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:

$$D\left(\pm v_{i\mathbf{k}}\right) \equiv e^{\pm\left(\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}b_{\mathbf{k}}^{\dagger}-\frac{v_{i\mathbf{k}}^{*}}{\omega_{\mathbf{k}}}b_{\mathbf{k}}\right)}. \tag{11}$$

$$B_{i\pm} \equiv e^{\pm\sum_{\mathbf{k}}\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\left(b_{\mathbf{k}}^{\dagger}-b_{\mathbf{k}}\right)} \tag{12}$$

$$= \prod_{\mathbf{k}} D\left(\pm v_{i\mathbf{k}}\right) \tag{13}$$

$$B_i \equiv \left\langle B_{i\pm}\right\rangle_{H_B} \tag{14}$$

$$= e^{-(1/2)\sum_{\mathbf{k}}\left(\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\right)^2\coth(\beta\omega_{\mathbf{k}}/2)} \tag{15}$$

$$B_{iz} \equiv \sum_{\mathbf{k}}\left(\left(g_{i\mathbf{k}}-v_{i\mathbf{k}}\right)b_{\mathbf{k}}^{\dagger}+\left(g_{i\mathbf{k}}-v_{i\mathbf{k}}\right)^{*}b_{\mathbf{k}}\right) \tag{16}$$

$$B_x = \frac{B_{1+}B_{0-}+B_{0+}B_{1-}-B_{10}-B_{10}^{*}}{2} \tag{17}$$

$$B_y = \frac{B_{0+}B_{1-}-B_{1+}B_{0-}+B_{10}-B_{10}^{*}}{2\mathrm{i}} \tag{18}$$

$$\left\langle B_z\right\rangle_{H_B} = 0 \tag{19}$$

$$R_i \equiv \sum_{\mathbf{k}}\left(\frac{\left|v_{i\mathbf{k}}\right|^2}{\omega_{\mathbf{k}}}-\left(g_{i\mathbf{k}}\frac{v_{i\mathbf{k}}^{*}}{\omega_{\mathbf{k}}}+g_{i\mathbf{k}}^{*}\frac{v_{i\mathbf{k}}}{\omega_{\mathbf{k}}}\right)\right) \tag{20}$$

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

$$\overline{H_T(t)} \equiv \overline{H_{\bar{S}}(t)}+\overline{H_{\bar{I}}}+\overline{H_{\bar{B}}} \tag{21}$$

$$\overline{H_{\bar{S}}(t)} \equiv (\varepsilon_0(t)+R_0)|0\rangle\langle 0|+(\varepsilon_1(t)+R_1)|1\rangle\langle 1|+\sigma_x\big(B_{10}^{\Re}(t)V_{10}(t)\big)-B_{10}^{\Im}(t)V_{10}(t)-\sigma_y\big(B_{10}^{\Re}(t)V_{10}^{\Im}(t)+B_{10}^{\Im}(t)V_{10}^{\Re}(t)\big) \tag{22}$$

$$\overline{H_{\bar{I}}} \equiv B_z|1\rangle\langle 1|+\Re\left(V_{10}(t)\right)\left(B_x\sigma_x+B_y\sigma_y\right)-\Im\left(V_{10}(t)\right)\left(B_x\sigma_y-B_y\sigma_x\right) \tag{23}$$

$$\overline{H_{\bar{B}}} \equiv \sum_{\mathbf{k}}\omega_{\mathbf{k}}b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}} \tag{24}$$

$$= H_B \tag{25}$$

III. FREE-ENERGY MINIMIZATION

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

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

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 $\{v_{\mathbf{k}}\}$:

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

This leads us to:

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

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 $\rho_{\mathrm{Total}}(0) = \rho_S(0) \otimes \rho_B(0)$, where $\rho_B(0) \equiv \rho_B^{\mathrm{Thermal}} \equiv \rho_B$ the transformed density operator is equal to:

$$\text{for } \rho_S(0) = |0\rangle\langle 0| : = |0\rangle\langle 0| \otimes B_{0+}\rho_B B_{0-} \tag{31}$$

$$\text{for } \rho_S(0) = |1\rangle\langle 1| : = |1\rangle\langle 1| \otimes B_{1+}\rho_B B_{1-} \tag{32}$$

$$\text{for } \rho_S(0) = |0\rangle\langle 1| : = |0\rangle\langle 1| \otimes B_{0+}\rho_B B_{1-} \tag{33}$$

$$\text{for } \rho_S(0) = |1\rangle\langle 0| : = |1\rangle\langle 0| \otimes B_{1+}\rho_B B_{0-} \tag{34}$$

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

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

$$U(t) \equiv \mathcal{T}\exp\left(-\mathrm{i}\int_0^t \mathrm{d}t' \overline{H_S}\left(t'\right)\right). \tag{36}$$

Therefore:

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

We define the following matrix:

$$\begin{pmatrix} A(t) \\ B \\ C(t) \end{pmatrix} = \begin{pmatrix} \sigma_x & \sigma_y & \frac{I-\sigma_z}{2} & \sigma_x & \sigma_y & \frac{I+\sigma_z}{2} \\ B_x & B_y & B_{1z} & B_y & B_x & B_{0z} \\ \Re\left(V_{10}(t)\right) & \Re\left(V_{10}(t)\right) & 1 & \Im\left(V_{10}(t)\right) & -\Im\left(V_{10}(t)\right) & 1 \end{pmatrix} \tag{38}$$

