Characterization of the Quasi-Stationary State of an Impurity Driven by Monochromatic Light I - The Effective Theory*

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December 21, 2013

Abstract

We consider an impurity (N-level atom) driven by monochromatic light in a host environment which is a fermionic thermal reservoir. The external light source is a time-periodic perturbation of the atomic Hamiltonian stimulating transitions between two atomic energy levels $E_1$ and $E_N$ and thus acts as an optical pump. The purpose of the present work is the analysis of the effective atomic dynamics resulting from the full microscopic time-evolution of the compound system. We prove, in particular, that the atomic dynamics of population relaxes for large times to a quasi-stationary state, that is, a stationary state up to small oscillations driven by the external light source. This state turns out to be uniquely determined by a balance condition. The latter is related to "generalized Einstein relations" of spontaneous/stimulated emission/absorption rates, which are conceptually similar to the phenomenological relations derived by Einstein in 1916. As an application we show from quantum mechanical first principles how an inversion of population of energy levels of an impurity in a crystal can appear. Our results are based on the spectral analysis of the generator of the evolution semigroup related to a non-autonomous Cauchy problem effectively describing the atomic dynamics.

1. Introduction

In the present paper and in a companion one [1] we study the dynamics of an impurity (an atom) in a crystal, or host environment, interacting with an external monochromatic light source serving as an optical pump. The host environment corresponds to free electrons in thermal equilibrium within the crystal whereas the atom is described by a $N$-level system, the pure states of which are the unit vectors of the finite dimensional Hilbert space $\mathbb{C}^d$ ($d \geq N \geq 1$). The external monochromatic light source is a time-periodic classical field stimulating transitions between the two energy levels $E_1$ and $E_N$. The microscopic non-autonomous dynamics of the full system, that is, impurity, host environment and external light source, is then described through a two-parameter group of automorphisms generated by a time-dependent symmetric derivation acting on the $C^*$-algebra of observables of the compound system. This dynamics is generally non-unitary when restricted to the atomic subalgebra. The restricted dynamics of the impurity shows a dissipative behavior, provided the coupling to the host environment (thermal reservoir) is effective. In the second part [1] of the present work we prove that the dynamics of the impurity is properly described – up to small corrections for a moderate reservoir–atom–pump interaction – by some effective non-autonomous time-evolution involving atomic degrees of freedom only. See Theorem 3.3.

The detailed study of this effective dynamics is the main goal of this paper. More precisely, we are interested in an effective description of the time-evolution of populations of the atomic energy levels, i.e., an effective atomic block-diagonal dynamics. To this end, we use evolution semigroup techniques transforming the non-autonomous Cauchy problem into an autonomous one on a suitable Banach space. The same method is also used in [1] to analyze the full microscopic dynamics. In fact, many key arguments of the present analysis recur in [1], albeit technically more involved. We remark that evolution semigroups to study the long-time behaviour of quantum systems have also been used by Abou–Salem and Fröhlich [2].

*To appear in Annales Henri Poincaré (www.springer.com). The final publication is available at www.springerlink.com.
Observe that similar models without external light source have been extensively analyzed, see for instance [3, 4, 5, 6] and references therein. One of the most important questions considered previously (see, e.g., [4, 5, 6, 7, 8]) are existence and asymptotic stability of stationary states, especially asymptotic stability of thermal equilibrium states. The latter refers to the so-called “return to equilibrium” which typically occurs in models involving one thermal reservoir at fixed temperature weakly coupled to an isolated atom. Note also that, as soon as there are several thermal reservoirs at distinct temperatures, the system does not possess a thermal equilibrium state, but rather a “non-equilibrium stationary state” (NESS) [9, 10]. In our setting we certainly cannot not expect a thermal equilibrium state to exist due to the optical pump.

However, it turns out that the atomic block-diagonal dynamics attains for large times a block-diagonal, quasi-stationary state $\rho$, that is, a stationary state up to small oscillations driven by the optical pump. Observables describing quantum coherences between atomic energy levels are not considered. We show that the structure of this quasi-stationary state is uniquely characterized by a rather intuitive balance condition involving population densities only (Theorem 1.7). This balance condition reads $(A + B)\rho = 0$. The operator $A$ does not depend on the pump and can be interpreted as a matrix of spontaneous transition rates, whereas $B$ is the matrix related to the stimulated transition rates, which are induced by the optical pump. We show that $A$ and $B$ satisfy some relations which are conceptually related to the well-known Einstein relations [11].

Einstein considered an atom interacting with a black body (i.e., broad band) radiation field and derived by phenomenological arguments that the stimulated transition rates $B$ between atomic energy levels are proportional to the radiation density as well as to the spontaneous transition rates $A$. In contrast, we consider the situation of a narrow band (i.e., monochromatic field) driving an atom in interaction with a thermal reservoir. In this case, we find that the effective stimulated processes are proportional to the intensity of the radiation in this model and that, apart from this simple dependency on the pump intensity, the natural decay rates of the atom (i.e., the dynamical properties of the atom as the pump is turned off) uniquely determines the stimulated transition rates $B$. The relevant dynamical parameters turn out to be the decoherence times of the atom, which are purely quantum mechanical objects. In a recent work [12], Berman, Merkli and Sigal show for a similar class of models (without pump term, however) that quantum coherences of the atom decay exponentially fast for well-defined time scales $t_{dec}$ of decoherence. We show that the (pump independent) parameters $t_{dec}$ and the intensity of the pump uniquely define effective stimulated rates.

As a result of our analysis we find that the time-evolution of the population density of the atomic energy levels, i.e., the block-diagonal dynamics, differs from the phenomenological time-evolution usually used in the physics literature (cf. [3]). The block-diagonal evolution is usually described by the so-called Pauli master equation, which is a phenomenological first order autonomous linear differential equation. Our analysis shows that the effective block-diagonal evolution derived from the microscopic model is well described by an integro-differential equation (Theorems 1.6 and 1.8). Nevertheless, the long-time behavior of the solutions of our equation is in generic situations the same as the one predicted by the Pauli master equation.

The model we study here is a basic model for pumping schemes of doped crystals as explained in [13]. One expects for large times a steady emission of photons with fixed frequency $E_{N''} - E_{N'}$, where $E_1 < \cdots < E_{N'} < \cdots < E_{N''} < \cdots < E_N$ are the (ordered) eigenvalues of the atomic Hamiltonian, i.e., the atomic energy levels. Such a steady emission should result from a stronger occupation of the energy atomic level $E_{N''}$ as compared to $E_{N'} < E_{N''}$. This effect, called inversion of population, is a central mechanism to obtain lasing materials (cf. [14]) and we show that the quasi-stationary state $\rho$ characterized by the balance condition may exhibit such an inversion of population.

In another recent work [15], Bach, Merkli, Pedra and Sigal investigate the possibility to control the decoherence time(s) $t_{dec}$ using a different external light source, which does not act as an optical pump but imposes oscillations of the atomic energy levels. In certain situations control of decoherence could thus enhance the inversion of the population. Consequently, measuring the threshold of the pump intensity needed for inversion of population could yield a simple experimental test for control of decoherence.

To our knowledge there is only one framework in which some aspect of laser phenomenology has been rigorously analyzed from first principles, namely for some versions of the Dicke model [16], see [17, 18, 19, 20]. In [17, 19], Hepp and Lieb study an interacting conservative system consisting of a reservoir (a radiation field), a finite number $N$ of two-level atoms, and finitely many quantized oscillators as “radiation-modes”. In [18, 19] the reservoir is absent in their study. In [17, 19] it is shown that this system undergoes a phase transition in the
limit $N \to \infty$ with the appearance of a coherent radiation driven by the reservoir, whereas [18] \cite{19} establish a transition from a normal to a superradiant phase as $N \to \infty$. More than 20 years later, Alli and Sewell use in \cite{20} a dissipative (i.e., non–conservative) version of the Dicke model to get similar results as $N \to \infty$. For more details, we recommend for instance \cite{21} Chap. 11]. In a more recent paper \cite{22} the dissipative model of \cite{20} is proven to be the Markovian approximation of the model considered in \cite{17}. On the other hand, solid state lasers are usually constructed with weakly doped crystals \cite{13}. Consequently, an infinite number of impurities is not fundamental for the inversion of population. Moreover, the phenomenology of lasers as described in physics textbooks is based on three– or four–level atoms \cite{13}, but Dicke–type models are based on two–level atoms which cannot explain the inversion of population at finite number of impurities. The assumption of impurities with three or four levels is also very realistic from the experimental point of view, as most of lasing materials used in the praxis are of this type \cite{13}. We focus on a comprehension of the pumping scheme in lasers which is (i) coherent with physics textbooks and experimental facts and (ii) uses first principles of quantum mechanics only. An important open problem remains however to find a realistic description of a cavity. A recent work in this direction is due to Bruneau and Pillet \cite{23}.

To resume, we focus here on the derivation and analysis of the structure of the quasi–stationary state of the corresponding time–dependent master equation for the impurity, whereas its link to the full microscopic dynamics is established in \cite{1}.

The paper is organized as follows. In Section 2 we introduce the microscopic model. Then, in Section 3 we define the effective master equation and specify its relation – proven in \cite{1} – to the microscopic model. In Section 4.2 we introduce the evolution semigroup of the non–autonomous effective master equation on a suitable enlarged Hilbert space. This leads to a study of an autonomous problem. The spectral analysis of the generator of such evolution semigroup performed in Sections 4.3–4.5 yields a pre–master equation and a balance condition, which characterizes the quasi–stationary state for large times in terms of generalized Einstein relations as explained in Section 5. Numerical simulations show in Section 4.1 an inversion of population with a dynamics far from the one described by the usual Pauli equations. Finally, Section 6 is an appendix consisting of a collection of some notions and results on completely positive (CP) semigroups used in our proofs. Moreover, it provides heuristics of the proof of the result of \cite{1}, some technical proofs as well as a detailed discussion of properties of the operator $B$ appearing in the balance condition, which is in general not a generator of a CP semigroup.

Notation 1.1 (Generic constants)

To simplify our notation, we denote by $C$, $c$, $c'$ any generic positive and finite constant. Note that these constants do not need be the same from one statement to another.

2. The host environment–impurity–light source microscopic model

Laser devices are based on several physical processes. One of them consists of pumping the electron densities of atomic energy levels to obtain a so–called inversion of population. It is achieved in many cases by using an external source of light. This procedure is called here optical pumping. Note however that pumping can also be implemented by means of chemical reactions, electric currents, and other methods. A further step is to use this inversion of population to obtain optical amplification through stimulated emission of photons. These two processes implement a gain medium, which is then put into a resonant optical cavity to obtain a laser, i.e., light amplification by stimulated emission of radiation. Here, we are only interested in the optical pumping performed in the gain medium.

Since the invention of lasers in 1958 by the physicists Schawlow, Townes and Bassov, a significant amount of gain mediums have been found. This includes semiconductors, liquids with dyes, gases such as carbon dioxide, argon, etc., and solids such as doped crystals and glasses. A large part of gain mediums are based on a host material, usually a solid or a liquid, containing impurities or dyes, which can be pumped. A typical example is given by the well–known Nd:YAG laser which uses a crystal doped with neodymium. Inversion of population is obtained in this family of laser devices by optical pumping. Such a gain medium is typically what we have in mind here.
An appropriately chosen impurity is crucial to get inversion of population by pumping and hence a positive optical gain. In particular, the pumping step should involve at least three or four atomic energy levels, as explained in any physics textbook on lasers. For instance, in the case of a monochromatic pump, neodymium impurities in Nd:YAG lasers are well described as four level systems.

Surprisingly, also dissipative processes are important in order to get an efficient optical pumping, in general. For instance, we could analyze the model for impurities in crystals which will be defined below with $\lambda = 0$ (meaning that the impurity does not interact with the surrounding environment) and $\eta \neq 0$ (meaning that the optical pump is turned on). For such a choice of parameters, the dynamics can be explicitly computed, showing that the atomic populations undergo Rabi oscillations and one cannot obtain a (quasi-) stable inversion of population and not even a positive inversion in time average. In solid-state lasers, the interaction with the host material, i.e., the crystal or the glass, provides a dissipative component to the (effective) atomic dynamics. The same should be found in a gain medium constituted of a liquid with a dye to be optically pumped.

The dissipative mechanism can be quite different from one gain medium to another. It can be due to many different types of interactions such as interactions with phonons, electrons in a crystal, etc. Nevertheless, its only important property for the pumping process is to damp Rabi oscillations of pumped atoms and to modify, in interplay with the optical pump, transition rates between atomic energy levels in order to get a (quasi-) stable inversion of population.

Therefore, we can conclude three things about the optical pumping, i.e., the stimulated process to get an inversion of population:

(a) To explain optical pumping, it suffices to use a generic dissipative mechanism. To this end, we consider here an interaction between a $N$–level atom (serving as a small system in a host environment) and a fermionic thermal (macroscopic) reservoir whose mathematical setting is standard, see, e.g., [24, 25]. This choice excludes interactions between impurities and bosonic particles like phonons. This could also be implemented, but for simplicity we refrain from considering it as it qualitatively leads to similar results. Note, however, that more physical models for host environments could be useful to get more precise quantitative results.

(b) To obtain an efficient optical pumping, the strengths $\tilde{\eta}, \tilde{\lambda} > 0$ of respectively the optical pump and the interaction with the host environment have to be of the same order, i.e., $\tilde{\eta} \sim \tilde{\lambda}$, see Section 2.7. Indeed, if $\tilde{\eta} \gg \tilde{\lambda}$, then Rabi oscillations with frequency of order $\tilde{\eta}$ are the dominating processes governing the dynamics of populations. This oscillations are then generally damped in a time–scale of order $\tilde{\lambda}^{-1}$ not depending much on the intensity of the pump. In the opposite situation, i.e., when $\tilde{\eta} \ll \tilde{\lambda}$, no inversion of population can appear as the system relaxes in this case to a state near the ground state (or a thermal equilibrium state) of the atom and all the energy provided by the (weak) optical pump is lost into the (very large) host environment.

(c) Finally, the inversion of population described in physics textbooks makes sense if the “$N$–level atom” picture stays valid. In other words, the interaction with the host environment must be a small perturbation of the Hamiltonian representing the $N$–level atom. By (b), the optical pump should also be seen as a perturbation of the $N$–level atom. In view of this observation it is appropriate to use (Kato’s) perturbation theory [26] of (discrete) eigenvalues of the $N$–level atom.

The host environment discussed in (a), i.e., the fermionic thermal reservoir, is described in details in Section 2.1. We then define the impurity and the external monochromatic light source in Sections 2.2–2.3, respectively. The coupled full system is set up in Sections 2.4–2.7.

### 2.1 The host environment as a fermionic thermal reservoir

Let $h_1 := L^2(\mathbb{R}^3, \mathbb{C})$ be the separable Hilbert space representing the one–particle space of the host environment (reservoir). The one–particle Hamiltonian $h_1$ is then defined by using a dispersion relation. Indeed, we choose some measurable, rotationally invariant function $E : \mathbb{R}^3 \to \mathbb{R}$, i.e., $E(p) = E(|p|)$, and define the multiplication operator $h_1 \equiv h_1(E)$ by $f(p) \mapsto E(p)f(p)$ on $h_1$. Physically, $E(p)$ represents the energy of one particle with momentum $p$ within the reservoir.
In the case where the host material of the gain medium is a crystal, observe that $L^2$ spaces on Brillouin zones as one–particle spaces are more realistic than $L^2(\mathbb{R}^3, \mathbb{C})$. As explained above (cf. Observation (a)), the results obtained would qualitatively be the same. For instance, the rotation symmetry assumed above for the dispersion relation $E$ is not essential to the analysis performed below. However, to satisfy the assumptions of Theorem 3.3, we require that $E$ behaves (at least near $p = 0$) like $|p|$ up to some diffeomorphism. For instance, the dispersion relation $E(p) \equiv |p|^2$ is allowed as $|p|$ and $|p|^2$ are clearly the same function up a diffeomorphism of $\mathbb{R}^3 \setminus \{0\}$.

The field algebra $V_R$ of the reservoir is the CAR $C^*$–algebra generated by the annihilation and creation operators $a(f), a^+(f) := a(f)^*, f \in \mathfrak{h}_1$, acting on the antisymmetric Fock space $\mathcal{F}_-(\mathfrak{h}_1)$ and satisfying the canonical anti–commutation relations (CAR):

$$a(f_1)a^+(f_2) + a^+(f_2)a(f_1) = \langle f_1, f_2 \rangle, \quad a(f_1)a(f_2) + a(f_2)a(f_1) = 0$$

for any $f_1, f_2 \in \mathfrak{h}_1$. $\langle \cdot, \cdot \rangle$ denotes the scalar product in $\mathfrak{h}_1$.

The (unperturbed) dynamics of the reservoir is given by the family $\{\tau_t^R\}_{t \in \mathbb{R}}$ of Bogoliubov automorphisms on the algebra $V_R$ uniquely defined by the condition:

$$\tau_t^R(a(f)) = a(e^{i\hbar t} f), \quad f \in \mathfrak{h}_1, \quad t \in \mathbb{R}.$$  

(2.1)

Physically, this means that the fermionic particles of the reservoir do not interact with each other, i.e., they form an ideal Fermi gas. The group $\tau^R := \{\tau_t^R\}_{t \in \mathbb{R}}$ of automorphisms is strongly continuous and hence, $(V_R, \tau^R)$ is a $C^*$–dynamical system. We denote its generator by $\delta_R$.

