Lecture notes on open quantum systems

Ahsan Nazir

I. INTRODUCTION

It is generally impossible to isolate a particular quantum system in which we are interested from its surroundings. Hence, for almost all quantum systems we would like to study, we must consider the influence of the external environment upon the system dynamics, in order to faithfully represent the real dynamical evolution of the physical system. However, we are usually unable to track, either theoretically or experimentally, the dynamical evolution of the full system-plus-environment, and neither would we generally want to, given the large amount of redundant information that would be generated. In this situation, we would therefore like to develop a description of open quantum systems, no longer undergoing unitary dynamics as in the closed system case, but instead described by a dynamical equation that accounts (usually approximately) for the influence of the surrounding environment on the system state, while removing the need to track the full environment evolution.

In these lectures we shall develop the basic techniques relevant for the master equation description of open quantum systems in terms of density operators. We shall also apply these techniques to a number of example problems to see how the effects of environmental noise are captured in various realistic situations.

A. Books on open quantum systems

Two particularly useful books with introductory material on this topic are:

- *The theory of open quantum systems*, by H.-P. Breuer and F. Petruccione (Oxford, 2002)
- *Statistical Methods in Quantum Optics 1: Master Equations and Fokker-Planck Equations*, by H. J. Carmichael (Springer 1999). See also the Springer website for an online version (accessible through Imperial College) of the precursor to this book, *An Open Systems Approach to Quantum Optics*, also by H. J. Carmichael.

B. Unitary evolution of closed quantum systems

By way of introduction, we shall first briefly recap the dynamics of closed quantum systems in terms of both pure states and the density operator. We shall also introduce the interaction picture representation. We shall then move on to the description of open quantum systems, and derive a general perturbative master equation governing the evolution of a small system of interest, under the influence of a large surrounding environment.

Consider a closed quantum system that can be described by a pure state $|\psi(t)\rangle$, evolving under the action of some (possibly time-dependent) Hamiltonian $H(t)$, as governed by the Schrödinger equation (note that we set $\hbar = 1$ throughout these notes):

$$\frac{\partial}{\partial t}|\psi(t)\rangle = -iH(t)|\psi(t)\rangle . \tag{1}$$

This has the formal solution

$$|\psi(t)\rangle = U(t,t_0)|\psi(t_0)\rangle , \tag{2}$$

where the *unitary* time-evolution operator is given in terms of a time-ordered exponential

$$U(t,t_0) = \underset{\leftarrow}{T}\exp\left[-i\int_{t_0}^{t} ds H(s)\right], \tag{3}$$

such that $U^\dagger U = UU^\dagger = \mathbb{1}$. Here, the time-ordering operator $\underset{\leftarrow}{T}$ orders products of time-dependent operators such that earliest times are to the right. We see that $U(t,t_0)$ generates unitary evolution, transforming the state $|\psi(t_0)\rangle$ at an initial time t_0, to the state $|\psi(t)\rangle$ at time t. In the case that the Hamiltonian is time-independent, $H(t) = H$, it can be reduced to the far simpler form

$$U(t,t_0) = \exp\left[-iH(t-t_0)\right]. \tag{4}$$

1. Density operator evolution

We may alternatively characterise the system state through the density operator, which in the present case of a pure state is given simply by

$$\rho(t) = |\psi(t)\rangle\langle\psi(t)| . \tag{5}$$

Differentiating Eq. (5) with respect to time, we find that the density operator evolves according to the equation

$$\frac{\partial}{\partial t}\rho(t) = -i[H(t),\rho(t)], \tag{6}$$

which is known as the Liouville-von Neumann equation. More generally, the density operator is useful because it allows us to go beyond a pure state description of quantum systems and describe *mixed* states, in terms of an ensemble of pure states. For a system described by an ensemble of states $\{|\psi_n(t)\rangle\}$, with probabilities $\{p_n\}$, the density operator is given by

$$\rho(t) = \sum_n p_n |\psi_n(t)\rangle\langle\psi_n(t)| . \tag{7}$$

Such a representation is clearly going to be important in describing open quantum systems, where the interaction of our system of interest with its surroundings introduces uncertainty into our knowledge of the system state. However, it can also be relevant for closed systems, for example if we have a situation where preparation of the initial state is uncertain.

Differentiating Eq. (7) with respect to time, we again recover the Liouville-von Neumann equation [Eq. (6)], which therefore governs the evolution of both pure and mixed state density operators for closed systems. Taking some initial density operator

$$\rho(t_0) = \sum_n p_n \left|\psi_n(t_0)\right\rangle \left\langle\psi_n(t_0)\right|, \tag{8}$$

we see that for a closed system the state at time t should be given by

$$\rho(t) = \sum_n p_n U(t,t_0) \left|\psi_n(t_0)\right\rangle \left\langle\psi_n(t_0)\right| U^\dagger(t,t_0), \tag{9}$$

such that the density operator evolves according to

$$\rho(t) = U(t,t_0)\rho(t_0)U^\dagger(t,t_0), \tag{10}$$

in terms of the unitary time-evolution operator $U(t,t_0)$ for a closed system. Thus, Eq. (10) gives us a formal way of writing the solution to Eq. (6).

Briefly, a few properties of the density operator are:

- It is Hermitian, i.e. $\rho^\dagger = \rho$.
- For pure states, $\rho^2 = \rho$.
- It has trace one, i.e. $\mathrm{tr}\rho = 1$.
- For mixed states $\mathrm{tr}\rho^2 < \mathrm{tr}\rho = 1$
- The expectation value of any operator $\hat{O}$ can be calculated through $\langle\hat{O}\rangle = \mathrm{tr}(\hat{O}\rho)$.

2. *The interaction picture*

In what follows, we shall frequently make use of the interaction picture in deriving master equations that govern the evolution of the density operator of an open quantum system. In this picture, we remove the contribution of part of the system-environment Hamiltonian (H_0) to the density operator evolution, by performing a unitary transformation into a new representation. We can then derive an equation for the system evolution due to what remains. Of course, the contribution due to H_0 is moved instead to the time evolution of the operators, and so must be reintroduced if we want to evaluate, for example, the expectation value of a particular observable of the system. However, we generally choose the partitioning of our Hamiltonian such that the evolution due to H_0 itself is simple to solve, and it is the dynamics generated by the rest of the Hamiltonian (H_I) which may be complicated enough that we have to approximate.

Consider partitioning the Hamiltonian of a system such that

$$H = H_0 + H_I, \tag{11}$$

where we take H to be time-independent for simplicity in the formalism. In the master equation derivation that follows, H_0 will be the sum of the Hamiltonians of the system and environment when their coupling is ignored, while H_I will represent the system-environment interaction. However, the partitioning need not always be done in this way.

