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:

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

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

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

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

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

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:

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

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:

$$\overline{H(t)} = \varepsilon_1\,(t)\,|1\rangle\langle 1| + \varepsilon_0\,(t)\,|0\rangle\langle 0| + V_{10}\,(t)\,|1\rangle\langle 0|B_+ + V_{01}\,(t)\,|0\rangle\langle 1|B_- + |1\rangle\langle 1|\sum_{\mathbf{k}} g_{\mathbf{k}}\left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}} - 2\frac{v_{\mathbf{k}}}{\omega_{\mathbf{k}}}\right) \tag{7}$$

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

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

$$\rho_B = \frac{e^{-\beta H_B}}{\mathrm{Tr}\left(e^{-\beta H_B}\right)} \tag{9}$$

With the following definitions and relations:

$$D\left(\pm v_{\mathbf{k}}\right) \equiv e^{\pm \frac{v_{\mathbf{k}}}{\omega_{\mathbf{k}}}\left(b_{\mathbf{k}}^{\dagger}-b_{\mathbf{k}}\right)}, \tag{10}$$

$$B_{\pm} \equiv e^{\pm \sum_{\mathbf{k}} \frac{v_{\mathbf{k}}}{\omega_{\mathbf{k}}}\left(b_{\mathbf{k}}^{\dagger}-b_{\mathbf{k}}\right)} \tag{11}$$

$$= \prod_{\mathbf{k}} D\left(\pm v_{\mathbf{k}}\right) \tag{12}$$

$$B \equiv \left\langle B_{\pm}\right\rangle_{H_B} \tag{13}$$

$$= e^{-(1/2)\sum_{\mathbf{k}}\left(\frac{v_{\mathbf{k}}}{\omega_{\mathbf{k}}}\right)^2 \coth(\beta\omega_{\mathbf{k}}/2)} \tag{14}$$

$$B_z \equiv \sum_{\mathbf{k}} \left(g_{\mathbf{k}} - v_{\mathbf{k}}\right)\left(b_{\mathbf{k}}^{\dagger} + b_{\mathbf{k}}\right) \tag{15}$$

$$B_x = \frac{B_+ + B_- - 2B}{2} \tag{16}$$

$$B_y = \frac{B_- - B_+}{2} \tag{17}$$

$$\left\langle B_z\right\rangle_{H_B} = 0 \tag{18}$$

$$R_1 \equiv \sum_{\mathbf{k}} \left(v_{\mathbf{k}} - 2 v_{\mathbf{k}} g_{\mathbf{k}}\right) \tag{19}$$

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

$$\overline{H_T(t)} \equiv \overline{H_{\bar{S}}(t)} + \overline{H_{\bar{I}}} + \overline{H_{\bar{B}}} \tag{20}$$

$$\overline{H_{\bar{S}}(t)} \equiv \left(\varepsilon_1(t) + R_1\right)|1\rangle\langle 1| + \varepsilon_0(t)|0\rangle\langle 0| + \frac{B\sigma_x}{2}\left(V_{10}(t) + V_{01}(t)\right) + \frac{\mathrm{i}B\sigma_y}{2}\left(V_{10}(t) - V_{01}(t)\right) \tag{21}$$

$$\overline{H_{\bar{I}}} \equiv B_z|1\rangle\langle 1| + \Re\left(V_{10}(t)\right)\left(B_x\sigma_x + B_y\sigma_y\right) - \Im\left(V_{10}(t)\right)\left(B_x\sigma_y - B_y\sigma_x\right) \tag{22}$$

$$\overline{H_{\bar{B}}} \equiv \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} \tag{23}$$

$$= H_B \tag{24}$$

III. FREE-ENERGY MINIMIZATION

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

$$A \leq A_{\mathrm{B}} \equiv -\frac{1}{\beta}\ln\left(\mathrm{Tr}\left(e^{-\beta\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}\right)\right) + \left\langle\overline{H_{\bar{I}}}\right\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}} + O\left(\left\langle\overline{H_{\bar{I}}^2}\right\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}}\right) \tag{25}$$

