

Quantum dynamics for a quantum dot qubit using a variational master equation for a time dependent Hamiltonian.

April 26, 2018

1 Variational transformation.

The Hamiltonian associated for a quantum dot qubit in presence of a bath can be written in the following way :

$$H = H_{\text{OQS}} + H_{\text{I}} + H_{\text{B}} \quad (1)$$

$$H_{\text{OQS}} = \delta_1(t) + \delta_x(t) \sigma_x + \delta_y(t) \sigma_y + \delta_z(t) \sigma_z \quad (2)$$

$$H_{\text{I}} = \sum_{\mathbf{k}} g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}) |1\rangle\langle 1| \quad (3)$$

$$H_{\text{B}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \quad (4)$$

$$H = \varepsilon_1(t) |1\rangle\langle 1| + \varepsilon_0(t) |0\rangle\langle 0| + V_{10}(t) |1\rangle\langle 0| + V_{01}(t) |0\rangle\langle 1| + \sum_{\mathbf{k}} g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}) |1\rangle\langle 1| + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \quad (5)$$

Let's consider the following canonical transformation:

$$e^{\pm V} = |0\rangle\langle 0| + |1\rangle\langle 1| \prod_{\mathbf{k}} D(\pm \alpha_{\mathbf{k}}) \quad (6)$$

Where $B_{\pm} = \prod_{\mathbf{k}} D(\pm \alpha_{\mathbf{k}}) = \prod_{\mathbf{k}} \exp\left(\pm \alpha_{\mathbf{k}} (b_{\mathbf{k}}^\dagger - b_{\mathbf{k}})\right)$ is a product of displacement operators and $\alpha_{\mathbf{k}} = f_{\mathbf{k}}/\omega_{\mathbf{k}}$ assumed to be real, here $\{f_{\mathbf{k}}\}$ represent the set of variational parameters that shall be chosen in order to minimize the free energy bound defined above. As we can see the operators B_+ and B_- are inverse of each other so $B_+ B_- = 1$.

Applying the canonical transformation (6) to H we can obtain the Hamiltonian transformed:

$$H_V = e^V H e^{-V} = \overline{H} \quad (7)$$

The results obtained from the application of the mentioned canonical transformation to each part of the Hamiltonian of the equation (5) can be evaluated using the linear nature of the transformation, for a given operator A we have the following result for the transformation:

$$\overline{A} = e^V A e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) A (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \quad (8)$$

We summarize the results obtained from (7) to the relevant parts of the Hamiltonian (5) using the formula (8):

$$\begin{aligned} \overline{|0\rangle \langle 0|} &= e^V |0\rangle \langle 0| e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |0\rangle \langle 0| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= B_+ |0\rangle \langle 0| B_- = |0\rangle \langle 0| \end{aligned} \quad (9)$$

$$\begin{aligned} \overline{|1\rangle \langle 1|} &= e^V |1\rangle \langle 1| e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |1\rangle \langle 1| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= B_+ |1\rangle \langle 1| B_- = |1\rangle \langle 1| \end{aligned} \quad (10)$$

$$\begin{aligned} \overline{|0\rangle \langle 1|} &= e^V |0\rangle \langle 1| e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |0\rangle \langle 1| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= (|0\rangle \langle 0|) |0\rangle \langle 1| (|1\rangle \langle 1| B_-) = |0\rangle \langle 1| B_- \end{aligned} \quad (11)$$

$$\begin{aligned} \overline{|1\rangle \langle 0|} &= e^V |1\rangle \langle 0| e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) |1\rangle \langle 0| (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= (|1\rangle \langle 1| B_+) |1\rangle \langle 0| (|0\rangle \langle 0|) = |1\rangle \langle 0| B_+ \end{aligned} \quad (12)$$

$$\begin{aligned} \overline{b_{\mathbf{k}}} &= e^V b_{\mathbf{k}} e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) b_{\mathbf{k}} (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= (|0\rangle \langle 0|) b_{\mathbf{k}} (|0\rangle \langle 0|) + (|1\rangle \langle 1| B_+) b_{\mathbf{k}} (|1\rangle \langle 1| B_-) = |0\rangle \langle 0| b_{\mathbf{k}} + |1\rangle \langle 1| (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \end{aligned} \quad (13)$$

$$\begin{aligned} \overline{b_{\mathbf{k}}^\dagger} &= e^V b_{\mathbf{k}}^\dagger e^{-V} = (|0\rangle \langle 0| + |1\rangle \langle 1| B_+) b_{\mathbf{k}}^\dagger (|0\rangle \langle 0| + |1\rangle \langle 1| B_-) \\ &= (|0\rangle \langle 0|) b_{\mathbf{k}}^\dagger (|0\rangle \langle 0|) + (|1\rangle \langle 1| B_+) b_{\mathbf{k}}^\dagger (|1\rangle \langle 1| B_-) = |0\rangle \langle 0| b_{\mathbf{k}}^\dagger + |1\rangle \langle 1| (b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}}) \end{aligned} \quad (14)$$

For this case we used the following relationships

$$B_+ b_{\mathbf{k}} B_- = b_{\mathbf{k}} - \alpha_{\mathbf{k}} \quad B_+ b_{\mathbf{k}}^\dagger B_- = b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}} \quad (15)$$

The transformation of H divided by relevant part is shown:

$$\overline{\varepsilon_1(t) |1\rangle \langle 1|} = \varepsilon_1(t) |1\rangle \langle 1| \quad (16)$$

$$\overline{\varepsilon_0(t) |0\rangle \langle 0|} = \varepsilon_0(t) |0\rangle \langle 0| \quad (17)$$

$$\overline{V_{10}(t) |1\rangle \langle 0|} = V_{10}(t) |1\rangle \langle 0| B_+ \quad (18)$$

$$\overline{V_{01}(t) |0\rangle \langle 1|} = V_{01}(t) |0\rangle \langle 1| B_- \quad (19)$$

$$\overline{g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}) |1\rangle \langle 1|} = g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}}) |1\rangle \langle 1| \quad (20)$$