Note that generators of $C^*$–dynamical systems are symmetric derivations. This means that the domain $\text{Dom}(\delta_R)$ of the generator $\delta_R$ is a dense sub--$*$–algebra of $V_R$ and, for all $A, B \in \text{Dom}(\delta_R)$,

$$\delta_R(A^*) = \delta_R(A^*), \quad \delta_R(AB) = \delta_R(A)B + A\delta_R(B).$$

Thermal equilibrium states of the reservoir are defined through the bounded positive operators

$$d_R := \frac{1}{1 + e^{\beta \hbar t}}$$

acting on $\mathfrak{h}_1$ for all inverse temperatures $\beta \in (0, \infty)$. Indeed, the so–called symbol $d_R$ uniquely defines a (faithful) quasi–free state

$$\omega_{d_R} := \omega_{d_R}$$

on the fermion algebra $V_R$ by the conditions $\omega_{d_R}(1_R) = 1$ and

$$\omega_{d_R}(a^+(f_1) \ldots a^+(f_m)a(g_1) \ldots a(g_n)) = \delta_{m,n} \det([\langle gj, dfk \rangle])_{j,k}$$

for all $\{f_j\}_{j=1}^m, \{g_j\}_{j=1}^n \subset \mathfrak{h}_1$. The positive, normalized linear functional $\omega_R$ is the unique $\beta$–KMS state of the $C^*$–dynamical system $(V_R, \tau^R)$ and is called the thermal equilibrium state of the reservoir at inverse temperature $\beta \in (0, \infty)$.

This definition of thermal states is rather abstract, but it can physically be motivated as follows: Confining the particles within a box of side length $L$ corresponds to the replacement of the momentum space $\mathbb{R}^3$ by $\frac{2\pi}{L} \mathbb{Z}^3$, i.e., $L^2(\mathbb{R}^3)$ by $L^2(\frac{2\pi}{L} \mathbb{Z}^3)$. In particular, the spectrum of $\mathfrak{h}_1$ (and hence of its fermionic second quantization $d\Gamma_-(\mathfrak{h}_1)$) becomes purely discrete. Additionally, the operators $e^{-\beta d\Gamma_-(\mathfrak{h}_1)}$ are in this case trace–class for all side lengths $L$. Hence, we can define Gibbs states

$$\vartheta^{(L)}_R(\cdot) := \frac{\text{Tr}(e^{-\beta d\Gamma_-(\mathfrak{h}_1)} \cdot \Omega^{(L)}(\cdot))}{\text{Tr}(e^{-\beta d\Gamma_-(\mathfrak{h}_1)})},$$

which have the thermal equilibrium state $\omega_R$ as unique weak–$*$ limit\(^1\) for $L \to \infty$. This follows, for instance, from the results of [23] Chapters 5.2 and 5.3 on KMS states.

\(^1\) This refers to the weak$^*$–topology on the locally convex real space $V_R^*$, the dual space of the separable Banach space $V_R$. 
2.2 The impurity as a $N$–level atom

The impurity (atom) is modeled by a finite quantum system, i.e., its observables are the self–adjoint elements of the finite dimensional $C^*$–algebra $\mathcal{B}(\mathbb{C}^d)$ of all linear operators on $\mathbb{C}^d$ for $d \in \mathbb{N}$ ($d \geq 2$).

In the sequel it is convenient to define left and right multiplication operators on $\mathcal{B}(\mathbb{C}^d)$: For any $A \in \mathcal{B}(\mathbb{C}^d)$ we define the linear operators $A_{\downarrow}$ and $A_{\downarrow}$ acting on $\mathcal{B}(\mathbb{C}^d)$ by

$$B \mapsto A_{\downarrow} B := AB \quad \text{and} \quad B \mapsto A_{\downarrow} B := BA \ .$$

The Hamiltonian of the atom is an arbitrary observable $H_{\text{at}} = H_{\text{at}}^{\ast} \in \mathcal{B}(\mathbb{C}^d)$ representing its total energy. We denote its eigenvalues and corresponding eigenspaces by $E_k \in \mathbb{R}$ and $\mathcal{H}_k \subset \mathbb{C}^d$ for $k \in \{1, \ldots, N\}$ ($N \geq 2$), respectively. $E_k$ is chosen such that $E_j < E_k$ whenever $j < k$. In other words, $E_k$ is the energy of the $k$th atomic level and vectors of $\mathcal{H}_k$ describe the sub–band structure of the corresponding energy level. The dimension $n_k$ of the eigenspace $\mathcal{H}_k$ is the degeneracy of the $k$th atomic level.

As usual, the Hamiltonian $H_{\text{at}}$ defines a free atomic dynamics, i.e., a continuous one–parameter group of automorphisms $\tau^t := \{ \tau^t_\cdot \}_{t \in \mathbb{R}}$ of the $C^*$–algebra $\mathcal{B}(\mathbb{C}^d)$ defined by

$$\tau^t_\cdot (A) := e^{it H_{\text{at}} A} e^{-it H_{\text{at}}} , \quad A \in \mathcal{B}(\mathbb{C}^d) ,$$

for all $t \in \mathbb{R}$.

Thermal equilibrium states of the free atom are Gibbs states $\omega_{\text{at}}$ given by the density matrix

$$\rho_g := \frac{e^{-\beta H_{\text{at}}}}{\operatorname{Tr}_{\mathbb{C}^d} (e^{-\beta H_{\text{at}}})}$$

for any inverse temperature $\beta \in (0, \infty)$.

In presence of the optical pump and the host environment (the thermal reservoir), the state of the atom is generally far from the Gibbs state $\omega_{\text{at}}$. We thus consider arbitrary atomic states $\omega_{\text{at}}$. For any state $\omega_{\text{at}}$ on $\mathcal{B}(\mathbb{C}^d)$, there is a unique trace–one positive operator $\rho_{\text{at}}$ on $\mathbb{C}^d$, the so–called density matrix of $\omega_{\text{at}}$, such that

$$\omega_{\text{at}}(A) = \operatorname{Tr}_{\mathbb{C}^d} (\rho_{\text{at}} A) \ , \quad A \in \mathcal{B}(\mathbb{C}^d) .$$

Note that any state $\omega_{\text{at}}$ on $\mathcal{B}(\mathbb{C}^d)$ can be represented as a vector state via its GNS representation $(\mathcal{H}_{\text{at}}, \pi_{\text{at}}, \Omega_{\text{at}})$, see, e.g., [24, Theorem 2.3.16]. If $\omega_{\text{at}}$ is faithful then $(\mathcal{H}_{\text{at}}, \pi_{\text{at}}, \Omega_{\text{at}})$ is explicitly given as follows. The Hilbert space $\mathcal{H}_{\text{at}}$ corresponds to the linear space $\mathcal{B}(\mathbb{C}^d)$ endowed with the Hilbert–Schmidt scalar product

$$\langle A, B \rangle_{\text{at}} := \operatorname{Tr}_{\mathbb{C}^d} (A^\ast B) \ , \quad A, B \in \mathcal{B}(\mathbb{C}^d) .$$

The representation $\pi_{\text{at}}$ is the left multiplication explained above in (2.3), i.e.,

$$\pi_{\text{at}} (A) = A_{\downarrow} , \quad A \in \mathcal{B}(\mathbb{C}^d) .$$

The cyclic vector of the GNS representation of $\omega_{\text{at}}$ is defined by using the density matrix $\rho_{\text{at}} \in \mathcal{B}(\mathbb{C}^d)$ of $\omega_{\text{at}}$ as

$$\Omega_{\text{at}} := \rho_{\text{at}}^{1/2} \in \mathcal{H}_{\text{at}} .$$

Using the cyclicity of the trace we obtain that

$$\omega_{\text{at}} (A) = \langle \Omega_{\text{at}}, A_{\downarrow} \Omega_{\text{at}} \rangle_{\text{at}} , \quad A \in \mathcal{B}(\mathbb{C}^d) .$$

This GNS representation $(\mathcal{H}_{\text{at}}, \pi_{\text{at}}, \Omega_{\text{at}})$ is known in the literature as the standard representation of the state $\omega_{\text{at}}$. See [27, Section 5.4].

The dynamics given by the continuous one–parameter group $\tau^t$ of automorphisms of the $C^*$–algebra $\mathcal{B}(\mathbb{C}^d)$ defined by (2.4) can be represented in the Schrödinger picture of Quantum Mechanics through the so–called (standard) Liouvillian operator

$$L_{\text{at}} := H_{\text{at}} - H_{\text{at}}^{\ast} = [H_{\text{at}}, \cdot] = L_{\text{at}}^{\ast}$$

acting on the Hilbert space $\mathcal{H}_{\text{at}}$. Indeed, it is easy to check that:
Lemma 2.1 (Schrödinger picture of $\tau_{\text{at}}$)
For all $t \in \mathbb{R}$,
\[ \omega_{\text{at}}(\tau_{\text{at}}^t(A)) = \langle \Omega_{\text{at}}(t), \pi_{\text{at}}(A) \Omega_{\text{at}}(t) \rangle_{\text{at}}, \quad A \in \mathcal{B}(\mathbb{C}^d), \]
where $\Omega_{\text{at}}(t) := e^{-it\mathcal{L}_{\text{at}}}\Omega_{\text{at}}$.

Finally, for $k \in \{1, \ldots, N\}$, note that the population of the $k$th atomic level in the state $\omega_{\text{at}}$ is defined by the expectation
\[ P_k(\rho_{\text{at}}) := \omega_{\text{at}}(1[H_{\text{at}} = E_k]) = \text{Tr}_{\mathbb{C}^d}(1[H_{\text{at}} = E_k]\rho_{\text{at}}) \geq 0, \quad (2.9) \]
where $1[H_{\text{at}} = E_k] \in \mathcal{B}(\mathbb{C}^d)$ is the orthogonal projection onto the eigenspace $\mathcal{H}_k$. If $\omega_{\text{at}} = \rho_{\text{at}}$ is the Gibbs state of the atom and the atomic energy levels are non–degenerated or the inverse temperature $\beta \in (0, \infty)$ is large enough then
\[ \forall j, k \in \{1, \ldots, N\}, \ j < k : \quad P_j(\rho_\eta) > P_k(\rho_\eta). \]
In contrast, we say that a state $\omega$ or a density matrix $\rho$ shows inversion of population if there are $j, k \in \{1, \ldots, N\}$ such that $j < k$, i.e., $E_j < E_k$, and $P_j(\rho) < P_k(\rho)$. In other words, inversion of population requires a higher energy level more populated than a lower one. Of course, this phenomenon can, in general, only appear in a state out of equilibrium and one usually uses external light sources to artificially pump electrons from a low energy level of the atom to a higher one.

2.3 The external monochromatic light source as a classical optical pump

The optical pump, i.e., the monochromatic photon field interacting with the atom, is described by the following time–periodic perturbation of the atomic Hamiltonian $H_{\text{at}}$:
\[ \eta \cos(\varpi t) H_p, \quad \varpi := E_N - E_1 > 0, \quad t \in \mathbb{R}. \quad (2.10) \]
Recall that $E_1 < \cdots < E_N$ denote the $N$ eigenvalues of $H_{\text{at}} = H_{\text{at}}^* \in \mathcal{B}(\mathbb{C}^d)$. Here,
\[ H_p := h_p + h_p^* \in \mathcal{B}(\mathbb{C}^d) \quad (2.11) \]
for some $h_p \in \mathcal{B}(\mathbb{C}^d)$ satisfying
\[ \ker (h_p) \perp \mathcal{H}_1 := \text{ran} (1[H_{\text{at}} = E_1]), \quad (2.12) \]
\[ \text{ran} (h_p) \subseteq \mathcal{H}_N := \text{ran} (1[H_{\text{at}} = E_N]) \quad (2.13). \]
In other words, the optical pump produces only transitions between the lowest and the highest atomic levels 1 and $N$, as described in standard textbooks on the physics of lasers.

From the physical point of view, the time–dependent optical pump may be regarded as a partial classical limit of a closed (autonomous) physical system involving a quantized pump modeled by a quantum harmonic oscillator. The corresponding initial state for this quantized pump should be chosen as being a coherent state. See, e.g., [28, 29].

Remark 2.1 (Non monochromatic light sources as classical optical pumps)
Results of this paper can easily be extended to non–monochromatic light sources as classical optical pumps. This case corresponds here to replace the cosine in (2.10) by some time–periodic and continuous function. However, in order to keep technical aspects as simple as possible, we refrain from considering this more general case.

2.4 The uncoupled reservoir–atom system

Define the $C^*$–algebra $\mathcal{V} := \mathcal{B}(\mathbb{C}^d) \otimes \mathcal{V}_R$. As both $C^*$–algebras $\mathcal{B}(\mathbb{C}^d)$ and $\mathcal{V}_R$ are already realized as algebras of bounded operators on Hilbert spaces and since $\mathcal{B}(\mathbb{C}^d)$ is finite dimensional, we do not have to specify the meaning of the tensor product. Observables of the reservoir–atom system are self–adjoint elements of $\mathcal{V}$. Its
free dynamics is described by the strongly continuous one–parameter group \( \tau := \{ \tau_t \}_{t \in \mathbb{R}} \) of automorphisms of \( \mathcal{V} \) defined by

\[
\tau_t := \tau^\tau_t \otimes \tau^R_t, \quad t \in \mathbb{R}.
\]  

(2.14)

This tensor product is well–defined and unique because the atomic algebra \( \mathcal{B}(\mathbb{C}^d) \) is finite dimensional. The generator of the free dynamics defined by \( \tau \) is a symmetric derivation, denoted by \( \delta \), which acts on a dense sub–\( \ast \)–algebra \( \text{Dom}(\delta) \) of \( \mathcal{V} \).

Let \( \omega_{\text{at}} \) be any initial (not necessarily Gibbs) state of the atom and define the initial state of the atom–reservoir system by

\[
\omega_0 := \omega_{\text{at}} \otimes \omega_R.
\]  

(2.15)

Again, the latter is well–defined and unique, by finite dimensionality of \( \mathcal{B}(\mathbb{C}^d) \). If \( \omega^\text{at} = \rho_{\text{at}} \) is the Gibbs state then \( \omega_0 \) is clearly a \( (\beta, \tau) \)–KMS state. Observe also that \( \rho_{\text{at}} \) is a faithful state and we assume without loss of generality that \( \omega^\text{at} \) is also a faithful state. Indeed, the set of faithful states is dense in the set of all states of the atom. Since the quasi–free state \( \omega_R \) of the reservoir is also faithful, this property carries over to the initial state \( \omega_0 \) of the composite system.

**Remark 2.2 (Coupled initial state of the atom–reservoir system)**

Considering impurities interacting with the host environment long before the pump is turned on, the initial state of the atom–reservoir system should, in principle, not be a product state as in (2.15). Instead, it should be a thermal equilibrium state of the coupled atom–reservoir system. However, in rather generic situations it can be shown that at small atom–reservoir couplings this thermal state is near the product state \( \omega_0 \) with \( \omega^\text{at} = \rho_{\text{at}} \) and the results would be the same up to sub–leading corrections. As the KMS states of the model considered here are unique (KMS states of bounded perturbations of a free fermion gas are unique), the latter follows from standard results on the stability of KMS states, see for instance [25, Section 5.4.1]. Indeed, we can even treat, by the same methods, any initial state of the composite system as soon as its relative entropy with respect to the product state (2.15) is finite. This feature is verified for the equilibrium thermal state of the composite system at weak coupling. In order to keep technical aspects as simple as possible, we will not consider this case.