If the interaction picture is to be physically equivalent to the Schödinger picture in which we have been working so far, then the expectation value of some observable $\hat{O}$ (which we take to have no intrinsic time dependence)

$$\langle\hat{O}(t)\rangle = \mathrm{tr}(\hat{O}\rho(t)) = \mathrm{tr}(\hat{O}U(t,t_0)\rho(t_0)U^\dagger(t,t_0)), \tag{12}$$

should not change upon transformation. Let's split the time-evolution operator as

$$U(t,t_0) = U_0(t,t_0)U_I(t,t_0), \tag{13}$$

where we define

$$U_0(t,t_0) \equiv \exp\left[-iH_0(t-t_0)\right]. \tag{14}$$

In Eq. (12), we now obtain

$$\langle\hat{O}(t)\rangle = \mathrm{tr}(\hat{O}U_0(t,t_0)U_I(t,t_0)\rho(t_0)U_I^\dagger(t,t_0)U_0^\dagger(t,t_0)), \tag{15}$$

which can be written

$$\langle\hat{O}(t)\rangle = \mathrm{tr}(\hat{O}_I(t)\rho_I(t)), \tag{16}$$

where we have made use of the cyclic property of the trace ($\mathrm{tr}(ABC) = \mathrm{tr}(BCA) = \mathrm{tr}(CAB)$), and have defined

$$\hat{O}_I(t) = U_0^\dagger(t,t_0)\hat{O}U_0(t,t_0), \tag{17}$$

and

$$\begin{aligned} \rho_I(t) &= U_I(t,t_0)\rho(t_0)U_I^\dagger(t,t_0), \\ &= U_0^\dagger(t,t_0)\rho(t)U_0(t,t_0). \end{aligned} \tag{18}$$

Thus, from Eq. (17), we see that in the interaction picture, operators evolve in time according to the free (time-independent) Hamiltonian H_0 as

$$\hat{O}_I(t) = \exp\left[iH_0(t-t_0)\right]\hat{O}\exp\left[-iH_0(t-t_0)\right]. \tag{19}$$

Differentiating Eq. (18) with respect to time, and using Eq. (13), it is possible to show that the evolution of the interaction picture density operator is governed by a Liouville-von Neumann equation of the form

$$\frac{\partial}{\partial t}\rho_I(t) = -i[H_I(t), \rho_I(t)], \tag{20}$$

as we might expect, where the interaction picture interaction Hamiltonian is given by

$$H_I(t) = U_0^\dagger(t,t_0) H_I U_0(t,t_0). \tag{21}$$

Finally, it is clear from Eqs. (18) and (20) that the interaction picture time-evolution operator must take the form

$$U_I(t,t_0) = \underset{\leftarrow}{T} \exp\left[-i\int_{t_0}^{t} ds H_I(s)\right], \tag{22}$$

since we can see that $\rho_I(t)$ obeys the Liouville-von Neumann equation, but with the total Hamiltonian replaced by $H_I(t)$.

II. OPEN QUANTUM SYSTEMS

Having recapped the dynamical evolution of closed quantum systems, let's now define an open quantum system S as a subsystem of a larger combined system $S+E$. Here, E represents another quantum system called the environment, to which S is coupled. An example might be an electron (the system) interacting with the vibrational modes of a solid (the environment), or an atom interacting with the surrounding electromagnetic field. We shall assume that the combined evolution of the system-plus-environment $(S+E)$ is closed, and so follows unitary Hamiltonian dynamics as we described previously. The state of the system S, however, will evolve not only according to its own internal dynamics, but also due to interactions with the environment E. In this situation, it is generally not possible to represent the system dynamics by a unitary evolution operator acting on S alone. Instead, we shall derive a master equation describing the dynamics of the *reduced* density operator $\rho_S(t) = \mathrm{tr}_E \rho(t)$, which represents the system state once we trace out the environment degrees of freedom (a partial trace).

To see why the reduced density operator is the object we wish to work with, we note that the Hilbert space of our combined system and environment is given by a tensor product

$$\mathcal{H} = \mathcal{H}_\mathcal{S} \otimes \mathcal{H}_\mathcal{E}, \tag{23}$$

since we have assumed the system and environment to be distinguishable, and hence we must specify a basis for both to give a full description. If the states $\{|\psi_i\rangle_S\}$ and $\{|\psi_j\rangle_E\}$ form a basis set in $\mathcal{H}_\mathcal{S}$ and $\mathcal{H}_\mathcal{E}$, respectively, then the set $\{|\psi_i\rangle_S \otimes |\psi_j\rangle_E\}$ forms a basis of the tensor product space $\mathcal{H}$. Now, any observable of the system S can be written in the form

$$\hat{O} = \hat{O}_S \otimes \mathbb{1}_E, \tag{24}$$

in the combined space $S+E$. Then,

$$\langle \hat{O}(t)\rangle = \mathrm{tr}_{S+E}(\hat{O}_S \otimes \mathbb{1}_E \rho(t)) = \mathrm{tr}_S(\hat{O}_S \rho_S(t)), \tag{25}$$

where $\rho_S(t) = \mathrm{tr}_E \rho(t)$, as before. Hence, the reduced density operator completely describes all accessible information about the system S, while having dimension much smaller than that of the combined system-environment density operator $\rho(t)$.

A. Markovian master equations

We have now outlined what we mean by an open system S, and we also know that it is the reduced density operator $\rho_S(t)$ whose dynamical evolution we would like to calculate in order to describe the system's dynamics under the environmental influence. Starting from the full Hamiltonian H of $S+E$, we shall now derive an approximate, perturbative master equation describing the system dynamics. The procedure we follow here is standard, and serves to highlight well the physical nature of the assumptions that go into deriving a practical master equation. However, it is far from rigorous, and those who would like to see a more rigorous derivation should refer to Chapter 9 of *The theory of open quantum systems*, by H.-P. Breuer and F. Petruccione (Oxford, 2002), where the projection operator technique is used, or Appendix A below for an alternative derivation.

