On the modeling of irreversibility by relaxator Liouville dynamics

János Hajdu and Martin Janßen*

Institut für Theoretische Physik, Universität zu Köln, Zülpicher Str. 77, D-50937 Köln

(Dated: September 4, 2025)

Abstract

A general approach to modeling irreversibility starting from microscopic reversibility is presented. The time t_s up to which relevant degrees of freedom of a system are tracked is extremely much shorter than the spectral resolution time t_e that would be necessary to resolve the spectrum of all degrees of freedom involved. A relaxator that breaks reversibility condenses in the Liouville operator of the relevant degrees of freedom. The irrelevant degrees of freedom act as an environment to the system. The irreversible relaxator Liouville equation contains memory effects and initial correlations of all degrees of freedom. Stationary states turn out to be generically unique and independent of the initial conditions and exceptions are due to degeneracies. Equilibrium states lie in the relaxator's kernel yielding a stationary Pauli master equation. Kinetic equations for one-particle densities are constructed as special cases of relaxator Liouville dynamics. Kubo's linear response theory is generalized to relaxator Liouville dynamics and related to irreversibility within the system. In a weak coupling approximation between system and environment the relaxator can be reduced to environmental correlations and bilinear system operators. Markov approximation turns the relaxator Liouville dynamics into a semi-group dynamics.

*Electronic address: mj@thp.uni-koeln.de

I. INTRODUCTION

The microscopic von Neumann equation, $\dot{\rho} = -\imath L\rho$, for quantum states ρ is reversible for $L\cdot = [H, \cdot]$ with Hermitian Hamiltonian H. To each solution, $\rho(t) = e^{-\imath Lt}\rho_0$, the inverse operation, $e^{-\imath L(-t)}\rho(t) = \rho_0$, describes the reversed motion. The von Neumann entropy, $S = -\operatorname{Tr}\rho\ln\rho$ (we use units in which $\hbar = k_B = 1$), of an arbitrary state ρ stays constant according to the microscopic von Neumann equation (see e.g. [1]). Similar conclusions hold in classical Hamiltonian mechanics (Liouville theorem).

Basic laws of thermodynamics state that macroscopic systems consisting of a huge number N of constituents reach a steady state after some characteristic relaxation time τ_r. Lots of initial states reach the same stationary state. In closed systems with fixed average energy the stationary state is an equilibrium state, very well described as a canonical Gibbs state, $e^{-H/T}/Z$ with temperature T and partition sum Z. Among all possible unit trace states with fixed average value of energy $E = \langle H\rangle$ the Gibbs state has the maximum von Neumann entropy. In closed systems with fixed average energy E the von Neumann entropy will change from its initial value to the maximum value of the Gibbs state. The reversed motion never happens. The condition of a macroscopic description by the Gibbs state as an equilibrium state relies on the condition of large N manifesting in very small relative fluctuations of thermodynamic state variables like energy E of the order of $1/\sqrt{N}$ (see e.g. [2]).

The irreversibility of macroscopic processes expressed by the basic thermodynamic laws seems to be in contradiction with the reversibility of the microscopic von Neumann equation. The debate of this issue goes back to the early days of statistical mechanics and is still with us (see e.g. [3]-[14]). To stick to the von Neumann equation and modify e.g. the definition of entropy and/or refer to the role of special initial conditions that are rare in some probabilistic sense cannot suffice to recover macroscopic irreversibility, because reversible motions cannot relax to a steady state in a manner independent of the initial state. The dynamics should tell us about the relaxation time τ_r to reach stationarity. To extract relaxation as an emerging qualitative feature from the von Neumann equation it needs a treatment under the condition of macroscopic N. It is long known that microscopic reversibility has one consequence which must be sacrificed for macroscopic irreversibility: recurrence of initial states after some recurrence time T_R. Thus, there is one obvious condition for macroscopic irreversibility, already formulated by Smoluchowski in 1916 [4]: the available time t_s to explore the system

has to be negligible compared to the recurrence time T_R. Typically, recurrence times grow double exponential with N and quickly reach super-astronomic time scales for many body systems (for a detailed discussion see [15]).

As we will argue now, the essential condition for irreversibility in macroscopic systems is the smallness of t_s with respect to the time scale t_e that is needed to explore the full spectral properties of all degrees of freedom involved. In macroscopic systems t_e scales exponentially with N (see below) while t_s does not depend on N. The condition $t_s \ll t_e$ guarantees that the Smoluchowski criterion is fulfilled with ease. The spectral exploration time t_e is the inverse of the typical spacing of eigenfrequencies Ω_n of L in a certain dynamic regime of frequencies Ω. The condition $t_s \ll t_e$ means that we can treat the spectrum of L as continuous with a spectral density $\gamma_L(\Omega)$, because we cannot resolve it when following the system over reasonable time scales of order t_s. One immediate consequence is that recurrence times are treated as infinite, because in a continuum of periods there is no finite common least multiplier. A second consequence corresponds to the relaxation of time dependent correlation functions $c_A(t) = \langle A(t)A\rangle$ of time dependent observables $A(t)$ with respect to a stationary state ρ, $c_A(t) = \sum_n P_n^{[A]} e^{\imath\Omega_n t}$, with $P_n^{[A]} > 0$. Such superposition with lots of different frequencies leads to almost cancellations and thus irreversible relaxation to a stationary value in a macroscopic system under a limited spectral resolution. With full spectral resolution of all terms in the superposition, one could find almost recurrence of some initial value after T_R, but on time scales $t_s \ll t_e \ll T_r$ the relaxation to stationarity is the appropriate picture [15]. Indeed, to take imperfect resolution into account one approximates the sum in $c_A(t)$ with an integral over the density $\gamma_L(\Omega)$, with $\gamma_L(\Omega) \cdot P^{[A]}(\Omega)$ of a finite range of order $1/\tau_A$. Then one finds the correlations to relax to stationarity on a scale of order τ_A. Thus, the condition of $t_s \ll t_e$ in macroscopic systems is the reason for treating the spectrum of all degrees of freedom as continuous with a spectral function $\gamma_L(\Omega)$. With the continuity of $\gamma_L(\Omega)$ macroscopic irreversibility shows up in correlation functions. In addition, the continuity and smoothness assumption of the spectral density is necessary in the formulation of equilibrium thermodynamics. The very definition of entropy relies on these conditions (see e.g. [2]), $S(E) = \ln(\gamma_H(E) \cdot \Delta)$, where $\gamma_H(E)$ is the density of energy eigenvalues at energy E and Δ is an arbitrary fixed energy window sufficiently separated from the lower value $1/t_e$ and the upper value $1/t_s$. Only then, temperature $T(E)$ can be identified by $1/T(E) := \partial_E S(E)$. Energy as a continuous parameter of the macroscopic

thermodynamic equilibrium state manifold could not be defined without the continuity condition of the spectral density $\gamma_H(E)$. Thermodynamics with Legendre transform relations between thermodynamic potentials derived from partition sums of different equilibrium ensembles relies on the smoothness condition as well and is justified up to relative fluctuations of order $1/\sqrt{N}$ in the thermodynamic potentials (see e.g. [16]). Since thermodynamic entropy $S(E)$ is an extensive quantity, the condition of continuity $t_s \ll t_e$ is easily justified as $t_e \propto \gamma_L(E) \propto e^{S(E)} \propto e^N$ which is exponentially larger in N than t_s. Since the spectral densities $\gamma_H(E)$ and $\gamma_L(\Omega)$ are related by convolution, $\gamma_L(\Omega) = (\gamma_H(-E) * \gamma_H(E))(\Omega)$, qualitative considerations of continuity and scaling with N apply equally to both.

In addition to pure reversible mechanics, thermodynamics requires in its foundation an unavoidable statistical element for large numbers N: the assumption of continuously distributed energy eigenvalues due to an unavoidable resolution limit of the eigenvalues. This condition of continuous spectrum suffices to explicitly construct an irreversible equation of motion for relevant variables from the microscopic von Neumann equation valid under the condition $t_s \ll t_e$. This will be shown in Sec. II. It is a consequent combination of projection techniques pioneered by Nakajima [17] and Zwanzig [18] and the resolvent method in the frequency picture pioneered by Fano [19]. The relevance of variables is conditioned by the fact that their spectrum under isolated conditions could be resolved on the time scale t_s. By the projection to relevant variables the dynamics is no longer local in time. Retardation effects for relevant variables show up via integrating out irrelevant variables and the resulting dynamics is non-Markovian. It is therefore advantageous to change to the frequency picture of dynamic equations (resolvent equations) via the Laplace transform. In the following $f(\Omega + \imath\epsilon)$ denotes the Laplace transform of time dependent function $f(t)$, $f(\Omega + \imath\epsilon) = \int_0^\infty dt f(t) e^{i\Omega t - \epsilon t}$. Here Ω and ϵ are real valued frequency and positive regularizing inverse time scale, respectively. For the von Neumann equation of all degrees of freedom this means

$$\rho_{\rm tot}(\Omega + \imath\epsilon) = iG_{\rm tot}(\Omega + \imath\epsilon)\rho_{\rm tot_0}\,, \tag{1}$$

where $\rho_{\rm tot_0}$ is an initial state and

$$G_{\rm tot}(z) = [z - L_{\rm tot}]^{-1} \tag{2}$$

is the resolvent of Liouville $L_{\rm tot}$ with poles at the real valued eigenfrequencies Ω_n symmetrically distributed around $\Omega = 0$. When the limit to a continuous spectrum of $L_{\rm tot}$ has been

taken, the limit $\epsilon \to 0^+$ can usually be performed. For systems with a many body localization (see e.g.[20]) phase with respect to a certain basis the global spectrum may be dense, but the typical local density of states vanishes for $N \to \infty$. Thus, typically $t_e \to 0$ and no equilibration can take place. Such many body localization phases will not be considered further in the course of this work.

The exact relaxator Liouville equation for relevant variable states under the condition of continuous total spectrum will be discussed in Section II. It has the same structure as (1). However, the initial state and the resolvent are restricted to the relevant degrees of freedom and depend on frequency showing memory effects. The initial state $\rho(\omega)_0$ describes a frequency dependent initial state for relevant variables incorporating initial correlations between all degrees of freedom. Furthermore, the resolvent is now

$$G(z) = [z - L(\omega)]^{-1} \tag{3}$$

for a frequency dependent non-Hermitian relaxator Liouville operator $L(\omega)$ acting on relevant variable states

$$\rho(\omega) = iG(\omega + \imath 0^+)\rho(\omega)_0 \,. \tag{4}$$

Equation (4) is the general equation of motion for relevant degrees of freedom in many body systems under the condition $t_s \ll t_e$.

In Section III we derive and discuss the general properties of relaxator Liouville dynamics thereby relating $L(\omega)$ to the Hamiltonian of relevant variables interacting with irrelevant variables. Although there are several approaches to non-Markovian irreversible dynamics (see e.g. [21]-[23]) we believe that the relaxator Liouville approach is most general and covers them in a systematic and transparent way, well suited for phenomenological, axiomatic, and constructive modelings. In Section IV kinetic equations (see e.g. [24]) for one-particle densities are constructed as a special application of Liouville relaxator dynamics where one-particle degrees of freedom are taken as the relevant ones. The corresponding relaxator determines the self-energy in the corresponding non-equilibrium Green's function NEGF (see e.g. [25],[26]). By incorporating driving fields for currents in the system we derive linear response susceptibilities for relaxator Liouville dynamics that generalize Kubo's linear response theory [8] and discuss different sources of irreversibility in Sec. V. By a weak coupling approximation in Sec. VI it is possible to express the relaxator Liouville in terms of isolated properties of relevant and irrelevant variables. We investigate the Markov approximation

to compare our general approach with the well known semi-group approach after [27] and [28] (for overviews see e.g. [29]-[33]) to irreversible systems in Sec. VII. Finally we give a summary in Sec. VIII.

II. INSTABILITY OF REVERSIBILITY

We consider an arbitrary isolated quantum system denoted as total system that allows to distinguish two groups of degrees of freedom, relevant and irrelevant, there numbers being N_s and N_e, respectively. The total Hilbert space is the product of the Hilbert spaces of relevant and irrelevant variables. We want to model the dynamics of the relevant variables up to a time scale t_s.

It is now a defining condition for the distinction between relevant and irrelevant degrees of freedom that the total system as well as the idealization of an isolated system of irrelevant variables has a spectral resolution time of the same order, denoted as t_e, which has to be larger than t_s by a factor which is exponential in $N_e/N_s \gg 1$. The total system has a huge number $N = N_e + N_s \approx N_e$ of degrees of freedom most of which are irrelevant. It has become popular to denote such systems of relevant variables in contact with a huge number of irrelevant variables as open system (in the sense of [29]). The relevant variables are denoted simply as system variables and the irrelevant ones as environment. Note however, that the notion of open system here only means a system of variables which is not isolated but interacts with the environment. It is possible, that such open system represents a closed system in the thermodynamic sense, i.e. a system that even in equilibrium exchanges energy with its environment but no matter. Both sets of variables (system and environment) may act within the same volume (e.g. in Brownian motion or in the scattering of electrons with phonons or with lattice imperfections and impurities) and may be even based on the same microscopic particles (e.g. in the relaxation to stationarity of a fluid of particles by self-interaction). It is not necessary that the system exchanges particles with the environment, but it is also not excluded. The belonging of chosen degrees of freedom to system or environment however is held fixed in the modeling. In case the environment surrounds the volume of the system we call it a reservoir (e.g. a small piece of matter subject to radiation). In case that the reservoir is in thermal equilibrium, we call it a heat bath.

The system's state is defined by virtue of a projector $\mathcal{P}$ on the Hilbert-Schmidt space of

linear operators over the Hilbert space of the total system,

$$\rho_{\mathcal{P}} := \mathcal{P}\rho_{\rm tot} = \left(\mathop{\rm Tr}_{\rm env}\rho_{\rm tot}\right)\otimes\rho_{\rm env} =: \rho\otimes\rho_{\rm env}\,, \tag{5}$$

where $\rho_{\rm env}$ is a fixed density operator of the environment and ρ is the searched for density operator of the system. By tracing out the environment the density operator ρ is of much lower dimension as $\rho_{\rm tot}$, but suffices to calculate the expectation value of any observable A of the system in state $\rho_{\rm tot}$,

$$\mathop{\rm Tr}_{\rm tot}\rho_{\rm tot}A\otimes 1_{\rm env} = \mathrm{Tr}\,\rho A\,. \tag{6}$$

We will use the following terminology. The Hilbert-Schmidt space of linear operators over the Hilbert space of the total system is known as the total Liouville space of system plus environment. The projected subspace by $\mathcal{P}$ is the open system's Liouville space, which we also denote as $\mathcal{P}$-space, the complement space to the open system's Liouville space as $\mathcal{Q}$-space. The density operator ρ is an element of the $\mathcal{P}$-space and can be represented by a $d\times d$ dimensional density matrix when the idealized isolated system's Hilbert space is d-dimensional. The complementary projector is denoted as $\mathcal{Q} := 1-\mathcal{P}$. Projector property $\mathcal{P}^2=\mathcal{P}$, $\mathcal{Q}^2=\mathcal{Q}$ and the complement property $\mathcal{P}\mathcal{Q}=\mathcal{Q}\mathcal{P}=0$ are fulfilled for any choice of the fixed normalized environmental density operator $\rho_{\rm env}$ in (5). Note that ρ is independent of the choice of $\rho_{\rm env}$ as long as one proceeds without further approximations involving a specific choice of $\rho_{\rm env}$.

One choice for $\rho_{\rm env}$ in an exact approach is singled out by requiring the projectors $\mathcal{P},\mathcal{Q}$ to be Hermitian with respect to the Hilbert-Schmidt metric,

$$(\sigma_{\rm tot}|\rho_{\rm tot}) \;:=\; \mathop{\rm Tr}_{\rm tot}\sigma_{\rm tot}^\dagger\rho_{\rm tot} = (\rho_{\rm tot}|\sigma_{\rm tot})^*\;, \tag{7}$$

$$(\rho_{\rm tot}|\rho_{\rm tot}) \;\geq 0\;\;, \tag{8}$$

$$(\sigma_{\rm tot}|\mathcal{P}\rho_{\rm tot})^* \;=\; \left(\rho_{\rm tot}|\mathcal{P}^\dagger\sigma_{\rm tot}\right)\,. \tag{9}$$

By writing the density operators in an expansion of system and environmental operators, $\rho_{\rm tot}=\sum_k \rho^{(k)}\otimes\rho_{\rm env}^{(k)}$, one can conclude: the Hermiticity $\mathcal{P}=\mathcal{P}^\dagger$ is valid on the total Liouville space only if $\rho_{\rm env}=d_{\rm env}^{-1}1_{\rm env}$, where $d_{\rm env}$ is the dimension of the environmental Hilbert space, while the Hermiticity of $\mathcal{P}$ on the $\mathcal{P}$-space is valid for any choice of $\rho_{\rm env}$. Hermiticity of $\mathcal{P}$ is passed on to $\mathcal{Q}=1-\mathcal{P}$. Therefore, we prefer the choice $\rho_{\rm env}=d_{\rm env}^{-1}1_{\rm env}$ as long as we perform exact calculations on ρ involving the total Liouville space. In Section VI we will

introduce a weak coupling approximation. Within this weak coupling approximation it is more appropriate to consider the environment to be already in a self-consistent stationary state $\rho_{\rm env}$ which, under equilibrium conditions, acts like a heat bath on the system. Whenever one takes the choice of $\rho_{\rm env}$ not being proportional to $1_{\rm env}$ one cannot rely on the Hermiticity of $\mathcal{P}, \mathcal{Q}$ on the total Liouville space.