### 2.5 The atom–reservoir interaction

The interaction between the atom and the fermionic thermal reservoir involves the so–called fermionic field operators defined, for all \( f \in \mathfrak{h}_1 \), by

\[
\Phi(f) := \frac{1}{\sqrt{2}}(a^+(f) + a(f)) = \Phi(f)^* \in \mathcal{B}(\mathcal{F}_-(\mathfrak{h}_1)).
\]

Choose now a finite collection \( \{ Q_{\ell} \}_{\ell=1}^m \subset \mathcal{B}(\mathbb{C}^d) \) of self–adjoint operators and an orthonormal (finite) system \( \{ f_{\ell} \}_{\ell=1}^m \subset \mathfrak{h}_1 \). Then, the atom–reservoir interaction is implemented by the bounded symmetric derivation

\[
\delta_{\text{at}, R} := i \sum_{\ell=1}^m [Q_{\ell} \otimes \Phi(f_{\ell}), \cdot] .
\]

Note that the orthonormality of the family \( \{ f_{\ell} \}_{\ell=1}^m \) does not inflict loss of generality. Indeed, for an arbitrary finite set \( \{ \tilde{Q}_{\ell} \}_{\ell=1}^m \subset \mathcal{B}(\mathbb{C}^d) \) of self–adjoint operators and (possibly not orthonormal) family \( \{ \tilde{f}_{\ell} \}_{\ell=1}^m \subset \mathfrak{h}_1 \), there are \( m \in \mathbb{N} \), a finite collection \( \{ Q_{\ell} \}_{\ell=1}^m \subset \mathcal{B}(\mathbb{C}^d) \) of self–adjoint operators and an orthonormal system \( \{ f_{\ell} \}_{\ell=1}^m \subset \mathfrak{h}_1 \) such that

\[
\sum_{\ell=1}^m Q_{\ell} \otimes \Phi(f_{\ell}) = \sum_{\ell=1}^m Q_{\ell} \otimes \Phi(f_{\ell}).
\]

As we seek to maintain mathematical rigor while keeping technical aspects as simple as possible, we assume some technically useful conditions on the family \( \{ f_{\ell} \}_{\ell=1}^m \subset \mathfrak{h}_1 \) guaranteeing the assumptions of Theorem 3.3 to be satisfied. Note that these conditions will by no means restrict the range of our analysis to rather physically meaningless submodels. First, \( \{ f_{\ell}(p) = \)}
f_\ell(|p|) for all \(p \in \mathbb{R}^3\) and \(\ell \in \{1, \ldots, m\}\) with \(f_\ell : \mathbb{R}_0^+ \rightarrow \mathbb{C}\). The rotational invariance of \(f_\ell\) for all \(\ell \in \{1, \ldots, m\}\) is only assumed for technical simplicity as more general choices of such functions would yield similar results, at least qualitatively. The second, rather technical assumption is an analyticity condition which is only required to prove Theorem 3.3. It is given here for completeness, but it can clearly be omitted as no proof of this paper uses it. This analyticity condition reads as follows: For all \(\ell \in \{1, \ldots, m\}\), the complex valued functions \(g_\ell\) and \(g^\#_\ell\) respectively defined by

\[
\forall x \in \mathbb{R}: \quad g_\ell(x) := |x| (1 + e^{-\beta x})^{-1/2} \begin{cases} f_\ell(x) & x \geq 0, \\ f_\ell(-x) & x < 0, \end{cases} \quad (2.16)
\]

and \(g^\#_\ell(x) := ig\ell(-x)\) on \(\mathbb{R}\) have an analytic continuation to the strip \(\mathbb{R} + i(-C, C)\), and satisfy

\[
\sup_{\vartheta \in (-C, C)} \left\{ \int_{\mathbb{R}} (|g_\ell(x + i\vartheta)| + |g^\#_\ell(x + i\vartheta)|)^2 dx \right\} < \infty
\]

for all \(\ell \in \{1, \ldots, m\}\). For instance, to satisfy these conditions one may choose for any \(\ell \in \{1, \ldots, m\}\), the function \(f_\ell\) as linear combinations of terms of the form \(|x|^{2k-1} \exp(-c x^2)\) with \(k \in \mathbb{N}_0\).

### 2.6 Dynamics of the coupled atom–reservoir–pump system

The full dynamics of the system involves the classical pump described in Section 2.3 which is implemented as a periodic perturbation of the dynamics of the atom–reservoir system described in Section 2.4. Indeed, let

\[
\delta_{\text{at,p}} := i[H_p \otimes 1_\mathcal{R}, \cdot]
\]

and \(\eta \in \mathbb{R}\). The coupled atom–reservoir–pump dynamics is then generated by the time–dependent symmetric derivation

\[
\delta^{(\lambda,\eta)}(t) := \delta + \eta \cos(\pi t)\delta_{\text{at,p}} + \lambda\delta_{\text{at,R}}, \quad t \in \mathbb{R}. \quad (2.17)
\]

Here, \(\lambda, \eta \in \mathbb{R}\) are the atom–reservoir and atom–pump coupling constants, respectively.

Observe that \(\delta^{(\lambda,\eta)}(t)\) acts on a dense sub–\(s\)–algebra \(\text{Dom}(\delta_t) = \text{Dom}(\delta)\) of \(\mathcal{V}\) which does not depend on \(t \in \mathbb{R}\). Indeed, \(\delta_{\text{at,R}}\) and \(\delta_{\text{at,p}}\) are bounded symmetric derivations and \(\delta^{(\lambda,\eta)}\) is the generator of a strongly continuous one–parameter group of automorphisms of \(\mathcal{V}\). As the map

\[
t \mapsto \delta^{(\lambda,\eta)}(t) - \delta^{(\lambda,\eta)}(0)
\]

is norm–continuous, \(\delta^{(\lambda,\eta)}(t)\) generates a strongly continuous two–parameter family \(\{\tau^{(\lambda,\eta)}_{t,s}\}_{t \geq s}\) of automorphisms of \(\mathcal{V}\) corresponding to the non–autonomous dynamics of the (coupled) atom–reservoir–pump system. The operator \(\tau^{(\lambda,\eta)}_{t,s}\) can even be explicitly constructed as a Dyson series because the operator \(\eta \cos(\pi t)\delta_{\text{at,p}}\) is bounded and \(\cos\) is a smooth function.

The time–evolution of the state of the full system is then given by

\[
\omega_t := \omega_0 \circ \tau^{(\lambda,\eta)}_{t,0} = (\omega_{\text{at}} \otimes \omega_{\mathcal{R}}) \circ \tau^{(\lambda,\eta)}_{t,0}, \quad t \in \mathbb{R}_0^+.
\]

The reduction of this state onto the atomic degrees of freedom yields a time–dependent atomic state defined by

\[
\omega_{\text{at}}(t)(A) := \omega_t(A \otimes 1_\mathcal{R}), \quad A \in \mathcal{B}(\mathcal{C}^d), \quad (2.18)
\]

for all \(t \in \mathbb{R}_0^+\).

### 2.7 Moderate optical pump and atom–reservoir interaction

As explained at the beginning of Section 2 (cf. Observation (c)), we are interested in the regime where \(|\lambda|, |\eta| << 1\). In other words, we take the atom–reservoir and atom–pump interactions as being small, but non–vanishing, perturbations of the free dynamics generated by the symmetric derivation \(\delta\). Moreover, we assume that the pump is moderate with respect to the atom–reservoir interaction in the following sense:
Assumption 1 (Moderate optical pump)

For any \( \lambda \in \mathbb{R} \) and some fixed constant \( C \in (0, \infty) \): \(|\eta| \leq C \lambda^2\).

Actually, in all our proofs, it would suffice to impose \(|\eta| \leq C|\lambda|\) for some sufficiently small constant \( C \in (0, \infty) \). However, as it will be shown below (cf. Remark 4.3), the contribution of the pump to the final atomic state is of order \( \eta^2/\lambda^4 \) whereas the contribution of the interaction with the reservoir is of order one (in the parameter \( \eta^2/\lambda^3 \)). Thus, imposing \(|\eta| \sim \lambda^2\) means physically that both the pump and the reservoir contribute in an essential way to the final state of the atom. The (relative) strengths \( \tilde{\eta}, \tilde{\lambda} > 0 \) of respectively the optical pump and the interaction with the host environment turn out to be equal to \( \tilde{\eta} := \eta^2/\lambda^2 \) and \( \tilde{\lambda} := \lambda^2 \). In particular, Assumption 1 means that \( \tilde{\eta} \sim \tilde{\lambda} \). See Observation (b) above. Consequently, we say in this context that the pump is weak whenever \(|\eta| < \lambda^2\), i.e., \( \tilde{\eta} < \tilde{\lambda} \).

In the opposite situation when for small \(|\eta|, |\lambda|, (\eta, \lambda) \in \mathbb{R}^2\), one has \(|\eta| >> \lambda^2\), i.e., \( \tilde{\eta} >> \tilde{\lambda} \), Rabi oscillations are the dominant processes driving the populations of the atomic energy levels. Indeed, a similar version of Corollary 4.5 is still valid in the strong pump regime \(|\eta| >> \lambda^2\) if \(|\eta| >> |\lambda|\). Using this result one can show that, in general, Rabi oscillations dominate the dynamics of populations at time–scales comparable to \(|\eta|^{-1}\) and that there is a global relaxation of populations at time–scale \( \lambda^{-2} \) not depending much on the pump intensity. By this last property we could also call the regime \(|\eta| >> \lambda^2\) saturated pumping. However, we are rather interested in the situation where pump and natural relaxations of the atom compete in a non trivial way with each other and the evolution of the full system is well described by some relaxing dynamics driving the atom to a state with a persisting inversion of population. See again the discussions at the beginning of Section 2 in particular Observation (b).

3. The effective atomic master equation

The aim of this paper is to analyze the atomic dynamics resulting from the restriction on \( \mathcal{B}(\mathbb{C}^d) \) of the full dynamics generated by the symmetric derivation \( \delta_t^{(\lambda, \eta)} \). See Equation (2.17). This corresponds to the family \( \{\omega_{at} (t)\} \in \mathbb{R}_+^+ \) of states defined by (2.18) or, equivalently, to the family \( \{\rho_{at} (t)\} \in \mathbb{R}_+^+ \) of density matrices. More precisely, we are interested in the time–behavior of observables related to atomic levels only, and not to correlations between different levels. This amounts to study the orthogonal projection \( \mathcal{P}_\mathcal{D} (\rho_{at} (t)) \) of the atomic density matrix \( \rho_{at} (t) \) on the subspace

\[
\mathcal{D} \equiv \mathcal{D}(H_{at}) := \mathcal{B}(\mathcal{H}_1) \oplus \cdots \oplus \mathcal{B}(\mathcal{H}_N) \subset \mathcal{H}_{at}
\]

of block–diagonal matrices. In other words, we analyze the density matrix

\[
P_\mathcal{D} (\rho_{at} (t)) = \sum_{k=1}^{N} \mathbf{1} [H_{at} = E_k] \rho_{at} (t) \mathbf{1} [H_{at} = E_k]
\]

for any \( t \in \mathbb{R}_+^+ \).

As proven in [1], the density matrix \( \rho_{at} (t) \) can be well approximated, uniformly in time, on the subspace \( \mathcal{D} \) by the solution of an effective non–autonomous initial value problem in \( \mathcal{B}(\mathbb{C}^d) \), the effective atomic master (or Kossakowski–Lindblad) equation described in Section 3.3. The corresponding time evolution is generated by the time–dependent Lindbladian \( \mathcal{L}_t^{(\lambda, \eta)} \) defined in Section 3.1 and its derivation from the microscopic model is discussed in [6.1]. In Section 3.2 we impose a condition, introduced in [30] Theorem 2] in the context of relaxing CP semigroups, on the dissipative part of this generator which is a non–commutative version of the irreducibility of classical Markov chains and ensures the existence of the (quasi–) stationary state for populations at large times.

3.1 The atomic time–dependent Lindbladian

Lindblad operators (or Lindbladians) are standard objects used to describe (generally dissipative) dynamics compatible with some phenomenologically reasonable prescriptions like complete positivity. As explained in [3], the typical example of application of such operators is related to the time–evolution of composite systems
made of a small (open) quantum system interacting with a macroscopic one (a reservoir). In this context, the
dynamics restricted to the small system is in many situations well–described by a semigroup \( \{ e^{L t} \}_{t \geq 0} \) which
is trace–preserving and completely positive, see Section 6.3. Generators \( L \) of such completely positive (CP) semigroups are called Lindblad operators or Lindbladians.

A first rigorous result in this direction is due to Davies \cite{31,32,33} in certain scaling limit, the so–called
weak coupling limit for similar interacting systems as ours with \( \eta = 0 \), i.e., without the optical pump or any
other time dependent term. See also \cite{27} and references therein. For more details on CP semigroups, we also
recommend Section 6.3. Note however that, in contrast to Davies’ approach, we never take the limit \( \lambda \to 0 \).
It suffices in our analysis to have a sufficiently small coupling constant due to the presence of the reservoir, and the
so–called weak coupling limit for similar interacting systems as ours with \( \eta \sim \lambda^2 \), by the moderate pump assumption) and the precise meaning of a weak–coupling limit is not clear from the
beginning. The dynamics we deal with is moreover non–autonomous.

The physical system considered here yields a non–autonomous effective time–evolution with a time–dependent,
genetically non anti–self–adjoint generator \( L \) of the form
\[
L(\rho) = -iL_{\text{at}}(\rho) - i[Q,\rho] + \lambda^2 \mathcal{L}_R(\rho), \quad \rho \in \mathcal{H}_{\text{at}}.
\]
See Section 6.1 for an heuristic derivation of this time–dependent Lindbladian from the microscopic model.

The first term defining \( L_{\text{at}}(\rho) \) is the Lindbladian of the free atomic dynamics. It is the anti–self–adjoint
operator
\[
L_{\text{at}}(\rho) := -iL_{\text{at}}(\rho) = -i[H_{\text{at}}, \rho] = -\mathcal{L}_{\text{at}}^*(\rho), \quad \rho \in \mathcal{H}_{\text{at}}.
\]
See Lemma 2.1. The second term of (3.3) encodes the influence of the optical pump. It is defined via the
Lindbladian
\[
L_{\text{p}}(\rho) := -i[H_{\text{p}}, \rho] = -\mathcal{L}_{\text{p}}^*(\rho), \quad \rho \in \mathcal{H}_{\text{at}}.
\]
The third term includes a dissipative part \( L_d \) corresponding to the non–unitary character of the
effective dynamics and so, \( \mathcal{L}_R \in \mathcal{B}(\mathcal{H}_{\text{at}}) \) is not anti–self–adjoint. In fact, the Lindbladian \( \mathcal{L}_R \) is related to the
second–order perturbation term coming from the atom–reservoir (electron field–impurity) interaction in a
suitable representation and equals
\[
\mathcal{L}_R(\rho) := -i[H_{\text{Lamb}}, \rho] + \mathcal{L}_d(\rho), \quad \rho \in \mathcal{H}_{\text{at}}.
\]
In order to define the so–called atomic Lamb shift \( H_{\text{Lamb}} \), which encodes the shift of the atomic energy levels
due to the presence of the reservoir, and the effective atomic dissipation \( L_d \), some preliminary definitions are
necessary: We denote the spectrum of any operator \( A \) by \( \sigma(A) \), its positive part by \( \sigma^+(A) := \sigma(A) \cap \mathbb{R}_+^+ \), and
define the sets
\[
\lambda_{\epsilon} := \{(j, k) : E_j - E_k = \epsilon \} \subset \{1, 2, \ldots, N\} \times \{1, 2, \ldots, N\}
\]
for each eigenvalue
\[
\epsilon \in \sigma(L_{\text{at}}) = \sigma(H_{\text{at}, \cdot}) = \{E_j - E_k : j, k \in \{1, 2, \ldots, N\}\}.
\]
Let \( \{V_{j,k}^{(\ell)}\}_{j,k,\ell} \subset \mathcal{B}(\mathbb{C}^d) \) be the family of operators defined by
\[
V_{j,k}^{(\ell)} := 1[H_{\text{at}}, \rho] Q_{\ell} 1[H_{\text{at}}, \rho] = V_{j,k}^{(\ell)}.
\]
for \( j, k \in \{1, 2, \ldots, N\} \) and \( \ell \in \{1, 2, \ldots, m\} \), and let \( \{f_{\ell}^{(\beta)}\}_{m=1}^M \) be the family of functions \( \mathbb{R} \to \mathbb{R}_0^+ \) defined by
\[
f_{\ell}^{(\beta)}(x) := 4 \pi \frac{|x f_{\ell}(|x|)|^2}{1 + e^{-\beta x}} = 4 \pi |g_{\ell}(x)|^2.
\]
at any fixed inverse temperature \( \beta \in (0, \infty) \) of the fermionic reservoir, see \cite{2.16}.

Remark 3.1 (Self–adjointness of the space spanned by \( \{V_{j,k}^{(\ell)}\}_{j,k,\ell} \))
Since \( Q_{\ell} = Q_{\ell}^* \), one has \( (V_{j,k}^{(\ell)})^* = V_{k,j}^{(\ell)} \) for any \( j, k \in \{1, 2, \ldots, N\} \) and \( \ell \in \{1, 2, \ldots, m\} \), and obviously,
\[
\text{span}\{V_{j,k}^{(\ell)}\}_{j,k,\ell} = \text{span}\{(V_{j,k}^{(\ell)})^*\}_{j,k,\ell} \subset \mathcal{H}_{\text{at}}.
\]
This fact is important when using Theorem 6.10.
Then, the atomic Lamb shift $H_{\text{Lamb}} \in \mathcal{B}(\mathbb{C}^d)$ is the self-adjoint operator defined by

$$H_{\text{Lamb}} := -\frac{1}{2} \sum_{c \in \sigma([H_{\text{at}}, \cdot]) \setminus \{0\}} \sum_{(j,k) \in \mathcal{L}_t} \sum_{\ell=1}^m d_{j,k}^{(\ell)} V_{j,k}^{(\ell)}$$

with the real coefficients

$$d_{j,k}^{(\ell)} := \mathcal{P}(f_{\ell}^{(\beta)} (\cdot + (E_k - E_j)))$$

being the principal part $\mathcal{P}(f)$ of functions $f \equiv f_{\ell}^{(\beta)} (\cdot + (E_k - E_j))$.