We begin by writing the system-environment Hamiltonian as

$$H = H_S + H_E + H_I, \tag{26}$$

where we assume that the interaction Hamiltonian H_I is the only part that involves both system and environment degrees of freedom. It will be this part of the Hamiltonian that is treated as a perturbation. To make this more explicit, we now take $H_0 = H_S + H_E$, and move into the interaction picture:

$$\tilde{H}_I(t) = e^{i(H_S+H_E)t} H_I e^{-i(H_S+H_E)t}, \tag{27}$$

where we set $t_0 = 0$. Here, a tilde is used to represent an operator that has been transformed into the interaction picture, i.e. $\tilde{O} = e^{iH_0 t} O e^{-iH_0 t}$. We assume that the combined system and environment is closed, hence within the interaction picture the density operator evolves according to

$$\frac{\partial}{\partial t}\tilde{\rho}(t) = -i[\tilde{H}_I(t), \tilde{\rho}(t)]. \tag{28}$$

This equation has the formal solution

$$\tilde{\rho}(t) = \rho(0) - i\int_0^t ds[\tilde{H}_I(s), \tilde{\rho}(s)], \tag{29}$$

which we substitute back into Eq. (28) to give

$$\frac{\partial}{\partial t}\tilde{\rho}(t) = -i[\tilde{H}_I(t), \rho(0)] - \int_0^t ds[\tilde{H}_I(t), [\tilde{H}_I(s), \tilde{\rho}(s)]]. \tag{30}$$

Note that by iterating this procedure, we could build up terms on the right-hand-side that contain larger and

larger products of H_I. Hence, though Eq. (30) is of course still exact (and so as difficult to solve as the original Liouville-von Neumann equation), we have put it into a convenient form that will allow us to make some reasonable simplifications. Before doing so, we take the trace over the environment, since it is the dynamics of the reduced density operator $\rho_S(t)$ that we would like to calculate:

$$\begin{aligned}\frac{\partial}{\partial t}\tilde{\rho}_S(t) &= -i\mathrm{tr}_E[\tilde{H}_I(t),\rho(0)] \\ &\quad -\int_0^t ds\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(s),\tilde{\rho}(s)]]. \quad (31)\end{aligned}$$

We now make a number of assumptions:

- We assume that the interaction between S and E is turned on at the initial time $t=0$, and that no correlations exist between S and E at this time. Hence, the initial density operator factorises as $\rho(0)=\rho_S(0)\otimes\rho_E(0)$.
- We shall now set the first term on the right-hand-side of Eq. (31) to zero, $\mathrm{tr}_E[\tilde{H}_I(t),\rho(0)]=0$. This is not really an approximation, as it can be ensured by removing a term $\mathrm{tr}_E[H_I\rho_E(0)]$ from the interaction Hamiltonian, and including it instead in the system Hamiltonian [1]. Eq. (31) now takes on the form

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^t ds\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(s),\tilde{\rho}(s)]]. \quad (32)$$

- Having stated that the density operator factorises at $t=0$, we now make the **Born approximation** and assume that the density operator factorises at *all* times as

$$\tilde{\rho}(t)\approx\tilde{\rho}_S(t)\rho_E, \quad (33)$$

  where the environment density operator is assumed to be time-independent, $\rho_E=\rho_E(0)$. Clearly, we would generally expect correlations to build up over time between S and E, given their mutual interaction. The Born approximation thus **relies on the interactions described by H_I being weak**, and also on the fact that the environment is a large system, whose state is virtually unaffected by coupling to the system S.

At this point, the master equation is given by

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^t ds\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(s),\tilde{\rho}_S(s)\rho_E]], \quad (34)$$

which, though significantly simplified, still has a time non-local form, and so could be difficult to work with. In particular, the future evolution of $\tilde{\rho}_S(t)$ depends on its past history through the integration over $\tilde{\rho}_S(s)$. We now introduce a further simplification, known as the **Markov approximation**, which will put the equation into a form such that the future behaviour of $\tilde{\rho}_S(t)$ depends only on its present state:

- We make the replacement $\tilde{\rho}_S(s)\to\tilde{\rho}_S(t)$ on the right-hand-side of Eq. (34). This relies on a separation of timescales between the environment memory (correlation) time (i.e. the timescale over which the environment preserves information about past system states) and the timescale for significant evolution of S. If the environment memory time is extremely short in comparison to the typical timescale for system evolution, then the replacement $\tilde{\rho}_S(s)\to\tilde{\rho}_S(t)$ is justified. Note that this seems to be consistent with the Born Approximation, as we already assumed that the environment is a large system whose state is barely affected by coupling to S. We now have:

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^t ds\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(s),\tilde{\rho}_S(t)\rho_E]]. \quad (35)$$

- This equation is still not truly Markovian, as it contains a reference to a particular starting time $t=0$ in the lower limit of integration. However, making the substitution $s\to t-\tau$:

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^t d\tau\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(t-\tau),\tilde{\rho}_S(t)\rho_E]], \quad (36)$$

  we can now extend the limit of integration in Eq. (35) to infinity, again justified by the separation of system and environment timescales [2].

In the interaction picture, our weak-coupling, Markovian master equation can then be written

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^\infty d\tau\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(t-\tau),\tilde{\rho}_S(t)\rho_E]], \quad (37)$$

approximately describing the open system evolution. This equation provides a general starting point for a large number of applications in open quantum systems theory, a few of which we shall explore in this course [3].

1. *Environment correlation functions*

To gain a little more insight into the structure of the master equation we have derived, it helps to be a bit more specific about the form of the interaction Hamiltonian H_I. Let's decompose H_I as

$$H_I=\sum_\alpha A_\alpha\otimes B_\alpha, \quad (38)$$

where A_α are system operators and B_α environment operators. Moving into the interaction picture, we find

$$\tilde{H}_I(t)=\sum_\alpha A_\alpha(t)\otimes B_\alpha(t), \quad (39)$$

where $A_\alpha(t)=e^{iH_St}A_\alpha e^{-iH_St}$ and $B_\alpha(t)=e^{iH_Et}B_\alpha e^{-iH_Et}$. Expanding out the commutators in

Eq. (37) gives

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\int_0^\infty d\tau \mathrm{tr}_E\Big\{\tilde{H}_I(t)\tilde{H}_I(t-\tau)\tilde{\rho}_S(t)\rho_E - \tilde{H}_I(t-\tau)\tilde{\rho}_S(t)\rho_E\tilde{H}_I(t) - \tilde{H}_I(t)\tilde{\rho}_S(t)\rho_E\tilde{H}_I(t-\tau) + \tilde{\rho}_S(t)\rho_E\tilde{H}_I(t-\tau)\tilde{H}_I(t)\Big\}. \quad (40)$$

Now, let us define environment correlation functions as

$$C_{\alpha\beta}(t,s) = \langle B_\alpha(t)B_\beta(s)\rangle_E = \mathrm{tr}_E(B_\alpha(t)B_\beta(s)\rho_E). \quad (41)$$

In the following, we shall assume that the environment is in a stationary (equilibrium) state, such that $[H_E, \rho_E] = 0$. In this case, we can show that

$$C_{\alpha\beta}(t,s) = \mathrm{tr}_E(B_\alpha(t-s)B_\beta\rho_E) \equiv C_{\alpha\beta}(t-s). \quad (42)$$