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

$$+ \omega_{\mathbf{k}} D(\alpha_{\mathbf{k}}) b_{\mathbf{k}}^\dagger D(-\alpha_{\mathbf{k}}) D(\alpha_{\mathbf{k}}) b_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) |1\rangle \langle 1| = \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} |0\rangle \langle 0| + \omega_{\mathbf{k}} (b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}}) (b_{\mathbf{k}} - 2\alpha_{\mathbf{k}}) |1\rangle \langle 1|$$

$$= \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} + \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 |1\rangle \langle 1| - \omega_{\mathbf{k}} \alpha_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}}) |1\rangle \langle 1| \quad (21)$$

Respect to H_{OQS} , H_I , H_B as shown in (2), (3) and (4) the transformed hamiltonian of each one of these terms is:

$$\overline{H_{\text{OQS}}} = \delta_x(t) \overline{\sigma_x} + \delta_y(t) \overline{\sigma_y} + \delta_z(t) \overline{\sigma_z} + \delta_1(t)$$

$$= \delta_x(t) (|1\rangle \langle 0| B_+ + |0\rangle \langle 1| B_-) + \delta_y(t) i (|0\rangle \langle 1| B_- - |1\rangle \langle 0| B_+) + \delta_z(t) (|1\rangle \langle 1| - |0\rangle \langle 0|) + \delta_1(t) \quad (22)$$

$$\begin{aligned} \overline{H_I} &= \sum_{\mathbf{k}} g_{\mathbf{k}} (\bar{b}_{\mathbf{k}}^\dagger + \bar{b}_{\mathbf{k}}) |1\rangle \langle 1| = \sum_{\mathbf{k}} g_{\mathbf{k}} \left((b_{\mathbf{k}}^\dagger - \alpha_{\mathbf{k}}) + (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) \right) |1\rangle \langle 1| \\ &= \sum_{\mathbf{k}} g_{\mathbf{k}} (b_{\mathbf{k}}^\dagger + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}}) |1\rangle \langle 1| \end{aligned} \quad (23)$$

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

$$\begin{aligned}
& + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} - \alpha_{\mathbf{k}} \right) (b_{\mathbf{k}} - \alpha_{\mathbf{k}}) = (|0\rangle \langle 0| + |1\rangle \langle 1|) \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) \\
& + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \alpha_{\mathbf{k}} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) + |1\rangle \langle 1| \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2
\end{aligned} \tag{24}$$

Finally merging the precedent expressions we obtain the transformed Hamiltonian:

$$\begin{aligned}
H_V = & \varepsilon_1(t) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) |1\rangle \langle 0| B_+ + V_{01}(t) |0\rangle \langle 1| B_- + \sum_{\mathbf{k}} g_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} - 2\alpha_{\mathbf{k}} \right) |1\rangle \langle 1| \\
& + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} - \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) |1\rangle \langle 1| + \sum_{\mathbf{k}} \omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 |1\rangle \langle 1|
\end{aligned} \tag{25}$$

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

$$H_V = \overline{H_{\text{OQS}}} + \overline{H_B} + \overline{H_I} \tag{26}$$

Let's define:

$$R_1 = \sum_{\mathbf{k}} (\omega_{\mathbf{k}} \alpha_{\mathbf{k}}^2 - 2\alpha_{\mathbf{k}} g_{\mathbf{k}}) \quad B_z = \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) \left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} \right) \tag{27}$$

For our model we consider a stationary bath state:

$$\rho_B = \exp(-\beta H_B) / \text{Tr}(\exp(-\beta H_B)) \tag{28}$$

for the inverse temperature $\beta = 1/k_B T$.

Under this model it's straightforward to show that $\langle B_z \rangle_{H_B} = \text{Tr}(B_z \exp(-\beta H_B)) / \text{Tr}(\exp(-\beta H_B)) = 0$. Furthermore we find the renormalization factor $\langle B_{\pm} \rangle_{H_B} = B$ to be:

$$B = \exp \left(- (1/2) \sum_{\mathbf{k}} (\alpha_{\mathbf{k}})^2 \coth(\beta \omega_{\mathbf{k}}/2) \right) \tag{29}$$

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 $\overline{H_{\text{OQS}}}$ to treat it nonperturbatively 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 $\overline{H_{\text{OQS}}}$. The final form of the terms of the splitted Hamiltonian H_V is:

$$\overline{H_{\text{OQS}}} = (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \sigma_+ + V_{01}(t) B \sigma_- \tag{30}$$

$$\overline{H_I} = \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) \left(b_{\mathbf{k}}^\dagger + b_{\mathbf{k}} \right) |1\rangle \langle 1| + V_{10}(t) (\sigma_+ B_+ - \sigma_+ B) + V_{01}(t) (\sigma_- B_- - \sigma_- B) \quad (31)$$

$$H_B = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \quad (32)$$

The following convention give us the form of the Pauli matrices to be used:

$$\sigma_x = |1\rangle \langle 0| + |0\rangle \langle 1| \quad (33)$$

$$\sigma_y = -i|1\rangle \langle 0| + i|0\rangle \langle 1| \quad (34)$$

$$\sigma_z = -|0\rangle \langle 0| + |1\rangle \langle 1| \quad (35)$$

Let's consider the following Hermitian combinations:

$$B_x = \frac{B_+ + B_- - 2B}{2} \quad (36)$$

$$B_y = \frac{B_- - B_+}{2i} \quad (37)$$

$$B_z = \sum_{\mathbf{k}} (g_{\mathbf{k}} - f_{\mathbf{k}}) \left(b_{\mathbf{k}}^\dagger + b_{\mathbf{k}} \right) \quad (38)$$

$$\sigma_+ = \frac{\sigma_x + i\sigma_y}{2} \quad (39)$$

$$\sigma_- = \frac{\sigma_x - i\sigma_y}{2} \quad (40)$$

Writing the equations (30) and (31) using the precedent combinations we obtain that:

$$\begin{aligned} \overline{H_{\text{OQS}}} &= (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \sigma_+ + V_{01}(t) B \sigma_- \\ &= (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + V_{10}(t) B \frac{\sigma_x + i\sigma_y}{2} + V_{01}(t) B \frac{\sigma_x - i\sigma_y}{2} \\ &= (\varepsilon_1(t) + R_1) |1\rangle \langle 1| + \varepsilon_0(t) |0\rangle \langle 0| + \frac{B\sigma_x}{2} (V_{10}(t) + V_{01}(t)) + \frac{iB\sigma_y}{2} (V_{10}(t) - V_{01}(t)) \end{aligned} \quad (41)$$

