}$, where *V.P.* denotes the Cauchy's principal value. These principal values are ignored because they lead to small renormalizations of the Hamiltonian. Furthermore we don't take account of value associated to the matrix $A_3(0)$ because the spectral density $J(\omega)$ is equal to zero when $\omega = 0$. Replacing in the equation (??) lead us to obtain:

$$\frac{d\rho_S(t)}{dt} = -i\frac{\tilde{\Omega}}{2} [\sigma_x, \rho_S(t)] - \frac{\pi}{8} J(\tilde{\Omega}) \left((n(\tilde{\Omega}) + 1) [\sigma_z, (\sigma_z + i\sigma_y) \rho_S(t)] + n(\tilde{\Omega}) [\sigma_z, (\sigma_z - i\sigma_y) \rho_S(t)] \right) \quad (239)$$

$$- \frac{\pi}{8} J(\tilde{\Omega}) \left((n(\tilde{\Omega}) + 1) [\rho_S(t) (\sigma_z + i\sigma_y), \sigma_z] + n(\tilde{\Omega}) [\rho_S(t) (\sigma_z - i\sigma_y), \sigma_z] \right) \quad (240)$$

This is the same result than the equation (S17), so we have proved that our general master equation allows to reproduce the results of the weak-coupling time-dependent. Now the master equation in the evolution time is given by

$$\frac{d\rho_S(t)}{dt} = -i\frac{\Omega(t)}{2} [\sigma_x, \rho_S(t)] - \frac{\pi}{8} J(\Omega(t)) ((n(\Omega(t)) + 1) [\sigma_z, (\sigma_z + i\sigma_y) \rho_S(t)] + n(\Omega(t)) [\sigma_z, (\sigma_z - i\sigma_y) \rho_S(t)]) \quad (241)$$

$$- \frac{\pi}{8} J(\Omega(t)) ((n(\Omega(t)) + 1) [\rho_S(t) (\sigma_z + i\sigma_y), \sigma_z] + n(\Omega(t)) [\rho_S(t) (\sigma_z - i\sigma_y), \sigma_z]) \quad (242)$$

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