Meanwhile, the non anti-self-adjoint operator $\mathfrak{L}_d \in \mathcal{B}(\mathfrak{H}_{\text{at}})$ describing the effective atomic dissipation is defined by

$$\mathfrak{L}_d := \frac{1}{2} \sum_{c \in \sigma([H_{\text{at}}, \cdot])} \sum_{(j,k) \in \mathcal{L}_t} \sum_{\ell=1}^m c_{j,k}^{(\ell)} \mathfrak{L}_{j,k}^{(\ell)}$$

where $c_{j,k}^{(\ell)} := \pi f_{\ell}^{(\beta)} (E_k - E_j)$ and

$$\mathfrak{L}_{j,k}^{(\ell)} (\rho) := 2V_{j,k}^{(\ell)} \rho V_{j,k}^{(\ell)*} - V_{j,k}^{(\ell)*} V_{j,k}^{(\ell)} \rho - \rho V_{j,k}^{(\ell)*} V_{j,k}^{(\ell)} , \quad \rho \in \mathfrak{H}_{\text{at}}.$$  

The terms $V_{j,k}^{(\ell)} \rho V_{j,k}^{(\ell)*}$ in these definitions correspond to transitions from the $k$th to the $j$th atomic levels, whereas the other terms guarantee the Markov property of the dynamics, i.e., the preservation of the trace of the density matrix.

Note that the functions $f_{\ell}^{(\beta)}$ satisfy the equality

$$f_{\ell}^{(\beta)} (-x) = e^{-\beta x} f_{\ell}^{(\beta)} (x), \quad x \in \mathbb{R},$$

whereas $V_{j,k}^{(\ell)} = V_{k,j}^{(\ell)*}$ because $Q_\varepsilon$ is self-adjoint, by assumption. Using these properties, the effective atomic dissipation equals

$$\mathfrak{L}_d = \frac{1}{2} \sum_{c \in \sigma([H_{\text{at}}, \cdot])} \sum_{(j,k) \in \mathcal{L}_t} \sum_{\ell=1}^m c_{j,k}^{(\ell)} \left( \mathfrak{L}_{j,k}^{(\ell)} + (1 - \delta_{\ell,0}) e^{-\beta \mathfrak{L}_{j,k}^{(\ell)}} \right).$$

Observe further that $[H_{\text{Lamb}}, \rho_\beta] = 0$ and

$$e^{it H_{\text{at}}} \mathfrak{L}_{j,k}^{(\ell)} e^{-it H_{\text{at}}} = e^{it \mathfrak{L}_{j,k}^{(\ell)}}$$

for all $(j,k) \in \mathcal{L}_t$, $c \in \sigma^+([H_{\text{at}}, \cdot])$. This is the standard form of a Lindbladian fulfilling the so-called (quantum) detailed balance condition in the sense of Alicki–Frigerio–Gorini–Kossakowski–Verri with respect to the atomic Gibbs state $\rho_{\text{at}}$. See \[34\] \[35\] which is reviewed in \[27\] Section 4.5]. One important consequence of this fact is that the atomic Gibbs state, for all $\lambda, t$, satisfies

$$\mathfrak{L}_{\ell}^{(\lambda,0)} (\rho_\beta) = 0.$$  

See, for instance, \[3\] Section III.4]. Here, the parameter $\beta$ of the density matrix $\rho_\beta$ (see \(2.5\)) is chosen to be the inverse temperature of the reservoir.

**Remark 3.2 (Lindbladians as generators of Markov CP semigroups)**

The operators $H_{\text{at}}, H_p, H_{\text{Lamb}} \in \mathcal{B}(\mathbb{C}^d)$ are self-adjoint and $\{c_{j,k}^{(\ell)}\}_{j,k,\ell}$ are non-negative numbers. Thus, at any fixed time $t \in \mathbb{R}$ and for all $(\lambda, \eta) \in \mathbb{R}^2$, the Lindbladians $\mathfrak{L}_{\ell}^{(\lambda,\eta)}$ and $\frac{\eta}{2} \mathfrak{L}_p + \lambda^2 \mathfrak{L}_R$ are generators of Markov CP semigroups, by Theorem \[6.7\].

### 3.2 Irreducibility of quantum Markov chains

In principle, even after having extracted (by some averaging procedure, for instance) the oscillations of frequency $\varpi$ caused by the presence of the pump, the family $\{P_D (\rho_{\text{at}} (t))\}_{t \in \mathbb{R}_0^+}$ of density matrices could have...
several accumulation points, limits depending on the initial state, or even be oscillating (Rabi oscillations) as $t \to \infty$. We would like to avoid this situation and spectral properties of the Lindbladian $\frac{\eta}{2}\mathcal{L}_p + \lambda^2\mathcal{L}_R$ turn out to be important in this sense, see Section 6.2. To this end, we require that 0 is a non-degenerated eigenvalue of $\frac{\eta}{2}\mathcal{L}_p + \lambda^2\mathcal{L}_R$ with some non-trivial real spectral gap, that is,

$$\max\left\{\Re\{w\} \mid w \in \sigma\left(\frac{\eta}{2}\mathcal{L}_p + \lambda^2\mathcal{L}_R\right) \setminus \{0\}\right\} \leq -\lambda^2C < 0$$

with $C \in (0, \infty)$ being some fixed constant not depending on $\lambda$ and $\eta$.

This is useful (and very likely even essential) to prove Theorem 5.3 because it yields uniform bounds in time and allows the study of the asymptotic dynamics of the atom. The following assumption on the dissipative part $\mathcal{L}_d$ suffices to ensure the spectral properties mentioned above (cf. Lemma 6.3).

**Assumption 2 (Irreducibility of quantum Markov chains)**

The family $\{V_{j,k}^{(\ell)}\}_{j,k,\ell} \subset \mathcal{B}(\mathbb{C}^d)$ of operators defined by (3.9) satisfies

$$\left(\bigcup_{(j,k,\ell) : c_{j,k}^{(\ell)} \neq 0} \{V_{j,k}^{(\ell)}\}\right)^\prime = \mathcal{B}(\mathbb{C}^d)$$

with $M^\prime$ being the bicommutant of $M \subset \mathcal{B}(\mathbb{C}^d)$. Recall that $c_{j,k}^{(\ell)} := \pi f_\ell(\beta)(E_k - E_j)$, see also (3.10).

The assumption above highlights the role played by dissipative effects of the fermionic reservoir on the atom in order to get an appropriate asymptotic evolution of populations of atomic levels. Actually, the existence and uniqueness of the final ($t \to \infty$) density matrix projected on the subspace $\mathcal{D}$ of block–diagonal matrices follows from this hypothesis (cf. Theorem 4.7). See also Observation (a) at the beginning of Section 2.

Assumption 2 is a non–commutative version of the irreducibility of classical Markov chains. To illustrate this, we consider the following example: Assume for simplicity that $n = m = 1$ and the degeneracy $n_k$ of the $k$th atomic level equals $n_k = 1$ for all $k \in \{1, \ldots, N = d\}$. Let $\{\varphi_k\}_{k=1}^N \subset \mathbb{C}^d$ be an orthonormal basis of eigenvectors of $H_{at}$ with $H_{at}\varphi_k = E_k\varphi_k$. If the self–adjoint operator $Q_1$ of the atom–reservoir interaction is defined by

$$Q_1\varphi_k = \sum_{j=1}^d \varphi_j, \quad k \in \{1, \ldots, d\},$$

then the family $\{V_{j,k}^{(1)}\}_{j,k=1}^d$ satisfies $V_{j,n}^{(1)}V_{n,k}^{(1)} = V_{j,k}^{(1)}$ for all $j, k, n$ and forms an orthonormal basis of $\mathcal{H}_{at}$. In the orthonormal basis $\{\varphi_k\}_{k=1}^N$, $V_{j,k}^{(1)}$ is the elementary matrix made of zeros except at the intersection of the $j$th row with the $k$th column where its matrix coefficient is 1. We assume the irreducibility of the family $\{c_{j,k}^{(1)}\}_{j,k=1}^d \subset \mathbb{R}_0^+$ of non–negative numbers in the sense that, for all $j \neq k$, there is a finite sequence $(j_1, k_1), \ldots, (j_n, k_n)$ such that $c_{j_1,k_1}, \ldots, c_{j_{n-1},k_{n-1}} \neq 0$, $j_1 = j$, $k_n = k$, and $k_l = j_{l+1}$ for $l \in \{1, 2, \ldots, n - 1\}$. Physically speaking it means that any arbitrary pair of atomic levels is connected by non–vanishing transitions. By using the commutator identity

$$[A, V_{j,k}^{(1)}] = [A, V_{j,n}^{(1)}V_{n,k}^{(1)}] = V_{j,n}^{(1)}[A, V_{n,k}^{(1)}] + [A, V_{j,n}^{(1)}]V_{n,k}^{(1)}$$

for all $j, n, k$ and the irreducibility of the family $\{c_{j,k}^{(1)}\}_{j,k=1}^d$ one can compute the commutant

$$\left(\bigcup_{(j,k) : c_{j,k}^{(1)} \neq 0} \{V_{j,k}^{(1)}\}\right)^\prime = \mathbb{C} \cdot 1_{\mathbb{C}^d},$$

from which Assumption 2 follows. This is in perfect analogy to well–known results about uniqueness of invariant states of (aperiodic irreducible) discrete Markov chains. See for instance [36, Chapter 18].

Assumption 2 concludes the list of required conditions and from now on, we assume Assumptions 1, 2 to be satisfied.
3.3 The effective master equation

We define now the effective atomic master equation on the Hilbert space $\mathcal{H}_{at}$ as the initial value problem

$$\forall t \geq 0 : \quad \frac{d}{dt} \rho(t) = \Sigma_t^{(\lambda, \eta)}(\rho(t)), \quad \rho(0) = \rho_{at}(0) \equiv \rho_{at} \in \mathcal{H}_{at}. \quad (3.15)$$

Recall that $\rho_{at}$ is the density matrix of the initial atomic state $\omega_{at}$ of the atom and $\rho_{at}(t)$ is the density matrix of the time–dependent state $\omega_{at}(t)$ defined by (2.18) for any $t \in \mathbb{R}_+^\ast$. Even if one imposes the condition $\rho_{at} \in \mathcal{D}$, note that $\rho_{at}(t)$ is generally not block–diagonal, i.e., $\rho_{at}(t) \notin \mathcal{D}$ for all $t \geq 0$. The same is true for the solution of (3.15).

The effective atomic master equation has a unique solution which, by finite dimensionality of $\mathcal{H}_{at}$, is explicitly given by a Dyson series. In particular, this initial value problem defines a two–parameter family denoted by $\{\hat{\tau}_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$. Since the Lindbladian $\Sigma_t^{(\lambda, \eta)}$ is continuous and generates a Markov CP semigroup at any fixed time $t \in \mathbb{R}$ (cf. Remark 3.2), the two–parameter family $\{\hat{\tau}_{t,s}^{(\lambda, \eta)}\}_{t \geq s} \subset \mathcal{B}(\mathcal{H}_{at})$ is continuous, completely positive and preserves the trace. The positivity and trace preservation imply that this family is uniformly norm bounded\footnote{This can be seen by using a decomposition of any $A \in \mathcal{H}_{at}$ in imaginary and real parts, each of them being also decomposed in positive and negative parts. Use then the trace–norm, which is equivalent to the norm on $\mathcal{H}_{at} \equiv \mathcal{B}(\mathbb{C}^d)$.}

$$\forall \lambda, \eta, \varpi, s, t \in \mathbb{R}, \quad t \geq s : \quad \|\hat{\tau}_{t,s}^{(\lambda, \eta)}\| \leq C$$

(3.16)

for some finite constant $C \in (0, \infty)$ not depending on $\lambda$, $\eta$, $\varpi$, $s$, and $t$. When the optical pump is absent, the dynamics becomes autonomous and the family $\{\hat{\tau}_{t,s}^{(\lambda, 0)}\}_{t \geq s}$ corresponds to an one–parameter semigroup denoted for simplicity by

$$\hat{\tau}_t^{(\lambda, 0)} := \hat{\tau}_{t,0}^{(\lambda, 0)}. \quad (3.17)$$

The main interest of the initial value problem (3.15) is that its (unique) solution $\rho(t)$ accurately approximates at small couplings the true density matrix $\rho_{at}(t)$ of the time–dependent state $\omega_{at}(t)$ on the subspace $\mathcal{D}$ of block–diagonal matrices for all $t \in \mathbb{R}_+^\ast$. Indeed, we prove in [1] the following assertion:

**Theorem 3.3 (Validity of the effective atomic master equation)**

*Assume that $\rho_{at} \in \mathcal{D}$. The unique solution $\{\rho(t)\}_{t \geq 0}$ of the effective atomic master equation (3.15) and the atomic density matrix $\{\rho_{at}(t)\}_{t \geq 0}$ satisfy the bound

$$\|P_\mathcal{D} (\rho_{at}(t) - \rho(t))\| \leq C_{\varpi} |\lambda|$$

for some constant $C_{\varpi} \in (0, \infty)$ depending on $\varpi$, but not on the initial state $\omega_{at}$ of the atom and the parameters $t$, $\lambda$, and $\eta$.*

**Sketch of the proof.** The proof of Theorem 3.3 is conceptually similar to what is done in Section 4 but technically much more involved:

- Similar to the one–parameter semigroup $\{T_\alpha\}_{\alpha \geq 0}$ defined below, we represent the non–autonomous evolution $\{U_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$ (cf. (6.2)) as an autonomous dynamics $\{e^{\alpha \hat{G}}\}_{\alpha \geq 0}$ on an enlarged Hilbert space $\mathcal{H}_{evo} \supset \mathcal{H}$ of periodic $\mathcal{H}$–valued functions (vectors of $\mathcal{H}$ are naturally identified with the constant functions in this case). The generator $\hat{G}$ of the new time–evolution is occasionally referred to as Howland or Floquet operator. $\mathcal{H}$ is the GNS–space of the initial state $\omega_0$ and $\{U_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$ is a suitable representation of the full microscopic dynamics $\tau_{t,0}^{(\lambda, \eta)}$ through a family of bounded operators on $\mathcal{H}$. See Section 6.1.1–4 for the explicit construction of $\mathcal{H}$ and $\{U_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$.

- Then, we perform an analytic deformation $\hat{G}(\theta)$ (more precisely, analytic translation) of the unbounded closed operator $\hat{G}$ and prove that the dynamics driven by $\{e^{\alpha \hat{G}}\}_{\alpha \geq 0}$ and $\{e^{\alpha \hat{G}(\theta)}\}_{\alpha \geq 0}$ are the same on the atomic subspace $\mathcal{H}_{at} \equiv \mathcal{H}_{at} \otimes \{\Omega_\mathbb{R}\} \subset \mathcal{H}_{evo}$. The use of analytic deformations is the reason for the analyticity condition stated at the end of Section 2.5 which is also related to (6.4).
• In contrast to \( \mathcal{G} \), whose eigenvalues are all embedded in the continuous spectrum, \( \mathcal{G}(\theta) \) has discrete spectrum. It turns out that the discrete eigenspace of \( \mathcal{G}(\theta) \) is the relevant one for the atomic dynamics. We then analyze the discrete spectrum and eigenspace of \( \mathcal{G}(\theta) \) through Kato’s perturbation theory for closed operators. If Assumption 1 holds to the leading order in \( \lambda \) and \( \eta \), that is, second order in \( \lambda \) and first order in \( \eta \), the operator \( \mathcal{G}(\theta) \) is up to purely imaginary constants — unitarily equivalent to \( \frac{\eta}{2} \mathcal{L}_p + \lambda^2 \mathcal{L}_R \) in finite dimensional invariant subspaces spanning the whole discrete subspace of \( \mathcal{G}(\theta) \).

• We are then in position, by using the inverse Laplace transform for \( C_0 \)-semigroups together with Riesz projections, to analyze the action of the semigroup \( \{e^{\lambda t\mathcal{G}(\theta)}\}_{\lambda \geq 0} \) on vectors of the atomic subspace. This analysis leads to a version of Corollary 4.5 for the full microscopic dynamics and Theorem 3.3 follows. \( \square \)

4. Effective atomic dynamics

In this Section we study the behavior of the solutions of the effective atomic master equation, that is, the initial value problem \( \ref{evo} \). Since an important issue of laser technology is to obtain an optical pumping of atomic energy levels, we want to understand the time behavior of its solution in relation with the phenomenological Pauli master equation found in standard textbooks on lasers. The small (order \( \lambda^2 \omega^{-1} \)) fast oscillations of the populations due to the cosine in the master equation preclude a perfect steady behavior at large times. It is thus convenient to remove them by averaging the non-autonomous dynamics over a moving period. This leads to a pre–master equation proven in Theorem 4.8 whereas the Pauli master equation only extracts the limiting behavior of populations at large times (see Section 5).

4.1 Methodology and Numerical Illustrations

To this end, we first represent this non–autonomous evolution as an autonomous dynamics on an enlarged, infinite dimensional Hilbert space

\[
\mathfrak{h}_{\text{evo}} \supsetneq \mathfrak{h}_{\text{at}} \equiv \mathcal{B}(\mathbb{C}^d) .
\]

See \( \ref{4.5} \) below. The latter emerges through an additional degree of freedom which is a new time variable denoted by \( \alpha \geq 0 \).