$$\begin{aligned}
\overline{H_I} &= B_z |1\rangle \langle 1| + V_{10}(t) (\sigma_+ B_+ - \sigma_- B_-) + V_{01}(t) (\sigma_- B_- - \sigma_+ B_+) \\
&= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) \left(\frac{\sigma_x + i\sigma_y}{2} B_+ - \frac{\sigma_x + i\sigma_y}{2} B_- + \frac{\sigma_x - i\sigma_y}{2} B_- - \frac{\sigma_x - i\sigma_y}{2} B_+ \right) \\
&\quad + i\Im(V_{10}(t)) \left(\frac{\sigma_x + i\sigma_y}{2} B_+ - \frac{\sigma_x + i\sigma_y}{2} B_- - \frac{\sigma_x - i\sigma_y}{2} B_- + \frac{\sigma_x - i\sigma_y}{2} B_+ \right) \\
&= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) (B_x \sigma_x + B_y \sigma_y) + i\Im(V_{10}(t)) (iB_x \sigma_y - iB_y \sigma_x) \\
&= B_z |1\rangle \langle 1| + \Re(V_{10}(t)) (B_x \sigma_x + B_y \sigma_y) - \Im(V_{10}(t)) (B_x \sigma_y - B_y \sigma_x) \quad (42)
\end{aligned}$$

2 Free-energy minimization.

We can find the set of variational parameter $\{f_{\mathbf{k}}\}$ imposing the minimization of the free energy of the transformed Hamiltonian. We will use the Feynman-Bogoliubov upper bound of the free energy A_B :

$$A_B = -\frac{1}{\beta} \ln \left(\text{Tr} \left(e^{-\beta(\overline{H_{\text{OQS}}} + H_B)} \right) \right) + \langle H_I \rangle_{H_0} + O \left(\langle H_I^2 \rangle_{H_0} \right) \quad (43)$$

to minimize and obtain the set of variational parameters. Neglecting the higher order terms and using $\langle H_I \rangle_{H_0} = 0$ we can obtain the following condition to obtain the set $\{f_{\mathbf{k}}\}$:

$$\frac{\partial A_B}{\partial f_{\mathbf{k}}} = 0 \quad (44)$$

Using this condition and given that $[\overline{H_{\text{OQS}}}, H_B] = 0$ then $e^{-\beta(\overline{H_{\text{OQS}}} + H_B)} = e^{-\beta \overline{H_{\text{OQS}}}} e^{-\beta H_B}$, furthermore $\text{Tr} \left(e^{-\beta \overline{H_{\text{OQS}}}} e^{-\beta H_B} \right) = \text{Tr} \left(e^{-\beta \overline{H_{\text{OQS}}}} \right) \text{Tr} \left(e^{-\beta H_B} \right)$ from the fact that $\overline{H_{\text{OQS}}}$ 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 (44) in the following way:

$$\frac{\partial \text{Tr} \left(e^{-\beta \overline{H_{\text{OQS}}}} \right)}{\partial f_{\mathbf{k}}} = 0 \quad (45)$$

The variational parameters are:

$$f_{\mathbf{k}} = \frac{g_{\mathbf{k}} \left(1 - \frac{\tanh(\frac{\beta\eta}{2})}{\eta} (\varepsilon_1(t) + R_1 - \varepsilon_0(t)) \right)}{1 - \frac{\tanh(\frac{\beta\eta}{2})}{\eta} \left(\varepsilon_1(t) + R_1 - \varepsilon_0(t) - \frac{2|V_{10}(t)|^2 B^2}{\omega_{\mathbf{k}}} \coth\left(\frac{\beta\omega_{\mathbf{k}}}{2}\right) \right)} \quad (46)$$

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

3 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 $\chi(0) = |0\rangle\langle 0| \otimes \rho_B$, the transformed density operator is equal to:

$$\begin{aligned} e^V \chi(0) e^{-V} &= \left(|0\rangle\langle 0| + |1\rangle\langle 1| \prod_{\mathbf{k}} D(\alpha_{\mathbf{k}}) \right) (|0\rangle\langle 0| \otimes \rho_B) \left(|0\rangle\langle 0| + |1\rangle\langle 1| \prod_{\mathbf{k}} D(-\alpha_{\mathbf{k}}) \right) \\ &= |0\rangle\langle 0| \otimes \rho_B = \chi(0) \end{aligned} \quad (47)$$

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 (42) as:

$$H_I(t) = \sum_i C_i(t) (A_i \otimes B_i(t)) \quad (48)$$

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

$$\frac{d\tilde{\rho}_V}{dt} = - \int_0^t ds \text{Tr}_B \left[\tilde{H}_I(t), \left[\tilde{H}_I(s), \tilde{\rho}_V(t) \rho_B \right] \right] \quad (49)$$

here $\rho_V(t) = \text{Tr}_B(e^V \chi(t) e^{-V})$ describes the QD excitonic degrees of freedom in the variational frames, with Tr_B denoting a trace over the environment degrees of freedom. From the equation (48) and the interaction picture applied on H_I we find:

$$\tilde{H}_I(t) = U_O^\dagger(t) H_I(t) U_O(t) \quad (50)$$

where $U_O(t) = U_{\text{OQS}}(t) e^{-iH_B t}$ with

$$U_{\text{OQS}}(t) = \mathcal{T} \exp \left(-i \int_0^t dv \overline{H_{\text{OQS}}}(v) \right) \quad (51)$$

In this case the Schrödinger and the interaction picture coincide at $t = 0$, we use the time-ordering operator \mathcal{T} because in general $\bar{H}_{\text{OQS}}(t)$ doesn't commute with itself at two different times. We write the interaction Hamiltonian as:

$$\tilde{H}_I(t) = \sum_i C_i(t) \left(\tilde{A}_i \otimes \tilde{B}_i(t) \right) \quad (52)$$

$$\tilde{A}_i(t) = U_{\text{OQS}}^\dagger(t) A_i U_{\text{OQS}}(t) \quad (53)$$

$$\tilde{B}_i(t) = e^{iH_B t} B_i(t) e^{-iH_B t} \quad (54)$$

Using the equation (42) and returning to the Schrödinger picture, we then obtain:

$$\begin{aligned} \frac{d\rho_V}{dt} = & -i[H_{\text{OQS}}(t), \rho_V(t)] - \sum_{ij} \int_0^t d\tau (C_i(t) C_j(t-\tau) A_{ij}(\tau) [A_i, \tilde{A}_j(t-\tau, t) \rho_V(t)] \\ & + C_j(t) C_i(t-\tau) A_{ji}(-\tau) [\rho_V(t) \tilde{A}_j(t-\tau, t), A_i]) \end{aligned} \quad (55)$$

