Module III, Lecture 10: Lindblad Superoperators

Currently accepted spin relaxation theories are often criticised for the apparent violation of one or more of the following requirements. Any master equation for the density matrix must:

1. Preserve the Hermitian property of the density matrix, so that all probabilities are real.
2. Preserve the trace of the density matrix, so that the sum of all probabilities over a complete orthogonal set of states is equal to 1.
3. Preserve positivity – the probability of all states must be non-negative.

It is easy to see that, unless some phenomenological modifications are introduced, perturbative relaxation theories do indeed violate at least Condition 2.

Definition: a linear map $T:\mathbb{C}^{n\times n}\to\mathbb{C}^{m\times m}$ is called a *positive map* if $T(A)\geq 0$ for all $A\geq 0$, where the ≥ 0 indicates that the operator is Hermitian and positive semidefinite. That is, a positive map preserves the Hermitian property and keeps the operator eigenvalues non-negative.

Definition: a linear map $T:\mathbb{C}^{n\times n}\to\mathbb{C}^{m\times m}$ is called *completely positive* if all maps of the form $I\otimes T$, where I is a unit matrix of any dimension, are also positive maps. An example of a positive map that is not completely positive is a transposition map.

Definition: a *semigroup* is a set equipped with an associative binary operation. It differs from a group in that the identity element might not exist and not every element might have an inverse.

Time propagators in systems with relaxation need not form a group – there might not be an inverse (prove this as an exercise), but they would always be a semigroup, because matrix-vector multiplication is associative.

Theorem 1: for a linear map $T:\mathbb{C}^{n\times n}\to\mathbb{C}^{n\times n}$ to be the generator of a dynamical semigroup of $\mathbb{C}^{n\times n}$, it is necessary and sufficient for the following conditions to hold:

$$\begin{aligned}&\mathrm{Tr}\left(P_n T P_k\right)\geq 0, \qquad n\neq k\\ &\sum_n \mathrm{Tr}\left(P_n T P_k\right)=0 \qquad \forall k\end{aligned}$$

where $\{P_k\}$ is a complete family of self-adjoint projectors in $\mathbb{C}^{n\times n}$.

The general form of a completely positive map $A\to T(A)$ is therefore the following:

$$T(X)=\sum_n A_n X A_n^\dagger$$

where A_n are operators from the same space as X. In the infinitesimal case, the most general generator of a completely positive density matrix evolution has the following form (see the original papers by Lindblad):

$$\frac{\partial\hat{\rho}}{\partial t}=-\frac{i}{\hbar}[\hat{H},\hat{\rho}]+\sum_{nm}h_{nm}\left(-\hat{\rho}\hat{L}_m^\dagger\hat{L}_n-\hat{L}_m^\dagger\hat{L}_n\rho+2\hat{L}_n\rho\hat{L}_m^\dagger\right)$$

Where $\{\hat{L}_n\}$ is an orthonormal basis of the operator space and the matrix h_{nm} must be time-independent and positive definite. This is useful to know, but this does not get us very far, because the expressions for h_{nm} are not provided.

A part of the answer may be gleaned from BRW theory, which may be re-written in a Lindblad form:

$$\int_0^\infty G_{kmpq}(\tau)[\hat{Q}_{km},[e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau},\hat{\rho}]]d\tau = \int_0^\infty G_{kmpq}(\tau)\begin{pmatrix} \hat{Q}_{km}e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\hat{\rho} - \hat{Q}_{km}\hat{\rho}e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau} - \\ -e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\hat{\rho}\hat{Q}_{km} + \hat{\rho}e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\hat{Q}_{km} \end{pmatrix} d\tau =$$

$$= \hat{Q}_{km}\left[\int_0^\infty G_{kmpq}(\tau)\left(e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\right)d\tau\right]\hat{\rho} - \hat{Q}_{km}\hat{\rho}\left[\int_0^\infty G_{kmpq}(\tau)\left(e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\right)d\tau\right] -$$

$$-\left[\int_0^\infty G_{kmpq}(\tau)\left(e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\right)d\tau\right]\hat{\rho}\hat{Q}_{km} + \hat{\rho}\left[\int_0^\infty G_{kmpq}(\tau)\left(e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\right)d\tau\right]\hat{Q}_{km} =$$

$$= \hat{A}\hat{B}\hat{\rho} - \hat{A}\hat{\rho}\hat{B} - \hat{B}\hat{\rho}\hat{A} + \hat{\rho}\hat{B}\hat{A}; \quad \hat{A} = \hat{Q}_{km}, \quad \hat{B} = \left[\int_0^\infty G_{kmpq}(\tau)\left(e^{i\hat{H}_0\tau}\hat{Q}_{pq}^\dagger e^{-i\hat{H}_0\tau}\right)d\tau\right]$$

This form can be brought to a Lindblad form by a linear transformation, but the coefficient matrix in the result would not be positive definite. We do therefore need a new *ab initio* treatment to obtain h_{nm}.