By iterating the “variation of constants formula”, i.e., the integral equation

\[
\forall s, t \in \mathbb{R}, \ t \geq s : \quad \hat{\tau}_{t,s}^{(\lambda,\eta)} = \hat{\tau}_{t-s}^{(0)} + \int_s^t \hat{\tau}_{t-v}^{(0)} \mathfrak{m}_v^{(\lambda,\eta)} \hat{\tau}_{v,s}^{(\lambda,\eta)} \, dv , \quad (4.1)
\]

with the \( 2\pi \omega^{-1} \)-periodic operator

\[
\mathfrak{m}_t^{(\lambda,\eta)} := \mathfrak{S}_t^{(\lambda,\eta)} - \mathfrak{S}_t^{(0,0)} = \eta \cos(\pi \omega t) \mathcal{L}_p + \lambda^2 \mathcal{L}_R \in \mathcal{B}(\mathfrak{h}_{\text{at}}) , \quad (4.2)
\]

we get a representation of \( \hat{\tau}_{t,s}^{(\lambda,\eta)} \) as an absolutely convergent (Dyson) series which shows that \( (t, s) \mapsto \hat{\tau}_{t,s}^{(\lambda,\eta)} \) is continuous and

\[
\forall k \in \mathbb{Z}, \ s, t \in \mathbb{R}, \ t \geq s : \quad \hat{\tau}_{t,s}^{(\lambda,\eta)} = \hat{\tau}_{t+2\pi \omega^{-1} k, s+2\pi \omega^{-1} k}^{(\lambda,\eta)} . \quad (4.3)
\]

In other words, the dynamics between times \( (\alpha + 2\pi \omega^{-1} k) \) and \( (t + \alpha + 2\pi \omega^{-1} k) \) does not depend on \( k \in \mathbb{Z} \). As \( \hat{\tau}_{t-s}^{(0,0)} \) does not affect populations, \( \ref{4.2} \) also shows that they do not change much within a period of the pump. As already explained, it is thus natural to average the non–autonomous dynamics over a moving period of length \( 2\pi \omega^{-1} \) to extract the leading dynamical behavior of populations, in particular the inversion of population. The latter is described in Section 4.2.

This first step of the analysis of the solutions of the atomic master equation is quite useful because it enables the analysis of the non–autonomous dynamics via an associated (evolution) semigroup denoted by \( \{T_\alpha\}_{\alpha \geq 0} \) corresponding to an autonomous dynamics. Observe that \( \{T_\alpha\}_{\alpha \geq 0} \) acts on an infinite dimensional Hilbert space \( \mathfrak{h}_{\text{evo}} \supsetneq \mathfrak{h}_{\text{at}} \) although the initial non–autonomous dynamics was finite dimensional. Nevertheless, since the Hilbert space \( \mathfrak{D} \subset \mathfrak{h}_{\text{at}} \) is an invariant subspace of \( \{T_\alpha\}_{\alpha \geq 0} \) when \( \lambda = \eta = 0 \), Kato’s perturbation theory \( \ref{20} \) shows, for sufficiently small coupling constants \( |\lambda| \) and \( |\eta| \), the existence of a finite dimensional invariant Hilbert
space $\mathcal{H}_0^{(\lambda, \eta)}$ of $\{ T_\alpha \}_{\alpha \geq 0}$ almost parallel to $\mathcal{D}$. In particular, concerning the dynamics of populations, we can finally pass to an autonomous finite dimensional dynamics. Note however that $\dim \mathcal{H}_0^{(\lambda, \eta)} > \dim \mathcal{D}$, as $\mathcal{D}$ is a subspace of the larger invariant space $\mathcal{H}_0^{(0,0)}$ of $\{ T_\alpha \}_{\alpha \geq 0}$ when $\lambda = \eta = 0$. This second step is performed in Sections 4.3–4.4.

As a first application, these results are then used at the end of Section 4.4 to study the large time behavior of $P_\mathcal{D}(\rho(t))$. We show in particular the existence of a density matrix $\rho_\infty$ (Theorem 4.6) which is uniquely determined by a balance condition (Theorem 4.7) and approximates $P_\mathcal{D}(\rho(t))$ properly in the limit $t \to \infty$ for small enough coupling constants $|\lambda|$, $|\eta|$. In Section 4.5 we derive an integro–differential equation (pre–master equation) on the subspace of block–diagonal density matrices $\mathcal{D} \subset \mathcal{H}_\text{at}$ effectively describing the physical evolution of the populations. The dynamics of populations is properly described by an integro–differential equation and not by an effective differential equation like a Pauli equation. This reflects the strict inequality $\dim \mathcal{H}_0^{(\lambda, \eta)} > \dim \mathcal{D}$.

The rigorous proofs can sometimes be tedious and require a number of definitions and notations. Therefore, we outline our study by giving now some numerical illustrations of these three master equations, that is:

(i) The effective atomic master equation (3.15).

(ii) The pre–master equation, see Theorem 4.8.

(iii) The phenomenological Pauli master equation (5.5) as explained in standard textbooks on lasers. See Section 5 for more details.

As explained in Sections 1–2, an important step of laser technology is to obtain an optical pumping of atomic energy levels. So, we focus here on the so–called inversion of population$^3$ As described in textbooks on laser physics, optical pumping is in many situations based on three– or four–level atoms [13], the second case being the more efficient of both. Therefore, we restrict our study on a non–degenerated four–level atom, i.e., $d = N = 4$. In this case, the effective atomic master equation (3.15) is a non–autonomous evolution equation on a 16–dimensional Hilbert space which can easily be treated by standard numerical methods. One can then understand better the different approximations performed in this section which lead to the pre–master and Pauli master equations.

In our example, the atomic Hamiltonian depends on the parameter $\varpi$ and equals

$$H_{\text{at}} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \frac{1}{2} \varpi & 0 & 0 \\ 0 & 0 & \frac{5}{6} \varpi & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}.$$

(4.4)

The atom–reservoir interaction is fixed by $m = 1$, the self–adjoint matrix

$$Q_1 = \begin{pmatrix} 0 & \frac{5}{6} & \frac{1}{3} & \frac{1}{18} \\ \frac{5}{6} & 0 & \frac{1}{3} & \frac{1}{18} \\ \frac{1}{3} & \frac{1}{3} & 0 & 1 \\ \frac{1}{12} & \frac{1}{12} & 0 & 1 \end{pmatrix}$$

and the coupling function $f_1$ defined on $\mathbb{R}^3$ by

$$f_1(p) = f_\ell(|p|) = \frac{1}{2\pi |p|} \exp \left( -\frac{|p|^2}{2} \right).$$

See Section 2.5 The optical pump is modeled here by the time–periodic perturbation

$$\eta \cos(\varpi \alpha) \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}, \quad \alpha \equiv t \in \mathbb{R},$$

$^3$Note that analytical studies concerning the inversion of population can easily be done, at least for $d = N = 4$, by using the balance equation 4.45.
of the atomic Hamiltonian $H_{\text{at}}$, see Section 2.3. The initial state of the atom $\rho_{\text{at}} \in \mathcal{D} \subset \mathcal{D}_{\text{at}}$ is the Gibbs state $\rho_g = P_\mathcal{D} (\rho_g)$, taken at the same inverse temperature $\beta$ as the fermionic reservoir. We set the frequency $\varpi$, the inverse temperature $\beta$ and the coupling constants $\lambda, \eta$ respectively equal to $\varpi = 3$, $\beta = 0.5$, $\lambda \approx 0.385$, and $\eta = \lambda^2 \approx 0.148$.

The population density of the $k$th atomic level, at any fixed time $\alpha \in \mathbb{R}^+_0$ and for any $k \in \{1, \ldots, 4\}$, is given by

$$d_k(\alpha) := p_k(\rho(\alpha)) = \text{Tr}_{\mathcal{C}_4} (\mathbf{1} [H_{\text{at}} = E_k] \rho(\alpha) \mathbf{1} [H_{\text{at}} = E_k]) \geq 0.$$  

Here, $\rho(\alpha)$ is the solution of the effective atomic master equation (3.15) and $d_k(\alpha)$ is directly related to the atomic populations because of Theorem 3.3. Similarly, populations can be defined for the solutions of the pre-master equation and the Pauli master equation. The plots of all these densities are given in figure 1 and clearly show a stable inversion of population as

$$d_3(\alpha) > d_1(\alpha) > d_4(\alpha) > d_2(\alpha)$$

for large enough $\alpha \leq 180$. As soon as the optical pump is turned off, the systems relaxes to the Gibbs state $\rho_g$.

Figure 1: Illustration of the populations as functions of $\alpha \equiv t \in \mathbb{R}^+_0$ at $\beta = 0.5$, $\lambda \approx 0.385$, $\eta = \lambda^2 \approx 0.148$ and $\varpi = 3$. Blue, green, orange, and red lines correspond to the populations of the 1st, the 2nd, the 3rd and the 4th atomic energy levels, respectively. The dashed dotted line marks the time $\alpha = 180$ when the pump is turned off, i.e., $\eta = 0$ for all times beyond this line. The four dashed lines mark the populations of the thermal equilibrium state, i.e., the Gibbs state $\rho_g$ at inverse temperature $\beta = 0.5$ of the thermal reservoir. The two figures on the top are computed from the master equation. The pre-master equation gives exactly the same picture at this time scale. The figure on the bottom is computed from the Pauli master equation.

As we can observe in figures 1 and 2 the qualitative difference in the behavior of the solutions of the master, pre-master and Pauli master equations for population densities are clear: The pre-master equation removes the small, but fast oscillations due to the cosine in the master equation (figure 2), and the Pauli master equation additionally cancels the Rabi (moderate) oscillations still present in the pre-master equation. (Compare the plots of figure 1).

Note that Rabi oscillations, which depends on the coupling constant $\eta$, are progressively suppressed by dissipative processes. Removing the atom–reservoir interaction by setting $\lambda = 0$ while keeping $\eta \approx 0.148$ we observe non-suppressed oscillations, see figure 3. In this case the stable inversion of population also disappears, as explained in the beginning of Section 2.

To conclude, our results permit purely quantum mechanical detailed studies of many–level optically active impurities used to produce lasing materials. For instance, they can be used to analyze the influence of temperature and other relevant physical parameters on inversion of population: At least in the example considered here, the inversion of population is decreasing with the temperature, i.e., is an increasing function of $\beta$ as shown figure 1 and the most efficient parameter to strengthen it is curiously the frequency $\varpi$ and not the parameter $\eta$, provided the last is neither too small nor too large. The effect of the degeneracy and the dynamics for very low frequencies can also be analyzed. The latter corresponds to a situation close to the adiabatic limit and shows an unusual behavior. We postpone this kind of studies to a further paper and start now the rigorous proofs related to these numerical observations.
Figure 2: Illustration of the populations as functions of $\alpha \equiv t \in [0, 2]$ at $\beta = 0.5$, $\lambda \simeq 0.385$, $\eta = \lambda^2 \simeq 0.148$ and $\varpi = 3$. Blue, green, orange, and red lines correspond to the populations of the 1st, the 2nd, the 3rd and the 4th atomic energy levels, respectively. They are computed from the master equation. The dashed dotted lines are the same objects computed from the pre–master equation.

Figure 3: Illustration of the populations as functions of $\alpha \equiv t \in [0, 300]$ at $\beta = 0.5$, $\lambda = 0$, $\eta \simeq 0.148$ and $\varpi = 3$. Blue, green, orange, and red lines correspond to the populations of the 1st, the 2nd, the 3rd and the 4th atomic energy levels, respectively. The dashed dotted line marks the time $\alpha = 180$ when the pump is turned off, i.e., $\eta = 0$ for all times beyond this line. The four dashed line mark the populations of the Gibbs state $\rho_g$ at inverse temperature $\beta = 0.5$ of the thermal reservoir. The figure is computed from the master equation. Note that, for $\alpha \geq 180$, the system do not relax to the Gibbs state $\rho_g$ at inverse temperature $\beta = 0.5$ of the thermal reservoir because there is no atom–reservoir interaction as $\lambda = 0$. 
From the non–autonomous master equation to an autonomous effective dynamics, evolution semigroups

Consider the Hilbert space

\[ \mathcal{H}_{\text{evo}} := L^2 \left( [0, 2\pi \varpi^{-1}], \mathcal{D}_{\text{at}} \right) \]

of time–dependent \(2\pi \varpi^{-1}\)–periodic \(\mathcal{D}_{\text{at}}\)–valued functions. The scalar product on \(\mathcal{H}_{\text{evo}}\) is naturally defined by

\[ \langle f, g \rangle_{\text{evo}} := \varpi \int_0^{2\pi} \langle f(t), g(t) \rangle_{\text{at}} \, dt = \varpi \int_0^{2\pi} \text{Tr}_{C^*} \left( (f(t))^* g(t) \right) \, dt \]

for all \(f, g \in \mathcal{H}_{\text{evo}}\), see (2.6). \(\mathcal{D}_{\text{at}} \subset \mathcal{H}_{\text{evo}}\) is seen as the subspace of constant functions of \(\mathcal{H}_{\text{evo}}\).

From the continuous two–parameter family \(\{\hat{\pi}_{t,s}^{(\lambda,\eta)}\}_{t \geq s}\) corresponding to the non–autonomous master equation (3.15) we can uniquely define a strongly continuous one–parameter semigroup \(\{T_\alpha\}_{\alpha \geq 0}\) by the condition

\[ \forall t \in [0, 2\pi \varpi^{-1}] \text{ a.e. : } \quad T_\alpha (f)(t) = \hat{\pi}_{t,t-\alpha}^{(\lambda,\eta)} f(t - \alpha) \]

for all \(\alpha \geq 0\) and \(f \in \mathcal{H}_{\text{evo}}\). Because of (4.3), \(T_\alpha\) is an operator acting on \(\mathcal{H}_{\text{evo}}\) for any \(\alpha \geq 0\). The strong continuity of \(\alpha \mapsto T_\alpha\) follows from the continuity of \((s, t) \mapsto \hat{\pi}_{t,s}^{(\lambda,\eta)}\), and the semigroup property of \(\{T_\alpha\}_{\alpha \geq 0}\) from the cocycle property of the two–parameter family \(\{\hat{\pi}_{t,s}^{(\lambda,\eta)}\}_{t \geq s}\). Moreover, by the norm boundedness (3.16) of the evolution family \(\{\hat{\pi}_{t,s}^{(\lambda,\eta)}\}_{t \geq s}\), the semigroup \(\{T_\alpha\}_{\alpha \geq 0} \subset \mathcal{B}(\mathcal{H}_{\text{evo}})\) is uniformly norm bounded:

\[ \forall \lambda, \eta \in \mathbb{R}, \alpha \geq 0 : \quad \| T_\alpha \| \leq C \]

for some finite constant \(C \in (0, \infty)\) not depending on \(\lambda, \eta, \) and \(\alpha\).

The generator of the strongly continuous semigroup \(\{T_\alpha\}_{\alpha \geq 0}\) is the closed unbounded operator

\[ G^{(\lambda,\eta)} := -\frac{d}{dt} + \mathcal{D}_{\text{evo}}^{(\lambda,\eta)} \]

sometimes referred to as Howland operator of the non–autonomous atomic dynamics, with dense domain

\[ \mathcal{D}(G^{(\lambda,\mu)}) := \left\{ \sum_{k=-\infty}^{\infty} a_k e^{ik\varpi t} : a_k \in \mathcal{D}_{\text{at}}, \quad \sum_{k=-\infty}^{\infty} \|k a_k\|_{\text{at}}^2 < \infty \right\} \subset \mathcal{H}_{\text{evo}} \]

Here,

\[ \frac{d}{dt} f(t) = \sum_{k=-\infty}^{\infty} ik a_k e^{ik\varpi t} \]
in the $L^2\left([0, 2\pi\varpi^{-1}], \mathfrak{H}_{\text{at}}\right)$ sense for all

$$f = \sum_{k=-\infty}^{\infty} a_k e^{i k \varpi t} \in \mathcal{D}(G^{(\lambda, \mu)}) ,$$

and $\mathfrak{L}_{\text{evo}}^{(\lambda, \eta)} \in B(\mathfrak{H}_{\text{evo}})$ is the bounded operator defined, for all $f \in \mathfrak{H}_{\text{evo}}$, by

$$\forall t \in [0, 2\pi\varpi^{-1}] \text{ a.e.: } \mathfrak{L}_{\text{evo}}^{(\lambda, \eta)}(f)(t) := \mathfrak{L}_{\text{evo}}^{(\lambda, \eta)}(f(t)) . \quad (4.10)$$

**Remark 4.1 (The uncoupled atom–reservoir case)**

The spectrum of $G^{(0, 0)}$ is purely discrete. Therefore, Kato’s perturbation theory [26] of discrete eigenvalues can be used to study the spectral properties of the generator $G^{(\lambda, \eta)}$ for small $|\lambda|$ and $|\eta|$.

The time–behavior of the solution $\rho(t) \in \mathfrak{H}_{\text{at}}$ of the non–autonomous master equation (3.15) can be studied on the subspace $\mathfrak{D} \subset B(\mathbb{C}^d) \equiv \mathfrak{H}_{\text{at}}$ of block–diagonal matrices (cf. (3.1)) by using the $C_0$–semigroup $\{T_\alpha\}_{\alpha \geq 0}$. More precisely, we prove in the following lemma that, for any (block–diagonal) $A \in \mathfrak{D}$, the scalar products of the form

$$\langle T_\alpha (\rho), A \rangle_{\text{evo}}$$

properly describe (i.e. up to small oscillations uniformly bounded in time) the time evolution of

$$\langle \rho(\alpha), A \rangle_{\text{at}} := \text{Tr} (\rho(\alpha)A)$$

whenever the pump frequency $\varpi$ is sufficiently large or the couplings $|\lambda|, |\eta|$ are sufficiently small.