Hence, inserting the decomposition of Eq. (39) into the master equation [Eq. (40)], and using the definition of the environment correlation functions in Eq. (42), we find

$$\frac{\partial}{\partial t}\tilde{\rho}_S(t) = -\sum_{\alpha\beta}\int_0^\infty d\tau\Big([A_\alpha(t), A_\beta(t-\tau)\tilde{\rho}_S(t)]C_{\alpha\beta}(\tau) + [\tilde{\rho}_S(t)A_\beta(t-\tau), A_\alpha(t)]C_{\beta\alpha}(-\tau)\Big). \quad (43)$$

Note that in getting to Eq. (43), we have again made use of the cylclic property of the trace, this time to write $\mathrm{tr}_E(B_\beta(t-\tau)\rho_E B_\alpha(t)) = \mathrm{tr}_E(B_\alpha(t)B_\beta(t-\tau)\rho_E) = C_{\alpha\beta}(\tau)$, and $\mathrm{tr}_E(B_\alpha(t)\rho_E B_\beta(t-\tau)) = \mathrm{tr}_E(B_\beta(t-\tau)B_\alpha(t)\rho_E) = C_{\beta\alpha}(-\tau)$.

Finally, let's transform Eq. (43) back to the Schrödinger picture using

$$\frac{\partial}{\partial t}\rho_S(t) = -i[H_S, \rho_S(t)] + e^{-iH_S t}\left(\frac{\partial}{\partial t}\tilde{\rho}_S(t)\right)e^{iH_S t}. \quad (44)$$

This gives the Schrödinger picture master equation

$$\frac{\partial}{\partial t}\rho_S(t) = -i[H_S, \rho_S(t)] - \sum_{\alpha\beta}\int_0^\infty d\tau\Big([A_\alpha, A_\beta(-\tau)\rho_S(t)]C_{\alpha\beta}(\tau) + [\rho_S(t)A_\beta(-\tau), A_\alpha]C_{\beta\alpha}(-\tau)\Big). \quad (45)$$

Here, the first term on the right-hand-side generates unitary system evolution due to H_S, while the remaining terms describe the environmental influence on the system state.

To go further, we now require a specific model, which we shall look at in the next section. However, the definition of the correlation functions in Eq. (41) makes it clear which environmental timescales must be short in order for the Markov approximation to be applied: the two-time correlation functions of the environment operators that couple to the system must decay on timescales much shorter than the timescale for the dynamics of the system for the Markov approximation to be valid.

B. Worked example: Damping of a two-level system - the optical master equation

We shall now derive a Born-Markov master equation for a specific model, that of a two-level system (our system of interest S) interacting with a bath of harmonic oscillators (the environment E). This model could represent, for example, the damping of a two-level atom (or quantum dot, etc.) due to its interactions with the surrounding (quantised) electromagnetic field. It is thus one of the basic, though extremely important, models in quantum optics.

We write the Hamiltonian as $H = H_S + H_E + H_I$, where the system Hamiltonian is

$$H_S = \frac{\epsilon}{2}\sigma_z = \frac{\epsilon}{2}(|e\rangle\langle e| - |g\rangle\langle g|), \quad (46)$$

which defines ϵ to be the energy splitting between the ground state ($|g\rangle$) and excited state ($|e\rangle$) of the two-level system. The environment Hamiltonian,

$$H_E = \sum_{\mathbf{k}}\omega_{\mathbf{k}} b_{\mathbf{k}}^\dagger b_{\mathbf{k}}, \quad (47)$$

comprises of a bath of harmonic oscillators of frequencies $\omega_{\mathbf{k}}$, with creation (annihilation) operators $b_{\mathbf{k}}^\dagger$ ($b_{\mathbf{k}}$) representing the field modes, and

$$H_I = \sum_{\mathbf{k}}(g_{\mathbf{k}}\sigma_+ b_{\mathbf{k}} + g_{\mathbf{k}}^*\sigma_- b_{\mathbf{k}}^\dagger), \quad (48)$$

is the atom-field coupling within the rotating-wave approximation [4], where the coupling constants are given by $g_{\mathbf{k}}$ ($g_{\mathbf{k}}^*$). Moving into the interaction picture, we find

$$\tilde{H}_I(t) = \sum_{\mathbf{k}}\left(g_{\mathbf{k}}\sigma_+ e^{i\epsilon t}b_{\mathbf{k}}e^{-i\omega_{\mathbf{k}}t} + g_{\mathbf{k}}^*\sigma_- e^{-i\epsilon t}b_{\mathbf{k}}^\dagger e^{i\omega_{\mathbf{k}}t}\right), \quad (49)$$

such that we may write $A_1(t) = \sigma_+ e^{i\epsilon t} = |e\rangle\langle g| e^{i\epsilon t}$, $A_2(t) = \sigma_- e^{-i\epsilon t} = |g\rangle\langle e| e^{-i\epsilon t}$, $B_1(t) = \sum_{\mathbf{k}} g_{\mathbf{k}} b_{\mathbf{k}} e^{-i\omega_{\mathbf{k}}t}$, and $B_2(t) = \sum_{\mathbf{k}} g_{\mathbf{k}}^* b_{\mathbf{k}}^\dagger e^{i\omega_{\mathbf{k}}t}$, using the decomposition presented in Eq. (39).

We now want to calculate the environment correlation functions $C_{\alpha\beta}(\tau)$. In order to do this, we must first define the environment state ρ_E. We shall consider here the case that the environment is assumed to be in thermal (canonical) equilibrium, such that

$$\rho_E = \frac{e^{-H_E/k_B T}}{\mathrm{tr}(e^{-H_E/k_B T})}, \quad (50)$$

where k_B is the Boltzmann constant, and T is the bath temperature. For a thermal equilibrium state ρ_E, the following averages over the environment operators are found:

$$\begin{aligned}
\langle b_{\mathbf{k}} b_{\mathbf{k}'}\rangle_E = \mathrm{tr}_E(b_{\mathbf{k}} b_{\mathbf{k}'}\rho_E) &= 0,\\
\langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}'}^\dagger\rangle_E = \mathrm{tr}_E(b_{\mathbf{k}}^\dagger b_{\mathbf{k}'}^\dagger\rho_E) &= 0,\\
\langle b_{\mathbf{k}} b_{\mathbf{k}'}^\dagger\rangle_E = \mathrm{tr}_E(b_{\mathbf{k}} b_{\mathbf{k}'}^\dagger\rho_E) &= \delta_{\mathbf{k}\mathbf{k}'}(1 + N(\omega_{\mathbf{k}'})),\\
\langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}'}\rangle_E = \mathrm{tr}_E(b_{\mathbf{k}}^\dagger b_{\mathbf{k}'}\rho_E) &= \delta_{\mathbf{k}\mathbf{k}'}N(\omega_{\mathbf{k}'}),
\end{aligned} \quad (51)$$