The total density operator will be decomposed as

$$\rho_{\rm tot} = \rho_{\mathcal{P}} + \Delta\rho^{\rm corr}\,,\ \Delta\rho^{\rm corr} := \mathcal{Q}\rho_{\rm tot}\,, \tag{10}$$

where $\Delta\rho^{\rm corr}$ captures correlations between system and environment. With the decomposition of the total Liouville

$$L_{\rm tot} = L_{\mathcal{P}} + L_{\mathcal{PQ}} + L_{\mathcal{QP}} + L_{\mathcal{Q}}\,, \tag{11}$$

the equation of motion (4) follows by algebraic means [34] and no approximation is involved. In the derivation it is essential that a projected total resolvent $\mathcal{P}G_{\rm tot}(z)\mathcal{P}$ can always be expressed by the resolvent of a Liouville operating solely on the Liouville space of the open system ($\mathcal{P}$-space),

$$\mathcal{P}G_{\rm tot}(z)\mathcal{P} = G(z) = [z - L(z)]^{-1}\,, \tag{12}$$

and the relaxator Liouville is algebraically given as

$$L(z) = L_{\mathcal{P}} + L_{\mathcal{PQ}}G_{\mathcal{Q}}(z)L_{\mathcal{QP}}\,. \tag{13}$$

It has the following intuitive interpretation: the first term $L_{\mathcal{P}}$ is an isolated system Liouville in the absence of an environment whereas the second term describes virtual processes in the system triggered by the environment, $L_{\mathcal{PQ}}G_{\mathcal{Q}}(z)L_{\mathcal{QP}}$, hopping to $\mathcal{Q}$-space, taking there a lift with isolated $\mathcal{Q}$-propagator and finally hopping back to $\mathcal{P}$-space, see Fig. 1. This second term is no longer Hermitian and breaks reversibility.

The initial state $\rho_0(z)$ in (4) is the initial state ρ_0 plus a virtual change of initial state within the system, caused by the initial correlation ($\Delta\rho_0^{\rm corr}$) that gets a lift by the isolated $\mathcal{Q}$-propagator and hops to $\mathcal{P}$-space [34] (see Fig. 2),

$$\Delta\rho_0^{\rm corr}(z) = L_{\mathcal{PQ}}G_{\mathcal{Q}}(z)\Delta\rho_0^{\rm corr}\,. \tag{14}$$

Equation (4) is an exact equation of motion for states ρ of the system, provided one knows $G_{\mathcal{Q}}(z)$. $G_{\mathcal{Q}}(z)$ can be considered as a propagator of an isolated system with Hermitian

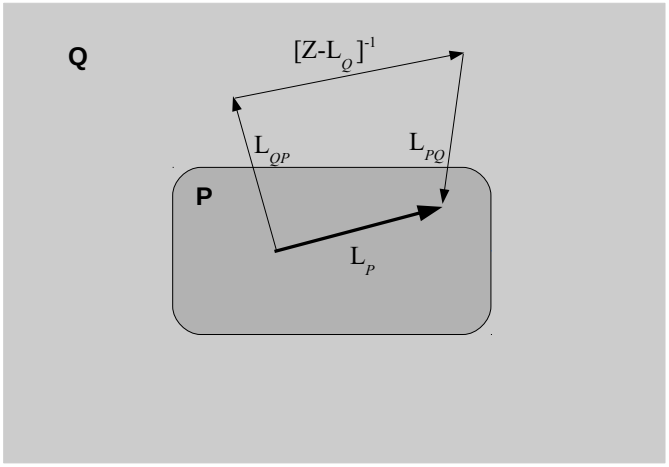

FIG. 1: Visualizing relaxator Liouville dynamics in $\mathcal{P}$-space by direct processes and virtual processes via hopping and propagation in $\mathcal{Q}$-space.

Liouville of the form $L_{\mathcal{Q}}\cdot = [H_{\mathcal{Q}}, \mathcal{Q}\cdot]$ with Hermitian $H_{\mathcal{Q}}$ and corresponding real valued spectrum E_a. If Hermiticity of $\mathcal{Q}$ itself is not guaranteed we can replace $L_{\mathcal{Q}}$ with the total Liouville $L_{\rm tot}$ since in (13) and in (14) the resolvent $G_{\mathcal{Q}}(z)$ is framed by two $\mathcal{Q}$-projectors.

One may object that $L(z)$ relies on the solution of the total system, expressed by $G_{\mathcal{Q}}(z)$. But we do not need the spectral fine structure of $G_{\mathcal{Q}}(z)$. We work with phenomenological and/or constructive modelings of $G_{\mathcal{Q}}(z)$ as an input device to extract general properties and quantitative properties of the dynamics of system states without knowing a detailed solution of the total system.

As to the general properties we decompose $G_{\mathcal{Q}}(z)$ into Hermitian and anti-Hermitian parts, for $\epsilon \to 0+$,

$$G_{\mathcal{Q}}(z) = [z - L_{\mathcal{Q}}]^{-1} = \mathrm{P}\,[\omega - L_{\mathcal{Q}}]^{-1} - \imath\pi\delta(\omega - L_{\mathcal{Q}})\,, \tag{15}$$

where P stands for the Cauchy value on integration. Due to the δ functional in (15), the Hermiticity of $L(z)$ is still valid for almost all values of ω, except for a number of zero measure. If, however, the spectrum Ω_n of complementary dynamics cannot be resolved on the scale of the system's exploration time $t_s \ll t_e$ it has to be treated as distributed with a positive, even spectral density $\gamma_{\mathcal{Q}}(\omega)$ and we arrive at

$$L(\omega) = L_{\mathcal{P}} + \mathcal{D}(\omega)\,. \tag{16}$$

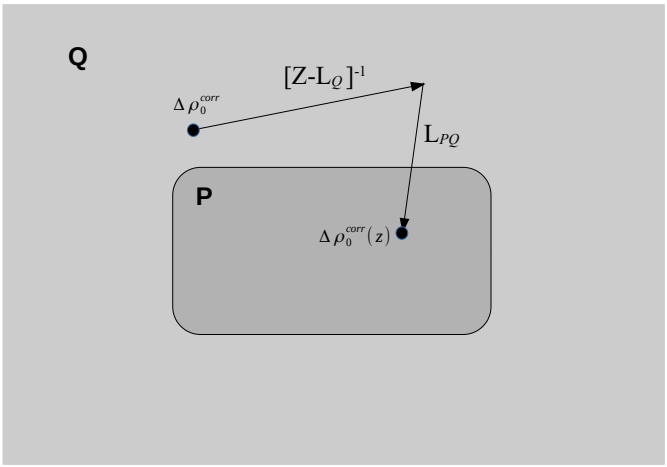

FIG. 2: Visualizing the contribution of virtual processes by initial correlations to the opens sytems's initial state. The correlation part $\Delta\rho_0^{\rm corr}$ of the initial state within the complementary $\mathcal{Q}$-space propagates there and hops to the $\mathcal{P}$-space to contribute to the system's initial state in $\mathcal{P}$-space with $\Delta\rho_0^{\rm corr}(z)$.

where the presence of the dissipator $\mathcal{D}(\omega)$ shows the irreversible character of the dynamics described by $L(\omega)$. The two contributions to the dissipator will be denoted as spectral shift $\Delta\mathcal{H}(\omega)$ (due to the resemblance with the Lamb shift) and as ($-\imath$ times) relaxator $\Gamma(\omega)$,

$$\mathcal{D}(\omega) = \Delta\mathcal{H}(\omega) - \imath\Gamma(\omega)\,, \tag{17}$$

$$\begin{aligned}\Gamma(\omega) &= \pi L_{\mathcal{PQ}}\delta(\omega - L_{\mathcal{Q}})L_{\mathcal{QP}} = \\ &= -\lim_{\epsilon\to 0}\frac{1}{2}L_{\mathcal{PQ}}\left(G_{\mathcal{Q}}(z) - G_{\mathcal{Q}}(z^*)\right)L_{\mathcal{QP}}\,,\end{aligned} \tag{18}$$

$$\Delta\mathcal{H}(\omega) = \frac{1}{\pi}\mathrm{P}\int \mathrm{d}\Omega\,\frac{\Gamma(\Omega)}{\omega - \Omega}\,. \tag{19}$$

Equation (19) shows a Kramers-Kronig relation between the spectral shift and the relaxator. With the convenient Hermitian choice of the projector $\mathcal{P}$ the spectral shift is a Hermitian super operator and the relaxator is a Hermitian and positive super operator.

The presence of the relaxator is the central new element of the dynamics for states in a macroscopic system on time scales $t_s \ll t_e$. The relaxator is given by a resolvent $G_{\mathcal{Q}}(z)$ within a continuous spectral resolution and the hopping super operators. It can also be given in terms of the spectral density for the complementary dynamics, $\gamma_{\mathcal{Q}}(\omega)$, corresponding

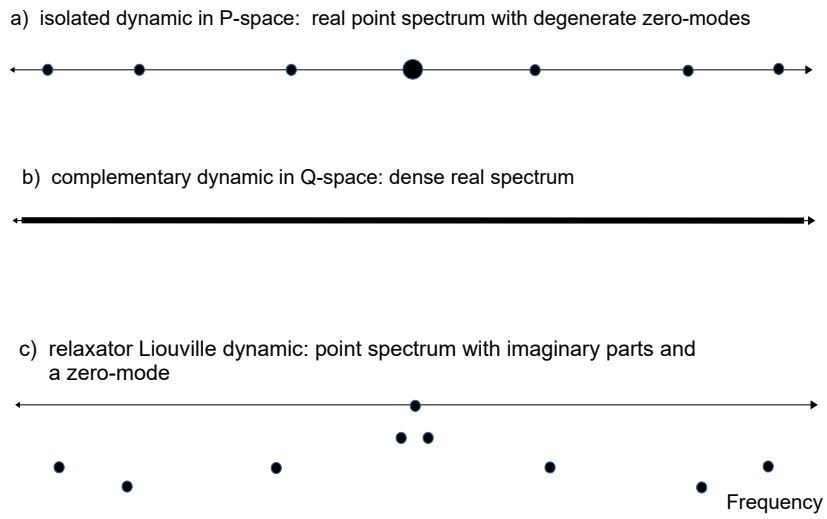

FIG. 3: Sketch of spectral properties of (a) the isolated system dynamic, (b) the complementary dynamic and (c) of the relaxator Liouville dynamic.

eigenspace projectors $\Pi^{\mathcal{Q}}(\omega)$, and the hopping super operators, $L_{\mathcal{PQ}}$, $L_{\mathcal{QP}}$ as

$$\Gamma(\omega) = \pi\gamma_{\mathcal{Q}}(\omega)L_{\mathcal{PQ}}\Pi^{\mathcal{Q}}(\omega)L_{\mathcal{QP}}\,. \tag{20}$$

So we can conclude as follows: provided the system's energy spectrum can be considered as discrete relative to the continuous spectrum of the complementary system, the $\mathcal{Q}$-propagator leads to non-positive imaginary contributions to the eigenvalues of the Liouville (see Fig. 3), indicating that the system undergoes a dynamic phase transition [35] to irreversibility, with relaxation and decoherence (see Sec. III) when coupled to an environment [36]. A similar characterization of irreversibility by complex eigenvalues of the time-evolution operator within the unit-circle in many-body systems has been put forward in [37].

It is worth mentioning that our criterion $t_s \ll t_e$ for irreversibility was used before by van Kampen [38] to derive from reversible quantum mechanics a classical master equation for the probability to find a system in a macro state [39]. Whereas van Kampen used coarse grained modeling over frequency levels of width $1/t_s \gg 1/t_e$ and brute force omission of fine structure in matrix elements of observables with respect to the total system's frequency spectrum to arrive at his goal, we used an exact projector formalism to derive the equation of motion (4) for the system density operator.

III. RELAXATOR LIOUVILLE DYNAMICS: GENERAL PROPERTIES

Equation (13) allows to estimate the relevant time scale of relaxation indicating irreversibility. The propagator of the complementary dynamics, $[z - L_{\mathcal{Q}}]^{-1} \propto t_e(\omega)$, scales with the spectral exploration time $t_e(\omega)$ of the complementary dynamics at frequency ω. The couplings $L_{\mathcal{PQ}} \propto L_{\mathcal{QP}} \propto r_{\rm se}$ scale with the rates $r_{\rm se}$ for transitions between $\mathcal{P}$- and $\mathcal{Q}$-spaces and thus yield the transition time $\tau_{\rm se}$ as the typical inverse transition rate for states to hop between system and environment. Since the relaxator $\Gamma(\omega)$ introduces relaxation we can estimate the relaxation time at frequency ω by dimensional analysis as

$$\tau_r(\omega) = a \cdot \tau_{\rm se} \frac{\tau_{\rm se}}{t_e(\omega)} \,, \tag{21}$$

with some dimensionless number a of $\mathcal{O}(1)$.

To discuss the importance of initial correlations expressed by (14) we look at its contribution to the initial expectation value of any reasonable observable A and ask when is it much smaller than the expectation value in the state ρ_0,

$$|\,{\rm Tr}\left\{AL_{\mathcal{PQ}}\left[z - L_{\mathcal{Q}}\right]^{-1}\Delta\rho_0^{\rm corr}\right\}| \ll |\,{\rm Tr}\left\{A\rho_0\right\}|\,. \tag{22}$$

We expect that the ratio of the two sides of (22) is set by the typical time scales associated with $L_{\mathcal{PQ}}$ and with $[z - L_{\mathcal{Q}}]^{-1}$, respectively. By dimensional analysis we find as a condition for neglecting initial correlations

$$\frac{t_e(\omega)}{\tau_{\rm se}} \ll 1\,. \tag{23}$$

As a consequence, the estimation (21) of the relaxation time tells that $\tau_r(\omega)$ has to be much larger than the transition time, so that we have the inequality

$$t_e(\omega) \ll \tau_{\rm se} \ll \tau_r(\omega) \tag{24}$$

as the characteristic condition for the initial correlations to be ignored. If the restrictive condition (24) is violated, we have to face a significant influence of the initial correlations during the time evolution of the system until the stationary state is reached. This implies to work with the equation of motion (4) to calculate quantities at intermediate times. A semigroup modeling ignores initial correlations and is not capable of the dynamics at intermediate times when (24) is violated. However, the stationary state ρ_∞ is not influenced by initial correlations and completely determined by the zero mode of $L(z = \imath 0^+)$ (see later in this

section). For the generic situation of a unique stationary state initial correlations are as unimportant as the initial state itself.

The Hermiticity of density operators $\rho^\dagger(t) = \rho(t)$ leads for arbitrary choice of $\rho_{\rm env}$ for the projector in (5) via the Laplace transform to the following properties of Hermiticity in the frequency domain:

$$(\rho(z))^\dagger \;=\; \rho(-z^*)\,, \tag{25}$$

$$[L(\omega)\rho(\omega)]^\dagger \;=\; -L(-\omega)\rho(-\omega)\,, \tag{26}$$

$$[\mathrm{H}(\omega)\rho(\omega)]^\dagger \;=\; -\mathrm{H}(-\omega)\rho(-\omega)\,, \tag{27}$$

$$[\Gamma(\omega)\rho(\omega)]^\dagger \;=\; \Gamma(-\omega)\rho(-\omega)\,. \tag{28}$$

For the convenient choice of a Hermitian projector $\mathcal{P}$ the spectral shift $\mathrm{H}(\omega)$ is a Hermitian super operator and the relaxator $\Gamma(\omega)$ is a positive super operator and an even function of ω. For our purposes this means for any density operator ρ

$$\mathrm{Tr}\,\rho\Gamma(\omega)\rho = \mathrm{Tr}\,\rho\Gamma(-\omega)\rho \geq 0\,. \tag{29}$$

With Hermitian projector $\mathcal{P}$ the spectral shift vanishes at $\omega = 0$,

$$\Delta\mathcal{H}(0) = 0 \tag{30}$$

due to the even character of the relaxator and the corresponding odd character of the spectral shift with respect to frequency ω.

A modeling of an open quantum system can thus be based on the choice of an isolated system Liouville $L_{\mathcal{P}}$ and a choice of a positive, even relaxator $\Gamma(\omega)$, which has to fulfill the probability conservation condition when applied to any state ρ,

$$\mathrm{Tr}\,\{L(\omega)\rho\} \;=\; 0\,, \tag{31}$$

$$\mathrm{Tr}\,\{\Gamma(\omega)\rho\} \;=\; -\imath\,\mathrm{Tr}\,\{\Delta\mathcal{H}(\omega)\rho\}\;. \tag{32}$$

Equations (31,32) express constraints for the relaxator Liouville dynamics.

The time dependent density operator is given by inverse Laplace transformation,

$$\rho(t) = \frac{\imath}{2\pi}\int_{-\infty+\imath 0^+}^{\infty+\imath 0^+} dz\, e^{-\imath z t} G(z)\rho_0(z)\,. \tag{33}$$

Although there is no quantum dynamical map from the initial ρ_0 to $\rho(t)$ within the open system in a strict sense (the part $\Delta\rho_0^{\rm corr}(z)$ depends on initial properties of the environment),

the generated dynamics (33) is as operative as the semi-group dynamics $\rho(t) = e^{-\imath Lt}\rho_0$ to which it reduces when initial correlations and memory effects can be neglected.

