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:

$$H_T(t) = H_S(t) + H_I + H_B, \tag{1}$$

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

$$H_I = |0\rangle\langle 0| \sum_{\mathbf{k}} \left(g_{0\mathbf{k}} b_{\mathbf{k}}^{\dagger} + g_{0\mathbf{k}}^{*} b_{\mathbf{k}} \right) + |1\rangle\langle 1| \sum_{\mathbf{k}} \left(g_{1\mathbf{k}} b_{\mathbf{k}}^{\dagger} + g_{1\mathbf{k}}^{*} b_{\mathbf{k}} \right), \tag{3}$$

$$H_B = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}. \tag{4}$$

II. UNITARY TRANSFORMATION INTO THE VARIATIONALLY OPTIMIZABLE FRAME

We will apply to $H(t)$, the unitary transformation defined by $e^{\pm V}$ where is the variationally optimizable anti-Hermitian operator:

$$V \equiv |0\rangle\langle 0| \sum_{\mathbf{k}} \left(\frac{v_{0\mathbf{k}}}{\omega_{\mathbf{k}}} b_{\mathbf{k}}^{\dagger} - \frac{v_{0\mathbf{k}}^{*}}{\omega_{\mathbf{k}}} b_{\mathbf{k}} \right) + |1\rangle\langle 1| \sum_{\mathbf{k}} \left(\frac{v_{1\mathbf{k}}}{\omega_{\mathbf{k}}} b_{\mathbf{k}}^{\dagger} - \frac{v_{1\mathbf{k}}^{*}}{\omega_{\mathbf{k}}} b_{\mathbf{k}} \right) \tag{5}$$

in terms of the variational scalar parameters $v_{\mathbf{k}}$, which will soon be optimized in order to give the most accurate possible master equation for the system's dynamics in the presence of this bath. Operators O in the variational frame will be written as:

$$\overline{O} \equiv \mathrm{e}^{V} O \mathrm{e}^{-V}. \tag{6}$$

We get:

$$\overline{H(t)} = \varepsilon_0\,(t)\,|0\rangle\langle 0| + \varepsilon_1\,(t)\,|1\rangle\langle 1| + V_{10}\,(t)\,|1\rangle\langle 0| B_{1+}B_{0-} + V_{01}\,(t)\,|0\rangle\langle 1| B_{0+}B_{1-} + \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} \tag{7}$$

$$+ \sum_{\mathbf{k}} \left(|1\rangle\langle 1| \left(\frac{|v_{1\mathbf{k}}|^2}{\omega_{\mathbf{k}}} - \left(v_{1\mathbf{k}}^{*} b_{\mathbf{k}} + v_{1\mathbf{k}} b_{\mathbf{k}}^{\dagger} \right) \right) + |0\rangle\langle 0| \left(\frac{|v_{0\mathbf{k}}|^2}{\omega_{\mathbf{k}}} - \left(v_{0\mathbf{k}}^{*} b_{\mathbf{k}} + v_{0\mathbf{k}} b_{\mathbf{k}}^{\dagger} \right) \right) \right) \tag{8}$$

$$+ \sum_{\mathbf{k}} \left(|0\rangle\langle 0| \left(g_{0\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} - \frac{v_{0\mathbf{k}}^{*}}{\omega_{\mathbf{k}}} \right) + g_{0\mathbf{k}}^{*} \left(b_{\mathbf{k}} - \frac{v_{0\mathbf{k}}}{\omega_{\mathbf{k}}} \right) \right) + |1\rangle\langle 1| \left(g_{1\mathbf{k}} \left(b_{\mathbf{k}}^{\dagger} - \frac{v_{1\mathbf{k}}^{*}}{\omega_{\mathbf{k}}} \right) + g_{1\mathbf{k}}^{*} \left(b_{\mathbf{k}} - \frac{v_{1\mathbf{k}}}{\omega_{\mathbf{k}}} \right) \right) \right) \tag{9}$$

We assume that the bath starts equilibrium with inverse temperature $\beta = 1/k_{\mathrm{B}}T$:

$$\rho_B = \frac{e^{-\beta H_B}}{\mathrm{Tr}\left(e^{-\beta H_B}\right)} \tag{10}$$