where

$$N(\omega) = (e^{\omega/k_B T} - 1)^{-1}, \tag{52}$$

is the Bose-Einstein occupation number. As an immediate consequence of Eqs. (51), we can see that $C_{11}(\tau) = C_{22}(\tau) = 0$, since these correlation functions involve tracing only over the products $b_{\mathbf{k}} b_{\mathbf{k}'}$ and $b^\dagger_{\mathbf{k}} b^\dagger_{\mathbf{k}'}$, respectively. The correlation function $C_{12}(\tau)$ is written

$$C_{12}(\tau) = \sum_{\mathbf{k},\mathbf{k}'} g_{\mathbf{k}} g^*_{\mathbf{k}'} e^{-i\omega_{\mathbf{k}}\tau} \langle b_{\mathbf{k}} b^\dagger_{\mathbf{k}'} \rangle_E, \tag{53}$$

which becomes

$$C_{12}(\tau) = \sum_{\mathbf{k}} |g_{\mathbf{k}}|^2 e^{-i\omega_{\mathbf{k}}\tau}(1 + N(\omega_{\mathbf{k}})), \tag{54}$$

again using Eqs. (51). Likewise, we obtain

$$C_{21}(\tau) = \sum_{\mathbf{k}} |g_{\mathbf{k}}|^2 e^{i\omega_{\mathbf{k}}\tau} N(\omega_{\mathbf{k}}). \tag{55}$$

To proceed further, we convert the sum over modes in Eqs. (54) and (55) into an integral over the density of states $D(\omega)$:

$$\begin{aligned} C_{12}(\tau) &= \int_0^\infty d\omega J(\omega) e^{-i\omega\tau}(1 + N(\omega)), \\ C_{21}(\tau) &= \int_0^\infty d\omega J(\omega) e^{i\omega\tau} N(\omega), \end{aligned} \tag{56}$$

where we have defined the spectral density

$$J(\omega) \equiv \sum_{\mathbf{k}} |g_{\mathbf{k}}|^2 \delta(\omega - \omega_{\mathbf{k}}) = D(\omega)|g(\omega)|^2, \tag{57}$$

which is a measure of the system-environment coupling strength, weighted by the environmental density of states.

Now, we substitute these correlation functions, along with the definitions of $A_1(t)$ and $A_2(t)$, into the Schrödinger picture Born-Markov master equation (obtained from Eq. (45)):

$$\begin{aligned} \frac{\partial}{\partial t}\rho_S(t) &= -i[H_S, \rho_S(t)] \\ &\quad - \int_0^\infty d\tau \Big([A_1, A_2(-\tau)\rho_S(t)] C_{12}(\tau) \\ &\quad + [\rho_S(t) A_2(-\tau), A_1] C_{21}(-\tau) \\ &\quad + [A_2, A_1(-\tau)\rho_S(t)] C_{21}(\tau) \\ &\quad + [\rho_S(t) A_1(-\tau), A_2] C_{12}(-\tau) \Big), \end{aligned} \tag{58}$$

which becomes

$$\begin{aligned} \frac{\partial}{\partial t}\rho_S(t) &= -i\frac{\epsilon}{2}[\sigma_z, \rho_S(t)] \\ &\quad - \int_0^\infty d\tau \Big([\sigma_+, \sigma_- \rho_S(t)] e^{i\epsilon\tau} C_{12}(\tau) \\ &\quad + [\rho_S(t)\sigma_-, \sigma_+] e^{i\epsilon\tau} C_{21}(-\tau) \\ &\quad + [\sigma_-, \sigma_+ \rho_S(t)] e^{-i\epsilon\tau} C_{21}(\tau) \\ &\quad + [\rho_S(t)\sigma_+, \sigma_-] e^{-i\epsilon\tau} C_{12}(-\tau) \Big). \end{aligned} \tag{59}$$

Now, let's consider the integrals over τ on the right-hand-side, for example

$$\begin{aligned} \int_0^\infty d\tau e^{i\epsilon\tau} C_{12}(\tau) &= \int_0^\infty d\tau \int_0^\infty d\omega \\ &\quad \left\{ e^{i(\epsilon-\omega)\tau} J(\omega)(1 + N(\omega)) \right\}, \\ &\equiv \Lambda_a(\epsilon), \end{aligned} \tag{60}$$

which we define to be $\Lambda_a(\epsilon)$. The time integral can now be performed with the help of the relation

$$\int_0^\infty dt e^{\pm i\epsilon t} = \pi\delta(\epsilon) \pm i\frac{P}{\epsilon}, \tag{61}$$

where P denotes the Cauchy principal value, and we may then also perform part of the frequency integral by using the resulting delta function. This leaves us with

$$\Lambda_a(\epsilon) = \gamma(\epsilon)(1 + N(\epsilon)) + i(\Delta_1(\epsilon) + \Delta_2(\epsilon)), \tag{62}$$

where we shall see that the real part will give rise to environment-induced decay of the system, and is defined in terms of the (zero-temperature) rate

$$\gamma(\epsilon) = \pi J(\epsilon), \tag{63}$$

while the imaginary parts cause environment-induced energy shifts, and are defined as

$$\begin{aligned} \Delta_1(\epsilon) &= P\int_0^\infty d\omega \frac{J(\omega)}{\epsilon - \omega}, \\ \Delta_2(\epsilon) &= P\int_0^\infty d\omega \frac{J(\omega)N(\omega)}{\epsilon - \omega}. \end{aligned} \tag{64}$$

Notice that both the (zero-temeprature) rate and the energy shifts are functions of the system energy splitting ϵ, and hence the environmental influence on the system is naturally dependent on this energy scale within the theory as well. In particular, the rate $\gamma(\epsilon)$ is directly proportional to the spectral density evaluated at ϵ, and hence we might expect the rate to vary quite significantly with the system energy splitting, provided that the spectral density does too. This depends, of course, on the details of the environment density of states and the frequency dependence of the couplings $g(\omega)$.