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

Here $\tilde{A}_j(t-\tau, t) = U_{\text{OQS}}^\dagger(t) U_{\text{OQS}}^\dagger(t-\tau) A_j U_{\text{OQS}}(t-\tau) U_{\text{OQS}}(t)$. The equation (55) 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:

$$A_{ij}(\tau) = \text{Tr}_B \left(\tilde{B}_i(t) \tilde{B}_j(s) \rho_B \right) = \text{Tr}_B \left(\tilde{B}_i(\tau) \tilde{B}_j(0) \rho_B \right) \quad (56)$$

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

$$A_{11}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \quad (57)$$

$$A_{22}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \quad (58)$$

$$A_{33}(\tau) = \int_0^\infty d\omega J(\omega) (1 - F(\omega, \tau))^2 G_+(\tau) \quad (59)$$

$$A_{32}(\tau) = B(\tau) \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega, \tau) (1 - F(\omega, \tau)) iG_-(\tau) \quad (60)$$

$$A_{23}(\tau) = -B(0) \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega, \tau) (1 - F(\omega, \tau)) iG_-(\tau) \quad (61)$$

$$A_{12}(\tau) = A_{21}(\tau) = A_{13}(\tau) = A_{31}(\tau) = 0 \quad (62)$$

In the continuum limit we will obtain the following expression:

$$B(\tau) = \exp \left(- (1/2) \int_0^\infty J(\omega) F(\omega, \tau)^2 \frac{\coth(\beta\omega/2) d\omega}{2} \right) \quad (63)$$

$$R(\tau) = \int_0^\infty J(\omega) F(\omega, \tau) \omega^{-1} (F(\omega, \tau) - 2) d\omega \quad (64)$$

Where the spectral density is $J(\omega) = \sum_k |g_{\mathbf{k}}|^2 \delta(\omega - \omega_{\mathbf{k}})$ and $f_{\mathbf{k}} = g_{\mathbf{k}} F(\omega_{\mathbf{k}})$, the phonon propagator given by:

$$\phi(\tau) = \int_0^\infty d\omega \frac{J(\omega)}{\omega^2} F(\omega, \tau)^2 G_+(\tau) \quad (65)$$

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 (56) allows us to write all the correlations functions as:

$$\Lambda(\tau) = \begin{pmatrix} A_{11}(\tau) & 0 & 0 & 0 & -A_{11}(\tau) \\ 0 & A_{22}(\tau) & A_{23}(\tau) & A_{22}(\tau) & 0 \\ 0 & A_{32}(\tau) & A_{33}(\tau) & A_{32}(\tau) & 0 \\ 0 & A_{22}(\tau) & A_{23}(\tau) & A_{22}(\tau) & 0 \\ -A_{11}(\tau) & 0 & 0 & 0 & A_{11}(\tau) \end{pmatrix} \quad (66)$$

The eigenvalues of the Hamiltonian H_{OQS} are given by the solution of the following algebraic equation:

$$\lambda^2 - \text{Tr}(\overline{H_{\text{OQS}}}) \lambda + \text{Det}(\overline{H_{\text{OQS}}}) = 0 \quad (67)$$

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 $H_{\text{OQS}} |\pm\rangle = \lambda_\pm |\pm\rangle$. Using this notation is possible to write $H_{\text{OQS}} = \lambda_+ |+\rangle \langle +| + \lambda_- |-\rangle \langle -|$.

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

$$\tilde{A}_i(\tau) = e^{i\overline{H_{\text{OQS}}}\tau} A_i e^{-i\overline{H_{\text{OQS}}}\tau} = \sum_{\zeta} e^{-i\zeta\tau} A_i(\zeta) \quad (68)$$

Where the sum is defined on the set of all the differences between the eigenvalues of the system, in our case $\zeta \in \{0, \pm\eta\}$. In order to show the explicit form of the matrices present in the RHS of the equation (68) for a general 2×2 matrix let's write the matrix A_i in the base $V = \{|+\rangle, |-\rangle\}$ in the following way:

$$A_i = \sum_{\alpha, \beta \in V} \langle \alpha | A_i | \beta \rangle |\alpha\rangle \langle \beta| \quad (69)$$

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

$$\begin{aligned}
e^{i\overline{H_{\text{OQS}}}\tau} &= e^{i(\lambda_+|+\rangle\langle +| + \lambda_-|-\rangle\langle -|)\tau} = e^{i\lambda_+|+\rangle\langle +|\tau} e^{i\lambda_-|-\rangle\langle -|\tau} \\
&= (|-\rangle\langle -| + e^{i\lambda_+\tau}|+\rangle\langle +|) (|+\rangle\langle +| + e^{i\lambda_-\tau}|-\rangle\langle -|) \\
&= e^{i\lambda_+\tau}|+\rangle\langle +| + e^{i\lambda_-\tau}|-\rangle\langle -|
\end{aligned} \tag{70}$$

Calculating (68) we find that:

$$\begin{aligned}
\tilde{A}_i(\tau) &= (e^{i\lambda_+\tau}|+\rangle\langle +| + e^{i\lambda_-\tau}|-\rangle\langle -|) \left(\sum_{\alpha, \beta \in V} \langle \alpha | A_i | \beta \rangle |\alpha\rangle\langle \beta| \right) (e^{-i\lambda_+\tau}|+\rangle\langle +| + e^{-i\lambda_-\tau}|-\rangle\langle -|) \\
&= \langle + | A_i | + \rangle |+\rangle\langle +| + e^{i\eta\tau} \langle + | A_i | - \rangle |+\rangle\langle -| + e^{-i\eta\tau} \langle - | A_i | + \rangle |-\rangle\langle +| + \langle - | A_i | - \rangle |-\rangle\langle -|
\end{aligned} \tag{71}$$