**Lemma 4.1 (Average dynamics over a moving period of length $2\pi\varpi^{-1}$)**

Assume that $\rho_{\text{at}} \in \mathfrak{D}$. The unique solution $\{\rho(t)\}_{t \geq 0}$ of the effective atomic master equation (3.15) satisfies the bound

$$|\langle \rho(\alpha), A \rangle_{\text{at}} - \langle T_\alpha (\rho_{\text{at}}), A \rangle_{\text{evo}}| \leq C \lambda^2 \varpi^{-1} \|A\| \quad (4.11)$$

for all (block–diagonal) $A \in \mathfrak{D}$, all $|\lambda| \leq 1$ and all $\alpha \in \mathbb{R}_0^+$. Here, $C \in (0, \infty)$ is a finite constant not depending on $\rho_{\text{at}}$, $A$, $\lambda$, $\eta$, $\varpi$, and $\alpha$.

**Proof.** By (4.6),

$$\langle \rho(\alpha), A \rangle_{\text{at}} - \langle T_\alpha (\rho_{\text{at}}), A \rangle_{\text{evo}} = \frac{\varpi}{2\pi} \int_0^{2\pi} \left\langle \hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} - \hat{\varphi}_{\alpha,-\alpha}^{(\lambda, \eta)}, (\rho_{\text{at}}), A \right\rangle_{\text{at}} \, dt . \quad (4.12)$$

Therefore, we need to estimate the integrand in this last equality for any $\alpha \in \mathbb{R}_0^+$ and $t \in [0, 2\pi\varpi^{-1})$. To this end, we choose, for any $\alpha \in \mathbb{R}_0^+$, $r(\alpha) \in 2\pi \varpi^{-1}\mathbb{N}_0$ such that

$$0 \leq r(\alpha) - \alpha \leq 2\pi \varpi^{-1} .$$

Using the $2\pi \varpi^{-1}$–periodicity (4.3) of the evolution family $\{\hat{\varphi}_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$ we obtain that

$$\hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} - \hat{\varphi}_{\alpha,-\alpha}^{(\lambda, \eta)} = \hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} - \hat{\varphi}_{\delta + \alpha, \delta}^{(\lambda, \eta)} \quad (4.13)$$

for any $t \in [0, 2\pi\varpi^{-1}]$ with

$$\delta := t + r(\alpha) - \alpha \in [0, 4\pi \varpi^{-1}] .$$

Using the cocycle property of the two–parameter family $\{\hat{\varphi}_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$ together with (4.13) we have

$$\hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} - \hat{\varphi}_{\alpha,-\alpha}^{(\lambda, \eta)} = \hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} - \hat{\varphi}_{\delta,0}^{(\lambda, \eta)} + (1 - \hat{\varphi}_{\alpha, \delta, \delta}^{(\lambda, \eta)}) \hat{\varphi}_{\alpha,0}^{(\lambda, \eta)} \quad (4.14)$$

with $\delta \in [0, 4\pi \varpi^{-1}]$. Note that $\|[H_{\text{at}}, \cdot]\| = \mathcal{O}(\varpi)$ ($\varpi = E_N - E_1 \in \sigma([H_{\text{at}}, \cdot])$ and thus, we cannot expect the norms

$$\|\hat{\varphi}_{\delta,0}^{(\lambda, \eta)} - 1\|$$

and

$$\|1 - \hat{\varphi}_{\alpha, \delta, \delta}^{(\lambda, \eta)}\|$$
to be small for large pump frequencies \( \omega >> 1 \) when \( \delta = \mathcal{O}(\omega^{-1}) \).

However, the integral equation (4.11) implies
\[
\hat{\tau}_{\delta}(\lambda, \eta) - 1 = \hat{\tau}_{\delta}^{(0, 0)} - 1 + \int_{s}^{s+\delta} \hat{\tau}_{\delta}^{(0, 0)} \omega_{s}^{(\lambda, \eta)} \hat{t}_{\nu, \tau}^{(\lambda, \eta)} \, dv ,
\]
for all \( s \in \mathbb{R} \) and \( \delta \in [0, 4\pi\omega^{-1}] \). Meanwhile, for all \( A \in \mathcal{D} \),
\[
(1 - \hat{\tau}_{\delta}^{(0, 0)})^\ast (A) = 0 ,
\]
as \( (\hat{\tau}_{\delta}^{(0, 0)})^\ast = e^{-\delta \omega_{\ast}} \) and \( \mathcal{E}_{\ast}(\mathcal{D}) = \{0\} \). Recall that the pump is moderate with respect to the atom–reservoir interaction, i.e., \( |\eta| \leq C\lambda^2 \) for some fixed constant \( C \in (0, \infty) \), see Assumption [1]. Hence, since the evolution family \( \{\hat{t}_{\nu, \tau}^{(\lambda, \eta)}\}_{t \geq s} \) is uniformly norm bounded (cf. (3.16)), we deduce from (4.12), (4.15) and (4.16) that
\[
\| (1 - \hat{\tau}_{\delta}^{(\lambda, \eta)})^\ast (A) \| \leq C\lambda^2 \omega^{-1} \| A \| ,
\]
for some constant \( C \in (0, \infty) \) not depending on \( A \in \mathcal{D}, \lambda, \eta, \omega, \alpha \) and \( \delta \in [0, 4\pi\omega^{-1}] \). By (3.16),
\[
\left| \left( (1 - \hat{\tau}_{\delta}^{(\lambda, \eta)})^{\ast} (\hat{\tau}_{\delta}^{(\lambda, \eta)} (\rho_{\ast}, A) \right) \right| \leq C\lambda^2 \omega^{-1} \| A \| ,
\]
for some constant \( C \in (0, \infty) \) not depending on \( \rho_{\ast}, A, \lambda \in [-1, 1], \eta, \omega, \alpha \) and \( \delta \in [0, 4\pi\omega^{-1}] \).

Similarly, as \( \rho_{\ast} \in \mathcal{D} \) and \( \| \rho_{\ast} \| = 1 \),
\[
\| (\hat{\tau}_{\delta}^{(\lambda, \eta)} - 1)(\rho_{\ast}) \| \leq C\lambda^2 \omega^{-1}
\]
for some constant \( C \in (0, \infty) \) not depending on \( \rho_{\ast}, A \in \mathcal{D}, \lambda \in [-1, 1], \eta, \omega, \alpha \) and \( \delta \in [0, 4\pi\omega^{-1}] \). Using this together with (3.16) we conclude that
\[
\left| \left( \hat{\tau}_{\delta}^{(\lambda, \eta)} - \hat{\tau}_{\delta}^{(\lambda, \eta)} (\rho_{\ast}, A) \right) \right| \leq C\lambda^2 \omega^{-1} \| A \| ,
\]
for some constant \( C \in (0, \infty) \) not depending on \( \rho_{\ast} \in \mathcal{D}, A \in \mathcal{D}, \lambda \in [-1, 1], \eta, \omega, \alpha \) and \( \delta \in [0, 4\pi\omega^{-1}] \).

From (4.14), (4.18) and (4.19) we obtain
\[
\forall t \in [0, 2\pi\omega^{-1}] : \left| \left( \hat{\tau}_{\delta}^{(\lambda, \eta)} - \hat{\tau}_{t, \nu, \tau}^{(\lambda, \eta)} (\rho_{\ast}, A) \right) \right| \leq C\lambda^2 \omega^{-1} \| A \|
\]
with some constant \( C \in (0, \infty) \) not depending on \( \rho_{\ast} \in \mathcal{D}, A \in \mathcal{D}, \lambda \in [-1, 1], \eta, \omega, t \) and \( \alpha \). Combining this with (4.12), estimate (4.11) follows.

\[\square\]

**Remark 4.2 (General atomic initial states)**

If the density matrix \( \rho_{\ast} \in \mathcal{H}_{\ast} \) of the initial state \( \omega_{\ast} \) is not block–diagonal, i.e., \( \rho_{\ast} \in \mathcal{H}_{\ast} \setminus \mathcal{D} \), then the assertion of Lemma [4.1] holds at large times. Indeed, the transient behavior of \( \{\rho_{\ast}(t)\}_{t \in \mathbb{R}^+_0} \) strongly depends on the quantum correlations of the initial atomic state, whereas its long time behavior does not depend on the initial conditions. The following bound can be shown for arbitrary density matrices \( \rho_{\ast} \in \mathcal{H}_{\ast} \) (i.e., \( \rho_{\ast} \in \mathcal{D} \) is not assumed):
\[
| \langle \rho_{\ast} (\alpha), A \rangle_{\text{evo}} - \langle T_{\alpha} (\rho_{\ast}), A \rangle_{\text{evo}} | \leq C(\lambda^2 \omega^{-1} + e^{-c\alpha}) \| A \| ,
\]
for some constants \( c, C \in (0, \infty) \) not depending on \( \rho_{\ast}, A, \lambda, \eta, \omega, \) and \( \alpha \). We omit the details and illustrate this fact with a numerical example: see figure [3]. Indeed, we focus on the study of the dynamics of populations and the initial state is in most cases of interest the Gibbs state \( \rho_{\ast} \in \mathcal{D} \). For sake of technical simplicity we assume that \( \rho_{\ast} \in \mathcal{D} \) and only observe at this point that all results below on the dynamics \( \{\rho_{\ast}(t)\}_{t \in \mathbb{R}^+_0} \) stay correct for all \( \rho_{\ast} \in \mathcal{H}_{\ast} \setminus \mathcal{D} \) up to a transient factor decaying as \( e^{-c\alpha} \) for some \( c > 0 \).
Figure 5: Illustration of the populations computed from the master equation as functions of $\alpha \equiv t \in \mathbb{R}_0^+$ at $\beta = 0.5$, $\lambda \simeq 0.385$, $\eta = \lambda^2 \simeq 0.148$ and $\varpi = 3$. Blue, green, orange, and red lines correspond to the populations of the 1st, the 2nd, the 3rd and the 4th atomic energy levels, respectively. The dashed dotted line marks the time $\alpha = 100$ when the pump is turned off, i.e., $\eta = 0$ for all times beyond this line. The four dashed lines are the populations computed from the pre–master equation. The figure is computed with initial matrix coefficients $(\rho_{at})_{j,k} = 0.5$ for $(j,k) \in \{(1,1), (4,4), (1,4), (4,1)\}$ and $(\rho_{at})_{j,k} = 0$ otherwise. I.e., $\rho_{at} \notin \mathcal{D}$.

4.3 Dimensional restriction of $\{T_\alpha\}_{\alpha \geq 0}$

As explained at the beginning of Section 4, the semigroup $\{T_\alpha\}_{\alpha \geq 0}$ acts on an infinite dimensional Hilbert space $\mathcal{H}_{\text{evo}}$, but the initial conditions we are interested in are constant functions, i.e., elements of the finite dimensional subspace $\mathcal{H}_{\text{at}} \subset \mathcal{H}_{\text{evo}}$. It turns out that $\mathcal{H}_{\text{at}}$ is contained in some finite dimensional subspace $\mathcal{H}_{\text{inv}}^{(0,0)}$ which is almost parallel to a finite dimensional subspace $\mathcal{H}_{\text{inv}}^{(\lambda,\eta)} \subset \mathcal{H}_{\text{evo}}$. The latter is invariant with respect to $\{T_\alpha\}_{\alpha \geq 0}$, see (4.26) below. As this semigroup is bounded (cf. (4.17)), the restriction of the autonomous dynamics to this invariant subspace describes – up to small errors – the evolution of the solution $\{\rho(t)\}_{t \geq 0}$ of the effective atomic master equation (3.15).

To define $\mathcal{H}_{\text{inv}}^{(\lambda,\eta)}$ precisely we need some preliminary definitions. We denote by

$$P_\epsilon^{(\lambda,\eta)} := \frac{1}{2\pi i} \oint_{|z+i\epsilon| = \frac{\eta}{4}} (z - G^{(\lambda,\eta)})^{-1} \, dz \quad (4.20)$$

the Riesz projection [26, Chapter II] associated with the generator $G^{(\lambda,\eta)}$ defined by (4.8). Here,

$$R := \min \{|\epsilon - \epsilon'| : \epsilon, \epsilon' \in \sigma([H_{\text{at}}, \cdot]), \epsilon \neq \epsilon'\} > 0 \quad (4.21)$$

and we assume that the atom–reservoir coupling $|\lambda|$ – and thus $|\eta|$, by Assumption 1 – is sufficiently small to ensure that the Riesz projection $P_\epsilon^{(\lambda,\eta)}$ is well–defined and has the same dimension as $P_\epsilon^{(0,0)}$. Then, for each $\epsilon \in \sigma([H_{\text{at}}, \cdot])$,

$$\mathcal{H}_{\text{inv}}^{(\lambda,\eta)} := P_\epsilon^{(\lambda,\eta)} \mathcal{H}_{\text{evo}} \quad (4.22)$$

is an invariant, finite dimensional subspace of the evolution semigroup $\{T_\alpha\}_{\alpha \geq 0}$. Consequently,

$$\mathcal{H}_{\text{inv}}^{(\lambda,\eta)} := \text{span} \left\{ \bigcup_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} \mathcal{H}_{\epsilon}^{(\lambda,\eta)} \right\} \quad (4.23)$$

is a finite dimensional invariant subspace of $\{T_\alpha\}_{\alpha \geq 0}$ and $\mathcal{H}_{\text{at}} \subset \mathcal{H}_{\text{inv}}^{(0,0)}$ and $P_\epsilon^{(\lambda,\eta)} - P_\epsilon^{(0,0)} = \mathcal{O}(\lambda^2)$. 

As a consequence, if |λ| is sufficiently small and w is large enough then the restriction of the semigroup \( \{T_\alpha\}_{\alpha \geq 0} \) to its invariant space \( \mathcal{H}_{\text{inv}}^{(\lambda,\eta)} \) accurately describes the time evolution of \( \{\langle \rho(\alpha), A \rangle_{\text{evo}}\}_{\alpha \geq 0} \):

**Lemma 4.2 (Finite dimensional effective autonomous dynamics)**

Assume that \( \rho_{\text{at}} \in \mathcal{D} \). The unique solution \( \{\rho(t)\}_{t \geq 0} \) of the effective atomic master equation (3.15) satisfies the bound

\[
\left| \langle \rho(\alpha), A \rangle_{\text{at}} - \sum_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} \left\langle \exp \left( \alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon \right) P^{(\lambda,\eta)}_\epsilon \rho_{\text{at}}, A \right\rangle_{\text{evo}} \right| \leq C \lambda^2 (1 + w^{-1}) \|A\|
\]

for any (block–diagonal) \( A \in \mathcal{D} \), \(|\lambda| << 1\) and \( \alpha \in \mathbb{R}_0^+ \). Here, \( C \in (0, \infty) \) is a finite constant not depending on \( \rho_{\text{at}}, A, \lambda, \eta, w, \) and \( \alpha \).

**Proof.** Consider the projection

\[
P_{\text{inv}}^{(\lambda,\eta)} := \sum_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} P^{(\lambda,\eta)}_\epsilon \tag{4.24}
\]

onto the invariant subspace \( \mathcal{H}_{\text{inv}}^{(\lambda,\eta)} \). Note that \( \mathcal{H}_{\text{at}} \subset \mathcal{H}_{\text{inv}}^{(0,0)} \) and

\[
\forall \rho \in \mathcal{H}_{\text{at}} : \quad P_{\text{inv}}^{(0,0)} \rho = \rho . \tag{4.25}
\]

By Assumption 1 and Kato’s perturbation theory [26] for discrete eigenvalues, there is a constant \( C \in (0, \infty) \) not depending on coupling constants \( \lambda \) and \( \eta \) such that

\[
\|P_{\text{inv}}^{(\lambda,\eta)} - P_{\text{inv}}^{(0,0)}\| \leq C \lambda^2 \tag{4.26}
\]

at small \(|\lambda|\). By (4.7), we also observe that the operator family

\[
\left\{ \exp \left( \alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon \right) \right\}_{\alpha \geq 0}
\]

is a bounded semigroup for any eigenvalue \( \epsilon \in \sigma([H_{\text{at}}, \cdot]) \) as it is a restriction of the bounded semigroup \( \{T_\alpha\}_{\alpha \geq 0} \) onto the invariant subspace \( \mathcal{H}_{\text{inv}}^{(\lambda,\eta)} \) (4.22). Indeed,

\[
\forall \alpha \geq 0, \quad \epsilon \in \sigma([H_{\text{at}}, \cdot]) : \quad \exp \left( \alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon \right) = T_\alpha P^{(\lambda,\eta)}_\epsilon + (1 - P^{(\lambda,\eta)}_\epsilon).
\]

Hence, by (4.24), (4.25) and (4.27), for any \( \rho \in \mathcal{H}_{\text{at}} \),

\[
T_\alpha (\rho) = T_\alpha \left( (1 - P_{\text{inv}}^{(\lambda,\eta)}) \rho_{\text{at}} \right) + T_\alpha \left( P_{\text{inv}}^{(\lambda,\eta)} \rho_{\text{at}} \right)
\]

\[
= T_\alpha \left( \left( P_{\text{inv}}^{(0,0)} - P_{\text{inv}}^{(\lambda,\eta)} \right) \rho \right) + \sum_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} \exp \left( \alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon \right) \left( P^{(\lambda,\eta)}_\epsilon \rho \right)
\]

which, combined with (4.7) and (4.26), in turn implies

\[
\left| \left\langle T_\alpha (\rho_{\text{at}}), A \right\rangle_{\text{evo}} - \sum_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} \left\langle \exp \left( \alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon \right) P^{(\lambda,\eta)}_\epsilon \rho_{\text{at}}, A \right\rangle_{\text{evo}} \right| \leq C \|A\| \lambda^2
\]

with \( C \in (0, \infty) \) not depending on \( \rho_{\text{at}}, A, \lambda, \eta, w, \) and \( \alpha \). The assertion follows now from Lemma 4.1. □

As a consequence, we can restrict the autonomous dynamics described by the evolution semigroup \( \{T_\alpha\}_{\alpha \geq 0} \) to the finite dimensional subspace \( \mathcal{H}_{\text{inv}}^{(\lambda,\eta)} \subset \mathcal{H}_{\text{evo}} \).
4.4 Effective block–diagonal dynamics

Since Theorem 3.3 only compares the orthogonal projections $P_\mathcal{D}(\rho_{\text{at}}(t))$ (4.2) and $P_\mathcal{D}(\rho(t))$ of the atomic density matrix $\rho_{\text{at}}(t)$ and $\rho(t)$, respectively, we are only interested in the effective block–diagonal dynamics defined by $\{P_\mathcal{D}(\rho(t))\}_{t \geq 0}$. As shown in the following lemma, this quantity is related to the finite dimensional, invariant subspace $\mathcal{S}_0^{(\lambda,\eta)}$ defined by (4.22) for $\epsilon = 0 \in \sigma([H_{\text{at}}, \cdot])$, see also (3.8).