From the spectrum of eigenvalues of the Liouville we can get the overall time behavior (see [40]). To this end one takes the spectral representation of the relaxator Liouville for the resolvent

$$\begin{aligned}\rho(z) &= \imath\,(z - L(z))^{-1}\,\rho_0(z) \\ &= -\imath\sum_{k=0}^{N}\frac{a_k(z)}{z-\lambda_k(z)}\mathsf{R}_k(z)\,, z = \omega + i\epsilon\,, \epsilon \to 0^+\,. \end{aligned} \tag{34}$$

Here $\mathsf{R}_k(z)$ is the right eigenoperator of the relaxator Liouville corresponding to eigenvalue $\lambda_k(z)$ and

$$a_k(z) = (\mathsf{L}_k(z) \mid \rho_0(z)) \tag{35}$$

is the Hilbert-Schmidt scalar product between the corresponding left eigenoperator $\mathsf{L}_k(z)$ and the effective initial state $\rho(z)$. Left and right eigenoperators are chosen bi-orthonormal (see [40]),

$$(\mathsf{L}_k(z) \mid \mathsf{R}_{k'}(z)) = \delta_{kk'}\,. \tag{36}$$

In the relaxator Liouville the limit $\epsilon \to 0^+$ has been performed and $L(\omega) = \mathrm{H}(\omega) - \imath\Gamma(\omega)$ has complex eigenvalues $\lambda_k(\omega)$ and left eigenoperators $\mathsf{L}_k(\omega)$ and right eigenoperators $\mathsf{R}_k(\omega)$. Now for non-singular $\mathsf{R}_k(\omega), a_k(\omega)$ we define the effective eigenvalues $z_k = \omega_k - \imath\delta_k$ with $\delta_k \geq 0$, by

$$\omega_k - \imath\delta_k = \lambda_k(\omega_k)\,. \tag{37}$$

Then isolated poles $\rho(z)$ appear at those effective eigenvalues $\omega_k - \imath\delta_k$ (for non-vanishing $a_k(\omega_k)$) and the inverse Laplace transform (33) can be performed with the method of residues yielding

$$\rho(t) = \rho_\infty + \sum_{k=1}^{N}\frac{1}{2}\left(A_k e^{-\imath\omega_k t} + A_k^\dagger e^{\imath\omega_k t}\right)e^{-\delta_k t}\,. \tag{38}$$

Here the stationary state ρ_∞ has been separated and traceless right eigenoperators $\mathsf{R}_k(z_k)$ with factors $a_k(z_k)$ and factors $1/(1 - \frac{\mathrm{d}\lambda}{\mathrm{d}z}(z_k))$ have been put together to traceless operators A_k and brought in a way making Hermiticity obvious. Hermiticity of $\rho(t)$ requires that effective eigenvalues occur in pairs, $z_k, -z_k^*$ for $\omega_k \neq 0$. Taking the matrix elements in a basis where ρ_∞ is diagonal,

$$\rho_{nm}(t) = p_n\delta_{nm} + \sum_{k=1}^{N}\frac{1}{2}\left((A_k)_{nm}\,e^{-\imath\omega_k t} + (A_k)_{mn}^*\,e^{\imath\omega_k t}\right)e^{-\delta_k t} \tag{39}$$

reveals that the generic time behavior in a finite non-isolated quantum system is a superposition of damped, maybe over-damped, oscillations. This is in contrast to a finite isolated quantum system which shows an almost-periodic time dependence, a superposition of oscillations without any damping. The relaxation time τ_r is given by the largest value of $1/\delta_k$ in case of well separated effective eigenvalues, or as a collective decay time in (39) when the effective eigenvalues become dense with respect to observational resolution, as discussed in Sec. I for correlation functions. It is worth mentioning that according to (39) $\rho(t)$ stays positive semi-definite if $\rho(0)$ is such density operator.

The most basic example for the relaxator Liouville dynamics is a modeling of a 2-dimensional Hilbert state space and, thus, a 4-dimensional Liouville space. We choose four effective eigenvalues in accordance with the constraints on the spectral properties: a zero eigenvalue of L corresponding to the stationary state ρ_∞, an overdamped mode with negative imaginary eigenvalue $-i\delta_0$ and corresponding traceless right and left eigenoperators R_0, L_0, respectively. Finally we take a pair of complex eigenvalues $\pm\omega_1 - i\delta_1$ ($\delta_1 > 0$) with corresponding traceless right and left eigenoperators R_1, L_1, respectively.

$$(z - L(z))^{-1}\cdot = \frac{-i}{z}\,\mathrm{Tr}\,\{\cdot\}\,\rho_\infty + \frac{\mathsf{R}_0\,\mathrm{Tr}\left\{\mathsf{L}_0^\dagger\cdot\right\}}{z+\imath\delta_0} + \frac{\mathsf{R}_1\,\mathrm{Tr}\left\{\mathsf{L}_1^\dagger\cdot\right\}}{z-\omega_1+\imath\delta_1} + \frac{\mathsf{L}_1\,\mathrm{Tr}\left\{\mathsf{R}_1^\dagger\cdot\right\}}{z+\omega_1+\imath\delta_1}\,. \tag{40}$$

In a frequency dependent signal $\mathrm{Tr}\,\{A\rho(z)\}$ of some observable A we will find the stationary limit

$$A(t\to\infty) = \lim_{z\to 0} -\imath z\,\mathrm{Tr}\,\{A\rho(z)\} = \mathrm{Tr}\,\{\rho_\infty A\} \tag{41}$$

and resonances at oscillations with frequencies

$$\omega = \pm\omega_1\,, \tag{42}$$

line width δ_1 and coefficients $a = \mathrm{Tr}\,\{A\mathsf{R}_1\}\,\mathrm{Tr}\left\{\mathsf{L}_1^\dagger\rho_0\right\}$, a^*, respectively. The line width can be associated with a finite lifetime τ_1 of oscillations with ω_1,

$$\tau_1 = 1/\delta_1,\,. \tag{43}$$

The smaller value $\delta_{\min}$ of δ_0, δ_1 dominates the relaxation of time signals $\mathrm{Tr}\,\{A\rho(t)\}$ to the stationary value with relaxation time $\tau_r = 1/\delta_{\min}$. In the Appendix B we treat explicitly the example of a qubit weakly coupled to a bath.

Now we turn to the relaxation to a stationary state ρ_∞ which cannot capture any information about the initial state ($a_0 = 1$ by probability conservation) and its calculation

without referring to the full time dependence (see [34]). States obeying the equation of motion (4) will generically reach a unique stationary state in the long time limit. Due to the previously discussed time dependence of damped oscillations the long time limit can be taken as a long time average, defined by

$$f_\infty = \lim_{\epsilon\to 0+} \epsilon \int_0^\infty dt\, f(t) e^{-\epsilon t} \tag{44}$$

for a quite arbitrary time dependent function $f(t)$. In the same sense it holds true that

$$f_\infty = \lim_{z\to 0} -\imath z f(z)\ , \tag{45}$$

where $f(z)$ is the Laplace transform of of $f(t)$, $f(z) = \int_0^\infty \mathrm{d}t\, e^{\imath z t} f(t)$. By applying this limit to the equation of motion (4) we find the following expression for the stationary state,

$$\rho_\infty = \lim_{z\to \imath 0^+} z\left[z - L(z)\right]^{-1} \rho_0(z)\,. \tag{46}$$

Initial correlations are non-singular for $z \to \imath 0^+$ in the initial state $\rho_0(z)$. Then the long time limit is determined by the $z \to \imath 0^+$ limit of $z/(z - L(z))$ which is non-vanishing due to the existence of zero modes of $L(\imath 0^+)$. Denoting the projector on the space of these zero modes by $\Pi^0_{L(\imath 0^+)}$, the long time limit of the density operator reads

$$\rho_\infty = \Pi^0_{L(\imath 0^+)} \rho_0(\imath 0^+)\,. \tag{47}$$

The relaxator Liouville must have a zero mode due to probability conservation which means $\mathrm{Tr}\,\rho(z) = \imath/z$ and $\mathrm{Tr}\,L(z)\rho = 0$ for every z and ρ. Thus, the relaxator Liouville has the unit operator 1 as a left eigenoperator with eigenvalue 0 and, consequently, there must exist a right eigenoperator with eigenvalue 0. In generic systems the zero mode will be non-degenerate and leads to a unique normalized stationary state, ρ_∞. The projector on the zero mode can be written as

$$\Pi^0_{L(\imath 0^+)} = |\rho_\infty)\,(1|\ , \tag{48}$$

where the notation implies the Hilbert-Schmidt scalar product. Since the scalar product of the left unit matrix with the initial state amounts to taking the trace, any influence of the initial state is getting lost in a unique stationary state ρ_∞. In the non-generic case of degenerate zero modes (48) will change to a sum of projectors on different eigenstate combinations of zero modes. It is obvious that in such case the scalar product of left eigenoperator with the initial state is not simply unity and these scalar products store information about

the initial state. Stable degenerate zero modes must be protected against perturbations by design or by some stabilizing natural mechanism. Only with stable degenerate zero modes initial conditions can influence the final stationary states. Such non-generic scenarios are discussed in [41] for systems with semi-group dynamics.

As the unique zero mode of the relaxator Liouville dynamics at vanishing frequency, $L := L(\omega = 0)$, the stationary state ρ_∞ must satisfy the equation

$$L\rho_\infty = [H, \rho_\infty] - \imath\Gamma\rho_\infty = 0\,, \tag{49}$$

where $\Gamma := \Gamma(\omega = 0)$ is the relaxator at $\omega = 0$ and H the Hermitian Hamiltonian corresponding to $L_{\mathcal{P}}$. Due to (30) no spectral shift operator needs to be considered in the equation for the stationary state for $\omega = 0$.

As a condition for the stationary state to be an equilibrium state, $\rho_{\rm eq}$, we require that both contributions in (49) vanish separately, such that energy is conserved in the stationary state,

$$[H, \rho_{\rm eq}] = 0\,. \tag{50}$$

In such equilibrium state $\rho_{\rm eq}$, characterized by (50), no global currents $I = \dot{X} = i\,[H, X]$ can go through the system on average, $\mathrm{Tr}\, I\rho_{\rm eq} = 0$. The energy conserved equilibrium state becomes unique in the relaxator Liouville dynamics by fulfilling a second equation,

$$\Gamma\rho_{\rm eq} = 0\,. \tag{51}$$

Thus, the stationary state $\rho_{\rm eq}$ is diagonal in the energy representation $H\,|n\rangle = \epsilon_n\,|n\rangle$ and lies in the kernel of the relaxator Γ.

To proceed in solving these equations we use the following representation of super operators: an operator X in any orthonormal basis $\{\,|n\rangle\}_{n=1\ldots d}$ with $\langle n|m\rangle = \delta_{mn}$ can be written as $X = \sum_{nm} X_{nm}\,|n\rangle\,\langle m|$ with $X_{nm} = \langle n|\,X\,|m\rangle$. Similarly, any super operator $\mathcal{A}$ can then be written as

$$\mathcal{A}\cdot = \sum_{mn,rs} \mathcal{A}_{rs,mn}\,|r\rangle\,\langle s|\,\langle m|\,\cdot\,|n\rangle\ , \tag{52}$$

with

$$\mathcal{A}_{rs,mn} = \langle r|\,\mathcal{A}\,(\,|m\rangle\,\langle n|\,)\,|s\rangle\ . \tag{53}$$

For Γ the probability conservation requires $\mathrm{Tr}\,\Gamma\cdot = 0$ leading to

$$\sum_m \Gamma_{mm,rs} = 0 \ \text{ for all } r, s\ . \tag{54}$$

The stationary state is diagonal in the energy representation, $\rho_{\rm eq} = \sum_n p_n \left|n\right\rangle \left\langle n\right|$ with non-negative probabilities p_n, and (51) reads

$$0 = \sum_n \Gamma_{rs,nn} p_n \quad \text{for all } r, s \,. \tag{55}$$

We specify to $r = s$ and decompose to insert probability conservation (54),

$$0 = \sum_{n\neq r} \Gamma_{rr,nn} p_n + \Gamma_{rr,rr} p_r = \sum_{n\neq r} \left(\Gamma_{rr,nn} p_n - \Gamma_{nn,rr} p_r\right) \,. \tag{56}$$

and arrive at a stationary Pauli master equation [5] with $W_{rn} := -\Gamma_{rr,nn}$ for $n \neq r$ and $\sum_{n\neq r} W_{nr} = -W_{rr} = \Gamma_{rr,rr} \geq 0$,

$$\sum_{n\neq r} \left(W_{rn} p_n - W_{nr} p_r\right) = 0 \,. \tag{57}$$

W_{rn} can indeed, for $n \neq r$, be expected to be non-negative transition rates by their relation to the positive relaxator, and particularly since $\sum_{n\neq r} W_{nr} = \Gamma_{rr,rr} \geq 0$. Non-negative transition rates W_{rn} describe a gain of probability at r by transitions from n. A relabeling of states should not change this character. Since some of the off-diagonal W_{nr} have to be non-negative we expect that all of them are non-negative. It can explicitly be shown e.g. in the weak coupling approximation (see Appendix A).

Equation (57) is commonly known to describe the stationary equation of irreversible evolution into equilibrium states when W_{rn} can be seen as transition rates for $r \neq n$ and when they fulfill the detailed balance condition,

$$W_{rn} p_n - W_{nr} p_r = 0 \quad \text{for} \quad r \neq n \,. \tag{58}$$

The detailed balance condition expresses micro reversibility of the dynamics: for the joint probability to find states r and n, separated by one time step, it does not matter which one of both was earlier. This is exactly expressed by (58) when W_{rn} are transition rates for one time step from state n to state r. Since $W_{rn} = -\Gamma_{rr,nn}$ is build from the unitary complementary dynamics it respects micro reversibility. From (58) we finally arrive at the stationary distribution by recursion,

$$p_n = \frac{W_{n0}}{W_{0n}} p_0 \,. \tag{59}$$

We further distinguish two situations: (A) idealized isolated system with d states in an energy shell of fixed energy and (B) a system with an environment in equilibrium with a huge number N of constituents.

In case (A) the transition rates within the energy shell are all equal. This is the case according to Fermi's Golden Rule as well as according to the principle of ignorance when the energy window Δ is limited by $1/t_e \ll \Delta \ll 1/t_s$. Therefore, one arrives at the micro canonical uniform distribution,

$$p_n \equiv 1/d\,. \tag{60}$$

In case (B) the transition rate to the energetically higher state is by the Boltzmann factor

$$\frac{W_{nr}}{W_{rn}} = ce^{-(\epsilon_n-\epsilon_r)/T} \tag{61}$$

smaller than for the reverse process. In (61) T is the temperature of the environment and c a constant. The condition (61) is expected to be fulfilled when N becomes macroscopic. The argument is robust and independent of many details of W_{nr}. The joint probability $p_r W_{nr}$ can be seen as a joint probability of system and environment traced out over the environment, $p_r W_{nr} = \sum_{\alpha\beta} p_{r\alpha} W_{n\beta;r\alpha}$ (see also (70) below). For macroscopic environment in equilibrium not influenced by the system we can assume $p_{r\alpha} = p_r p_\alpha$ to factorize and $W_{n\beta;r\alpha}$ to be constant transition rates within an energy shell at total energy $E_{\rm tot} = \epsilon_r + E_\alpha = \epsilon_n + E_\beta$. Since p_α can be related to the entropy $S(E_\alpha)$ as $p_\alpha \propto e^{-S(E_{\rm tot}-\epsilon_r)}$, to find the dependence on ϵ_r we expand S to linear order as the dominant one for large N and find $W_{nr} \propto e^{\epsilon_r/T}$ with $T^{-1} := \partial_E S(E)$ defining the environmental temperature. The equilibrium distribution of the system then turns out to be the canonical distribution,

$$p_n = \frac{e^{-\epsilon_n/T}}{\sum_{k=1}^{d} e^{-\epsilon_k/T}}\,, \tag{62}$$

with temperature T inherited from the environment. In Appendix A we show the property (61) explicitly within in the weak coupling approximation.

We close this section on general properties of the relaxator Liouville dynamics by presenting it in terms of a Hamiltonian modeling. The total Hamiltonian $H_{\rm tot}$ consists of three parts, H representing the isolated system, $H_{\rm env}$ representing the isolated environment, and an interaction part, $H_{\rm int}$,

$$H_{\rm tot} = H + H_{\rm env} + H_{\rm int}\,. \tag{63}$$

To get more details of the representation we take the most general form of interaction,

$$H_{\rm int} = \sum_k S_k \otimes B_k\,, \tag{64}$$

where S_k are Hermitian operators on the system's Hilbert space and B_k are Hermitian operators on the environment's Hilbert space with expectation values $\langle B_k \rangle_{\rm env} := {\rm Tr}_{\rm env}\, B_k \rho_{\rm env}$ and deviations from their expectation values $\Delta B_k := B_k - \langle B_k \rangle_{\rm env}$. We are then led to the following expressions

$$L_{\mathcal{P}} \rho \otimes \rho_{\rm env} = [H_{\mathcal{P}}, \rho] \otimes \rho_{\rm env}\,, H_{\mathcal{P}} := H + \sum_k \langle B_k \rangle_{\rm env} S_k\,, \tag{65}$$

$$L_{\mathcal{Q}} \mathcal{Q} = [H_{\mathcal{Q}}, \mathcal{Q}] := [H_{\rm env} + H + H_{\rm int}, \mathcal{Q}] - \left(\underset{\rm env}{\rm Tr} [H_{\rm int}, \mathcal{Q}] \right) \otimes \rho_{\rm env}\,, \tag{66}$$

$$L_{\mathcal{Q}\mathcal{P}} \rho \otimes \rho_{\rm env} = \sum_k [S_k \otimes \Delta B_k, \rho \otimes \rho_{\rm env}]\,, \tag{67}$$

$$L_{\mathcal{P}\mathcal{Q}} \cdot = \left(\underset{\rm env}{\rm Tr} \sum_k [S_k \otimes \Delta B_k, \cdot] \right) \otimes \rho_{\rm env}\,. \tag{68}$$

The relaxator Liouville $L(\omega)$ reads

$$L(\omega)\rho = [H_{\mathcal{P}}, \rho] + \\ + \underset{\rm env}{\rm Tr} \left[\sum_k S_k \otimes \Delta B_k, G_{\mathcal{Q}}(z) \sum_{k'} [S_{k'} \otimes \Delta B_{k'}, \rho \otimes \rho_{\rm env}] \right]\,, \tag{69}$$

and the relaxator is

$$\Gamma(\omega)\rho = \underset{\rm env}{\rm Tr} \left[\sum_k S_k \otimes \Delta B_k, \pi \delta(z - L_{\mathcal{Q}}) \sum_{k'} [S_{k'} \otimes \Delta B_{k'}, \rho \otimes \rho_{\rm env}] \right]\,. \tag{70}$$

In these expressions ϵ is send to $0+$ after taking the trace over the environment with a continuous spectrum.