Here $\eta = \lambda_+ - \lambda_-$. Comparing the RHS of the equations (68) and (71) we obtain the form of the expansion matrices of the Fourier decomposition for a general 2×2 matrix:

$$A_i(0) = \langle + | A_i | + \rangle |+\rangle\langle +| + \langle - | A_i | - \rangle |-\rangle\langle -| \tag{72}$$

$$A_i(\zeta) = \langle + | A_i | - \rangle |+\rangle\langle -| \tag{73}$$

$$A_i(-\zeta) = \langle - | A_i | + \rangle |-\rangle\langle +| \tag{74}$$

Extending the Fourier decomposition we will obtain

$$\tilde{A}_j(t - \tau, t) = U_{\text{OQS}}(t - \tau) U_{\text{OQS}}^\dagger(t) A_j U_{\text{OQS}}(t) U_{\text{OQS}}^\dagger(t - \tau) = \sum_{\zeta, \zeta'} e^{-i\zeta\tau + i\tau(\zeta - \zeta')} A_i(\zeta, \zeta') \tag{75}$$

where ζ' belongs to the set of the differences of the eigenvalues of the Hamiltonian $H_{\text{OQS}}(t - \tau)$.

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 (68) to write the master equation (55) in the following neater form:

$$\begin{aligned}
\frac{d\rho_V}{dt} &= -i[H_{\text{OQS}}(t), \rho_V(t)] \\
&- \frac{1}{2} \sum_{ij} \sum_{\zeta, \zeta'} \gamma_{ij}(\zeta, \zeta', t) \left[A_i, A_j(\zeta, \zeta') \rho_V(t) - \rho_V(t) A_j^\dagger(\zeta, \zeta') \right] \\
&- \sum_{ij} \sum_{\zeta, \zeta'} S_{ij}(\zeta, \zeta', t) \left[A_i, A_j(\zeta, \zeta') \rho_V(t) + \rho_V(t) A_j^\dagger(\zeta, \zeta') \right]
\end{aligned} \tag{76}$$

as we can see the equation (76) 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

$$K_{ij}(\zeta, \zeta', t) = \int_0^t C_i(t) C_j(t - \tau) \Lambda_{ij}(\tau) e^{i\zeta\tau} e^{-it(\zeta - \zeta')} d\tau \quad (77)$$

If we extend the upper limit of integration to ∞ in the equation (76) 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.

4 Limit cases.

In order to show the plausibility of the master equation (76) 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.

4.1 Time-independent variational quantum master equation

At first let's show that the master equation (76) 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 (5) is given by:

$$H = \left(\delta + \sum_j g_k (b_k^\dagger + b_k) \right) |1\rangle \langle 1| + \frac{\Omega}{2} \sigma_x + \sum_k \omega_k b_k^\dagger b_k \quad (78)$$

After performing the transformation (7) on the Hamiltonian (78) it's possible to split that result in the following set of Hamiltonians:

$$\overline{H_{\text{OQS}}} = (\delta + R) |1\rangle \langle 1| + \frac{\Omega_r}{2} \sigma_x \quad (79)$$

$$\overline{H_I} = B_z |1\rangle \langle 1| + \frac{\Omega}{2} (B_x \sigma_x + B_y \sigma_y) \quad (80)$$

$$H_B = \sum_k \omega_k b_k^\dagger b_k \quad (81)$$

The Hamiltonian (78) differs from the transformed Hamiltonian H_{OQS} of the reference by a term proportional to the identity as $(\delta + R) |1\rangle \langle 1| - \frac{\delta}{2} \mathbb{I} = \frac{R}{2} \mathbb{I} + \frac{\varepsilon}{2} \sigma_z$. In this Hamiltonian we can write $A_i = \sigma_x$, $A_2 = \sigma_y$ 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 H_{OQS} .

$$\lambda_+ = \frac{R+\eta}{2} \quad |+\rangle = \frac{1}{\sqrt{(\varepsilon+\eta)^2 + \Omega_r^2}} \begin{pmatrix} \varepsilon+\eta \\ \Omega_r \end{pmatrix} \quad (82)$$

$$\lambda_- = \frac{R-\eta}{2} \quad |-\rangle = \frac{1}{\sqrt{(\varepsilon+\eta)^2 + \Omega_r^2}} \begin{pmatrix} -\Omega_r \\ \varepsilon+\eta \end{pmatrix} \quad (83)$$

Using this basis we can find the decomposition matrices using the equation (69), the time-independence of $U_{\text{OQS}}(t)U_{\text{OQS}}^\dagger(t-\tau) = U_{\text{OQS}}(\tau)$ to conclude that the matrices obtained don't need to be expanded in additional basis and the fact that $|+\rangle = \cos(\theta)|1\rangle + \sin(\theta)|0\rangle$ and $|-\rangle = -\sin(\theta)|1\rangle + \cos(\theta)|0\rangle$ with $\sin(\theta) = \frac{\Omega_r}{\sqrt{(\varepsilon+\eta)^2 + \Omega_r^2}}$ and $\cos(\theta) = \frac{\varepsilon+\eta}{\sqrt{(\varepsilon+\eta)^2 + \Omega_r^2}}$:

$$\langle + | \sigma_x | + \rangle = (\cos(\theta) \quad \sin(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = 2 \sin(\theta) \cos(\theta) = \sin(2\theta) \quad (84)$$

$$\langle - | \sigma_x | - \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} = -2 \sin(\theta) \cos(\theta) = -\sin(2\theta) \quad (85)$$

$$\langle - | \sigma_x | + \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = \cos^2(\theta) - \sin^2(\theta) = \cos(2\theta) \quad (86)$$

$$\langle + | \sigma_y | + \rangle = (\cos(\theta) \quad \sin(\theta)) \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = i \sin(\theta) \cos(\theta) - i \sin(\theta) \cos(\theta) = 0 \quad (87)$$

$$\langle - | \sigma_y | - \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} = -i \sin(\theta) \cos(\theta) + i \sin(\theta) \cos(\theta) = 0 \quad (88)$$

$$\langle - | \sigma_y | + \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = i \cos^2(\theta) + i \sin^2(\theta) = i \quad (89)$$

$$\langle + | \frac{1+\sigma_z}{2} | + \rangle = (\cos(\theta) \quad \sin(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = \cos^2(\theta) \quad (90)$$

$$\langle - | \frac{1+\sigma_z}{2} | - \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} -\sin(\theta) \\ \cos(\theta) \end{pmatrix} = \sin^2(\theta) \quad (91)$$

$$\langle - | \frac{1 + \sigma_z}{2} | + \rangle = (-\sin(\theta) \quad \cos(\theta)) \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} \cos(\theta) \\ \sin(\theta) \end{pmatrix} = -\sin(\theta) \cos(\theta) \quad (92)$$