Lemma 4.3 (Effective block–diagonal dynamics – I)

Assume that $\rho_{\text{at}} \in \mathcal{D}$. The effective block–diagonal density matrix $\{P_\mathcal{D}(\rho(t))\}_{t \geq 0}$ satisfies the bound

$$\left| \langle P_\mathcal{D}(\rho(\alpha)), A \rangle_{\text{at}} - \langle \exp (\alpha P^{(\lambda,\eta)}_0 G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) P^{(0,0)}_0 \rho_{\text{at}}, A \rangle_{\text{evo}} \right| \leq C \lambda^2 (1 + \varpi^{-1}) \| A \|$$

for any $A \in \mathcal{S}_{\text{at}}$, $|\lambda|$ sufficiently small and $\alpha \in \mathbb{R}_0^+$. Here, $C \in (0, \infty)$ is a finite constant not depending on $\rho_{\text{at}}$, $A$, $\lambda$, $\eta$, $\varpi$, and $\alpha$.

Proof. The orthogonal projection $P_\mathcal{D}$ acts in $\mathcal{S}_{\text{at}}$ and naturally induces an orthogonal projection, again denoted by $P_\mathcal{D}$, in the Hilbert space $\mathcal{H}_{\text{evo}}$ by:

$$\forall f \in \mathcal{H}_{\text{evo}}, \; t \in [0, 2\pi\varpi^{-1}) : \quad P_\mathcal{D}(f)(t) := P_\mathcal{D}(f(t)).$$

In particular, for any $A \in \mathcal{S}_{\text{at}}$, $\epsilon \in \sigma([H_{\text{at}}, \cdot])$ and $\alpha \in \mathbb{R}_0^+$,

$$\left\langle \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon) P^{(\lambda,\eta)}_\epsilon \rho_{\text{at}}, P_\mathcal{D}(A) \right\rangle_{\text{evo}}$$

$$= \langle P_\mathcal{D} \left( P^{(\lambda,\eta)}_\epsilon \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon) \rho_{\text{at}}, A \right) \rangle_{\text{evo}}$$

and similarly,

$$\langle \rho(\alpha), P_\mathcal{D}(A) \rangle_{\text{at}} = \langle P_\mathcal{D}(\rho(\alpha)), A \rangle_{\text{at}}.$$

With the last two equalities we use now Lemma 4.2 to obtain the bound

$$\left| \langle P_\mathcal{D}(\rho(\alpha)), A \rangle_{\text{at}} - \langle P_\mathcal{D} \left( P^{(\lambda,\eta)}_\epsilon \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_\epsilon) \rho_{\text{at}} \right), A \rangle_{\text{evo}} \right| \leq C \lambda^2 (1 + \varpi^{-1}) \| A \|, \quad (4.28)$$

where $C \in (0, \infty)$ is some constant not depending on $\rho_{\text{at}}$, $A$, $\lambda$, $\eta$, $\varpi$, and $\alpha$. For any $\epsilon \in \sigma([H_{\text{at}}, \cdot])$, note that

$$P_\mathcal{D} P^{(\lambda,\eta)}_\epsilon = P^{(\lambda,\eta)}_\epsilon P^{(0,0)}_0 + P_\mathcal{D}(P^{(\lambda,\eta)}_\epsilon - P^{(0,0)}_\epsilon) = \delta_{\epsilon,0} P_\mathcal{D} + P_\mathcal{D}(P^{(\lambda,\eta)}_\epsilon - P^{(0,0)}_\epsilon)$$

with $\delta_{\epsilon,\epsilon'}$ being the Kronecker delta. Similar to (4.26), there is a constant $C \in (0, \infty)$ not depending on the coupling constants $\lambda$ and $\eta$ such that

$$\max_{\epsilon \in \sigma([H_{\text{at}}, \cdot])} \| P^{(\lambda,\eta)}_\epsilon - P^{(0,0)}_\epsilon \| \leq C \lambda^2$$

(4.30)

for $|\lambda|$ sufficiently small. From (4.7) and (4.28)–(4.30)

$$\left| \langle P_\mathcal{D}(\rho(\alpha)), A \rangle_{\text{at}} - \langle P_\mathcal{D} \left( \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) \rho_{\text{at}} \right), A \rangle_{\text{evo}} \right| \leq C \lambda^2 (1 + \varpi^{-1}) \| A \| \quad (4.31)$$

with $C \in (0, \infty)$ not depending on $\rho_{\text{at}}$, $A$, $\lambda$, $\eta$, $\varpi$, and $\alpha$. Finally, for all $A \in \mathcal{S}_{\text{at}}$, observe that

$$\langle P_\mathcal{D} \left( \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) \rho_{\text{at}} \right), A \rangle_{\text{evo}}$$

$$= \langle \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) \rho_{\text{at}}, P_\mathcal{D}(A) \rangle_{\text{evo}}$$

$$= \langle \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) \rho_{\text{at}}, P^{(0,0)}_0(A) \rangle_{\text{evo}}$$

$$= \langle P^{(0,0)}_0 \exp (\alpha G^{(\lambda,\eta)} P^{(\lambda,\eta)}_0) \rho_{\text{at}}, A \rangle_{\text{evo}}.$$
Recall that \( t_{\mathcal{W}} \) depending on the pump frequency \( k, k' \) oscillating terms present in it, but it can be explicitly defined as follows.

Using (4.7), (4.27), (4.30) and (4.31) the assertion follows. □

The invariant spaces \( \mathcal{H}_0^{(\lambda, \eta)} \) associated with the projectors \( P_0^{(\lambda, \eta)} \) are, however, not explicit enough for practical purposes. Therefore, the next step is to represent the effective block–diagonal dynamics onto the explicitly known eigenspace \( \mathcal{H}_0^{(0,0)} \). To this end, we denote the restriction of \( G^{(\lambda, \eta)} \) onto the space \( \mathcal{H}_0^{(0,0)} \) by

\[
\Lambda^{(\lambda, \eta)} := P_0^{(0,0)} G^{(\lambda, \eta)} P_0^{(0,0)}. \tag{4.32}
\]

Observe that the semigroups generated by \( \Lambda^{(\lambda, \eta)} \) and \( P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} \) are very close to each other for small coupling constants \( |\lambda| \) and \( |\eta| \):

**Theorem 4.4 (Uniform norm estimates on semigroups)**

For any \( \varpi \in \mathbb{R} \), \( \varepsilon \in (0,1) \), and any \( \alpha \in \mathbb{R}_0^+ \), there is a constant \( C_{\varpi, \varepsilon} \in (0, \infty) \) not depending on \( \lambda, \eta, \) and \( \alpha \) such that

\[
\left\| \exp \left( \alpha \Lambda^{(\lambda, \eta)} \right) - \exp \left( \alpha P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} \right) \right\| \leq C_{\varpi, \varepsilon} |\lambda|^{2(1-\varepsilon)}.
\]

The proof of Theorem 4.4 needs some technical preparations. For the sake of clarity we defer it to Section 6.2. We stress that \( C_{\varpi, \varepsilon} \) can vary considerably with the pump frequency \( \varpi \).

Thus, Lemma 4.3 and Theorem 4.4 imply that the evolution of \( P_D (\rho (t)) \) can be approximated by the semigroup generated by \( \Lambda^{(\lambda, \eta)} \):

**Corollary 4.5 (Effective block–diagonal dynamics – II)**

Assume that \( \rho_{\text{at}} \in D \). The effective block–diagonal density matrix \( \{ D (\rho (t)) \}_{t \geq 0} \) satisfies the bound

\[
\left| \left\langle D (\rho (\alpha)), A \right\rangle_{\text{at}} - \left\langle P_0^{(0,0)} \exp \left( \alpha \Lambda^{(\lambda, \eta)} \right) P_0^{(0,0)} \rho_{\text{at}}, A \right\rangle_{\text{evo}} \right| \leq C_{\varpi, \varepsilon} |\lambda|^{2(1-\varepsilon)} \| A \|,
\]

for any \( A \in \mathcal{H}_{\text{at}} \), \( |\lambda| \) sufficiently small, any \( \varepsilon \in (0,1) \) and \( \alpha \in \mathbb{R}_0^+ \). Here, \( C_{\varpi, \varepsilon} \in (0, \infty) \) is a finite constant depending on the pump frequency \( \varpi \) and \( \varepsilon \) but not on \( \rho_{\text{at}}, A, \lambda, \eta, \) and \( \alpha \).

In Corollary 4.5 we approximate the effective block–diagonal dynamics by some time–evolution on the finite dimensional subspace \( \mathcal{H}_0^{(0,0)} \subsetneq \mathcal{H}_{\text{evo}} \). This Hilbert space is not a subspace of \( \mathcal{H}_{\text{at}} \subsetneq \mathcal{H}_{\text{evo}} \) because of oscillating terms present in it, but it can be explicitly defined as follows.

Recall that the eigenspaces of the atomic Hamiltonian \( H_{\text{at}} \in B(\mathbb{C}^d) \), associated with the eigenvalues \( E_k \) for \( k \in \{1, \ldots, N\} \), and their dimensions are denoted by \( \mathcal{H}_k \subset \mathbb{C}^d \) and \( n_k \in \mathbb{N} \), respectively. See Section 2.2. By taking any arbitrary orthonormal basis \( \{ e^{(k)}_n \}_{n=1}^{n_k} \) of \( \mathcal{H}_k \) for each \( k \in \{1, \ldots, N\} \) we define the elements

\[
W_{(k,n)}^{(k',n')} \in \mathcal{H}_{\text{at}} = B(\mathbb{C}^d)
\]

for any \( k, k' \in \{1, \ldots, N\}, n \in \{1, \ldots, n_k \} \) and \( n' \in \{1, \ldots, n_{k'} \} \) by

\[
\forall k'' \in \{1, \ldots, N\}, \quad n'' \in \{1, \ldots, n_{k''} \}: \quad W_{(k,n)}^{(k',n')} e^{(k'')}_{n''} := \delta_{n,n''} \delta_{k,k''} e^{(k')}_{n'} \tag{4.33}
\]

Then, straightforward computations show that the Hilbert space \( \mathcal{H}_0^{(0,0)} \) equals

\[
\mathcal{H}_0^{(0,0)} = \text{span} \left\{ e^{-it(E_{k'} - E_k) W_{(k,n)}^{(k',n')}} | (k, k') \in t_{\mathcal{W}} \cup t_0 \cup t_{\mathcal{W}}, \; n \in \{1, \ldots, n_k\}, \right. \quad n' \in \{1, \ldots, n_{k'}\} \}\right). \tag{4.34}
\]

Recall that \( t_{m\mathcal{W}} \) is the set defined by \( \mathcal{W} \) for \( \varepsilon = m\mathcal{W} \in \sigma([H_{\text{at}}, \cdot]) \) and \( m \in \{-1, 0, 1\} \), that is explicitly,

\[
t_{\mathcal{W}} = \{(1, N)\}, \quad t_0 = \{(j,j) : j \in \{1, 2, \ldots, N\}\}, \quad t_{\mathcal{W}} = \{(N,1)\}, \tag{4.34}
\]
because \( \varpi := E_N - E_1 > 0 \), see (2.10).

Obviously,  
\[
\mathcal{S}_0^{(0,0)} \cap \mathcal{S}_{\text{at}} = P_\mathcal{D} (\mathcal{S}_{\text{at}})
\]
and \( \mathcal{S}_0^{(0,0)} \not\subset \mathcal{S}_{\text{at}} \). Nevertheless, we can remove the oscillating terms by defining a unitary map \( U_0 \) from \( \mathcal{S}_0^{(0,0)} \) to the atomic subspace:

\[
\tilde{\mathcal{S}}_0^{(0,0)} := \text{span}\left\{ W_{(k',n')}^{(k,n)} \mid (k',k') \in \mathcal{T}_0 \cup \mathcal{T}_1, n \in \{1, \ldots, n_k\}, n' \in \{1, \ldots, n_{k'}\} \right\} \subset \mathcal{S}_{\text{at}} \quad (4.35)
\]
as follows:

\[
U_0 \left( e^{-i(t(E_k - E_{k'})} W_{(k,n)}^{(k',n')} \right) := W_{(k,n)}^{(k',n')} \in \tilde{\mathcal{S}}_0^{(0,0)}
\]
for any \( k, k' \in \{1, \ldots, N\}, n \in \{1, \ldots, n_k\} \) and \( n' \in \{1, \ldots, n_{k'}\} \). Clearly, \( P_\mathcal{D} (\mathcal{S}_{\text{at}}) \subset \tilde{\mathcal{S}}_0^{(0,0)} \) as \( U_0 P_\mathcal{D} (\mathcal{S}_{\text{at}}) = P_\mathcal{D} (\mathcal{S}_{\text{at}}) \).

Hence, by Corollary 4.3, the behavior of \( P_\mathcal{D} (\rho (t)) \) can be studied through the semigroup acting on \( \tilde{\mathcal{S}}_0^{(0,0)} \subset \mathcal{S}_{\text{at}} \) and generated by the operator

\[
\tilde{\Lambda}^{(\lambda,\eta)} := U_0 \Lambda^{(\lambda,\eta)} U_0^* .
\]
This follows, for any initial density matrix \( \rho_{\text{at}} \in \mathcal{S}_{\text{at}}, \) from the equality

\[
P_\mathcal{D} \left( \exp \left( \alpha \Lambda^{(\lambda,\eta)} \right) P_0^{(0,0)} (\rho_{\text{at}}) \right) = P_\mathcal{D} \left( U_0 \exp \left( \alpha \tilde{\Lambda}^{(\lambda,\eta)} \right) U_0 P_\mathcal{D} (\rho_{\text{at}}) \right) \\
= P_\mathcal{D} \left( \exp \left( \alpha \Lambda^{(\lambda,\eta)} \right) P_\mathcal{D} (\rho_{\text{at}}) \right) .
\]

As a first application of the above results we are now in position to study the large time behavior of the effective block–diagonal density matrix \( P_\mathcal{D} (\rho (t)) \):

**Theorem 4.6 (Large time behavior of \( P_\mathcal{D} (\rho (t)) \))**

(i) For all \( \rho \in \tilde{\mathcal{S}}_0^{(0,0)} \),

\[
\tilde{\Lambda}^{(\lambda,\eta)} (\rho) = \frac{\eta}{2} \mathcal{L}_\rho (\rho) + \lambda^2 \mathcal{L}_\mathcal{R} (\rho) \in \tilde{\mathcal{S}}_0^{(0,0)}
\]
with \( \mathcal{L}_\rho \) and \( \mathcal{L}_\mathcal{R} \) defined by (3.5) and (3.6), respectively.

(ii) Assume that \( \rho_{\text{at}} \in \mathcal{D} \). There is a unique density matrix \( \tilde{\rho}_\infty \in \tilde{\mathcal{S}}_0^{(0,0)} \) such that \( \tilde{\Lambda}^{(\lambda,\eta)} (\tilde{\rho}_\infty) = 0 \) and

\[
\limsup_{\alpha \to \infty} \| P_\mathcal{D} (\rho (\alpha)) - P_\mathcal{D} (\tilde{\rho}_\infty) \| \leq C_{\varpi,\varepsilon} |\lambda|^{2(1-\varepsilon)}
\]
for all \( |\lambda| \) sufficiently small and any \( \varepsilon \in (0,1) \). Here, \( C_{\varpi,\varepsilon} \in (0, \infty) \) is a finite constant depending on \( \varpi \) and \( \varepsilon \) but not on \( \rho_{\text{at}}, \lambda, \eta \).