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 $\langle\overline{H_{\bar{I}}}\rangle_{\overline{H_{\bar{S}}(t)+H_{\bar{B}}}} = 0$ we can obtain the following condition to obtain the set $\{v_{\mathbf{k}}\}$:

$$\frac{\partial A_{\mathrm{B}}}{\partial v_{\mathbf{k}}} = 0. \tag{26}$$

This leads us to:

$$v_{\mathbf{k}} = \frac{g_{\mathbf{k}}\left(1 - \frac{\tanh\left(\frac{\beta\eta}{2}\right)}{\eta}\left(\varepsilon_1(t) + R_1 - \varepsilon_0(t)\right)\right)}{1 - \frac{\tanh\left(\frac{\beta\eta}{2}\right)}{\eta}\left(\varepsilon_1(t) + R_1 - \varepsilon_0(t) - \frac{2|V_{10}(t)|^2 B^2}{\omega_{\mathbf{k}}}\coth\left(\frac{\beta\omega_{\mathbf{k}}}{2}\right)\right)}, \tag{27}$$

with the following definitions:

$$\eta = \sqrt{\left(\mathrm{Tr}\left(\overline{H_{\bar{S}}(t)}\right)\right)^2 - 4\mathrm{Det}\left(\overline{H_{\bar{S}}(t)}\right)} \tag{28}$$

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:

$$\overline{|0\rangle\langle 0| \otimes \rho_B} = |0\rangle\langle 0| \otimes \rho_B \tag{29}$$
$$\overline{|1\rangle\langle 1| \otimes \rho_B} = |1\rangle\langle 1| \otimes B_+\rho_B B_- \tag{30}$$
$$\overline{|0\rangle\langle 1| \otimes \rho_B} = |0\rangle\langle 1| \otimes \rho_B B_- \tag{31}$$
$$\overline{|1\rangle\langle 0| \otimes \rho_B} = |1\rangle\langle 0| \otimes B\rho_{+B} \tag{32}$$

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

$$\widetilde{O} \equiv U^\dagger(t) O U(t) \tag{33}$$
$$U(t) \equiv \mathcal{T}\exp\left(-\mathrm{i}\int_0^t \mathrm{d}v \overline{H_S}(v)\right). \tag{34}$$

Therefore:

$$\widetilde{\overline{\rho_S}}(t) = U^\dagger(t)\overline{\rho_S}(t)U(t), \text{ where} \tag{35}$$

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:

$$\overline{H_{\bar{I}}}(t) = \sum_i C_i(t)\left(A_i \otimes B_i(t)\right) \tag{36}$$
$$\widetilde{H_I}(t) = \sum_i C_i(t)\left(\widetilde{A_i}(t) \otimes \widetilde{B_i}(t)\right) \tag{37}$$

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

$$\frac{\mathrm{d}\widetilde{\overline{\rho_S}}(t)}{\mathrm{d}t} = -\int_0^t \mathrm{Tr}_B\left[\widetilde{\overline{H_{\bar{I}}}}(t), \left[\widetilde{\overline{H_{\bar{I}}}}(s), \widetilde{\overline{\rho_S}}(t)\rho_B\right]\right]\mathrm{d}s \tag{38}$$
$$= -\int_0^t \sum_{ij}\left(C_i(t)C_j(s)\left(\Lambda_{ij}(\tau)\left[\widetilde{A_i}(t), \widetilde{A_j}(s)\widetilde{\overline{\rho_S}}(t)\right] + \Lambda_{ji}(-\tau)\left[\widetilde{\overline{\rho_S}}(t)\widetilde{A_j}(s), \widetilde{A_i}\right]\right)\right)\mathrm{d}s \tag{39}$$

where:

$$\Lambda(\tau) = \begin{pmatrix} \Lambda_{11}(\tau) & 0 & 0 & 0 & -\Lambda_{11}(\tau) \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ 0 & \Lambda_{32}(\tau) & \Lambda_{33}(\tau) & \Lambda_{32}(\tau) & 0 \\ 0 & \Lambda_{22}(\tau) & \Lambda_{23}(\tau) & \Lambda_{22}(\tau) & 0 \\ -\Lambda_{11}(\tau) & 0 & 0 & 0 & \Lambda_{11}(\tau) \end{pmatrix}, \tag{40}$$