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

$$A_1(0) = \sin(2\theta) (|+\rangle \langle +| - |-\rangle \langle -|) \quad (93)$$

$$A_1(\eta) = \cos(2\theta) |-\rangle \langle +| \quad (94)$$

$$A_2(0) = 0 \quad (95)$$

$$A_2(\eta) = i |-\rangle \langle +| \quad (96)$$

$$A_3(0) = \cos^2(\theta) |+\rangle \langle +| + \sin^2(\theta) |-\rangle \langle -| \quad (97)$$

$$A_3(\eta) = -\sin(\theta) \cos(\theta) |-\rangle \langle +| \quad (98)$$

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 (56) in the following way:

$$\Lambda'_{ij}(\tau) = C_i(t) C_j(t - \tau) \Lambda_{ij}(\tau) \quad (99)$$

Using the notation of the master equation (76), 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 H_{OQS} is time-independent then $B(t) = B$. Taking the equations(57) – (62) we find that the correlation functions of the reference [1] written in terms of the RHS of the equation (56) are equal to:

$$\begin{aligned} A'_{11}(\tau) &= \left(\frac{\Omega}{2}\right)^2 \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \left(\frac{\Omega}{2}\right)^2 \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \\ &= \frac{(\Omega B)^2}{8} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) = \frac{\Omega_r^2}{8} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) \end{aligned} \quad (100)$$

$$\begin{aligned} A'_{22}(\tau) &= \left(\frac{\Omega}{2}\right)^2 \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \left(\frac{\Omega}{2}\right)^2 \frac{B(\tau) B(0)}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \\ &= \frac{\Omega_r^2}{8} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) \end{aligned} \quad (101)$$

$$\Lambda'_{33}(\tau) = \int_0^\infty d\omega J(\omega) (1 - F(\omega))^2 G_+(\tau) \quad (102)$$

$$\Lambda'_{32}(\tau) = \frac{\Omega}{2} B(\tau) \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega) (1 - F(\omega)) iG_-(\tau) = \frac{\Omega_r}{2} \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega) (1 - F(\omega)) iG_-(\tau) \quad (103)$$

$$\Lambda_{23}(\tau) = -B(0) \frac{\Omega}{2} \int_0^\infty d\omega \frac{J(\omega)}{\omega} F(\omega) (1 - F(\omega)) iG_-(\tau) = -\Lambda'_{32}(\tau) \quad (104)$$

$$\Lambda'_{12}(\tau) = \Lambda'_{21}(\tau) = \Lambda'_{13}(\tau) = \Lambda'_{31}(\tau) = 0 \quad (105)$$

Finally taking the Hamiltonian (78) 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 (30) that $\text{Det}(\overline{H_{\text{OQS}}}) = -\left(\frac{\Omega B}{2}\right)^2 = -\frac{\Omega_r^2}{4}$, $\text{Tr}(\overline{H_{\text{OQS}}}) = \delta + R = \epsilon$. Now $\eta = \sqrt{(\text{Tr}(\overline{H_{\text{OQS}}}))^2 - 4\text{Det}(\overline{H_{\text{OQS}}})} = \sqrt{\epsilon^2 + \Omega_r^2}$ and using the equation (33) we have that:

$$f_k = \frac{g_k \left(1 - \frac{\epsilon \tanh(\frac{\beta\eta}{2})}{\eta}\right)}{1 - \frac{\tanh(\frac{\beta\eta}{2})}{\eta} \left(\epsilon - \frac{\Omega_r^2 \coth(\frac{\beta\omega_k}{2})}{2\omega_k}\right)} = \frac{g_k \left(1 - \frac{\epsilon \tanh(\frac{\beta\eta}{2})}{\eta}\right)}{1 - \frac{\epsilon \tanh(\frac{\beta\eta}{2})}{\eta} \left(1 - \frac{\Omega_r^2 \coth(\frac{\beta\omega_k}{2})}{2\epsilon\omega_k}\right)} \quad (106)$$

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 (76) under a time-independent approach providing similar results.

Given that the Hamiltonian of this system is time-independent, then $U_{\text{OQS}}(t) U_{\text{OQS}}^\dagger(t - \tau) = U_{\text{OQS}}(\tau)$. From the equation (76) and using the fact that $\tilde{A}_j(t - \tau, t) = U_{\text{OQS}}(\tau) A_j U_{\text{OQS}}(-\tau) = \sum_\zeta e^{i\zeta\tau} A_i(-\zeta) = \sum_\zeta e^{-i\zeta\tau} A_i(\zeta)$ then ζ' can be put equal to ζ because the matrices $U_{\text{OQS}}(t)$ and $U_{\text{OQS}}(t - \tau)$ commute. The master equation is equal to:

$$\begin{aligned} \frac{d\rho_V}{dt} &= -i[H_{\text{OQS}}(t), \rho_V(t)] \\ &- \frac{1}{2} \sum_{ij} \sum_{\zeta} \gamma_{ij}(\zeta, t) \left[A_i, A_j(\zeta) \rho_V(t) - \rho_V(t) A_j^\dagger(\zeta) \right] \\ &- \sum_{ij} \sum_{\zeta} S_{ij}(\zeta, t) \left[A_i, A_j(\zeta) \rho_V(t) + \rho_V(t) A_j^\dagger(\zeta) \right] \end{aligned} \quad (107)$$

where $A_j^\dagger(\zeta) = A(-\zeta)$, as we can see the equation (107) 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

$$K_{ij}(\zeta, t) = \int_0^t C_i(t) C_j(t - \tau) \Lambda_{ij}(\tau) e^{i\zeta\tau} d\tau \quad (108)$$