The Hamiltonian of the complementary dynamics, $H_{\mathcal{Q}}$, has an eigenvalue representation, $H_{\mathcal{Q}} \mid m\alpha\rangle = E_{m\alpha} \mid m\alpha\rangle$ where the factors $\mid m\rangle$ belong to the system's Hilbert space and the factors $\mid \alpha\rangle$ to the environment's Hilbert space. The resolvent can be written in this eigenvalue representation and the relaxator Liouville of (69) reads in the $H_{\mathcal{Q}}$ eigenvalue representation

$$L(\omega)\rho = [H_{\mathcal{P}}, \rho] + \mathcal{D}(\omega)\rho\,, \tag{71}$$

$$\mathcal{D}(\omega)\rho = \sum_{k,k',m,n,\alpha,\beta} \left\{ \frac{(\Delta B_k)_{\beta\alpha} \left(\Delta B_{k'} \rho_{\rm env}\right)_{\alpha\beta}}{\omega + \imath\epsilon - (E_{m\alpha} - E_{n\beta})} [S_k, P_m S_{k'} \rho P_n] + \right. \\ \left. + \frac{(\Delta B_k)_{\beta\alpha} \left(\rho_{\rm env} \Delta B_{k'}\right)_{\alpha\beta}}{\omega + \imath\epsilon - (E_{m\alpha} - E_{n\beta})} [P_m \rho S_{k'} P_n, S_k] \right\}\,, \tag{72}$$

where $P_m := \mid m\rangle \langle m \mid$ and $\epsilon \to 0^+$ after summing over α, β with a continuous spectrum of states.

With a representation of $\Delta\rho_0$ by a superposition of system times environment operators,

$$\Delta\rho_0 = \sum_l \Delta\rho_{\rm s0}^{(l)} \otimes \Delta\rho_{\rm e0}^{(l)} \,, \tag{73}$$

we can calculate the correlation contribution to the initial state as

$$\Delta\rho_0(\omega) = \sum_{k,l,\alpha,m,\beta,n} \frac{(\Delta B_k)_{\beta\alpha}(\Delta\rho_{\rm e0}^{(l)})_{\alpha\beta}\left[S_k, P_m\Delta\rho_{\rm s0}^{(l)}P_n\right]}{\omega + \imath\epsilon - (E_{m\alpha} - E_{n\beta})} \,, \tag{74}$$

where $\epsilon \to 0^+$ after summing over α, β with a continuous spectrum of states.

Equation (71) with (72) is a general representation of the relaxator Liouville in terms of a decomposed Hamiltonian for the total system, ready for further investigations. For a frequency resolution of order $1/t_s \gg 1/t_e$ we may factorize the contributions in (72) to $\mathcal{D}(\omega)$ coming from the environment and the system, respectively. Such factorization of the form

$$\Gamma(\omega)\rho = \sum_{\tilde\omega}\sum_{kk'} \gamma_{kk'}(\omega+\tilde\omega)\,[S_k, (S_{k'}\rho)(\tilde\omega)] + h.c. \tag{75}$$

with an environmental correlation function $\gamma_{kk'}(\Omega)$ becomes possible if we can rewrite the total energies $E_{\alpha m}$ as additive with respect to system and environment, $E_{m\alpha} \approx E_\alpha + \epsilon_m$ and treat interaction energies between system and environment as small compared to the resolution scale $1/t_s$. This will be undertaken in Sec VI.

IV. KINETIC EQUATIONS FROM RELAXATOR LIOUVILLE DYNAMICS

So far, the relaxator Liouville equation (4) was derived for an arbitrary open system. To extract the full time dependence of a many particle state, $\rho = \rho_{1\ldots N}$, is not only impossible in practice, but also often not of interest; it contains to much information to be manageable. Therefore, one prefers to have an equation for the time evolution of the reduced density operator for just one particle or a few, say s, particles out of the N. In particular, the one-particle density operator suffices to calculate expectation values of additive quantities like kinetic energy and current.

Dynamic equations for one-particle distributions or density operators are called kinetic equations. Historically the Boltzmann equation [3] was the first of these and with it began the study of irreversible processes, including the relaxation to equilibrium. The Boltzmann equation and more general quantum kinetic equations can be derived from the BBGKY

hierarchy (derived by Bogoliubov, Born, Green, Kirkwood, Yvon and others in the 1930's to 1940's) for s-particle reduced density operators (for an overview see [24]) after appropriate decoupling approximations. We take the following definition for the s-particle reduced density operator $\rho_{1\ldots s}$

$$\rho_{1\ldots s} := \operatorname*{Tr}_{s+1\ldots N} \rho_{1\ldots N} \; ; \; \operatorname*{Tr}_{1\ldots s} \rho_{1\ldots s} = 1 \, . \tag{76}$$

Other definitions in the literature differ by normalization conventions.

An alternative way to the BBGKY route to arrive at kinetic equations uses so called nonequilibrium Green functions (NEGF) (invented by Schwinger, Feynman and Vernon, Kadanoff and Baym, Keldysh [42] and others in the 1960's). These are nonequilibrium expectation values for one quantum object (particle) to propagate from one event 1 to another event 2. In the NEGF approach, the idea of a self-energy Σ is exploited. It takes the many particle influences on the one-particle propagator into account on its way from 1 to 2. For an introduction to NEGF see [25], Chap. 13 in [24] and [26]. Usually, in the NEGF approach, the self-energy is constructed by approximate methods to incorporate as much of the interaction between many particles as possible or needed. The self-energy can then show finite broadened levels shifted from stationary one-particle levels.

The route to kinetic equations we take with relaxator Liouville dynamics is close in spirit to the NEGF method, but the construction of the self-energy is formally exact owing to a projector method. Our procedure has the advantage to immediately leading us to a closed equation for the one-particle density operator via a one-particle propagator. It makes the origin of irreversible behavior apparent by the relaxator appearing in this equation. The corresponding spectral shift and relaxator in our approach correspond to the self-energy in the NEGF approach.

We take a projector onto one-partice states in the following form

$$\mathcal{P}^{[1]} \rho_{1\ldots N} = \rho_1 \otimes \rho^0_{2\ldots N} \, . \tag{77}$$

where ρ_1 is constructed from tracing out all particles but one, as defined in Eq. (76). The $N-1$-particle operator $\rho^0_{2\ldots N}$, with $\mathrm{Tr}_{2\ldots N} \, \rho^0_{2\ldots N} = 1$, is taken as a fixed factor, independent of the chosen $\rho_{1\ldots N}$. By omission of this fixed factor in the end, a reduced equation of motion on a one-particle Liouville space will result.

It is easy to show that an application of $\mathcal{P}^{[1]}$ to $\mathcal{P}^{[1]} \rho_{1\ldots N}$ does not change the projection.

This establishes $\mathcal{P}^{[1]} = {\mathcal{P}^{[1]}}^2$ as a projector. The complement projector is defined by

$$\mathcal{Q}^{[/1]} = 1 - \mathcal{P}^{[1]} \tag{78}$$

and fulfills the desired projector and complement rules,

$${\mathcal{Q}^{[/1]}}^2 = 1 - \mathcal{P}^{[1]} = \mathcal{Q}^{[/1]} \; ; \; \mathcal{P}^{[1]}\mathcal{Q}^{[/1]} = \mathcal{Q}^{[/1]}\mathcal{P}^{[1]} = 0 \, . \tag{79}$$

By the same algebraic reasoning as used in [34] one can then show that for a given N-particle Liouville $L_{_{1\ldots N}}$ we have

$$\mathcal{P}^{[1]}(z - L_{_{1\ldots N}})^{-1}\mathcal{P}^{[1]} = (z - L_1(z))^{-1} \tag{80}$$

which acts, like $L_1(z)$, only on the one-particle Liouville space ($\mathcal{P}^{[1]}$-space). The one-particle relaxator Liouville dynamics is then defined with the help of $L_1(z)$,

$$\rho_1(z) = (z - L_1(z))^{-1}\rho_{1;0}(z) \, . \tag{81}$$

The expressions for $L_1(z)$ and the initial state $\rho_{1;0}(z)$, containing initial many-particle correlations, take the form

$$L_1(z) \; = \; L_1 + L_{\mathcal{P}^{[1]}\mathcal{Q}^{[/1]}} \left[z - L_{\mathcal{Q}^{[/1]}}\right]^{-1} L_{\mathcal{Q}^{[/1]}\mathcal{P}^{[1]}} \, , \tag{82}$$

$$\rho_{1;0}(z) \; = \; \rho_1(t=0) + L_{\mathcal{P}^{[1]}\mathcal{Q}^{[/1]}} \left[z - L_{\mathcal{Q}^{[/1]}}\right]^{-1} \rho_{_{1\ldots N}}(t=0) \, , \tag{83}$$

with $L_1 := \mathcal{P}^{[1]}L_{_{1\ldots N}}\mathcal{P}^{[1]}$, $L_{\mathcal{P}^{[1]}\mathcal{Q}^{[/1]}} := \mathcal{P}^{[1]}L_{_{1\ldots N}}\mathcal{Q}^{[/1]}$, $L_{\mathcal{Q}^{[/1]}\mathcal{P}^{[1]}} := \mathcal{Q}^{[/1]}L_{_{1\ldots N}}\mathcal{P}^{[1]}$, and $L_{\mathcal{Q}^{[/1]}} := \mathcal{Q}^{[/1]}L_{_{1\ldots N}}$. The time dependent one-particle density operator is given by the inverse Laplace transformation,

$$\rho_1(t) = \frac{\imath}{2\pi} \int_{-\infty+\imath 0^+}^{\infty+\imath 0^+} dz \, e^{-\imath z t} \left[z - L_1(z)\right]^{-1} \rho_{1;0}(z) \, . \tag{84}$$

We can identify the source for relaxation of the one-particle density operator as the interaction with the other $N-1$ particles: as soon as the spectrum of the complementary dynamics (described by $L_{\mathcal{Q}^{[/1]}}$) becomes dense relative to the spectrum of the isolated one-particle dynamics (described by L_1), it is a source of irreversibility and a relaxator can be defined for the kinetic equation (by the same reasoning that led to Eq. (16)) by

$$\Gamma_1(\omega) = L_{\mathcal{P}^{[1]}\mathcal{Q}^{[/1]}} \delta\left(\omega - L_{\mathcal{Q}^{[/1]}}\right) L_{\mathcal{Q}^{[/1]}\mathcal{P}^{[1]}} \, . \tag{85}$$

To set up the kinetic equation for one particle, one has to find the propagator $(z - L_{\mathcal{Q}^{[/1]}})^{-1}$. Although $L_{\mathcal{Q}^{[/1]}}$ defines an interacting many-body problem as complicated as the

original $L_{1\ldots N}$ problem, for the kinetic equation we don't need the spectral details of $L_{\mathcal{Q}^{[/1]}}$ for investigation times up to t_s. We can therefore introduce approximations, e.g. in a first step treating $L_{2\ldots N}$ as an effective additive single-particle Liouville, and still reach the goal for a kinetic equation for particle 1, capturing irreversible relaxation to a stationary state of (82).

V. LINEAR RESPONSE FOR RELAXATOR LIOUVILLE DYNAMICS

Linear response theory addresses deviations in expectation values of some observable B to linear order in a given external, in general time dependent, driving field $F(t)$, coupled to the system by some observable A. It is assumed that the field causes a linear shift of the system's energy,

$$H \to H(t) = H - A \cdot F(t)\,. \tag{86}$$

For a time translational invariant and causal linear response the resulting expectation value of B can be written as

$$\langle B\rangle_t = \langle B\rangle_{\rm st} + \int_{-\infty}^{t} dt'\,\chi_{BA}(t-t')F(t')\,, \tag{87}$$

where $\langle B\rangle_{\rm st}$ is the stationary expectation value of B in the absence of the field and $\chi_{BA}(t'-t)$ is the phenomenological definition of the dynamical susceptibility, the quantity of interest in linear response theory. Causality requires that $\chi_{BA}(t-t') = 0$ for $t < t'$.

The linear response deviation $\Delta B(t) := (\langle B\rangle_t - \langle B\rangle_{\rm st})$ and the field $F(t)$ can be analyzed with respect to their frequency content. By the convolution property of the Laplace transform and the condition of causality one concludes the linear response relation in frequencies $z = \omega + \imath\epsilon\,, \epsilon \to 0^+$,

$$\Delta B(z) = \chi_{BA}(z)F(z)\,, \tag{88}$$

where the functions with argument z stand for the Laplace transform of the functions with time argument, e.g. $\chi_{BA}(z) = \int_0^\infty d(t-t')\chi_{BA}(t-t')e^{\imath z(t-t')}$ with $z = \omega + \imath\epsilon$ for $\epsilon \to 0^+$. The introduction of $\epsilon > 0$ and the use of the Laplace transform is mathematically adopted to the causal structure of the response: fields $F(t')$ influence the signal at time t only when $t' \le t$.

Before turning to set up a linear response theory for open systems we recall the dynamical susceptibility formula found by Kubo [8] to make a comparison easier. The Kubo formula is

derived for thermally isolated Hamiltonian systems subject to weak external driving fields. Kubo showed in his famous paper that dynamic susceptibilities can be expressed in terms of correlation functions in the stationary state in the absence of the driving field. The Kubo formula can be viewed as a generalization of the previously known fluctuation-dissipation theorem (see his review article [43]). It reads

$$\chi_{BA}(z) = -\imath \int_0^\infty \mathrm{d}s\, e^{\imath z s} \,\mathrm{Tr}\left\{\rho_{\mathrm{eq}}\left[A, B(s)\right]\right\}\,, z = \omega + \imath\epsilon\,, \epsilon \to 0^+\,, \tag{89}$$

for the dynamic susceptibility of an isolated system, where $B(s) = e^{\imath H s} B e^{-\imath H s} = e^{\imath L s} B$. With

$$\mathrm{Tr}\left\{\rho_{\mathrm{eq}}\left[A, B(s)\right]\right\} = -\,\mathrm{Tr}\left\{\left(\left[e^{-\imath L s}\delta L\right]\rho_{\mathrm{eq}}\right) B\right\} \tag{90}$$

and carrying out the time integral sets (89) in the compact resolvent form,

$$\chi_{BA}(z) = -\,\mathrm{Tr}\left\{\left(\left[G(z)\delta L\right]\rho_{\mathrm{eq}}\right) B\right\}\,, \epsilon \to 0^+\,. \tag{91}$$

Here $G(z) = [z - L]^{-1}$ and $\delta L \cdot := [A, \cdot]$. With Equation (91) it is easy to see that $\chi_{BA}(z)$ is analytic in the upper z-plane and has singularities at the spectrum of L. The limit $\epsilon \to 0^+$ thus requires a careful analysis of the spectrum of L. Typically, a limit to an infinite number of variables or parameters has to be performed first to reach a reasonable limit $\chi_{BA}(\omega + \imath 0^+)$.

The Kubo formula has proved to be very successful. Irreversibility indeed shows up in dynamic susceptibilities like dissipative conductances. However, when performing measurements of dynamic susceptibilities, the initial state is not necessarily an equilibrium state, and the number of variables is huge, but finite. In the following we will alternatively start from an open quantum system, where we have gained insight how irreversibility and stationarity emerges. It is a systematic approach to linear response in open systems based on the general relaxator Liouville dynamics and more general than other approaches [44]-[46] to open systems that rely on semi-group or time local quantum master equations. In [47] an alternative general approach to linear response in open systems is presented. It is based on the Feynman-Vernon influence functional [48] and hierarchical equations of motion for propagator and state, which have to be solved. We prefer the relaxator Liouville approach for which we know already general properties of the propagator and state as outlined in Sec. III.