Then we have:

$$\overline{H_{\bar{I}}}(t) = \sum_i C_i(t)\left(A_i \otimes B_i(t)\right) \tag{39}$$

$$\widetilde{H_I}(t) = \sum_i C_i(t)\left(\widetilde{A_i}(t) \otimes \widetilde{B_i}(t)\right) \tag{40}$$

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

$$\frac{\mathrm{d}\widetilde{\overline{\rho_S}}(t)}{\mathrm{d}t} = -\int_0^t \mathrm{Tr}_B\left[\widetilde{\overline{H_{\bar{I}}}}(t), \left[\widetilde{\overline{H_{\bar{I}}}}(s), \widetilde{\overline{\rho_S}}(t)\rho_B\right]\right]\mathrm{d}s \tag{41}$$

$$= -\int_0^t \sum_{ij}\left(C_i(t)C_j(s)\left(\Lambda_{ij}(\tau)\left[\widetilde{A_i}(t), \widetilde{A_j}(s)\widetilde{\overline{\rho_S}}(t)\right] + \Lambda_{ji}(-\tau)\left[\widetilde{\overline{\rho_S}}(t)\widetilde{A_j}(s), \widetilde{A_i}\right]\right)\right)\mathrm{d}s \tag{42}$$

where:

$$\Lambda(\tau)=\begin{pmatrix}\Lambda_{11}(\tau) & 0 & 0 & 0 & -\Lambda_{11}(\tau)\\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0\\ 0 & \Lambda_{32}(\tau) & \Lambda_{33}(\tau) & \Lambda_{32}(\tau) & 0\\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0\\ -\Lambda_{11}(\tau) & 0 & 0 & 0 & \Lambda_{11}(\tau)\end{pmatrix}, \tag{43}$$

$$\Lambda_{11}(\tau)=\frac{B(\tau)\,B(0)}{2}\left(\mathrm{e}^{\phi(\tau)}+\mathrm{e}^{-\phi(\tau)}-2\right) \tag{44}$$

$$\Lambda_{22}(\tau)=\frac{B(\tau)\,B(0)}{2}\left(\mathrm{e}^{\phi(\tau)}+\mathrm{e}^{-\phi(\tau)}\right) \tag{45}$$

$$\Lambda_{33}(\tau)=\int_0^\infty \mathrm{d}\omega J(\omega)\,(1-F(\omega))^2\,G_+(\tau) \tag{46}$$

$$\Lambda_{32}(\tau)=B(\tau)\int_0^\infty \mathrm{d}\omega\frac{J(\omega)}{\omega}F(\omega)\,(1-F(\omega))\,\mathrm{i}G_-(\tau) \tag{47}$$

$$\Lambda_{23}(\tau)=-B(0)\int_0^\infty \mathrm{d}\omega\frac{J(\omega)}{\omega}F(\omega,\tau)\,(1-F(\omega,\tau))\,\mathrm{i}G_-(\tau) \tag{48}$$

with the phonon propagator given by:

$$\phi(\tau)=\int_0^\infty \mathrm{d}\omega\frac{J(\omega)}{\omega^2}F(\omega)^2\,G_+(\tau), \tag{49}$$

$$G_\pm(\tau)=(n(\omega)+1)\,\mathrm{e}^{-\mathrm{i}\tau\omega}\pm n(\omega)\,e^{-\mathrm{i}\tau\omega} \tag{50}$$

$$n(\omega)=\left(\mathrm{e}^{\beta\omega}-1\right)^{-1}. \tag{51}$$

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t}=-\mathrm{i}\left[H_S(t),\overline{\rho_S}(t)\right]-\sum_{ij}\int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\left[A_i,\widetilde{A_j}(t-\tau,t)\,\overline{\rho_S}(t)\right]+C_j(t)\,C_i(t-\tau)\,\Lambda_{ji}(-\tau)\left[\overline{\rho_S}(t)\widetilde{A_j}(t-\tau,t),A_i\right]\mathrm{d}\tau. \tag{52}$$

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:

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

$$\widetilde{A_j}(t)=\sum_{w(t)} e^{-\mathrm{i}w(t)\tau}A_j(w(t)) \tag{54}$$

$$\widetilde{A_j}(t-\tau,t)=\sum_{w(t),w'(t-\tau)} e^{-\mathrm{i}w(t)t}e^{\mathrm{i}w'(t-\tau)}A'_j(w(t),w'(t-\tau)) \tag{55}$$

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:

$$A_i(0)=\langle+|\,A_i\,|+\rangle\,|+\rangle\,\langle+|+\langle-|\,A_i\,|-\rangle\,|-\rangle\,\langle-| \tag{56}$$

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

$$A_i(-w)=\langle-|\,A_i\,|+\rangle\,|-\rangle\,\langle+|. \tag{58}$$

We define the following response functions:

$$K_{ijww'}(t)=\int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\,e^{\mathrm{i}w\tau}e^{-\mathrm{i}t\left(w-w'\right)}\mathrm{d}\tau \tag{59}$$

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

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t} = -\mathrm{i}\left[H_S\left(t\right), \overline{\rho_S}\right] - \sum_{ijww'} K_{ijww'}(t)\left[A_i, A_{jww'}\overline{\rho_S(t)} - \overline{\rho_S}\left(t\right)A_{jww'}^{\dagger}\right] \tag{60}$$

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

$$\dot{\rho} = -\mathrm{i}\left[H_S\left(t\right), \rho\right] - \sum_{ijww'} K_{ijww'}(t)\left(\left[A_i, A_{jww'}\rho\right] - \left[A_i, \rho A_{jww'}^{\dagger}\right]\right) \tag{62}$$

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