With the following definitions and relations:

$$
\begin{aligned}
D\left(\pm v_{i\mathbf{k}}\right) &\equiv e^{\pm\left(\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}b_{\mathbf{k}}^{\dagger}-\frac{v_{i\mathbf{k}}^{*}}{\omega_{\mathbf{k}}}b_{\mathbf{k}}\right)}. && (11)\\
B_{i\pm} &\equiv e^{\pm\sum_{\mathbf{k}}\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\left(b_{\mathbf{k}}^{\dagger}-b_{\mathbf{k}}\right)} && (12)\\
&= \prod_{\mathbf{k}} D\left(\pm v_{i\mathbf{k}}\right) && (13)\\
B_i &\equiv \left\langle B_{i\pm}\right\rangle_{H_B} && (14)\\
&= e^{-(1/2)\sum_{\mathbf{k}}\left(\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\right)^2\coth(\beta\omega_{\mathbf{k}}/2)} && (15)\\
B_{iz} &\equiv \sum_{\mathbf{k}}\left(\left(g_{i\mathbf{k}}-v_{i\mathbf{k}}\right)b_{\mathbf{k}}^{\dagger}+\left(g_{i\mathbf{k}}-v_{i\mathbf{k}}\right)^{*}b_{\mathbf{k}}\right) && (16)\\
B_x &= \frac{B_{1+}B_{0-}+B_{0+}B_{1-}-B_{10}-B_{10}^{*}}{2} && (17)\\
B_y &= \frac{B_{0+}B_{1-}-B_{1+}B_{0-}+B_{10}-B_{10}^{*}}{2\mathrm{i}} && (18)\\
\left\langle B_z\right\rangle_{H_B} &= 0 && (19)\\
R_i &\equiv \sum_{\mathbf{k}}\left(\frac{\left|v_{i\mathbf{k}}\right|^2}{\omega_{\mathbf{k}}}-\left(g_{i\mathbf{k}}\frac{v_{i\mathbf{k}}^{*}}{\omega_{\mathbf{k}}}+g_{i\mathbf{k}}^{*}\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\right)\right) && (20)
\end{aligned}
$$

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

$$
\begin{aligned}
\overline{H_T(t)} &\equiv \overline{H_{\bar{S}}(t)}+\overline{H_{\bar{I}}}+\overline{H_{\bar{B}}} && (21)\\
\overline{H_{\bar{S}}(t)} &\equiv (\varepsilon_0(t)+R_0)|0\rangle\langle 0|+(\varepsilon_1(t)+R_1)|1\rangle\langle 1|+\sigma_x\big(B_{10}^{\Re}(t)V_{10}(t)\big)-B_{10}^{\Im}(t)V_{10}(t)-\sigma_y\big(B_{10}^{\Re}(t)V_{10}^{\Im}(t)+B_{10}^{\Im}(t)V_{10}^{\Re}(t)\big) && (22)\\
\overline{H_{\bar{I}}} &\equiv B_z|1\rangle\langle 1|+\Re\left(V_{10}(t)\right)\left(B_x\sigma_x+B_y\sigma_y\right)-\Im\left(V_{10}(t)\right)\left(B_x\sigma_y-B_y\sigma_x\right) && (23)\\
\overline{H_{\bar{B}}} &\equiv \sum_{\mathbf{k}}\omega_{\mathbf{k}}b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}} && (24)\\
&= H_B && (25)
\end{aligned}
$$

III. FREE-ENERGY MINIMIZATION

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

$$
A \leq A_{\mathrm{B}} \equiv -\frac{1}{\beta}\ln\left(\mathrm{Tr}\left(e^{-\beta\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}\right)\right)+\left\langle\overline{H_{\bar{I}}}\right\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}+O\left(\left\langle\overline{H_{\bar{I}}^{2}}\right\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}\right) \qquad (26)
$$

We will optimize the set of variational parameters $\{v_{\mathbf{k}}\}$ 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 $\left\langle\overline{H_{\bar{I}}}\right\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}=0$ we can obtain the following condition to obtain the set $\{v_{\mathbf{k}}\}$:

$$
\frac{\partial A_{\mathrm{B}}}{\partial v_{\mathbf{k}}}=0. \qquad (27)
$$

This leads us to:

$$
v_{i\mathbf{k}}=\frac{g_{i\mathbf{k}}\left(1-\frac{\tanh\left(\frac{\beta\eta(t)}{2}\right)}{\eta(t)}\left(2\varepsilon_i(t)+2R_i-\varepsilon(t)\right)\right)+2\frac{\tanh\left(\frac{\beta\eta(t)}{2}\right)}{\eta(t)}\frac{v_{i'\mathbf{k}}}{\omega_{\mathbf{k}}}\left|B_{10}\right|^2\left|V_{10}(t)\right|^2\coth\left(\beta\omega_{\mathbf{k}}/2\right)}{1-\frac{\tanh\left(\frac{\beta\eta(t)}{2}\right)}{\eta(t)}\left(\varepsilon(t)-2\left(\varepsilon(t)-\varepsilon_i(t)-R_i\right)-\frac{2|V_{10}(t)|^2|B_{10}|^2\coth(\beta\omega_{\mathbf{k}}/2)}{\omega_{\mathbf{k}}}\right)}, \qquad (28)
$$