We consider an open system consisting of system and its environment as in Sec. II. The

total Liouville is time dependent by coupling to an external driving field,

$$L_{\rm tot}(t) = L_{\rm tot} - \delta L F(t)\,. \tag{92}$$

Note, δL leaves the environment untouched. The Laplace transform of the von Neumann total equation of motion $\dot\rho_{\rm tot}(t) = -\imath L_{\rm tot}(t)\rho_{\rm tot}(t)$ reads

$$-\rho_{\rm tot}(t=0) - \imath z\rho_{\rm tot}(z) = -\imath L_{\rm tot}\rho_{\rm tot}(z) + \imath\sum_{\tilde\omega}\delta L\rho_{\rm tot}(z-\tilde\omega)F_{\tilde\omega}\,, \tag{93}$$

where the field $F(t)$ is given by its spectral representation

$$F(t) = \sum_{\tilde\omega} F_{\tilde\omega}e^{-\imath\tilde\omega t}\,. \tag{94}$$

Written in the form

$$\rho_{\rm tot}(z) = \imath(z-L_{\rm tot})^{-1}\rho_{\rm tot}(t=0) - \sum_{\tilde\omega}(z-L_{\rm tot})^{-1}\delta L\rho_{\rm tot}(z-\tilde\omega)F_{\tilde\omega}\,. \tag{95}$$

(93) is ready for iterating in powers of $\delta L F_{\tilde\omega}$. But before doing this we project to the system and use the separation $t_s \ll t_e$ and arrive, along the lines outlined in Sec. II, at an equation for the system state $\rho(z)$ incorporating memory and initial correlations with non-Hermitian Liouville $L(z)$ as in (16),

$$\rho(z) = \imath(z-L(z))^{-1}\rho_0(z) - \sum_{\tilde\omega}(z-L(z))^{-1}\delta L\rho(z-\tilde\omega)F_{\tilde\omega}\,. \tag{96}$$

Now, we expand this equation in powers of $\delta L F_{\tilde\omega}$ and estimate the regime of reliable linear response by requiring that the linear contribution to $\rho(z)$ is smaller than the contribution without the field. Since $(z-L(z))^{-1}$ scales with the system's spectral exploration time t_s and the coupling Liouville scales with a characteristic rate as the inverse of a field induced transition time $t_{\delta{\rm F}_\omega}$, the condition for the linear response theory to apply reads

$$t_s \ll t_{\delta{\rm F}_\omega}\,. \tag{97}$$

In other words, rates of change by the driving field (e.g. velocities v) have to be much smaller than the rate of change by system exploring processes,

$$v_{\delta{\rm F}_\omega} \ll v_{\rm s}\,. \tag{98}$$

Under these conditions we find to linear order

$$\rho(z) = \imath(z-L(z))^{-1}\rho_0(z) - \imath\sum_{\tilde\omega}(z-L(z))^{-1}\delta L(z-\tilde\omega-L(z-\tilde\omega))^{-1}\rho_0(z-\tilde\omega)F_{\tilde\omega}\,. \tag{99}$$

Now we introduce $\xi = z - \tilde{\omega}$ as new variable for later use and write (96) as

$$\rho(\xi+\tilde{\omega}) = \imath(\xi+\tilde{\omega}-L(\xi+\tilde{\omega}))^{-1}\rho_0(\xi+\tilde{\omega}) - \imath\sum_{\tilde{\omega}}(\xi+\tilde{\omega}-L(\xi+\tilde{\omega}))^{-1}\delta L(\xi-L(\xi))^{-1}\rho_0(\xi)F_{\tilde{\omega}}\,. \tag{100}$$

We can now use the Laplace transform of the function $f(t) = a\cdot e^{-i\tilde{\omega}t}$ is $f(z) = \imath a/(z-\tilde{\omega}) = \imath a/\xi$ to read off the coefficient a of the oscillation with $\tilde{\omega}$ from $\lim_{\xi\to 0} -\imath\xi f(\xi+\tilde{\omega})$,

$$\lim_{\xi\to 0} -\imath\xi\rho(\xi+\tilde{\omega}) = 0 - \sum_{\tilde{\omega}}(\tilde{\omega}-L(\tilde{\omega}))^{-1}\delta L \lim_{\xi\to 0}\xi(\xi-L(\xi))^{-1}\rho_0(\xi)F_{\tilde{\omega}}\,. \tag{101}$$

As shown in Sec. III, $\lim_{\xi\to 0}\xi(\xi-L(\xi))^{-1}\rho_0(\xi) = \rho_\infty$ and we arrive at the dynamic susceptibility formula for open systems,

$$\chi_{BA}(z) = -\operatorname{Tr}\left\{\left([G(z)\delta L]\,\rho_\infty\right)B\right\}, z = \omega+\imath\epsilon\,, \epsilon\to 0^+\,. \tag{102}$$

with $G(z) = [z-L(z)]^{-1}$. By comparing (102) and (91) we come to a number of conclusions.

1. The dynamic susceptibility in an open system is formally the same correlation expression as the one given by the Kubo formula. However, the state $\rho_{\rm eq}$ in the Kubo formula is changed to the stationary state ρ_∞ in the open system, and the resolvent of an isolated system's Liouville L is changed to an open system's relaxator Liouville $L(z)$. The structure of the susceptibility expression in [47] is also similar to (102), but the resolvent and state in that expression have to be found by solving a set of hierarchical equations of motion. However, we can draw conclusions from the general properties of the relaxator Liouville dynamics.

2. The equilibrium state $\rho_{\rm eq}$ in the Kubo formula is the initial state which is unchanged by the isolated dynamics in the absence of the driving field. In the open system treatment the initial state is not necessarily an equilibrium state, but can be arbitrary, as the dynamic evolution approaches a unique (except for non-generic degeneracies) stationary state ρ_∞ in the absence of the driving field. For heat bath conditions the stationary state is the canonical equilibrium state $\rho_{\rm eq}$ in accordance with the Kubo formula.

3. For finite isolated systems the Kubo susceptibility shows singular resonant behavior when the frequency ω approaches eigenfrequencies in the limit $\epsilon\to 0^+$. In particular,

the dissipative imaginary part vanishes for almost all frequencies but a number of zero measure. In the case of an open system the limit $\epsilon \to 0^+$ can be performed and the susceptibility stays regular as a function of frequency ω, because the system contains sufficient damping of isolated resonances due to environmental contact.

4. The resolvent $G(z)$ is analytic in the upper $z = \omega + i\epsilon$ plane in both cases. The Kubo formula is a limiting case of the open system formula when $\mathrm{H}(\omega)\cdot$ reduces to $[H, \cdot]$ and $\Gamma(\omega)$ reduces to constant ϵ and $\rho_\infty = \rho_{\mathrm{eq}}$. In the open system case the Hermitian part is odd with respect to ω and the anti-Hermitian part is even with respect to ω, and the operators $\Gamma(\omega)$ and $\mathrm{H}(\omega)$ are related to each other by a Kramers-Kronig relation (19).

5. In both cases, the real part $\chi'_{BA}(\omega)$ and the imaginary part $\chi''_{BA}(\omega)$ of the dynamic susceptibility fulfill a Kramers-Kronig relation (as a consequence of causality)

$$\chi'_{BA}(\omega) = +\mathrm{P}\int_{-\infty}^{\infty} \frac{\mathrm{d}\nu}{\pi} \frac{\chi''_{BA}(\omega)}{\nu - \omega} \,, \tag{103}$$

$$\chi''_{BA}(\omega) = -\mathrm{P}\int_{-\infty}^{\infty} \frac{\mathrm{d}\nu}{\pi} \frac{\chi'_{BA}(\omega)}{\nu - \omega} \,. \tag{104}$$

6. The Kubo dynamic susceptibility is an appropriate approximation for the open system dynamic susceptibility, if the stationary state ρ_∞ is the equilibrium state ρ_{eq} and if the average level spacing $\Delta\omega$ of real parts of effective eigenvalues of $L(z)$ as well as the exploring frequency ω is much larger than the inverse relaxation time $1/\tau_r$ corresponding to $L(z)$. In such regime, now denoted as Kubo regime, the discrete nature of the spectrum of $L(z)$ cannot be resolved by the linear response susceptibility. In the Kubo regime the resolvent in the system's energy representation (frequency eigenvalues ω_{mn}) can be replaced by

$$G(\omega)_{mm,nn} = (\omega - \omega_{mn} + \imath/\tau_r)^{-1} \,, \tag{105}$$

with $\tau_r \to \infty$ and dense spectrum of frequency levels ω_{mn}. The Kubo regime is adopted to a transport situation, where a macroscopic system is subject to driving fields (or attached macroscopic contacts with different potentials) and responding currents occur. The available time to explore the system is $\propto 1/\omega$. In the Kubo regime it is much shorter than the time needed to explore the spectral properties of the system,

t_s. Starting from the Kubo regime, corrections to the Kubo susceptibility will appear with $\mathcal{O}\left(\Delta\omega/\omega, (\tau_r \cdot \Delta\omega)^{-1}\right)$.

7. A regime, opposite to the Kubo regime, is reached when the frequency ω can resolve individual level spacings in a finite system, $\omega \ll \Delta\omega_{\rm s}$, now denoted as regime of isolated levels. Then the open system's dynamic susceptibility probes individual resonances of the system's propagator at levels ω_k and their life times $1/\delta_k$. When ω is close to one of well separated resonances, $\omega = \omega_k(\omega)$, and δ_k is also smaller than the separation distance

$$\chi_{AB}(\omega) \approx f_k^{AB}(\omega) \left(\omega - \omega_k(\omega) + \imath\delta_k(\omega)\right)^{-1} \tag{106}$$

measures an individual resonance at effective level ω_k with broadening δ_k (corresponding to finite life time $1/\delta_k$) and strength factor

$$f_k^{AB}(\omega) = -\sum_{n,m} \left(\rho_{\infty n} - \rho_{\infty m}\right) (AB)_{nmk}(\omega) \,. \tag{107}$$

Here $(AB)_{nmk}(\omega)$ is a short hand notation,

$$(AB)_{nmk}(\omega) := \mathrm{Tr}\left\{\mathsf{L}_k^\dagger(\omega) A\right\} \mathsf{R}_k(\omega)_{nm} B_{mn} \,. \tag{108}$$

In the regime of isolated levels the susceptibilities probe the environmental influence on the frequency spectrum and can help to extract level shifts and finite lifetimes.

8. $L(\omega)$ defines the time evolution via $G(z)$ in the susceptibility. $L_H\cdot = [H, \cdot]$ defines via $\dot{A} = \imath L_H A = \imath[H, A]$ the time derivative of operators within the system. H also defines the canonical equilibrium state. Usually $L(\omega)$ and L_H do not commute and with notable relaxation due to $L(\omega)$ one has to face corrections to the Onsager relations [52]. When deviating from the Kubo regime corrections to the Onsager relations start with corrections of order $\mathcal{O}\left(\Delta\omega/\omega, (\Delta\omega \cdot \tau_r)^{-1}\right)$.

Dissipation of energy is known to be expressed by the imaginary part $\chi''_{BA}(\omega)$. To discriminate the origin of dissipation in the isolated level regime and the Kubo regime, respectively, we use the matrix representation in the diagonal basis of ρ_∞,

$$\chi''_{BA}(\omega) = \sum_{k;m} \frac{\mathrm{Re}\,\left\{\langle AB\rangle_{k;m}(\omega)\right\} \cdot \delta_k(\omega) - \mathrm{Im}\,\left\{\langle AB\rangle_{k;m}(\omega)\right\} \cdot \left(\omega - \omega_k(\omega)\right)}{\left(\omega - \omega_k(\omega)\right)^2 + \left(\delta_k(\omega)\right)^2} \,, \tag{109}$$

where $\langle AB\rangle_{k;m}(\omega)$ is short hand for

$$\langle AB\rangle_{k;m}(\omega) := \rho_{\infty,m}\cdot \mathrm{Tr}\left\{L_k^\dagger(\omega)A\right\}\cdot\left([\mathsf{R}_k(\omega),B]\right)_{mm}\,. \tag{110}$$

In the regime of isolated levels in a finite system we put ω closely to a resonance, $\omega = \omega_k(\omega)$. Then the dissipation is proportional to the lifetime $\tau_k(\omega) = 1/\delta_k(\omega)$ of the level,

$$\chi''_{BA}(\omega) \approx \left(\sum_m \mathrm{Re}\ \left\{\langle AB\rangle_{k;m}(\omega)\right\}\right)\cdot\tau_k(\omega)\,. \tag{111}$$

Thus, relaxing irreversible behavior characterized by finite lifetimes of eigenfrequencies goes along with dissipative response.

In the Kubo regime the right and left eigenoperators R_k and L_k are given by dyads of energy eigenstates, $R_{uv} =\mid u\rangle\langle v\mid$ and $L^\dagger_{uv} =\mid v\rangle\langle u\mid$, with $\omega_{uv} = \epsilon_u - \epsilon_v$. Thus, in the Kubo regime we have

$$\begin{aligned}\chi''_{BA}(\omega) \;=\; & \sum_{uv}\left(\rho_{\mathrm{eq},u}-\rho_{\mathrm{eq},v}\right)\left[\pi\delta\left(\omega-(\epsilon_u-\epsilon_v)\right)\cdot\mathrm{Re}\ \left\{A_{uv}B_{vu}\right\}\right.\\ -\;& \left.\mathrm{P}\left(\frac{1}{\omega-(\epsilon_u-\epsilon_v)}\right)\cdot\mathrm{Im}\ \left\{A_{uv}B_{vu}\right\}\right]\,.\end{aligned} \tag{112}$$

In particular, for $A = B$, corresponding to a one component field induced dissipation, (112) simplifies, by using $\rho_{\mathrm{eq},u} = e^{-\epsilon_u/T}/Z$, to

$$\chi''_{AA}(\omega) = \pi\left(e^{-\omega/T}-1\right)\sum_{uv}\rho_{\mathrm{eq},v}\delta\left(\omega-(\epsilon_u-\epsilon_v)\right)\cdot|A_{uv}|^2\,. \tag{113}$$

Irreversible behavior is thus captured by the Kubo susceptibility, if the spectrum of the Liouville has to be treated as continuous due to the impossibility to resolve levels with even a small frequency $\omega \gg \Delta\omega_s$. As mentioned before, this is typical for a transport situation where currents enter and leave the system with scattering boundary conditions or with large reservoirs which cannot - in affordable time - react back to the system's dynamic. In such situation of irreversible behavior in the Kubo regime, time dependent correlation functions typically decay on a characteristic time scale (as discussed in Sec. I) which we denote as τ_{BA} for the time dependent correlation function $\chi_{BA}(t)$, the Laplace transform of which defines the susceptibility $\chi_{BA}(\omega)$. A convenient parametrization for the time dependent correlation function $\chi_{BA}(t)$ is given by

$$\chi_{BA}(t) = \imath\,[BA]_0\,e^{-t/\tau_{BA}-\imath t\omega_{BA}}\,, \tag{114}$$

where $[BA]_0$ is an initial complex correlation strength and ω_{BA} captures a possible oscillatory behavior of correlations with a characteristic frequency ω_{BA}. The time τ_{BA} is not to be mixed up with the relaxation time τ_r to reach stationarity in the absence of the driving field. The time τ_{BA} is due to internal interactions (scattering) yielding relaxation of correlations within affordable times $t \sim 1/\omega$. The relaxation time τ_r induced by a coupling to an environment is, in the Kubo regime, a much larger time scale and and could be sent to infinity in the end of calculations. By (114)

$$\chi_{BA}(\omega) = -\left[BA\right]_0 \left(\omega - \omega_{BA} + \imath/\tau_{BA}\right)^{-1} . \tag{115}$$

Generalizing to frequency dependent parameters $\tau_{BA}(\omega)$ and $\omega_{BA}(\omega)$ allows for a wide range of applications of (115) (e.g. a discussion of conductivity in weak and strong magnetic fields [49], [50]). The dissipative part of the dynamic susceptibility reads

$$\chi''_{BA}(\omega) = \frac{\{\mathrm{Re}\ \left[BA\right]_0 - \mathrm{Im}\ \left[BA\right]_0 (\omega - \omega_{BA})\tau_{BA}\} \cdot \tau_{BA}}{1 + \left((\omega - \omega_{BA})\tau_{BA}\right)^2} . \tag{116}$$

In the frequency regime of dominating decay of correlations, $\mid \omega - \omega_{BA} \mid \tau_{BA} \ll 1$, the dissipation is proportional to the decay time of correlations

$$\chi''_{BA}(\omega) \approx \mathrm{Re}\ \left[BA\right]_0 \cdot \tau_{BA} . \tag{117}$$

As mentioned in Sec. I phases of matter may exist where a dense spectrum of frequencies does not go along with decay of correlation functions and dissipation. In such systems of localization with respect to a certain basis (for a review see [20]) the local spacing between frequencies is huge, states are long-lived and the system cannot dissipate the energy of an external field within affordable time.

To summarize our findings within the linear response theory in open systems: there are two distinguishable routes to irreversibility and dissipation: (a) By coupling the system to an environment (e.g. to an external bath or to internal irrelevant variables) discrete frequency levels acquire finite lifetimes (and frequency shifts) which can be resolved within affordable times by linear response measurements. The dissipation is proportional to the lifetime of individual levels at resonating conditions (see (111)). The occurrence of lifetimes for discrete levels is caused by a collective action of densely distributed frequency levels of the complementary dynamics. (b) Inverse lifetimes and spacings of individual levels are to small to be resolved within affordable times by linear response measurements. Irreversibility

and dissipation are related to the collective action of densely distributed frequency levels within the system, leading to decay of correlations within affordable times of linear response measurements. In the frequency regime of dominating decay of correlations, captured already by the Kubo formula, the dissipation is proportional to the decay time of correlations (see (117)). For both routes the reason for irreversibility is the separation of time scales. The affordable time to measure a response of the open system to a perturbation is very much smaller than the time to explore the frequency content of either the complementary dynamic or of the system's dynamic.

VI. WEAK COUPLING APPROXIMATION

As indicated at the end of Sec. III we now consider an open system weakly coupled to its environment. The weakness condition is related to the fact that we cannot resolve the full spectrum of the environment as well as of the total system. We will neglect interaction energies that are small compared with the affordable resolution scale of $1/t_s$. To this end, we decompose the eigenfrequencies of $L_{\mathcal{Q}}$ as

$$\begin{aligned} E_{m\alpha} - E_{n\beta} &= \langle m\alpha \mid [H_{\mathcal{Q}}, \mid m\alpha\rangle \langle n\beta \mid] \mid n\beta\rangle = \\ &= (E_\alpha - E_\beta) + (\epsilon_m - \epsilon_n) + \delta\omega_{m\alpha n\beta}\,, \end{aligned} \tag{118}$$

where $E_\alpha := \langle \alpha \mid H_{\rm env} \mid \alpha\rangle$, $\epsilon_n := \langle n \mid H \mid n\rangle$ and

$$\delta\omega_{m\alpha n\beta} = \sum_k (S_k)_{mm} \left\{(B_k)_{\alpha\alpha} - (B_k)_{\beta\alpha}(\rho_{\rm env})_{\alpha\beta}\right\} - (S_k)_{nn} \left\{(B_k)_{\beta\beta} - (B_k)_{\beta\alpha}(\rho_{\rm env})_{\alpha\beta}\right\}\,. \tag{119}$$

The resolvent of complementary dynamics in the eigenvalue representation is peaked at $\omega = E_{m\alpha} - E_{n\beta}$ and, according to (118), the energy difference consists of additive contributions from the environment and the system,

$$E_{m\alpha} - E_{n\beta} = (E_\alpha - E_\beta) + (\epsilon_m - \epsilon_n)\,, \tag{120}$$

as long as the contribution from the interaction, $\delta\omega_{\alpha n\beta m}$, can be neglected on a resolution scale of order $1/t_s$,

$$\delta\omega_{\alpha n\beta m} \ll 1/t_s\,. \tag{121}$$

The approximation, expressed by (120) and (121), is denoted as the weak coupling approximation between environment and system.