In a similar manner to above, we may define

$$\Lambda_b(\epsilon) = \int_0^\infty d\tau e^{i\epsilon\tau} C_{21}(-\tau), \tag{65}$$

which we find can be written

$$\Lambda_b(\epsilon) = \gamma(\epsilon)N(\epsilon) + i\Delta_2(\epsilon), \tag{66}$$

while the remaining integrals can be given as $\int_0^\infty d\tau e^{-i\epsilon\tau} C_{12}(-\tau) = \Lambda^*_a(\epsilon)$ and $\int_0^\infty d\tau e^{-i\epsilon\tau} C_{21}(\tau) = \Lambda^*_b(\epsilon)$. Substituting into Eq. (59), and expanding out

the commutators, we then arrive at what is know as the **optical master equation**:

$$\begin{aligned}\frac{\partial}{\partial t}\rho_S &= -i\frac{\epsilon'}{2}[\sigma_z,\rho_S] \\ &+\gamma(\epsilon)(N(\epsilon)+1)(2\sigma_-\rho_S\sigma_+ - \{\sigma_+\sigma_-,\rho_S\}) \\ &+\gamma(\epsilon)N(\epsilon)(2\sigma_+\rho_S\sigma_- - \{\sigma_-\sigma_+,\rho_S\}),\end{aligned} \tag{67}$$

where $\epsilon' = \epsilon + \Delta_1(\epsilon) + 2\Delta_2(\epsilon)$ is the shifted system energy splitting, and $\{a,b\} = ab + ba$ is the anticommutator.

Let's now interpret each term in the optical master equation. The first term gives rise to coherent system dynamics at an environment-shifted energy scale ϵ'; this energy shift is known as the Lamb shift [5]. The second term describes decay of the two-level system from the excited state to the ground state due to the emission of radiation into the electromagnetic environment, at a rate proportional to $\gamma(\epsilon)(N(\epsilon)+1)$. This term therefore captures both the spontaneous and stimulated emission process, with rates proportional to $\gamma(\epsilon)$ and $\gamma(\epsilon)N(\epsilon)$, respectively. The spontaneous emission process can always occur if there are available states to decay into, and is therefore independent of the occupation number $N(\epsilon)$, while the stimulated emission process requires occupation of environmental states at energy matching the system level splitting ϵ. Likewise, the final term in Eq. (67) describes absorption processes, and is given by a rate proportional to $\gamma(\epsilon)N(\epsilon)$, which also requires occupied environmental states matching the system energy ϵ.

C. Solving the master equation

In the basis $\{|e\rangle, |g\rangle\}$ we can write the reduced system density operator in matrix form as

$$\rho_S(t) = \begin{pmatrix} \rho_{ee}(t) & \rho_{eg}(t) \\ \rho_{ge}(t) & \rho_{gg}(t) \end{pmatrix}, \tag{68}$$

where $\rho_{ij}(t) = \langle i|\,\rho_S(t)\,|j\rangle$, for $i,j \in e,g$. Thus, using

$$\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},\ \sigma_+ = \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix},\ \sigma_- = \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix}, \tag{69}$$

we get the following set of evolution equations for the density matrix elements from the optical master equation:

$$\begin{aligned}\dot\rho_{ee}(t) &= -2\gamma(N+1)\rho_{ee}(t) + 2\gamma N\rho_{gg}(t), \\ \dot\rho_{gg}(t) &= -2\gamma N\rho_{gg}(t) + 2\gamma(N+1)\rho_{ee}(t), \\ \dot\rho_{eg}(t) &= -[\gamma(2N+1) + i\epsilon']\rho_{eg}(t), \\ \dot\rho_{ge}(t) &= -[\gamma(2N+1) - i\epsilon']\rho_{ge}(t),\end{aligned} \tag{70}$$

where $\gamma \equiv \gamma(\epsilon)$ and $N \equiv N(\epsilon)$. The first two of these equations are the Einstein rate equations, and hence again demonstrate the physical interpretation of the damping terms in the optical master equation as emission and absorption processes. The last two equations are complex conjugates of each other, where the terms $\pm i\epsilon'$ give rise to precession about the z-axis.

In the zero temperature limit, $N \to 0$, and the solutions to Eqs. (70) take on particularly simple forms:

$$\begin{aligned}\rho_{ee}(t) &= e^{-2\gamma t}\rho_{ee}(0), \\ \rho_{gg}(t) &= 1 - e^{-2\gamma t}\rho_{ee}(0), \\ \rho_{eg}(t) &= e^{-i\epsilon' t}e^{-\gamma t}\rho_{eg}(0), \\ \rho_{ge}(t) &= e^{i\epsilon' t}e^{-\gamma t}\rho_{ge}(0),\end{aligned} \tag{71}$$

Notice that the population relaxation rate is twice the damping rate of the coherences. Often, the inverse of the population relaxation rate is defined as the population relaxation time $T_1 = 1/(2\gamma)$, while, in the absence of any other decoherence processes, the dephasing time is given by $T_2 = 1/\gamma = 2T_1$.

At finite temperature, the solutions to Eqs. (70) are still straightforward, though we shall not give them explicitly here. In this case, the population relaxation rate is given by $2\gamma(2N+1)$, while the dephasing rate is $\gamma(2N+1)$, so the relation $T_2 = 2T_1$ still holds, in the absence of other dephasing processes. Setting $\dot\rho_{ij}(t) = 0$ on the right-hand-side of Eqs. (70) gives the stead-state solution

$$\begin{aligned}\rho_{ee}(\infty) &= \frac{N}{1+2N}, \\ \rho_{gg}(\infty) &= \frac{1+N}{1+2N}, \\ \rho_{eg}(\infty) &= 0, \\ \rho_{ge}(\infty) &= 0,\end{aligned} \tag{72}$$

which is a thermal distribution between the states $|g\rangle$ and $|e\rangle$.

1. *Bloch equations*

An alternative representation of the solutions to the optical master equation is provided by the Bloch equations. Since

$$\begin{aligned}\langle\sigma_z\rangle_t = \mathrm{tr}(\sigma_z\rho(t)) &= \rho_{ee}(t) - \rho_{gg}(t), \\ \langle\sigma_+\rangle_t = \mathrm{tr}(\sigma_+\rho(t)) &= \rho_{ge}(t), \\ \langle\sigma_-\rangle_t = \mathrm{tr}(\sigma_-\rho(t)) &= \rho_{eg}(t),\end{aligned} \tag{73}$$

and $\rho_{ee}(t) + \rho_{gg}(t) = 1$, we can re-write Eqs. (70) as operator expectation value (Bloch) equations instead:

$$\begin{aligned}\dot{\langle\sigma_z\rangle}_t &= -2\gamma[\langle\sigma_z\rangle_t(2N+1)+1], \\ \dot{\langle\sigma_-\rangle}_t &= -[\gamma(2N+1)+i\epsilon']\langle\sigma_-\rangle_t, \\ \dot{\langle\sigma_+\rangle}_t &= -[\gamma(2N+1)-i\epsilon']\langle\sigma_+\rangle_t,\end{aligned} \tag{74}$$

with solutions

$$\begin{aligned}\langle\sigma_z\rangle_t &= \frac{e^{-2\gamma(1+2N)t}(1+\langle\sigma_z\rangle_0(1+2N))-1}{1+2N},\\ \langle\sigma_-\rangle_t &= e^{-i\epsilon' t}e^{-\gamma(1+2N)t}\langle\sigma_-\rangle_0,\\ \langle\sigma_+\rangle_t &= e^{i\epsilon' t}e^{-\gamma(1+2N)t}\langle\sigma_+\rangle_0, \end{aligned} \tag{75}$$

which provides a slightly more compact way of expressing the system dynamics than the density matrix elements.