with the following definitions:

$$\eta \equiv \sqrt{\left(\mathrm{Tr}\left(\overline{H_{\bar{S}}(t)}\right)\right)^2 - 4\mathrm{Det}\left(\overline{H_{\bar{S}}(t)}\right)} \tag{29}$$

$$\varepsilon(t) \equiv \mathrm{Tr}\left(\overline{H_{\bar{S}}(t)}\right) \tag{30}$$

IV. 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 $\rho_{\mathrm{Total}}(0) = \rho_S(0) \otimes \rho_B$, the transformed density operator is equal to:

$$\overline{|0\rangle\langle 0| \otimes \rho_B} = |0\rangle\langle 0| \otimes \rho_B \tag{31}$$

$$\overline{|1\rangle\langle 1| \otimes \rho_B} = |1\rangle\langle 1| \otimes B_+ \rho_B B_- \tag{32}$$

$$\overline{|0\rangle\langle 1| \otimes \rho_B} = |0\rangle\langle 1| \otimes \rho_B B_- \tag{33}$$

$$\overline{|1\rangle\langle 0| \otimes \rho_B} = |1\rangle\langle 0| \otimes B \rho_{+B} \tag{34}$$

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

$$\widetilde{O} \equiv U^\dagger(t) O U(t) \tag{35}$$

$$U(t) \equiv \mathcal{T}\exp\left(-\mathrm{i}\int_0^t \mathrm{d}v \overline{H_S}(v)\right). \tag{36}$$

Therefore:

$$\widetilde{\overline{\rho_S}}(t) = U^\dagger(t)\overline{\rho_S}(t)U(t), \text{ where} \tag{37}$$

We define $A_1 = \sigma_x$, $A_2 = \sigma_y$, $A_3 = \frac{I+\sigma_z}{2} = |1\rangle\langle 1|$, $A_4 = \sigma_x$ 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 $C_4(t) = \Im(V_{10}(t)) = -C_5(t)$. Therefore we have:

$$\overline{H_{\bar{I}}}(t) = \sum_i C_i(t)(A_i \otimes B_i(t)) \tag{38}$$

$$\widetilde{H_I}(t) = \sum_i C_i(t)\left(\widetilde{A_i}(t) \otimes \widetilde{B_i}(t)\right) \tag{39}$$

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

$$\frac{\mathrm{d}\widetilde{\overline{\rho_S}}(t)}{\mathrm{d}t} = -\int_0^t \mathrm{Tr}_B\left[\widetilde{\overline{H_{\bar{I}}}}(t), \left[\widetilde{\overline{H_{\bar{I}}}}(s), \widetilde{\overline{\rho_S}}(t)\rho_B\right]\right]\mathrm{d}s \tag{40}$$

$$= -\int_0^t \sum_{ij}\left(C_i(t)C_j(s)\left(\Lambda_{ij}(\tau)\left[\widetilde{A_i}(t), \widetilde{A_j}(s)\widetilde{\overline{\rho_S}}(t)\right] + \Lambda_{ji}(-\tau)\left[\widetilde{\overline{\rho_S}}(t)\widetilde{A_j}(s), \widetilde{A_i}\right]\right)\right)\mathrm{d}s \tag{41}$$

where:

$$\Lambda(\tau) = \begin{pmatrix} \Lambda_{11}(\tau) & 0 & 0 & 0 & -\Lambda_{11}(\tau) \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ 0 & \Lambda_{32}(\tau) & \Lambda_{33}(\tau) & \Lambda_{32}(\tau) & 0 \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ -\Lambda_{11}(\tau) & 0 & 0 & 0 & \Lambda_{11}(\tau) \end{pmatrix}, \tag{42}$$

$$\Lambda_{11}(\tau) = \frac{B(\tau)\,B(0)}{2}\left(\mathrm{e}^{\phi(\tau)} + \mathrm{e}^{-\phi(\tau)} - 2\right) \tag{43}$$

$$\Lambda_{22}(\tau) = \frac{B(\tau)\,B(0)}{2}\left(\mathrm{e}^{\phi(\tau)} + \mathrm{e}^{-\phi(\tau)}\right) \tag{44}$$