$$\Lambda_{11}(\tau) = \frac{B(\tau)B(0)}{2}\left(\mathrm{e}^{\phi(\tau)} + \mathrm{e}^{-\phi(\tau)} - 2\right) \tag{41}$$

$$\Lambda_{22}(\tau) = \frac{B(\tau)B(0)}{2}\left(\mathrm{e}^{\phi(\tau)} + \mathrm{e}^{-\phi(\tau)}\right) \tag{42}$$

$$\Lambda_{33}(\tau) = \int_0^\infty \mathrm{d}\omega J(\omega)(1 - F(\omega))^2 G_+(\tau) \tag{43}$$

$$\Lambda_{32}(\tau) = B(\tau)\int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega} F(\omega)(1 - F(\omega))\,\mathrm{i}G_-(\tau) \tag{44}$$

$$\Lambda_{23}(\tau) = -B(0)\int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega} F(\omega,\tau)(1 - F(\omega,\tau))\,\mathrm{i}G_-(\tau) \tag{45}$$

with the phonon propagator given by:

$$\phi(\tau) = \int_0^\infty \mathrm{d}\omega \frac{J(\omega)}{\omega^2} F(\omega)^2 G_+(\tau), \tag{46}$$

$$G_\pm(\tau) = (n(\omega) + 1)\,\mathrm{e}^{-\mathrm{i}\tau\omega} \pm n(\omega)\,e^{-\mathrm{i}\tau\omega} \tag{47}$$

$$n(\omega) = \left(\mathrm{e}^{\beta\omega} - 1\right)^{-1}. \tag{48}$$

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t} = -\mathrm{i}\left[H_S(t), \overline{\rho_S}(t)\right] - \sum_{ij}\int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\left[A_i, \widetilde{A_j}(t-\tau,t)\,\overline{\rho_S}(t)\right] + C_j(t)\,C_i(t-\tau)\,\Lambda_{ji}(-\tau)\left[\overline{\rho_S}(t)\widetilde{A_j}(t-\tau,t), A_i\right]\mathrm{d}\tau. \tag{49}$$

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:

$$\widetilde{A_j}(\tau) = \sum_w e^{-\mathrm{i}w\tau} A_j(w) \tag{50}$$

$$\widetilde{A_j}(t) = \sum_{w(t)} e^{-\mathrm{i}w(t)\tau} A_j(w(t)) \tag{51}$$

$$\widetilde{A_j}(t-\tau,t) = \sum_{w(t),w'(t-\tau)} e^{-\mathrm{i}w(t)t} e^{\mathrm{i}w'(t-\tau)} A'_j\left(w(t), w'(t-\tau)\right) \tag{52}$$

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:

$$A_i(0) = \langle +|A_i|+\rangle\,|+\rangle\langle +| + \langle -|A_i|-\rangle\,|-\rangle\langle -| \tag{53}$$

$$A_i(w) = \langle +|A_i|-\rangle\,|+\rangle\langle -| \tag{54}$$

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

We define the following response functions:

$$K_{ijww'}(t) = \int_0^t C_i(t)\,C_j(t-\tau)\,\Lambda_{ij}(\tau)\,\mathrm{e}^{\mathrm{i}w\tau}\mathrm{e}^{-\mathrm{i}t\left(w-w'\right)}\mathrm{d}\tau \tag{56}$$

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

$$\frac{\mathrm{d}\overline{\rho_S}(t)}{\mathrm{d}t} = -\mathrm{i}\left[H_S(t), \overline{\rho_S}\right] - \sum_{ijww'} K_{ijww'}(t)\left[A_i, A_{jww'}\overline{\rho_S(t)} - \overline{\rho_S}(t)\,A^\dagger_{jww'}\right] \tag{57}$$

$$\dot{\rho} = -\mathrm{i}\left[H_S\left(t\right),\rho\right] - \sum_{ijww'} K_{ijww'}(t)\left[A_i, A_{jww'}\rho - \rho A^{\dagger}_{jww'}\right] \tag{58}$$

$$\dot{\rho} = -\mathrm{i}\left[H_S\left(t\right),\rho\right] - \sum_{ijww'} K_{ijww'}(t)\left(\left[A_i, A_{jww'}\rho\right] - \left[A_i, \rho A^{\dagger}_{jww'}\right]\right) \tag{59}$$

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