4.2 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 (38) that $B_z = 0$. The Hamiltonian studied is given by:

$$H = \left(\delta + \sum_{\mathbf{k}} \left(g_{\mathbf{k}} b_{\mathbf{k}}^\dagger + g_{\mathbf{k}}^* b_{\mathbf{k}} \right) \right) |1\rangle \langle 1| + \frac{\Omega(t)}{2} \sigma_x + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \quad (109)$$

Now given that $f_{\mathbf{k}} \approx g_{\mathbf{k}}$ then $B(\tau) = B$, so B is independent of the time. In order to reproduce the Hamiltonian of the equation (109) using the Hamiltonian of the equation (5) 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 (41) and (42) we obtain the following transformed Hamiltonians:

$$\overline{H_{\text{OQS}}} = (\delta + R_1) |1\rangle \langle 1| + \frac{B\sigma_x}{2} \Omega(t) \quad (110)$$

$$\overline{H_I} = \frac{\Omega(t)}{2} (B_x \sigma_x + B_y \sigma_y) \quad (111)$$

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 so $\delta + R_1 = \delta'$. If $F(\omega_{\mathbf{k}}) = 1$ and using the equations (57) – (62) we can deduce that the only terms that survive are $\Lambda_{11}(\tau)$ and $\Lambda_{22}(\tau)$. The phonon propagator for this case is:

$$\phi(\tau) = \int_0^\infty \frac{J(\omega)}{\omega^2} G_+(\tau) d\omega \quad (112)$$

Writing

$$G_+(\tau) = (n(\omega) + 1) e^{-i\tau\omega} + n(\omega) e^{-i\tau\omega} = (n(\omega) + 1 + n(\omega)) \cos(\omega\tau) + i(n(\omega) - n(\omega) - 1) \sin(\omega\tau)$$

$$= (2n(\omega) + 1) \cos(\omega\tau) - i \sin(\omega\tau) = \left(2 \frac{1}{e^{\beta\omega} - 1} + 1 \right) \cos(\omega\tau) - i \sin(\omega\tau)$$

$$= \left(\frac{e^{\beta\omega} + 1}{e^{\beta\omega} - 1} \right) \cos(\omega\tau) - i \sin(\omega\tau) = \coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\tau) - i \sin(\omega\tau) \quad (113)$$

So the phonon propagator is given by:

$$\phi(\tau) = \int_0^\infty \frac{J(\omega)}{\omega^2} \left(\coth\left(\frac{\beta\omega}{2}\right) \cos(\omega\tau) - i \sin(\omega\tau) \right) d\omega \quad (114)$$

Writing the interaction Hamiltonian (109) in the similar way to the equation (48) 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 (55) 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 H_{OQS} is equal to:

$$\overline{H_{\text{OQS}}} = \frac{\delta'}{2}\sigma_z + \frac{B\sigma_x}{2}\Omega(t) \quad (115)$$

As we can see the function B is a time-independent function because we consider that $g_{\mathbf{k}}$ doesn't depend of the time. In this case the relevant correlation functions are given by:

$$A_{11}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \frac{B^2}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} - 2 \right) = \Lambda_x(\tau) = \Lambda_x(-\tau) \quad (116)$$

$$A_{22}(\tau) = \text{Tr}_B \left(\tilde{B}_1(\tau) \tilde{B}_1(0) \rho_B \right) = \frac{B^2}{2} \left(e^{\phi(\tau)} + e^{-\phi(\tau)} \right) = \Lambda_y(\tau) = \Lambda_y(-\tau) \quad (117)$$

These functions match with the equations $\Lambda_x(\tau)$ and $\Lambda_y(\tau)$ of the reference [2]. The master equation for this section based on the equation (55) is:

$$\begin{aligned} \frac{d\rho_V}{dt} = & -i \left[\frac{\delta'}{2}\sigma_z + \frac{\Omega_r(t)\sigma_x}{2}, \rho_V(t) \right] - \sum_{i=1}^2 \int_0^t d\tau (C_i(t) C_i(t-\tau) \Lambda_{ii}(\tau) [A_i, \tilde{A}_i(t-\tau, t) \rho_V(t)] \\ & + C_i(t) C_i(t-\tau) \Lambda_{ii}(-\tau) [\rho_V(t) \tilde{A}_i(t-\tau, t), A_i]) \end{aligned} \quad (118)$$

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 (113) and (114) on the equation (115) we obtain that:

$$\begin{aligned} \frac{d\rho_V}{dt} = & -\frac{i}{2} [\delta'\sigma_z + \Omega_r(t)\sigma_x, \rho_V(t)] - \frac{\Omega(t)}{4} \int_0^t d\tau \Omega(t-\tau) ([\sigma_x, \tilde{\sigma}_x(t-\tau, t) \rho_V(t)] \Lambda_x(\tau) \\ & + [\sigma_y, \tilde{\sigma}_y(t-\tau, t) \rho_V(t)] \Lambda_y(\tau) + H.c.) \end{aligned} \quad (119)$$

Here $H.c.$ means Hermitian conjugate, as we can see $[A_j, \tilde{A}_i(t-\tau, t) \rho_V(t)]^\dagger = [\rho_V(t) \tilde{A}_i(t-\tau, t), A_j]$, $\Lambda_x(\tau) = \Lambda_x(-\tau)$ and $\Lambda_y(\tau) = \Lambda_y(-\tau)$, so the inclusion of the Hermitian conjugate is valid and the result obtained is the same master equation (21) of the reference [2].