III. CONTROLLING OPEN QUANTUM SYSTEMS

To recap, we have now derived a perturbative master equation governing the evolution of a general open quantum system within the Born-Markov approximations. This relied on a weak-coupling approximation (i.e. perturbation in $\tilde{H}_I(t)$), and also introduced a coarse-graining, such that timescales smaller than the environmental correlation time are not properly resolved. We applied this formalism to the study of the interaction between a two-level system and the surrounding electromagnetic field, and derived the optical master equation governing the two-level system evolution. This equation captured emission and absorption processes due to the exchange of photons between the system and environment.

Let's now consider the case that we drive the system externally, in order to exert some control on its state. For example, we may wish to drive coherent Rabi oscillations between the two system states, or to prepare a particular system state. In this situation, we would like to know how the environmental influence hampers our ability to coherently control the system evolution.

We consider again $H = H_S + H_E + H_I$, where the driven system Hamiltonian (in the dipole approximation) is now given by

$$H_S = \frac{\epsilon}{2}\sigma_z + \Omega\cos\omega_l t\sigma_x, \tag{76}$$

while we assume that H_B and H_I are unchanged. Here, the term $\Omega\cos\omega_l t\sigma_x$ represents a *classical* external driving field applied to the two-level system (e.g. a laser), with Rabi frequency Ω (measuring the interaction strength), and frequency ω_l (see, e.g., Carmichael's books for justification of this form of Hamiltonian). In the absence of the environment, the driving field acts to induce coherent transitions between the two system states. Moving to a rotating frame using

$$H' = UHU^\dagger - iU\frac{\partial}{\partial t}U^\dagger, \tag{77}$$

where we take $U = e^{i\omega_l\sigma_z t/2}$, we find

$$H'_S = \frac{\nu}{2}\sigma_z + \frac{\Omega}{2}(e^{i\omega_l t}+e^{-i\omega_l t})(e^{i\omega_l t}\sigma_+ + e^{-i\omega_l t}\sigma_-), \tag{78}$$

where $\nu = \epsilon - \omega_l$ is the detuning of the driving frequency from resonance. Making the rotating-wave approximation, we ignore fast oscillating terms, to find

$$H'_S \approx \frac{\nu}{2}\sigma_z + \frac{\Omega}{2}\sigma_x, \tag{79}$$

then transform back to the original picture to give

$$H_S = \frac{\epsilon}{2}\sigma_z + \frac{\Omega}{2}(e^{-i\omega_l t}\sigma_+ + e^{i\omega_l t}\sigma_-). \tag{80}$$

The reason for moving back into the original picture is that in general, for optical frequencies, the condition $\epsilon \gg \Omega$ is usually satisfied (for example, for a quantum dot the energy splitting is of order 1 eV, while the obtainable Rabi frequencies are generally of order 1 meV or smaller). Thus, we may approximate the transformation into the interaction picture as

$$\tilde{H}_I(t) = U_0^\dagger H_I U_0 \approx e^{i(\epsilon/2)\sigma_z t}e^{iH_E t}H_I e^{-iH_E t}e^{-i(\epsilon/2)\sigma_z t}, \tag{81}$$

such that the derivation of the optical master equation proceeds in exactly the same manner as in the previous section. Thus, our driven system master equation simply becomes

$$\begin{aligned}\frac{\partial}{\partial t}\rho_S &= -i\frac{\epsilon'}{2}[\sigma_z,\rho_S] - i\frac{\Omega}{2}[(e^{-i\omega_l t}\sigma_+ + e^{i\omega_l t}\sigma_-),\rho_S]\\ &\quad +\gamma(\epsilon)(N(\epsilon)+1)(2\sigma_-\rho_S\sigma_+ - \{\sigma_+\sigma_-,\rho_S\})\\ &\quad +\gamma(\epsilon)N(\epsilon)(2\sigma_+\rho_S\sigma_- - \{\sigma_-\sigma_+,\rho_S\}),\end{aligned} \tag{82}$$

which, when transformed into the rotating-frame again using $U = e^{i\omega_l\sigma_z t/2}$, becomes

$$\begin{aligned}\frac{\partial}{\partial t}\rho'_S &= -i\frac{\nu'}{2}[\sigma_z,\rho'_S] - i\frac{\Omega}{2}[\sigma_x,\rho'_S]\\ &\quad +\gamma(\epsilon)(N(\epsilon)+1)(2\sigma_-\rho'_S\sigma_+ - \{\sigma_+\sigma_-,\rho'_S\})\\ &\quad +\gamma(\epsilon)N(\epsilon)(2\sigma_+\rho'_S\sigma_- - \{\sigma_-\sigma_+,\rho'_S\}),\end{aligned} \tag{83}$$

where $\rho'_S = U\rho_S U^\dagger$, and $\nu' = \epsilon' - \omega_l$.

For this master equation, our Bloch equations then become

$$\begin{aligned}\langle\dot{\sigma_z}\rangle_t &= i\Omega(\langle\sigma_-\rangle_t - \langle\sigma_+\rangle_t) - 2\gamma[\langle\sigma_z\rangle_t(2N+1)+1],\\ \langle\dot{\sigma_-}\rangle_t &= i\frac{\Omega}{2}\langle\sigma_z\rangle_t - [\gamma(2N+1)+i\nu']\langle\sigma_-\rangle_t,\\ \langle\dot{\sigma_+}\rangle_t &= -i\frac{\Omega}{2}\langle\sigma_z\rangle_t - [\gamma(2N+1)-i\nu']\langle\sigma_+\rangle_t,\end{aligned} \tag{84}$$

which are known as the **optical Bloch equations**. They are sometimes written in the equivalent form

$$\begin{aligned}\langle\dot{\sigma_x}\rangle_t &= -\gamma(2N+1)\langle\sigma_x\rangle_t - \nu'\langle\sigma_y\rangle_t,\\ \langle\dot{\sigma_y}\rangle_t &= \nu'\langle\sigma_x\rangle_t - \gamma(2N+1)\langle\sigma_y\rangle_t - \Omega\langle\sigma_z\rangle_t,\\ \langle\dot{\sigma_z}\rangle_t &= \Omega\langle\sigma_y\rangle_t - 2\gamma[\langle\sigma_z\rangle_t(2N+1)+1].\end{aligned} \tag{85}$$