In the weak coupling approximation the dissipator $\mathcal{D}(\omega)$ of (72) can be written as

$$\mathcal{D}(\omega)\rho = \sum_{\tilde{\omega}} \sum_{kk'} \left\{ g_{kk'}(\omega+\tilde{\omega}) \left[S_k, (S_{k'}\rho)(\tilde{\omega})\right] - g^*_{kk'}(-\omega-\tilde{\omega}) \left[S_k, (S_{k'}\rho)(-\tilde{\omega})\right]^\dagger \right\} , \quad (122)$$

where the environmental correlation function $g_{kk'}(\Omega)$ is defined by

$$g_{kk'}(\Omega) := \lim_{\epsilon\to 0^+} \sum_{\alpha,\beta} \frac{(\Delta B_k)_{\beta\alpha} (\Delta B_{k'}\rho_{\rm env})_{\alpha\beta}}{\Omega - (E_\alpha - E_\beta) + \imath\epsilon} , \quad (123)$$

and the frequency $\tilde{\omega}$ refers to an energy representation of system operators,

$$X(\tilde{\omega}) := \sum_{\substack{m,\,n \\ \epsilon_n - \epsilon_m = \tilde{\omega}}} P_m X P_n = X^\dagger(-\tilde{\omega}) . \quad (124)$$

By decomposing the denominator in (123) in real and imaginary part,

$$(\Omega - (E_\alpha - E_\beta) + \imath\epsilon)^{-1} = \mathrm{P}\frac{1}{\Omega - (E_\alpha - E_\beta)} - \imath\pi\delta\left(\Omega - (E_\alpha - E_\beta)\right) , \quad (125)$$

we can decompose the correlation function $g_{kk'}(\Omega)$ accordingly,

$$g_{kk'}(\Omega) = s_{kk'}(\Omega) - \imath\gamma_{kk'}(\Omega) . \quad (126)$$

The environmental correlation functions $g_{kk'}(\Omega) = s_{kk'}(\Omega) - \imath\gamma_{kk'}(\Omega)$ can be written as

$$g_{kk'}(\Omega) = \lim_{\epsilon\to 0^+} \left\langle \Delta B_k G^+_{\rm env}(\Omega) \Delta B_{k'} \right\rangle_{\rm env} , \quad (127)$$

$$s_{kk'}(\Omega) = \left\langle \Delta B_k \mathrm{P}(\Omega - L_{\rm env})^{-1} \Delta B_{k'} \right\rangle_{\rm env} , \quad (128)$$

$$\gamma_{kk'}(\Omega) = \left\langle \Delta B_k \pi\delta(\Omega - L_{\rm env}) \Delta B_{k'} \right\rangle_{\rm env} = \quad (129)$$

$$= \frac{1}{2}\int_{-\infty}^{\infty} \mathrm{d}t\, e^{i\Omega t} \left\langle \Delta B_k(t) \Delta B_{k'} \right\rangle_{\rm env} . \quad (130)$$

where $G^+_{\rm env}(\Omega) := (\Omega + \imath\epsilon - L_{\rm env})^{-1}$ is the resolvent with respect to an environmental dynamics within the weak coupling approximation and $X(t) = e^{\imath L_{\rm env} t} X$ the corresponding Heisenberg operator of X.

Note that $\gamma_{kk'}(\omega)$ determines the full $g_{kk'}(\omega)$, since

$$g_{kk'}(\Omega) = \lim_{\epsilon\to 0^+} \frac{1}{\pi} \int d\Theta\, \frac{\gamma_{kk'}(\Theta)}{\Omega - \Theta + \imath\epsilon} . \quad (131)$$

The s correlation function corresponds to $\Delta\mathcal{H}(\omega)$ and the γ correlation function corresponds to $\Gamma(\omega)$,

$$\Delta\mathcal{H}(\omega)\rho = \sum_{\tilde{\omega}} \sum_{kk'} s_{kk'}(\omega+\tilde{\omega}) \left[S_k, (S_{k'}\rho)(\tilde{\omega})\right] - s^*_{kk'}(-\omega-\tilde{\omega}) \left[S_k, (S_{k'}\rho)(-\tilde{\omega})\right]^\dagger , \quad (132)$$

$$\Gamma(\omega)\rho = \sum_{\tilde{\omega}} \sum_{kk'} \gamma_{kk'}(\omega+\tilde{\omega}) \left[S_k, (S_{k'}\rho)(\tilde{\omega})\right] + \gamma^*_{kk'}(-\omega-\tilde{\omega}) \left[S_k, (S_{k'}\rho)(-\tilde{\omega})\right]^\dagger . \quad (133)$$

The probability conservation condition $\mathrm{Tr}\, L(\omega)\rho = 0$ is obviously fulfilled in the weak coupling approximation by $L(\omega)$ in (122) due to the commutator structure. In addition we have $\mathrm{Tr}\, \Delta\mathcal{H}(\omega)\rho = 0 = \mathrm{Tr}\, \Gamma(\omega)\rho$ due to the commutator structure in (132) and in (133). Furthermore, the conditions (see (27), (28)) $(\Delta\mathcal{H}(\omega)\rho)^\dagger = -\Delta\mathcal{H}(-\omega)\rho$ and $(\Gamma(\omega)\rho)^\dagger = \Gamma(-\omega)\rho$ are fulfilled. They keep the time dependent density operator Hermitian.

For consistency of the weak coupling approximation ρ_{env} is assumed to be diagonal in the eigenvalue representation, $(\rho_{\mathrm{env}})_{\alpha\beta} = (\rho_{\mathrm{env}})_\alpha \delta_{\alpha\beta}$. It means that ρ_{env} is not only stationary with respect to L_{env} but also with respect to the complementary dynamics $L_{\mathcal{Q}}$ in the weak coupling approximation. As can be inferred from its definition $s_{kk'}(\omega)$ then forms a Hermitian matrix and $\gamma_{kk'}(\omega)$ is a Hermitian and positive matrix,

$$s^*_{kk'}(\omega) \;=\; s_{k'k}(\omega)\,, \tag{134}$$

$$\gamma^*_{kk'}(\omega) \;=\; \gamma_{k'k}(\omega)\,, \tag{135}$$

$$\sum_{kk'} \gamma_{kk'}(\omega) v_k v^*_{k'} \;\geq\; 0\,. \tag{136}$$

With the choice $\rho_{\mathrm{env}} = d_{\mathrm{env}}^{-1} 1_{\mathrm{env}}$ $\Delta\mathcal{H}(\omega)$ and $\Gamma(\omega)$ are still Hermitian super operators in the weak coupling approximation, $\gamma_{kk'}(\Omega) = \gamma_{k'k}(-\Omega)$ and $\mathrm{Tr}\, \rho(\Gamma(\omega)\rho) = \mathrm{Tr}\, \rho(\Gamma(-\omega)\rho) \geq 0$ holds true. However this choice is no longer appropriate in the weak coupling approximation as the environment is assumed to be in a self-consistent stationary state ρ_{env} no longer affected by the coupling to the system. Thus, ρ_{env} should be taken as the long time limit stationary state of the environment, e.g. an equilibrium state. In case the environment persists in thermal equilibrium, described by the canonical density matrix $\langle\alpha \mid \rho_{\mathrm{env}} \mid \beta\rangle = e^{-(E_\beta - F)/T}\delta_{\alpha\beta}$ with temperature T and canonical free energy F of the environment, it is referred to as the heat bath. As can be inferred from the definitions (128) and (129), the heat bath correlation functions fulfill the Kubo-Martin-Schwinger (KMS) condition of canonical equilibrium (see e.g. [51]),

$$s_{k'k}(-\Omega) \;=\; -e^{-\Omega/T} s_{kk'}(\Omega)\,, \tag{137}$$

$$\gamma_{k'k}(-\Omega) \;=\; e^{-\Omega/T} \gamma_{kk'}(\Omega)\,. \tag{138}$$

In the weak coupling approximation the initial state incorporating correlations, as given by (74), can be written as

$$\Delta\rho_0(\omega) = \sum_{\tilde\omega} \sum_{k,l} g^0_{kl}(\omega + \tilde\omega) \left[S_k, \Delta\rho^{(l)}_{\mathrm{s}0}(\tilde\omega)\right]\,, \tag{139}$$

with the environmental correlation function $g^0_{kl}(\Omega)$

$$g^0_{kl}(\Omega) := \lim_{\epsilon\to 0^+} \sum_{\alpha\beta} \frac{(\Delta B_k)_{\beta\alpha}(\Delta\rho^{(l)}_{e0})_{\alpha\beta}}{\Omega + \imath\epsilon - (E_\alpha - E_\beta)} \,. \tag{140}$$

The constraint $\mathrm{Tr}\,\Delta\rho_0(\omega) = 0$ is fulfilled in the weak coupling approximation.

As an application of our expression for the relaxator Liouville in the weak coupling approximation (122) we consider the paradigmatic spin-boson model. The isolated system consists of the smallest possible entity, a two-level system (qubit or spin). The final expression (152) for the relaxator Liouville can be generalized to arbitrary level number systems by adjusting bare H and the jump-operators S_k. It can then be related to other models like matter coupled to quantized radiation fields in quantum optics (see e.g. Sec. 3.4.1 in [29]) or to Brownian motion of particles as in the Caldeira Leggett model (see e.g. Sec. 3.6 in [29] and references therein). The essence of the relaxator Liouville is, in the weak coupling approximation, contained in the correlation functions $\gamma_{kk'}(\Omega) = \langle \Delta B_k \pi\delta(\Omega - L_{\mathrm{env}})\Delta B_{k'}\rangle_{\mathrm{env}}$ of the environmental coupling operators B_k and the action of the jump-operators S_k on ρ, $[S_k, (S_{k'}\rho)(\tilde{\omega})]$.

To get explicit expressions we considers a bath of independent bosonic modes of frequency $\omega_\alpha > 0$.

$$H_{\mathrm{env}} = \sum_\alpha \omega_\alpha a^\dagger_\alpha a_\alpha \,,\ [a_\alpha, a_{\alpha'}] = 0 = \left[a^\dagger_\alpha, a^\dagger_{\alpha'}\right] \,;\, \left[a_\alpha, a^\dagger_{\alpha'}\right] = \delta_{\alpha\alpha'} \,. \tag{141}$$

The coupling Hamiltonian is

$$H_{\mathrm{int}} = -S \otimes B \,, \tag{142}$$

where S is called dipole operator of the system and B is a field of bath operators.

It is tempting to take B linearly in the creation and annihilation operators for simplicity,

$$B = \sum_\alpha \kappa(\Omega_\alpha) a_\alpha + \kappa^*(\Omega_\alpha) a^\dagger_\alpha \,, \tag{143}$$

with coupling strengths $\kappa(\Omega_\alpha)$. Such linear coupling to independent bath modes is expected to be valid if the relevant wave length λ of modes under consideration is much larger than the characteristic size d of the dipole, $\lambda \gg d$. If characteristic d becomes larger than the relevant λ, the field B should be replaced by a more general field involving at least bilinear terms of creation and annihilation operators. Within the linear coupling to independent

bath modes one finds

$$\gamma(\Omega) = \pi\nu(|\Omega|)|\kappa(|\Omega|)|^2 \cdot \begin{cases} (1+N(\Omega)) & \text{if } \Omega > 0 \\ N(|\Omega|) & \text{if } \Omega < 0 \end{cases} . \tag{144}$$

Here $N(\Omega)$ is the average equilibrium number of modes. However, there is a problematic property of the linear coupling to independent bath modes: the correlation function $\gamma(\Omega)$ has no characteristic time scale involved besides the time scale inherent in $\rho_{\rm env}$ (e.g. the temperature time scale T^{-1} for environmental equilibrium) and the time scale inherent in the density of levels $\nu(|\Omega|)$ (which is the environmental inverse level spacing $t_e(|\Omega|)$). The characteristic time scale of an environmental correlation time τ_e is missing as an ingredient in a bath of independent modes and can only be introduced phenomenologically. Typically, for independent modes in the limit of large volume the density of states becomes a power low $\sim \Omega^p$ with $p \geq 1$ such that $\gamma(0) \to 0$ (due to $\lim_{\omega\to 0} t_e(\omega) = \infty$) in the limit of continuous environmental spectrum. However, for a realistic long time correlation function with decay on a scale τ_e one should find $\gamma(0) \propto \tau_e \neq 0$. Therefore, one shouldn't take the correlation function of (144) literally, but as a means of parametrization to be adjusted phenomenologically or to be improved by couplings which allow to introduce characteristic correlation time scales of the environment. For a deeper discussion of limitations of the linear coupling to independent bath modes we refer to [30] and [53]. In the following we will not rely on the linear coupling expression (144) but stick to the general properties of $\gamma(\Omega)$ for a bath in canonical equilibrium.

We specify the two-level system expressed by the ground state $\mid g\rangle$ and the exited state $\mid e\rangle$ of system Hamiltonian with excitation energy $\omega_0 > 0$,

$$H = \frac{1}{2}\omega_0\sigma_3 \,, \tag{145}$$

where $\sigma_3 := P_e - P_g := \mid e\rangle\langle e \mid - \mid g\rangle\langle g \mid$. For the coupling to the bath modes we take an arbitrary dipole operator,

$$S = S_g P_g + S_e P_e + S_{eg}\sigma_+ + S_{ge}\sigma_- \,, \tag{146}$$

where $\sigma_+ := \mid e\rangle\langle g \mid$ excites from the ground state to the excited state and $\sigma_- := \mid g\rangle\langle e \mid = \sigma_+^\dagger$ decreases an excited state to the ground state. Since S is Hermitian we have S_g, S_e real and $S_{ge}^* = S_{eg}$.

To evaluate level shift (132) and relaxator (133) within this setting we have to calculate $[S,(S\rho)(\tilde{\omega})]$. Taking an arbitrary operator ρ,

$$\rho = \rho_g P_g + \rho_e P_e + \rho_{eg}\sigma_+ + \rho_{ge}\sigma_- \,, \tag{147}$$

and using the algebra of $\sigma_+, \sigma_-, P_e, P_g$ we find

$$[S,(S\rho)(0)] \;=\; (S_{eg}\sigma_+ - S_{ge}\sigma_-)\left[(S_g\rho_g + S_{ge}\rho_{eg}) - (S_e\rho_e + S_{eg}\rho_{ge})\right] \,, \tag{148}$$

$$[S,(S\rho)(\omega_0)] \;=\; \left[(S_g - S_e)\,\sigma_- + S_{eg}\sigma_3\right](S_g\rho_{ge} + S_{ge}\rho_e) \,, \tag{149}$$

$$[S,(S\rho)(-\omega_0)] \;=\; \left[(S_e - S_g)\,\sigma_+ - S_{ge}\sigma_3\right](S_e\rho_{eg} + S_{eg}\rho_g) \,. \tag{150}$$

From Equations (148)-(150) we can already conclude that, for off-diagonal S, the dynamics of populations ρ_g, ρ_e decouples from the dynamics of coherences ρ_{eg}, ρ_{ge}. For diagonal S, the populations are unaffected by the coupling ($H_{\rm int}$ commutes with H) and only coherences will change and finally relax to 0 (pure decoherence).

To write the relaxator Liouville explicitly we consider the bath in canonical equilibrium with temperature T, such that the bath correlation function $g(\Omega) = s(\Omega) - \imath\gamma(\Omega)$ fulfills the KMS condition,

$$g^*(-\Omega) = -g(\Omega)e^{-\Omega/T} \,. \tag{151}$$

The relaxator Liouville in weak coupling (71),(122) then reads

$$\begin{aligned} L(\omega)\rho \;=\;& \omega_0\,(\rho_{eg}\sigma_+ - \rho_{ge}\sigma_-) \\ &+\; g(\omega)\,(S_{eg}\sigma_+ - S_{ge}\sigma_-)\left\{A(S,\rho) - e^{-\omega/T}A^*(S,\rho)\right\} \\ &+\; g(\omega+\omega_0)\,((S_g - S_e)\sigma_- + S_{eg}\sigma_3)\left\{B(S,\rho) - e^{-(\omega+\omega_0)/T}C^*(S,\rho)\right\} \\ &-\; g(\omega-\omega_0)\,((S_g - S_e)\sigma_+ + S_{ge}\sigma_3)\left\{C(S,\rho) - e^{-(\omega-\omega_0)/T}B^*(S,\rho)\right\} \,, \end{aligned} \tag{152}$$

where

$$A(S,\rho) \;:=\; (S_g\rho_g + S_{ge}\rho_{eg}) - (S_e\rho_e + S_{eg}\rho_{ge}) \,, \tag{153}$$

$$B(S,\rho) \;:=\; S_g\rho_{ge} + S_{ge}\rho_e \,, \tag{154}$$

$$C(S,\rho) \;:=\; S_e\rho_{eg} + S_{eg}\rho_g \,. \tag{155}$$

In Equation (152) $g(\Omega)$ is related to $\gamma(\Omega)$ by (131).