4.3 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 of $\Lambda_{ij}(\tau)$ the only term that survives is $\Lambda_{33}(\tau)$. Taking $\hbar = 1$ the Hamiltonian of the reference can be written in the form:

$$H = \Delta |1\rangle \langle 1| + \frac{\Omega(t)}{2} (|1\rangle \langle 0| + |0\rangle \langle 1|) + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + |1\rangle \langle 1| \sum_{\mathbf{k}} \left(g_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} + g_{\mathbf{k}}^* b_{\mathbf{k}} \right) \quad (120)$$

Using the equation (75) from the fact that the Hamiltonian is time-independent in the evolution time allow us to write:

$$\begin{aligned} \frac{d\rho_V}{dt} = & -i[H_{\text{OQS}}(t), \rho_V(t)] - \frac{1}{2} \sum_{\zeta} \gamma_{33}(\zeta, t) \left[A_3, A_3(\zeta) \rho_V(t) - \rho_V(t) A_3^{\dagger}(\zeta) \right] \\ & - \sum_{\zeta} S_{33}(\zeta, t) \left[A_3, A_3(\zeta) \rho_V(t) + \rho_V(t) A_3^{\dagger}(\zeta) \right] \end{aligned} \quad (121)$$

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

$$A_{33}(\tau) = \int_0^{\infty} d\omega J(\omega) G_+(\tau) \quad (122)$$

$$A_{33}(-\tau) = \int_0^{\infty} d\omega J(\omega) G_+(-\tau) \quad (123)$$

In our case $A_3 = \frac{\mathbb{I} + \sigma_z}{2}$, the equation (121) can be transformed in the following equation

$$\frac{d\rho_V}{dt} = -i[H_{\text{OQS}}(t), \rho_V(t)] - \left(\sum_{\zeta} K_{33}(\zeta, t) [A_3, A_3(\zeta) \rho_V(t)] + \text{H.c.} \right) \quad (124)$$

Where *H.c.* denotes hermitian conjugate. 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 will remember that $H_{\text{OQS}} = \frac{\Omega(t)}{2} (|1\rangle \langle 0| + |0\rangle \langle 1|)$, this Hamiltonian have the following eigenvalues and eigenvectors:

$$\lambda_+ = \frac{\tilde{\Omega}}{2}, \quad |+\rangle = \frac{1}{\sqrt{2}} (|1\rangle + |0\rangle) \quad \lambda_- = -\frac{\tilde{\Omega}}{2}, \quad |-\rangle = \frac{1}{\sqrt{2}} (-|1\rangle + |0\rangle) \quad (125)$$

The elements of the decomposition matrices are:

$$\langle + | \frac{1 + \sigma_z}{2} | + \rangle = \frac{1}{2} \begin{pmatrix} 1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} = \frac{1}{2} \quad (126)$$

$$\langle - | \frac{1 + \sigma_z}{2} | - \rangle = \frac{1}{2} \begin{pmatrix} -1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} -1 \\ 1 \end{pmatrix} = \frac{1}{2} \quad (127)$$

$$\langle - | \frac{1 + \sigma_z}{2} | + \rangle = \frac{1}{2} \begin{pmatrix} -1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \\ 1 \end{pmatrix} = -\frac{1}{2} \quad (128)$$

The decomposition matrices are

$$A_3(0) = \frac{1}{2} | + \rangle \langle + | + \frac{1}{2} | - \rangle \langle - | = \frac{\mathbb{I}}{2} \quad (129)$$

$$A_3(\eta) = -\frac{1}{2} | - \rangle \langle + | = \frac{1}{4} (\sigma_z + i\sigma_y) \quad (130)$$

$$A_3(-\eta) = -\frac{1}{2} | + \rangle \langle - | = \frac{1}{4} (\sigma_z - i\sigma_y) \quad (131)$$

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

$$\begin{aligned} \frac{d\rho_V}{dt} = & -i\frac{\tilde{\Omega}}{2} [\sigma_x, \rho_V(t)] - (K_{33}(\tilde{\Omega}, t) \left[\frac{\sigma_z}{2}, \frac{1}{4} (\sigma_z + i\sigma_y) \rho_V(t) \right] \\ & + K_{33}(-\tilde{\Omega}, t) \left[\frac{\sigma_z}{2}, \frac{1}{4} (\sigma_z - i\sigma_y) \rho_V(t) \right] + H.c.) \end{aligned} \quad (132)$$

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

$$\begin{aligned} K_{33}(\tilde{\Omega}) &= \int_0^\infty \int_0^\infty J(\omega) G_+(\tau) e^{i\tilde{\Omega}\tau} d\tau d\omega \\ &= \int_0^\infty \int_0^\infty J(\omega) e^{i\tilde{\Omega}\tau} ((n(\omega) + 1) e^{-i\tau\omega} + n(\omega) e^{i\tau\omega}) d\tau d\omega \\ &= \int_0^\infty \int_0^\infty J(\omega) e^{i\tilde{\Omega}\tau} (n(\omega) + 1) e^{-i\tau\omega} d\tau d\omega = \int_0^\infty \int_0^\infty J(\omega) (n(\omega) + 1) e^{i\tilde{\Omega}\tau - i\tau\omega} d\tau d\omega \\ &= \int_0^\infty J(\omega) (n(\omega) + 1) \pi \delta(\tilde{\Omega} - \omega) d\omega = J(\tilde{\Omega}) (n(\tilde{\Omega}) + 1) \pi \end{aligned} \quad (133)$$

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

$$\begin{aligned}
&= \int_0^\infty \int_0^\infty J(\omega) e^{-i\tilde{\Omega}\tau} ((n(\omega) + 1) e^{-i\tau\omega} + n(\omega) e^{i\tau\omega}) d\tau d\omega \\
&= \int_0^\infty \int_0^\infty J(\omega) e^{-i\tilde{\Omega}\tau} n(\omega) e^{i\tau\omega} d\tau d\omega = \int_0^\infty \int_0^\infty J(\omega) n(\omega) e^{-i\tilde{\Omega}\tau + i\tau\omega} d\tau d\omega \\
&= \int_0^\infty J(\omega) n(\omega) \pi \delta(-\tilde{\Omega} + \omega) d\omega = J(\tilde{\Omega}) n(\tilde{\Omega}) \pi \quad (134)
\end{aligned}$$

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 (132) lead us to obtain:

$$\begin{aligned}
\frac{d\rho_V}{dt} &= -i \frac{\tilde{\Omega}}{2} [\sigma_x, \rho_V(t)] \\
&- \frac{\pi}{8} J(\tilde{\Omega}) \left((n(\tilde{\Omega}) + 1) [\sigma_z, (\sigma_z + i\sigma_y) \rho_V(t)] + n(\tilde{\Omega}) [\sigma_z, (\sigma_z - i\sigma_y) \rho_V(t)] + H.c. \right) \quad (135)
\end{aligned}$$

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.

5 Bibliography.

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