Considering the simplest case of resonant driving, $\nu' = 0$, and zero temperature, $N = 0$, we solve the Bloch equations for the initial state $\langle\sigma_z\rangle_0 = -1$, $\langle\sigma_-\rangle_0 = 0$, $\langle\sigma_+\rangle_0 = 0$, i.e. all population initially in the system ground-state, to find that the population difference obeys

$$\langle\sigma_z\rangle_t = \frac{-2\gamma^2}{2\gamma^2+\Omega^2}\left[1+\frac{\Omega^2}{2\gamma^2}e^{-\frac{3\gamma}{2}t}\left(\cos\chi t+\frac{3\gamma}{2\chi}\sin\chi t\right)\right], \tag{86}$$

where $\chi = \sqrt{\Omega^2-\gamma^2/4}$ is a *generalised Rabi frequency*. The driven system thus performs exponentially damped Rabi oscillations between the states $|g\rangle$ and $|e\rangle$, tending towards a steady state $\langle\sigma_z\rangle_\infty = -2\gamma^2/(2\gamma^2+\Omega^2)$ in the long time limit. Note that as $\Omega \to 0$, we recover $\langle\sigma_z\rangle_\infty \to -1$, as expected at zero temperature in the absence of driving. In the opposite limit, when Ω becomes very large compared to γ (but still much smaller than ϵ), $\langle\sigma_z\rangle_\infty \to 0$, and population is shared almost equally between the ground and excited states in the long time limit.

Appendix A: Alternative derivation of the time-local master equation

Here we follow the master equation derivation given in Chapter 6 of *Quantum Optics*, by D. F. Walls and G. J. Milburn (Springer, 2008). Our starting point is Eq. (28), with formal solution as given in Eq. (29). We can iterate this solution to generate a series expansion in terms of $\rho(0)$:

$$\begin{aligned}\tilde{\rho}(t) =&\rho(0)+\sum_{n=1}^{\infty}(-i)^n\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{n-1}}dt_n\\ &\times[\tilde{H}_I(t_1),[\tilde{H}_I(t_2),\cdots[\tilde{H}_I(t_n),\rho(0)]]\cdots].\end{aligned} \tag{A1}$$

Taking a trace over the environmental degrees of freedom, we find

$$\begin{aligned}\tilde{\rho}_S(t) =&\rho_S(0)+\sum_{n=1}^{\infty}(-i)^n\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{n-1}}dt_n\times\\ &\mathrm{tr}_E[\tilde{H}_I(t_1),[\tilde{H}_I(t_2),\cdots[\tilde{H}_I(t_n),\rho_S(0)\rho_E(0)]]\cdots],\end{aligned} \tag{A2}$$

where we assume $\rho(0) = \rho_S(0)\rho_E(0)$ at the initial time only. Now, we may write Eq. (A2) in the form

$$\begin{aligned}\tilde{\rho}_S(t) &= (1+W_1(t)+W_2(t)+\cdots)\rho_S(0),\\ &= W(t)\rho_S(0),\end{aligned} \tag{A3}$$

where

$$\begin{aligned}W_n(t) = (-i)^n&\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{n-1}}dt_n\\ &\times\mathrm{tr}_E[\tilde{H}_I(t_1),[\tilde{H}_I(t_2),\cdots[\tilde{H}_I(t_n),(\cdot)\rho_E(0)]]\cdots],\end{aligned} \tag{A4}$$

are superoperators acting on the initial system density operator. Differentiating with respect to time, we have

$$\begin{aligned}\frac{\partial}{\partial t}\tilde{\rho}_S(t) &= (\dot{W}_1(t)+\dot{W}_2(t)+\cdots)\rho_S(0),\\ &= (\dot{W}_1(t)+\dot{W}_2(t)+\cdots)W(t)^{-1}\tilde{\rho}_S(t),\end{aligned} \tag{A5}$$

where we have used Eq. (A3) and assumed that $W(t)$ is invertible. Now, we again consider the case that $\mathrm{tr}_E(\tilde{H}_I(t)\rho_E(0)) = 0$, which means that $W_1(t) = 0$ as well. Thus, to second order, Eq. (A5) becomes

$$\begin{aligned}\frac{\partial}{\partial t}\tilde{\rho}_S(t) &= \dot{W}_2(t)\tilde{\rho}_S(t),\\ &= -\int_0^t dt_1\mathrm{tr}_E[\tilde{H}_I(t),[\tilde{H}_I(t_1),\tilde{\rho}_S(t)\rho_E(0)]],\end{aligned} \tag{A6}$$

which we see has precisely the form of the Born-Markov master equation given in Eq. (35). Note that, unlike the arguments that lead to Eq. (35), the formalism outlined here provides a natural starting point to construct higher order master equations, if necessary.

---

[1] From Eq. (38), we may write $\mathrm{tr}_E[H_I\rho_E(0)] = \sum_\alpha A_\alpha\langle B_\alpha\rangle$, where $\langle B_\alpha\rangle = \mathrm{tr}_E[B_\alpha\rho_E(0)]$. Now, let $H_I' = H_I - \mathrm{tr}_E[H_I\rho_E(0)] = \sum_\alpha A_\alpha(B_\alpha - \langle B_\alpha\rangle)$. Provided that $[H_E,\rho_E(0)] = 0$, i.e. $\rho_E(0)$ is a stationary state, then we see that $\mathrm{tr}_E[\tilde{H}_I'(t),\rho_E(0)] = 0$, as required.

[2] The master equation is now essentially coarse-grained, such that timescales shorter than the environment correlation time cannot be properly resolved.

[3] In quantum optics, a further approximation is often made, known as the secular (or sometimes rotating-wave) approximation, which is valid when the typical timescale for system evolution (due to H_S) is short compared to the relaxation timescale induced by the environment. This allows the master equation to be cast into what is known as Lindblad form. See Section 3.3 of Breuer and Petruccione for more details.

[4] The rotating-wave approximation is generally valid at optical frequencies. In this case, the rotating-wave approximation is equivalent to the secular approximation mentioned in footnote [2], and so our master equation will end up being in Lindblad form. However, it need not generally be true that the two approximations are equivalent.

[5] Actually, the Lamb shift is not fully captured within the rotating-wave approximation, and extra terms would be present if we did not make it (see e.g. Carmichael's books).