It is worth stressing that the relaxator Liouville (152) has the canonical distribution $\rho_{\rm c} = e^{-(H-F)/T}$ with the temperature T inherited from the bath as its stationary solution

for $\omega \to 0$,

$$L(\omega \to 0)\rho_{\rm c} = 0\,. \tag{156}$$

This follows from $\rho_{{\rm c}eg} = \rho_{{\rm c}ge} = 0$ and $\rho_{{\rm c}e} = \rho_{{\rm c}g}e^{-\omega_0/T}$ for arbitrary S. This conclusion is valid due to the weak coupling condition and the KMS condition of the bath, but holds for general couplings S and otherwise arbitrary non-Markovian relaxator Liouville.

In Appendix B we calculate the spectral properties of $L(\omega)$ sufficient for the full time dependence and explicit expressions for the relaxation times of a qubit weakly coupled to a bath from (152).

VII. MARKOV APPROXIMATION: SEMI-GROUP DYNAMICS

In the absence of memory and initial correlations the time evolution possesses the semi-group property, i.e. $\dot\rho = -iL\rho$, with L not depending on time nor on frequency and the initial state ρ_0 is assumed to be uncorrelated with the environment [30]. The absence of memory for relaxator Liouville dynamics means that $L(\omega)$ becomes independent of ω, at least approximately. Weak memory effects correspond to a weakly varying $L(\omega)$. To find a criterion we consider the weak coupling condition (122) where the frequency dependence is contained in the environmental correlation function $\gamma_{kk'}(\omega)$. If its temporal correlation function has a typical range of the environmental correlation time τ_e then $\gamma_{kk'}(\omega)$ will have a characteristic range of $1/\tau_e$. Thus, memory effects are negligible not only in the long time limit but also for investigation times $1/\omega$ much larger than τ_e. To have negligible memory effects throughout the whole time evolution of the open system the condition

$$\tau_e \ll \delta t_s \tag{157}$$

has to hold, where δt_s is the temporal resolution time of the system. As the Markovian limit of the relaxator Liouville we take

$$L := L(\omega = 0) \tag{158}$$

as a semi-group-generator, because we want to be sure that its stationary limit is as close as possible with that of the original relaxator Liouville. The condition for the absence of memory (157) is quite restrictive and often not fulfilled in a realistic system when times between δt_s and τ_r are addressed. One may ask why the semi-group approach that assumes

the absence of memory (and absence of initial correlations) is still quite successful in describing several features of open systems. We think the answer can be given by looking at the solution (34) of the relaxator Liouville dynamics in its spectral resolution. The solution (38) shows a superposition of damped oscillations which could as well be described as the solution of a frequency independent Liouville L with its eigenfrequencies adopted to the effective eigenfrequencies z_k (37) of the full $L(z)$. In particular, when fast oscillatory behavior is averaged out, the Markov approximation, still capturing relaxation, becomes better.

To make contact with previously known modeling we switch to the Markov limit of our relaxator Liouville within the weak coupling approximation, expressed by (132) and (133),

$$L\cdot = L(\omega = 0)\cdot = [H_{\mathcal{P}}, \cdot] + \Delta\mathcal{H}(0)\cdot - \imath\Gamma(0)\cdot \ . \tag{159}$$

By using the Hermiticity properties of the bath correlation functions and rearrangements of sums we find level shift and relaxator in the Markov limit of the weak coupling approximation,

$$\Delta\mathcal{H}(0)\rho \ = \ \sum_{\tilde{\omega}} \sum_{kk'} s_{kk'}(\tilde{\omega}) \left([S_k, (S_{k'}\rho)(\tilde{\omega})] - [(\rho S_k)(-\tilde{\omega}), S_{k'}] \right) \ , \tag{160}$$

$$\Gamma(0)\rho \ = \ \sum_{\tilde{\omega}} \sum_{kk'} \gamma_{kk'}(\tilde{\omega}) \left([S_k, (S_{k'}\rho)(\tilde{\omega})] + [(\rho S_k)(-\tilde{\omega}), S_{k'}] \right) \ . \tag{161}$$

To make contact with the modeling known as the Born-Markov approximation with secular approximation (see e.g. Chap. 3.31 in [29]) we perform an additional approximation reminiscent of the secular approximation. In a secular approximation, also known as rotating wave approximation, one focuses on those contributions to terms like $S_k(S_{k'}\rho)(\tilde{\omega})$ which do not contain strongly fluctuating phase factors and treats them as canceling each other. The secular approximation puts further limitations on the resolution of the time evolution of the density operator as the typical periods of oscillations are considered to be small compared to the observational time t before relaxation sets in and such oscillations are averaged out. To this end we rewrite such terms exactly in the eigenfrequency representation (see (124)) as

$$S_k(S_{k'}\rho)(\tilde{\omega}) = \sum_{\omega',\omega''} S_k(\omega') S_{k'}(\omega'') \rho(\tilde{\omega} - \omega'') \, . \tag{162}$$

Those contributions that fulfill $\tilde{\omega} = \omega''$ and $\omega' = -\omega''$ are free of strongly fluctuating phase factors and constitute the secular approximation for (162),

$$S_k(S_{k'}\rho)(\tilde{\omega}) \approx S_k^\dagger(\tilde{\omega}) S_{k'}(\tilde{\omega})\rho \, , \tag{163}$$

where $\rho(0)$ was replaced by ρ. Performing the secular approximation on (160) and (161) we arrive at level shift and relaxator in our weak coupling approximation in the Markov limit with secular approximation,

$$\Delta\mathcal{H}_{\rm sec}\rho \;=\; [H_{\rm LS},\rho]\;, H_{\rm LS} := \sum_{\tilde\omega}\sum_{kk'} s_{kk'}(\tilde\omega) S_k^\dagger(\tilde\omega) S_{k'}(\tilde\omega)\,, \tag{164}$$

$$\Gamma_{\rm sec}\rho \;=\; \sum_{\tilde\omega}\sum_{kk'} \gamma_{kk'}(\tilde\omega)\left[\left\{S_k^\dagger(\tilde\omega)S_{k'}(\tilde\omega),\rho\right\} - 2S_{k'}(\tilde\omega)\rho S_k^\dagger(\tilde\omega)\right]\,. \tag{165}$$

These expressions are identical with those of the well known Born-Markov master equation in secular approximation (see e.g. Chap. 3.31 in [29]). The level shift serves as a simple shift of levels as $H_{\rm LS}$ commutes with H and the relaxator is in Lindblad form and thus generates a so called completely positive semi-group dynamics. In the diagonal representation of ρ one can show that the relaxator is positive, $\mathrm{Tr}\,\rho\Gamma_{\rm sec}\rho \geq 0$.

As an application of the Markovian limit without secular approximation we consider the Markovian limit of the spin boson model as given in (152). For the semi-group dynamics of ρ we have

$$\begin{aligned}
\dot\rho = -\imath L(0)\rho \;=\;& -\imath\omega_0\left(\rho_{eg}\sigma_+ - \rho_{ge}\sigma_-\right)\\
&-\; 2\gamma(0)\left(S_{eg}\sigma_+ - S_{ge}\sigma_-\right) D(S,\rho)\\
&-\; \imath g(\omega_0)\left((S_g - S_e)\sigma_- + S_{eg}\sigma_3\right)\left\{B(S,\rho) - e^{-\omega_0/T}C^*(S,\rho)\right\}\\
&+\; \imath g^*(\omega_0)\left((S_g - S_e)\sigma_+ + S_{ge}\sigma_3\right)\left\{B^*(S,\rho) - e^{-\omega_0/T}C(S,\rho)\right\}\,,
\end{aligned} \tag{166}$$

where

$$D(S,\rho) = S_{ge}\rho_{eg} - S_{eg}\rho_{ge}\,. \tag{167}$$

Equation (166) leads to linear differential equations for the components of ρ,

$$\dot\rho_e \;=\; 2\mathrm{Im}\;\left\{g(\omega_0)\left[a_e(S,T)\left(\rho_e - \rho_{\mathrm{c},e}\right) + b_e(S,T)\rho_{ge}\right]\right\} \tag{168}$$

$$\begin{aligned}
\dot{\rho_{eg}} \;=\;& \left(-\imath\omega_0 + \imath g^*(\omega_0)a_{eg}(S,T)\right)\rho_{eg}\\
&+\; \imath g^*(\omega_0)b_{eg}(S,T)\left(\rho_e - \rho_{\mathrm{c},e}\right)\\
&-\; 2\imath\gamma(0)\left(|S_{eg}|^2\rho_{ge} - S_{eg}^2\rho_{eg}^*\right)\,,
\end{aligned} \tag{169}$$

where

$$a_e(S,T) \;:=\; |S_{eg}|^2\left(1 + e^{-\omega_0/T}\right)\;,\quad b_e(S,T) := S_{eg}\left(S_g - S_e e^{-\omega_0/T}\right)\,, \tag{170}$$

$$a_{eg}(S,T) \;:=\; (S_g - S_e)\left(S_g - S_e e^{-\omega_0/T}\right)\;,\; b_{eg}(S,T) := (S_g - S_e)\,S_{eg}\left(1 + e^{-\omega_0/T}\right)\,, \tag{171}$$

and

$$\rho_{\mathrm{c},e} = \frac{e^{-\omega_0/T}}{1+e^{-\omega_0/T}} \tag{172}$$

is the equilibrium canonical population in the excited state.

From Equations (168)-(171) we conclude for the qubit weakly coupled to a heat bath in the Markov approximation: as soon as there is some off-diagonal coupling, $S_{eg} \neq 0$, which means excitation and emission processes, the state will relax to the canonical equilibrium state with relaxation time

$$\tau_r = \left[2\gamma(\omega_0)|S_{eg}|^2\left(1+e^{-\omega_0/T}\right)\right]^{-1} . \tag{173}$$

The dying out of the coherence ρ_{eg} depends on the diagonal elements of S and in the generic case of a non-vanishing $\gamma(0)$ also on the off-diagonal elements. In case off a purely diagonal S, the populations are unaffected by symmetry and the coupling leads to a scenario of pure decoherence with decoherence time

$$\tau_{\mathrm{dec}} = \left[\gamma(\omega_0)\left(S_g - S_e\right)\left(S_g - S_e e^{-\omega_0/T}\right)\right]^{-1} . \tag{174}$$

In the non-generic case of purely off-diagonal S and vanishing $\gamma(0)$ we would have a degenerate asymptotic state: relaxation of populations to equilibrium with relaxation time τ_r and oscillations of coherences with frequency ω_0. In a secular approximation calculation such oscillations cannot be captured.

VIII. SUMMARY

We developed the relaxator Liouville dynamics (Eqs. (4),(16) - (19)) for relevant degrees of freedom in a macroscopic system from a microscopic von Neumann equation for an isolated total system of all degrees of freedom. The irrelevant degrees of freedom act as an environment on the relevant degrees of freedom. The relaxator $\Gamma(\omega)$ indicates the onset of irreversibility. It can be expressed by a Hamiltonian modeling of system environment coupling (Eq. (70). For a weak coupling the relaxator Liouville is determined by environmental correlation functions and hopping operators in the system (Eq. (122)). The relaxator Liouville dynamics has generically a unique stationary state (Eq. (48)) which is reached after a characteristic relaxation time that can be calculated from the relaxator Liouville's effective

eigenvalues (Eq. (39)). For equilibrium conditions the system reaches canonical thermal equilibrium with a temperature adopted to the environment.

We derived a closed kinetic equation for the one-particle density operator (Eq. (81)) as a special form of relaxator Liouville dynamics where one-particle degrees of freedom are the relevant ones. The origin of irreversible behavior can be identified via the relaxator (Eq. (85)).

For situations of current carrying stationary states in the presence of perturbing external fields we derived linear response expressions for dynamic susceptibilities (Eq. (102)). The derived susceptibilities coincide with Kubo's susceptibilities in a regime where the energy eigenstates of the unperturbed system have undetectable large lifetimes and the probing frequency is much larger than the typical level spacing. Despite the practically infinite lifetimes of energy eigenstates, the response can show an collective irreversible character as the spectrum of the system becomes dense relative to the frequency. The corresponding time scale of irreversibility, a decay time of correlations within the system, can be extracted from the susceptibility (Eq. (117)).

We identified Markov semi-group dynamics as limiting modelings of our more general relaxator Liouville dynamics ((160),(161)).

As the source of irreversibility we have identified time scale separations. For a system in contact with a surrounding bath the time to explore the spectral content of the bath is much larger than the time to explore the spectral content of the system. For a macroscopic system in general the dynamics of the so-called relevant variables becomes irreversible when the time to explore its spectral content is much smaller than the time to explore the spectral content of the complementary dynamics. In transport situations with driving fields, irreversibility and dissipation of energy emerges when they are macroscopic or subject to macroscopic contacts such that the level spacing of the system cannot be resolved with the frequency of the applied field. In other words: the time to explore the system's spectral content is much larger as any time we can afford to measure the response of the system to applied fields.

Acknowledgements The authors acknowledge valuable comments during the process of writing from Victor Albert (2018), Heinz-Peter Breuer (2018) , John Chalker (2024), Rochus Klesse (2017), Dimitry Polyakov (2025), János Polonyi (2017, 2025) and Peter Wölfle (2025).

Conflict of Interest The authors declare no conflict of interest.

Data Availability Statement Data sharing is not applicable to this article as no new

data were created or analyzed in this study.

Keywords irreversibility, non-equilibrium, relaxation, quantum Master, non-Markovian, kinetic equations, linear response

Appendix A: Approaching canonical equilibrium in weak coupling to a bath

We consider the relaxator within the weak coupling approximation (161) for $\omega = 0$ and calculate its elements $\Gamma_{rr,nn}$ according to (53)

$$\begin{aligned}\Gamma_{rr,mm}(\omega=0) &= (\langle r|\Gamma(0)P_n|r\rangle) = \\ &= \sum_{\tilde{\omega}}\sum_{k,k'}\gamma_{kk'}(\tilde{\omega})\left\{\langle r \mid [S_k,(S_{k'}P_n)(\tilde{\omega})] + [(P_nS_k)(-\tilde{\omega}),S_{k'}] \mid r\rangle\right\} . \quad \text{(A1)}\end{aligned}$$

Since P_n commutes with other P_q, the eigenfrequency representations in (A1) read $(S_{k'}P_n)(\tilde{\omega}) = S_{k'}(\tilde{\omega})P_n$ and $(P_nS_k)(-\tilde{\omega}) = P_nS_k(-\tilde{\omega})$. For $n \neq r$ we find the restriction $\tilde{\omega} = \epsilon_n - \epsilon_r$ in

$$\Gamma_{rr,mm}(\omega=0) = -\sum_{\tilde{\omega}}\sum_{k,k'}\gamma_{kk'}(\tilde{\omega})\left\{\langle r|S_k|n\rangle\langle n|S_{k'}|r\rangle + \langle r|S_{k'}|n\rangle\langle n|S_k|r\rangle\right\}. \quad \text{(A2)}$$

The resulting transition rate $W_{rn} = -\Gamma_{rr,nn}(\omega=0) \geq 0$ $(n \neq r)$ due to the positive matrix $\gamma_{kk'}$. By the KMS condition (138) on the bath correlation function $\gamma_{kk'}(\tilde{\omega})$ with $\tilde{\omega} = \epsilon_n - \epsilon_r$ for the bath in canonical equilibrium at temperature T we conclude

$$W_{nr} = e^{-(\epsilon_n-\epsilon_r)/T}W_{rn}\,, \quad \text{(A3)}$$

which is the condition (61) for reaching canonical equilibrium in the system with the bath temperature T.

Appendix B: qubit weakly coupled to a bath: spectral properties and relaxation times

As a prototypical example for non-Markovian dynamics we consider the relaxator Liouville of a qubit weakly coupled to a bath as given by (152). The time evolution is, according to (39), characterized by the effective eigenvalues z_k, the stationary state and coefficients depending on the initial state. We look at two special cases: (I) The coupling operator S is purely diagonal and (II) the coupling operator S is purely off-diagonal.