We shall make the standard assumptions that the time scale Δt on which we are operating is "short" on the scale of coherent dynamics, but "long" compared to the environment state memory time:

$$\left\|\hat{H}_0\right\|^{-1} \gg \Delta t \gg \tau_{\mathrm{C}}$$

We shall also assume that the evolution of the density matrix only depends on the current state of the system, and not on its previous history:

$$\frac{\partial\hat{\rho}(t)}{\partial t} = \hat{\hat{L}}\hat{\rho}(t) \quad \Rightarrow \quad \hat{\rho}(t) = \exp\left(\hat{\hat{L}}t\right)\hat{\rho}(0)$$

where $\hat{\hat{L}}$ is some as yet unknown generator of the propagator semigroup (a semigroup is a group that might not have an inverse defined for each element). The action by any superoperator in Liouville space, including the action of the exponential propagator above, may be written as:

$$\exp\left(\hat{\hat{L}}t\right)\hat{\rho} = \sum_{ij} c_{ij}(t)\hat{F}_i\hat{\rho}\hat{F}_j^\dagger$$

where $\{\hat{F}_n\}$ is a complete orthornormal operator basis and the $N^2 \times N^2$ matrix $c_{ij}(t)$ is Hermitian positive definite. The basis $\{\hat{F}_n\}$ has to have the identity operator $\hat{E}$ in it, and we shall choose $\hat{F}_{N^2} = \sqrt{1/N}\hat{E}$. This choice would make the other $N^2 - 1$ operators in the basis traceless. Because the generator $\hat{\hat{L}}$ is related to the infinitesimal propagator, we get:

$$\hat{\hat{L}}\hat{\rho} = \lim_{\Delta t \to 0} \frac{\exp\left(\hat{\hat{L}}\Delta t\right)\hat{\rho} - \hat{\rho}}{\Delta t} =$$

$$= \lim_{\Delta t \to 0}\left(\frac{N^{-1}c_{N^2,N^2}\left(\Delta t\right) - 1}{\Delta t}\hat{\rho} + \frac{1}{\sqrt{N}}\sum_{i=1}^{N^2-1}\left(\frac{c_{i,N^2}\left(\Delta t\right)}{\Delta t}\hat{F}_i\hat{\rho} + \frac{c_{N^2,i}\left(\Delta t\right)}{\Delta t}\hat{\rho}\hat{F}_i^{\dagger} \right) + \sum_{ij=1}^{N^2-1}\frac{c_{ij}\left(\Delta t\right)}{\Delta t}\hat{F}_i\hat{\rho}\hat{F}_j^{\dagger} \right)$$

in which the coefficients $c_{ij}\left(t\right)$ can, in principle, be obtained (it would not be easy) from the generalized cumulant expansion for the effective propagator. We shall group the terms in this equation as follows:

$$a_{N^2,N^2} = \lim_{\Delta t \to 0}\frac{c_{N^2,N^2}\left(\Delta t\right) - N}{\Delta t}, \quad a_{i,N^2} = \lim_{\Delta t \to 0}\frac{c_{i,N^2}\left(\Delta t\right)}{\Delta t}, \quad a_{ij} = \lim_{\Delta t \to 0}\frac{c_{ij}\left(\Delta t\right)}{\Delta t}$$

$$F = \frac{1}{\sqrt{N}}\sum_{i=1}^{N^2-1}a_{i,N^2}\hat{F}_i, \quad G = \frac{1}{2N}a_{N^2,N^2}\hat{E} + \frac{1}{2}\left(\hat{F} + \hat{F}^{\dagger}\right), \quad H = \frac{1}{2i}\left(\hat{F}^{\dagger} - \hat{F}\right)$$

With that in place, we have:

$$\hat{\hat{L}}\hat{\rho} = -i[\hat{H},\hat{\rho}] + \left(\hat{G}\hat{\rho} + \hat{\rho}\hat{G}\right) + \sum_{ij=1}^{N^2-1}a_{ij}\hat{F}_i\hat{\rho}\hat{F}_j^{\dagger}$$

Because we assume that the trace of the density matrix is preserved during the time evolution, we must have

$$\frac{\partial}{\partial t}\mathrm{Tr}\left(\hat{\rho}\right) = \mathrm{Tr}\left(\hat{\hat{L}}\hat{\rho}\right) = \mathrm{Tr}\left(2\hat{G}\hat{\rho} + \sum_{ij=1}^{N^2-1}a_{ij}\hat{F}_i\hat{\rho}\hat{F}_i^{\dagger} \right) = 0$$

(remember that the trace of a commutator is zero, so the $[\hat{H},\hat{\rho}]$ term does not enter, and that cyclic permutations are allowed under the trace). It follows that

$$\hat{G} = -\frac{1}{2}\sum_{ij=1}^{N^2-1}a_{ij}\hat{F}_i\hat{\rho}\hat{F}_j^{\dagger}$$

and the result is the famous Lindblad form for the generator of the propagator semigroup:

$$\hat{\hat{L}}\hat{\rho} = -i[\hat{H},\hat{\rho}] + \sum_{ij=1}^{N^2-1}a_{ij}\left(\hat{F}_i\hat{\rho}\hat{F}_j^{\dagger} - \frac{1}{2}\left(\hat{F}_j^{\dagger}\hat{F}_i\hat{\rho} + \hat{\rho}\hat{F}_j^{\dagger}\hat{F}_i\right) \right)$$

The matrix a_{ij} may in principle be diagonalized, yielding (for some other operator basis $\{\hat{A}_n\}$):

$$\hat{\hat{L}}\hat{\rho} = -i[\hat{H},\hat{\rho}] + \sum_{k=1}^{N^2-1}\gamma_k\left(\hat{A}_k\hat{\rho}\hat{A}_k^{\dagger} - \frac{1}{2}\left(\hat{A}_k^{\dagger}\hat{A}_k\hat{\rho} + \hat{\rho}\hat{A}_k^{\dagger}\hat{A}_k\right) \right)$$

Lindblad theory is largely an abstract exercise in Lie groups. The practical task of finding the expressions for the basis sets and the coefficients in a specific physical system amounts essentially to performing the treatment using one of the relaxation theories we have discussed earlier in the course.