$$\Lambda_{33}(\tau) = \int_0^\infty \mathrm{d}\omega J(\omega)\,(1 - F(\omega))^2\,G_+(\tau) \tag{45}$$

$$\Lambda_{32}(\tau) = B(\tau)\int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega} F(\omega)\,(1 - F(\omega))\,\mathrm{i}G_-(\tau) \tag{46}$$

$$\Lambda_{23}(\tau) = -B(0)\int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega} F(\omega,\tau)\,(1 - F(\omega,\tau))\,\mathrm{i}G_-(\tau) \tag{47}$$

with the phonon propagator given by:

$$\phi(\tau) = \int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega^2} F(\omega)^2\,G_+(\tau), \tag{48}$$

$$G_\pm(\tau) = (n(\omega) + 1)\,\mathrm{e}^{-\mathrm{i}\tau\omega} \pm n(\omega)\,e^{-\mathrm{i}\tau\omega} \tag{49}$$

$$n(\omega) = \left(\mathrm{e}^{\beta\omega} - 1\right)^{-1}. \tag{50}$$

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t} = -\mathrm{i}\left[H_S(t), \overline{\rho_S}(t)\right] - \sum_{ij}\int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\left[A_i, \widetilde{A_j}(t-\tau,t)\,\overline{\rho_S}(t)\right] + C_j(t)\,C_i(t-\tau)\,\Lambda_{ji}(-\tau)\left[\overline{\rho_S}(t)\widetilde{A_j}(t-\tau,t), A_i\right]\mathrm{d}\tau. \tag{51}$$

We still have interaction picture versions of A_j, so we will decompose $\widetilde{A_j}(\tau)$ in terms of the Schroedinger picture version A_i:

$$\widetilde{A_j}(\tau) = \sum_w e^{-\mathrm{i}w\tau} A_j(w) \tag{52}$$

$$\widetilde{A_j}(t) = \sum_{w(t)} e^{-\mathrm{i}w(t)\tau} A_j(w(t)) \tag{53}$$

$$\widetilde{A_j}(t-\tau,t) = \sum_{w(t),w'(t-\tau)} e^{-\mathrm{i}w(t)t} e^{\mathrm{i}w'(t-\tau)} A'_j(w(t), w'(t-\tau)) \tag{54}$$

Where the sum is defined on the set of all the differences between the eigenvalues of the system, in our case $w \in \{0, \pm\eta\}$. We also have that $w(t)$ belongs to the set of differences of eigenvalues that depends of the time. As we can see the eigenvectors are time dependent as well. Also, $w'(t-\tau)$ and $w(t)$ belong to the set of the differences of the eigenvalues of the Hamiltonian $H_S(t-\tau)$ and $H_S(t)$ respectively. In matrix form, these are:

$$A_i(0) = \langle +|\,A_i\,|+\rangle\,|+\rangle\langle +| + \langle -|\,A_i\,|-\rangle\,|-\rangle\langle -| \tag{55}$$

$$A_i(w) = \langle +|\,A_i\,|-\rangle\,|+\rangle\langle -| \tag{56}$$

$$A_i(-w) = \langle -|\,A_i\,|+\rangle\,|-\rangle\langle +|. \tag{57}$$

We define the following response functions:

$$K_{ijww'}(t) = \int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\,\mathrm{e}^{\mathrm{i}w\tau} e^{-\mathrm{i}t\left(w-w'\right)}\mathrm{d}\tau \tag{58}$$

Finally we end up with our final master equation in the variationally optimized frame in the Schroedinger picture:

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t} = -\mathrm{i}\left[H_S(t), \overline{\rho_S}\right] - \sum_{ijww'} K_{ijww'}(t)\left[A_i, A_{jww'}\overline{\rho_S(t)} - \overline{\rho_S}(t)\,A^\dagger_{jww'}\right] \tag{59}$$

$$\dot{\rho} = -\mathrm{i}\left[H_S\left(t\right),\rho\right] - \sum_{ijww'} K_{ijww'}(t)\left[A_i, A_{jww'}\rho - \rho A^{\dagger}_{jww'}\right] \tag{60}$$

$$\dot{\rho} = -\mathrm{i}\left[H_S\left(t\right),\rho\right] - \sum_{ijww'} K_{ijww'}(t)\left(\left[A_i, A_{jww'}\rho\right] - \left[A_i, \rho A^{\dagger}_{jww'}\right]\right) \tag{61}$$

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