In case (I) $S = S_e P_e + S_g + P_g$ commutes with the system Hamiltonian $H = \frac{1}{2}\omega_0(P_e - P_g)$. This leads to pure decoherence as the populations $\rho_e = 1 - \rho_g$ are not affected by the dynamics. Every diagonal density operator $\rho = \rho_e(P_e - P_g) + P_g$ is a right eigenoperator of $L(\omega)$ with vanishing eigenvalue, $\lambda = 0$. The eigenvalue 0 is twofold degenerate. As two $\lambda = 0$ eigenoperators of a bi-orthonormal set of 4 eigenoperators, we can choose:

$$\mathsf{R}_0 = \rho_e(P_e - P_g) + P_g \,, \mathsf{R}_1 = \sigma_3 \,. \tag{B1}$$

The corresponding left eigenoperators follow from the bi-orthonormal properties as

$$\mathsf{L}_0 = 1 \,, \mathsf{L}_1 = -\rho_e(P_g + P_e) + P_e \,. \tag{B2}$$

We can choose ρ_e in (B1) as the initial population in the excited state. Equation (39) confirms that the populations stay constant during the time evolution. From Equation (152) for case (I) we conclude that excitation and absorption form the two remaining right eigenoperators,

$$\mathsf{R}_2 = \sigma_+ \,, \mathsf{R}_3 = \sigma_- \,. \tag{B3}$$

By comparison with the eigenvalue condition we find the corresponding eigenvalues,

$$\begin{aligned} \lambda_2(\omega) &= \omega_0 + g(\omega - \omega_0)\,(S_e - S_g)\left(S_e - S_g e^{-(\omega-\omega_0)/T}\right) =: \lambda_+(\omega) \,, \\ \lambda_3(\omega) &= -\omega_0 + g(\omega + \omega_0)\,(S_g - S_e)\left(S_g - S_e e^{-(\omega+\omega_0)/T}\right) =: \lambda_-(\omega) \,. \end{aligned} \tag{B4}$$

The left eigenoperators follow from the bi-orthonormal properties as

$$\mathsf{L}_2 = \sigma_+ = \mathsf{R}_2 \,, \mathsf{L}_3 = \sigma_- = \mathsf{R}_3 \,. \tag{B5}$$

The relaxation of the coherences is described by a damped oscillation characterized by two effective complex eigenvalues

$$z_k = \omega_k - \imath\delta_k = \lambda_k(\omega_k) \,, k = +, - \,, \tag{B6}$$

where Hermiticity of $\rho(t)$ requires

$$z_+ = -z_-^* \,. \tag{B7}$$

The real part, $\omega_+ = -\omega_-$, is the solution of the real part of (B6) involving $s(\omega_+ - \omega_0)$, describing the oscillatory behavior. The relaxation is described by $\delta_+ = \delta_-$ and involves $\gamma(\omega_+ - \omega_0)$. δ_+ sets the decoherence time scale,

$$\tau_{\rm dec} = \left[\gamma(\omega_+ - \omega_0)\,(S_e - S_g)\left(S_e - S_g e^{-(\omega_+-\omega_0)/T}\right)\right]^{-1} \,. \tag{B8}$$

Since $s(0) = 0$ holds for symmetry reasons, the solution $\omega_+ = \omega_0$ solves the real part of (B6) and we find

$$\tau_{\rm dec} = \left[\gamma(0)\,(S_e - S_g)^2\right]^{-1} . \tag{B9}$$

A vanishing of $\gamma(0)$ would prohibit decoherence. However, as discussed before, a vanishing $\gamma(0)$ is unrealistic for a heat bath. Equation (B8) in comparison to (174) shows a characteristic difference between the non-Markovian relaxator Liouville dynamics and the Markov limit to that dynamics: while in the Markov case the limit $\omega \to 0$ comes first and then the eigenvalue of $L(0)$ counts, in the non-Markovian approach the effective eigenvalue of the frequency dependent relaxator Liouville counts. A difference between the decoherence times of (B8) and(174) will not be detectable if ω_0 is much smaller than the inverse of the environmental correlation time τ_e (see the discussion in Sec. VII).

Switching to case (II), $S = S_{ge}\sigma_- + S_{eg}\sigma_+$ we write the relaxator Liouville of (152) as

$$\begin{aligned} L(\omega)\rho \;=\; & \omega_0\,(\rho_{eg}\sigma_+ - \rho_{ge}\sigma_-) \\ & +\; g(\omega)\,(S_{ge}\rho_{eg} - S_{eg}\rho_{ge})\left(1 + e^{-\omega/T}\right)(S_{eg}\sigma_+ - S_{ge}\sigma_-) \\ & +\; |S_{eg}|^2\left\{g(\omega+\omega_0)\left(\rho_e - \rho_g e^{-(\omega+\omega_0)/T}\right) - g(\omega-\omega_0)\left(\rho_g - \rho_e e^{-(\omega-\omega_0)/T}\right)\right\}\sigma_3\,. \end{aligned} \tag{B10}$$

The $\lambda_0(\omega) = 0$ right eigenoperator with unit trace can be found by looking for ρ_e for which the factor in front of σ_3 in (B10) vanishes. Obviously σ_3 is a right eigenvector with non vanishing eigenvalue $\lambda_1(\omega)$. Thus, we have

$$\mathsf{R}_0(\omega) = \rho_e(\omega)(P_e - P_g) + P_g\,, \lambda_0(\omega) = 0\,, \mathsf{R}_1(\omega) = \sigma_3\,, \tag{B11}$$

with

$$\rho_e(\omega) = \frac{g(\omega+\omega_0)e^{-(\omega+\omega_0)/T} + g(\omega-\omega_0)}{g(\omega+\omega_0)\,(1+e^{-(\omega+\omega_0)/T}) + g(\omega-\omega_0)\,(1+e^{-(\omega-\omega_0)/T})}\,. \tag{B12}$$

The effective eigenvalue $z_0 = 0$ requires $\omega_{k=0} = 0$ and at this value we have the stationary canonical equilibrium density operator with

$$\rho_{e,\infty} = \rho_e(0) = \frac{e^{-\omega_0/T}}{1+e^{-\omega_0/T}} = \rho_{{\rm can},e}\,. \tag{B13}$$

The eigenvalue $\lambda_1(\omega)$ can be seen from the prefactor of σ_3 in (B10) and reads

$$\lambda_1(\omega) = |S_{eg}|^2\left\{g(\omega+\omega_0)\left(1+e^{-(\omega+\omega_0)/T}\right) + g(\omega-\omega_0)\left(1+e^{-(\omega-\omega_0)/T}\right)\right\}\,. \tag{B14}$$

The real part of the effective eigenvalue equation yields a vanishing frequency $\omega_1 = 0$ but a finite δ_1 which defines the relaxation time for the populations,

$$\tau_{r,\mathrm{diag}} = \left[2|S_{eg}|^2\gamma(\omega_0)\left(1+e^{-\omega_0/T}\right)\right]^{-1} . \tag{B15}$$

This unsurprisingly coincides with the result in the Markov limit (173).

The off-diagonal right eigenoperators and corresponding eigenvalues can be found by a standard matrix eigenvalue routine for $L(\omega)\rho$ in the $\sigma_\pm$ basis. We find the eigenvalues as

$$\lambda_{2,3}(\omega) = f(\omega,T,S) \pm (\omega_0 + \delta z_0(\omega,T,S)) , \tag{B16}$$

where

$$f(\omega,T,S) := g(\omega)\left(1+e^{-\omega/T}\right)|S_{ge}|^2 , \tag{B17}$$

$$\omega_0 + \delta z_0(\omega,T,S) := \omega_0\left(1+\frac{|f|^4}{\omega_0^4}+2\frac{|f|^2\cos(2\varphi)}{\omega_0^2}\right)^{1/4} e^{-\imath\tilde{\varphi}/2} , \tag{B18}$$

$$\tan(\varphi) := \frac{s}{\gamma} , \tan(\tilde{\varphi}) := \frac{|f|^2\sin(2\varphi)}{\omega_0^2+|f|^2\cos(2\varphi)} . \tag{B19}$$

The right eigenoperators read

$$\mathsf{R}_2(\omega) = \frac{1}{\left(1+|\frac{\delta z_0}{\omega_0+f}|^2\right)^{1/2}}\left(\sigma_+ + \frac{\delta z_0}{\omega_0+f}\sigma_-\right) , \tag{B20}$$

$$\mathsf{R}_3(\omega) = \frac{1}{\left(1+|\frac{\delta z_0}{\omega_0+f}|^2\right)^{1/2}}\left(\sigma_- - \frac{\delta z_0}{\omega_0+f}\sigma_+\right) . \tag{B21}$$

The corresponding left eigenoperator $L_2 = c_2\sigma_+ + d_3\sigma_-$ for two given right eigenoperators $R_{2,3} = a_{2,3}\sigma_+ + b_{2,3}\sigma_-$ can be constructed by the following scheme due to the bi-orthonormal properties

$$c_2^* = \frac{b_3}{a_2b_3-a_3b_2} , \; d_2^* = \frac{-a_3}{a_2b_3-a_3b_2} . \tag{B22}$$

The effective eigenvalues $z_2 = -z_3^*$ are found by solving the real part equation

$$\omega_2 = \omega_0 + \mathrm{Re}\ \left(f(\omega_2) + \delta z_0(\omega_2)\right) \tag{B23}$$

and calculating the imaginary part that defines the relaxation time for coherences,

$$\tau_{\mathrm{dec}} = \left[\gamma(\omega_2)(1+e^{-\omega_2/T})|S_{eg}|^2 + \mathrm{Im}\ \delta z_0(\omega_2)\right]^{-1} . \tag{B24}$$

Note, that here the decoherence time is different from the Markov limit value as ω_2 is shifted from ω_0. For weak coupling the shift amounts to a perturbation of $\mathcal{O}(|f(\omega_0)|/\omega_0)$ and for

short environmental correlation times $\tau_e \ll \omega_0^{-1}$ the difference in decoherence times will be negligible.

---

[1] B. Zwiebach, *Mastering Quantum Mechanics: Essentials, Theory, and Applications*, MIT Press, Cambridge, MA (2022).

[2] L. D. Landau, E. M Lifshitz, *Statistical Physics. Vol. 5 (3rd ed.)*, Butterworth-Heinemann, (1980).

[3] L Boltzmann, *Weitere Studien über das Wärmegleichgewicht unter Gasmolekülen*, in: Sitzungsberichte der Kaiserlichen Akademie der Wissenschaften **66**, 275 (1872), reprinted in L. Boltzmann, *Wissenschaftliche Abhandlungen, Bd. 1., p. 316*, J. A. Barth, Leipzig (1909).

[4] M.v. Smoluchowski, *Drei Vorträge über Diffusion, Brownsche Bewegung und Koagulation von Kolloidteilchen*, Physik. Zeit. **17**, 557 (1916).

[5] W. Pauli, *Festschrift zum 60. Geburtstage A. Sommerfelds, p.30* Hirzel, Leipzig (1928).

[6] V. Weisskopf and E. Wigner, *Berechnung der natürlichen Linienbreite auf Grund der Diracschen Lichttheorie*, Z. Phys. **63**, 54 (1930); *Über die natürliche Linienbreite in der Strahlung des harmonischen Oszillators*, Z. Phys. **65**, 18 (1930).

[7] L.v. Hove, *Quantum-Mechanical Perturbations Giving Rise to a Statistical Transport Equation*, Physica **21**, 517 (1955), Physica **23**, 441 (1957), Physica **25**, 268 (1959).

[8] R. Kubo, *Statistical Mechanical Theory of Irreversible Processes. I.* J. Phys. Soc. Japan **12**, 570 (1957).

[9] I. Prigogine, *Non-Equilibrium Statistical Mechanics*, Interscience Publishers, New York, (1963).

[10] R. Zwanzig, *Non Equilibrium Statistical Mechanics*, Oxford University Press, Oxford, (2001).

[11] J. Uffink, *Compendium of the foundations of classical statistical physics*, in: Jeremy Butterfield & John Earman (eds.), *Handbook of the Philosophy of Physics, p. 923*, Elsevier, (2007).

[12] T.B. Batalhão, S. Gherardini, J.P. Santos, G.T. Landi, and M. Paternostro, *Characterizing Irreversibility in Open Quantum Systems*, in: F. Binder, L.A. Correa, C. Gogolin, J. Anders, G. Adesso (eds.), *Thermodynamics in the Quantum Regime*, Springer, Cham, (2018), arXiv:1806.08441 [quant-ph].

[13] B.W. Roberts, *Reversing the Arrow of Time*, Cambridge University Press, Cambridge, (2022),

DOI 10.1017/9781009122139.

[14] C. Callender (ed.), *The Oxford Handbook of Philosophy of Time*, Oxford University Press Inc., New York (2011).

[15] L.C. Venuti, *The recurrence time in quantum mechanics*, arXiv:1509.04352v2 [quant-ph] (2015).

[16] R.K.P. Zia, E.F. Redish, and S.R. McKay, *Making Sense of the Legendre Transform*, arXiv:0806.1147 [physics.ed-ph] (2009).

[17] S. Nakajima, *On Quantum Theory of Transport Phenomena*, Prog. Theor. Phys. **20**, 948 (1958).

[18] R. Zwanzig, *Ensemble Method in the Theory of Irreversibility*, J. Chem. Phys. **33**, 1338 (1960).

[19] U. Fano, *Liouville Representation of Quantum Mechanics with Application to Relaxation Processes* in: E. R. Caianiello, *Lectures on the Many Body Problem Vol. 2*, p. 217, Academic Press, New York, (1964).

[20] D.A. Abanin, E. Altman, I. Bloch, and M. Serbyn, *Colloquium: Many-body localization, thermalization, and entanglement*, Rev. Mod. Phys. **91**, 021001 (2019).

[21] H.P. Breuer, E.-M. Laine, J. Piilo, and B. Vacchini *Non-Markovian dynamics in open quantum system*, Rev. Mod. Phys. **88**, 021002 (2016), arXiv:1505.01385v1[quant-ph].

[22] D. Maldonado-Mundo, P. Öhberg, B. W. Lovett, and E. Andersson, *Investigating the generality of time-local master equations*, Phys. Rev. A **86**, 042107 (2012), DOI https://doi.org/10.1103/PhysRevA.86.042107.

[23] W.-M. Zhang, P.-Y. Lo, H.-N. Xiong, M. W.-Y. Tu, and F. Nori, *General Non-Markovian Dynamics of Open Quantum Systems*, PRL **109**, 170402 (2012).

[24] M. Bonitz, *Quantum Kinetic Theory (2nd ed.)*, Teubner-Texte zur Physik, Springer, Cham, (2016).

[25] R. van Leeuwen and N.E. Dahlen *An Introduction to Nonequilibrium Green Functions*, lecture of 3rd International Workshop and School on Time-Dependent Density-Functional Theory: Prospects and Applications, https://www.tddft.org/TDDFT2008/lectures/RL_LN.pdf (2006) (accessed 2023/07/24).

[26] K.Y. Camsari, S. Chowdhury and S. Datta, *The Non-Equilibrium Green Function (NEGF) Method*, in: Springer Handbook of Semiconductor Devices, p. 1583, Springer, (2023), arXiv:2008.01275v2 [cond-mat.mes-hall].

[27] G. Lindblad, *On the generators of quantum dynamical semi-groups*, Commun. Math. Phys. **48**, 119 (1976).

[28] V. Gorini, A. Kossakowski and E.C.G. Sudarshan, *Completely Positive Dynamical Semigroups of N Level Systems*, Journ. of Math. Phys. **17**, 821 (1976).

[29] H.P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems*, Oxford University Press, Oxford, (2002).

[30] R. Alicki, *Invitation to quantum dynamical semi-groups*, arXiv:0205188[quant-ph] (2002).

[31] H. P. Breuer, and F. Petruccione, *Concepts and methods in the theory of open quantum systems* in: F.Benatti, and R.Floreanini (Eds.), *Irreversible Quantum Dynamics*, p. pp 65, Lecture Notes in Physics, Vol. 622, Springer, Berlin, Heidelberg, (2003), arXiv:quant-ph/0302047[quant-ph].

[32] A. Rivas, and F.S. Huelga, *Open Quantum Systems. An Introduction*, Springer Berlin, Heidelberg, arXiv:1104.5242v2 [quant-ph] (2012).

[33] D. Lidar, *Lecture Notes on the Theory of Open Quantum Systems*, arXiv:1902.00967v2 (2020).

[34] M. Janßen, *Equation of Motion for Open Quantum Systems incorporating Memory and Initial Correlations*, arXiv:1810.06458v1[quant-ph] (2018).

[35] J. Polonyi, *Dynamical breakdown of time reversal invariance and causality*, Phys. Rev. D **84**, 105021 (2011), arXiv:1109.2228 [hep-th].

[36] M. Janßen, *Generated Dynamics of Markov and Quantum Processes*, Springer-Verlag, Berlin Heidelberg, (2016).

[37] T. Yoshimura, and L. Sa, *Theory of Irreversibility in Quantum Many-Body Systems*, arXiv:2501.06183v3 [cond-mat.stat-mech] (2025).

[38] N.G. van Kampen, *Stochastic Processes in Physics and Chemistry, 2nd edn.*, North-Holland,Amsterdam, (1992).

[39] N.G. van Kampen, *Internal Noise*, Chapter XVII 7 in [38].

[40] M. Janßen, *On Generated Dynamics for Open Quantum Systems: Spectral Analysis of Effective Liouville*, arXiv:1707.09660[quant-ph] (2017).

[41] V.V. Albert, *Lindbladians with multiple steady states: theory and applications*, arXiv:1802.00010[quant-ph] (2018).

[42] J. Schwinger, J. Math. Phys. (N.Y.) **2**, 407 (1961); L.P. Kadanoff, G. Baym, *Quantum Statistical Mechanics*, Benjamin, New York, 1962 R. P. Feynman, F. L. Vernon, Ann. Phys. (N.Y.)

24, 118 (1963); L.V. Keldysh, Sov. Phys. JETP **20**, 1018 (1965).

[43] R. Kubo, *The fluctuation-dissipation theorem*, Rep. Prog. Phys. **29**, 255 (1966).

[44] M. Ban, S. Kitajima, T. Arimitsu, and F.Shibata, *Linear response theory for open systems: Quantum master equation approach*, Phys. Rev. A **95**, 022126 (2017).

[45] L. Pan, X. Chen, Y. Chen, and H. Zhai, *Non-Hermitian linear response theory*, Nature Physics **16**, 767 (2020).

[46] J.E. Avron, M. Fraas, and G.M. Graf, *Adiabatic response for Lindblad dynamics*, J. Stat Phys. **148**, 800 (2012).

[47] J. H. Wei, and Y. Yan *Linear response theory for quantum open systems*, arXiv:1108.5955 [cond-mat.mes-hall] (2011).

[48] R. P. Feynman, and F. L. Vernon, *The theory of a general quantum system interacting with a linear dissipative system*, in [42]

[49] P. Gies and J. Hajdu, *Shubnikov-de haas effect in kinetic approximation*, Z. Phys. B **60**, 231 (1985).

[50] O. Viehweger, *A self-consistent approach to the localization-delocalization problem of the quantum Hall effect*, Z. Phys. B **83**, 45 (1991).

[51] R. Haag, N.M. Hugenholtz, and N. Winnink, *On the Equilibrium States in Quantum Statistical Mechanics*, Commun. math. Phys. **5**, 215 (1967).

[52] L. Onsager, *Reciprocal Relations in Irreversible Processes.*, Phys. Rev. **37**, 405 (1931), Phys. Rev. **38**, 2265 (1931).

[53] K. Hornberger, *Introduction to decoherence theory*, Lect. Notes Phys. **768**, 221 (2009).