**Proof.** (i) For any \( \rho \in \tilde{\mathcal{S}}_0^{(0,0)} \) and \( t \in [0,2\pi\varpi^{-1}] \) a.e., note that

\[
- \frac{d}{dt} [U_0 \rho \ast t] + \mathcal{L}_{\text{at}} [U_0 \rho \ast t] (t) = 0 .
\]

Then, by construction,

\[
\left[ \tilde{G}^{(\lambda,\eta)} P_0^{(0,0)} U_0^* \right] (t) = \frac{\eta}{2} \left( e^{i\pi \varpi t} + e^{-i\pi \varpi t} \right) \mathcal{L}_\rho \left[ U_0 \rho \ast t \right] (t) + \lambda^2 \mathcal{L}_\mathcal{R} \left[ U_0^* \rho \ast t \right] (t)
\]
for any \( \rho \in \tilde{\mathcal{S}}_0^{(0,0)} \) and \( t \in [0,2\pi\varpi^{-1}] \) a.e., see (3.3), (4.8), (4.10) and (4.20), (4.22). Using the explicit expressions of (3.12) and (3.13) one easily checks \( \mathcal{L}_\mathcal{R} U_0 \ast t = 0 \), which obviously implies that

\[
U_0 \mathcal{L}_\mathcal{R} U_0^* = \mathcal{L}_\mathcal{R} .
\]
On the other hand, we have

\[
(1 - 1[\mathfrak{L}_p = 0]) [U_0^t \rho] (t) = \begin{pmatrix}
P_{1,1} (\rho) & e^{it\pi} P_{1,2} (\rho) \\
0 & 0 \\
e^{-it\pi} P_{1,2} (\rho) & P_{1,1} (\rho)
\end{pmatrix},
\]

where \(1[\mathfrak{L}_p = 0]\) is the projection onto the kernel of \(\mathfrak{L}_p\) and \(P_{j,k}\) are the orthogonal projections

\[
P_{j,k} := 1[H_{at} = E_j] 1[H_{at} = E_k] \in \mathcal{B}(\mathcal{H}_{at})
\]

for all \(j, k \in \{1, \ldots, N\}\). Hence, we obtain

\[
P_0^{(0,0)} (e^{it\pi} + e^{-it\pi}) \mathfrak{L}_p [U_0^t \rho] (t)
= i \begin{pmatrix}
P_{1,1} (\rho) & h_p - h_p^* P_{1,2} (\rho) & e^{it\pi} (P_{1,1} (\rho) h_p^* - h_p^* P_{1,2} (\rho)) \\
0 & 0 & 0 \\
e^{-it\pi} (P_{1,2} (\rho) h_p^* - h_p^* P_{1,2} (\rho)) & 0 & P_{1,1} (\rho) h_p^* - h_p^* P_{1,2} (\rho)
\end{pmatrix}.
\]

Multiplying now \(U_0\) from the left in \(\ref{4.40}\) and using the computations above we arrive at

\[
U_0 P_0^{(0,0)} \mathcal{G}(\lambda, \eta) P_0^{(0,0)} U_0^* \rho = \frac{\eta}{2} \mathfrak{L}_p (\rho) + \lambda^2 \mathfrak{L}_R (\rho)
\]

for any \(\rho \in \tilde{\mathcal{H}}_0^{(0,0)}\).

(ii) Note that the bounded operator \(\tilde{\Lambda}^{(\lambda, \eta)}\) given by the equality of the first assertion (i) makes sense for all atomic density matrices \(\rho \in \mathcal{H}_{at}\), whereas \(\tilde{\mathcal{H}}_0^{(0,0)} \subset \mathcal{H}_{at}\) is an invariant space of \(\tilde{\Lambda}^{(\lambda, \eta)} \in \mathcal{B}(\mathcal{H}_{at})\). By Remark 3.1, Assumption 2, Theorem 6.10, and assertion (i), \(\tilde{\Lambda}^{(\lambda, \eta)} \in \mathcal{B}(\mathcal{H}_{at})\) is the generator of a relaxing Markov CP semigroup for all \(\lambda, \eta \in \mathbb{R}^2, \lambda \neq 0\). See Definition 6.9 and Remark 3.2. I.e., there is a unique density matrix \(\tilde{\rho}_\infty \in \mathcal{H}_{at}\) such that, for any \(\rho \in \mathcal{H}_{at}\),

\[
\lim_{\alpha \to \infty} \left( \exp \left( \alpha \tilde{\Lambda}^{(\lambda, \eta)} \right) \rho \right) = \tilde{\rho}_\infty.
\]

It follows that \(\tilde{\rho}_\infty \in \mathcal{H}_{at}\) is the unique density matrix satisfying \(\tilde{\Lambda}^{(\lambda, \eta)} (\tilde{\rho}_\infty) = 0\), see Theorem 6.10. As \(\tilde{\mathcal{H}}_0^{(0,0)}\) is an invariant space of \(\tilde{\Lambda}^{(\lambda, \eta)} \in \mathcal{B}(\mathcal{H}_{at})\) containing density matrices, one must have \(\tilde{\rho}_\infty \in \tilde{\mathcal{H}}_0^{(0,0)}\). Using \(\ref{4.38}\) and the fact that \(P_\mathcal{D} (\rho_{at}) \in \mathcal{H}_{at}\) is also a density matrix, we obtain

\[
\lim_{\alpha \to \infty} P_0^{(0,0)} \exp \left( \alpha \tilde{\Lambda}^{(\lambda, \eta)} \right) P_0^{(0,0)} (\rho_{at}) = P_\mathcal{D} (\tilde{\rho}_\infty).
\]

The inequality \(\ref{4.39}\) then results from Corollary 4.5 and the finite dimensionality of the Hilbert space \(\mathcal{H}_{at}\). □

It now remains to characterize more precisely the block diagonal projection \(\rho_\infty := P_\mathcal{D} (\tilde{\rho}_\infty) \in \mathcal{D}\) of the density matrix \(\tilde{\rho}_\infty\) of Theorem 4.6 (ii). This is done via a balance condition for populations in Theorem 4.7 below.

We first define the orthogonal projection

\[
P_\mathcal{D}^\perp := P_{\tilde{\mathcal{H}}_0^{(0,0)}} - P_\mathcal{D} \in \mathcal{B}(\mathcal{H}_{at})
\]

and the operator

\[
\mathcal{C} := P_\mathcal{D}^\perp \left( \frac{\eta}{2} \mathfrak{L}_p + \lambda^2 \mathfrak{L}_R \right) P_\mathcal{D}^\perp \in \mathcal{B}(\mathcal{H}_{at}).
\]

i.e., \(\mathcal{C} = P_\mathcal{D}^\perp \tilde{\Lambda}^{(\lambda, \eta)} P_\mathcal{D}^\perp\).

The operator \(\mathcal{C} \equiv \mathcal{C}_\lambda\) only depends on \(\lambda \in \mathbb{R}\): Observe that \(P_\mathcal{D}\) and \(P_\mathcal{D}^\perp\) are orthogonal projections onto the Hilbert space \(\tilde{\mathcal{H}}_0^{(0,0)} \subset \mathcal{H}_{at}\) for which \(\text{ran}(P_\mathcal{D})\) and \(\text{ran}(P_\mathcal{D}^\perp)\) are invariant spaces of \(\mathfrak{L}_R\). The operator \(\mathfrak{L}_p\) however maps the subspace \(\text{ran}(P_\mathcal{D})\) of block diagonal matrices to the subspace \(\text{ran}(P_\mathcal{D}^\perp)\) of off–diagonal matrices in \(\tilde{\mathcal{H}}_0^{(0,0)}\) and vice versa. In particular,

\[
\mathfrak{L}_p P_\mathcal{D} = P_\mathcal{D}^\perp \mathfrak{L}_p P_\mathcal{D} \quad \text{and} \quad P_\mathcal{D} \mathfrak{L}_p P_\mathcal{D} = 0,
\]
and hence,
\[ \mathcal{C} \equiv \mathcal{C}_\lambda = \lambda^2 P_\lambda^\dagger \mathcal{L}_R P_\lambda^\dagger \quad \text{and} \quad \mathcal{C}^* \equiv \mathcal{C}_\lambda^* = \lambda^2 P_\lambda^\dagger \mathcal{L}_R^* P_\lambda^\dagger \]

for all \( \lambda, \eta \in \mathbb{R} \).

Recall that, by Remarks 3.1 and Theorem 6.10, the atom–reservoir Lindbladian \( \mathcal{L}_R \) is the generator of a relaxing Markov CP semigroup on \( \mathfrak{h}_{\text{at}} \), and all non–zero elements \( p \in \sigma(\mathcal{L}_R) \setminus \{0\} \) have a strictly negative real part \( \text{Re} \,(p) < 0 \). On the other hand, explicit computations show that the density matrix \( \rho_g = P_\mathcal{D} (\rho_g) \) of the Gibbs state \( \rho_{\text{gat}} \) belongs to the kernel of \( \mathcal{L}_R \), i.e., \( \mathcal{L}_R (\rho_g) = 0 \), provided that the parameter \( \beta \in (0, \infty) \) in (2.5) is the inverse temperature of the reservoir. Thus,
\[ \ker \mathcal{L}_R = \mathbb{C} \cdot \mathbf{g}_{\text{at}} \subset \mathcal{D} \quad \text{and} \quad \ker \mathcal{L}_R^* = \mathbb{C} \cdot 1_{\mathcal{C}^2} \subset \mathcal{D} . \]

Note that the second equality is an obvious consequence of Theorem 6.7.

It follows that the operator \( \mathcal{C} \) and its adjoint \( \mathcal{C}^* \) are both invertible on the subspace \( \text{ran}(P_\mathcal{D}) \). Therefore, using (4.33) and standard results on Feshbach maps [37, Theorem 2.1, Remark 2.2], we deduce that \( \rho \mapsto P_\mathcal{D} (\rho) \) defines a one–to–one map from
\[ \ker \tilde{\Lambda}^{(\lambda, \eta)} \cap \tilde{\mathfrak{h}}_0 \subset \mathcal{D} \quad \text{to the space} \quad \ker \left( \lambda^2 \mathcal{L}_R + \frac{\eta^2}{4 \lambda^2} \mathfrak{B}_{p, R} \right) \cap \mathcal{D} , \]

with
\[ \mathfrak{B}_{p, R} := - \lambda^2 P_\mathcal{D} \mathcal{L}_p \left( \mathcal{C}_{\text{ran}(P_\mathcal{D})} \right)^{-1} \mathcal{L}_p P_\mathcal{D} \in \mathcal{B}(\mathfrak{h}_{\text{at}}) . \]  

(4.44)

By uniqueness of the density matrix \( \tilde{\rho}_\infty \in \tilde{\mathfrak{h}}_0 \) satisfying \( \tilde{\Lambda}^{(\lambda, \eta)} (\tilde{\rho}_\infty) = 0 \) (Theorem 4.6 (ii)), the following unique characterization of the populations \( \rho_\infty = P_\mathcal{D} (\tilde{\rho}_\infty) \) holds:

**Theorem 4.7 (Characterization of \( \rho_\infty \) via a balance condition)**

\( \rho_\infty = P_\mathcal{D} (\tilde{\rho}_\infty) \) is the unique (block diagonal) density matrix \( \rho_\infty \in \mathcal{D} \) satisfying the balance condition
\[ \mathcal{L}_R (\rho_\infty) + \frac{\eta^2}{4 \lambda^2} \mathfrak{B}_{p, R} (\rho_\infty) = 0 . \]  

(4.45)

As explained above, all non–zero elements \( p \in \sigma(\mathcal{L}_R) \setminus \{0\} \) have a strictly negative real part \( \text{Re} \,(p) < 0 \). In particular, there are constants \( C, c \in (0, \infty) \) such that
\[ \| e^{s \mathcal{C}_\lambda} P_\mathcal{D} \| \leq C e^{-sc} . \]  

(4.46)

As a consequence, by using (4.33) and expressing resolvents of generator of semigroups through Laplace transform, we can rewrite the operator \( \mathfrak{B}_{p, R} \), which describes the pump contribution to the (quasi–) steady populations, as
\[ \mathfrak{B}_{p, R} = \int_0^\infty \mathcal{L}_p e^{s \mathcal{C}_\lambda} \mathcal{L}_p P_\mathcal{D} \hspace{1em} ds \in \mathcal{B}(\mathfrak{h}_{\text{at}}) . \]  

(4.47)

This formulation is important to get the balance condition from a dynamical principle.

### 4.5 The pre–master equation and the balance condition

We analyze in the present section the time–evolution of density matrices
\[ \rho_\mathcal{D} (\alpha) := P_\mathcal{D} \exp \left( \alpha \tilde{\Lambda}^{(\lambda, \eta)} \right) P_\mathcal{D} (\rho_{\text{at}}) \in P_\mathcal{D} (\tilde{\mathfrak{h}}_0^{(0,0)}) . \]  

(4.48)

Since
\[ \| P_\mathcal{D} (\rho (\alpha)) - P_\mathcal{D} (\alpha) \| \leq C_{\infty, \epsilon} |\lambda|^{2(1-\epsilon)} \]
with $C_{\pi,\varepsilon} \in (0, \infty)$, $\varepsilon \in (0, 1)$, not depending on $\rho_{at}$, $A$, $\lambda$, $\eta$, and $\alpha$ (cf. Corollary 4.5 and (4.38)), the density matrix $\rho_D(\alpha)$ accurately describes the real evolution of atomic populations at small couplings. One important consequence of this analysis is the derivation, after Theorem 4.8, of the balance condition (4.45) from a dynamical principle. Indeed, $\rho_D(\alpha)$ satisfies an integro-differential equation, called pre-master equation, and Theorem 4.7 is equivalent to the fact that $\rho_{\infty}$ is the unique stationary state of the Markov approximation of this integro-differential equation.

**Theorem 4.8 (The pre-master equation)**
The family $\{\rho_D(\alpha)\}_{\alpha \geq 0} \subset D$ of block-diagonal density matrices obeys the integro-differential equation

$$\forall \alpha \geq 0 : \frac{d}{d\alpha} \rho_D(\alpha) = \lambda^2 \mathcal{L}_R (\rho_D(\alpha)) + \eta^2 \frac{\lambda^2}{4} \int_0^\alpha \mathcal{L}_p e^{s \mathcal{L}_R} \mathcal{L}_p (\rho_D(\alpha - s \lambda^{-2})) \, ds$$

(4.49)

with $\rho_D(0) = \rho_{at} \in D$.

**Proof.** The proof of this assertion is standard (see for instance [38, Chapter 7]) and is given here for completeness. The two-fold iteration of “variation of constants formula” yields the equality

$$e^{\alpha \tilde{\Lambda}^{(\lambda, \eta)}} = e^{\alpha \lambda^2 \mathcal{L}_R} + \eta \int_0^\alpha e^{(\alpha - s) \lambda^2 \mathcal{L}_R} \mathcal{L}_p e^{s \lambda^2 \mathcal{L}_R} \, ds$$

(4.50)

$$+ \frac{\eta^2}{4} \int_0^\alpha ds_1 \int_0^{s_1} ds_2 \, e^{(\alpha - s_1) \lambda^2 \mathcal{L}_R} \mathcal{L}_p e^{(s_1 - s_2) \lambda^2 \mathcal{L}_R} \mathcal{L}_p e^{s_2 \tilde{\Lambda}^{(\lambda, \eta)}}.$$

Using that

$$\mathcal{L}_R(P_D(\tilde{\delta}_0^{(0,0)})) \subset P_D(\tilde{\delta}_0^{(0,0)}), \quad \mathcal{L}_R(P_D^+(\tilde{\delta}_0^{(0,0)})) \subset P_D^+(\tilde{\delta}_0^{(0,0)})$$

and

$$\mathcal{L}_p(P_D(\tilde{\delta}_0^{(0,0)})) \subset P_D^+(\tilde{\delta}_0^{(0,0)}), \quad \mathcal{L}_p(P_D^+(\tilde{\delta}_0^{(0,0)})) \subset P_D(\tilde{\delta}_0^{(0,0)})$$

we readily deduce from (4.50) that

$$P_D e^{\alpha \tilde{\Lambda}^{(\lambda, \eta)}} P_D = e^{\alpha \lambda^2 \mathcal{L}_R} P_D$$

$$+ \frac{\eta^2}{4} \int_0^\alpha ds_1 \int_0^{s_1} ds_2 \, e^{(\alpha - s_1) \lambda^2 \mathcal{L}_R} \mathcal{L}_p e^{(s_1 - s_2) \lambda^2 \mathcal{L}_R} \mathcal{L}_p P_D e^{s_2 \tilde{\Lambda}^{(\lambda, \eta)}} P_D.$$

Deriving this last equation we get

$$\frac{d}{d\alpha} \rho_D(\alpha) = \lambda^2 \mathcal{L}_R (\rho_D(\alpha)) + \frac{\eta^2}{4} \int_0^\alpha \mathcal{L}_p e^{s \lambda^2 \mathcal{L}_R} \mathcal{L}_p (\rho_D(\alpha - s)) \, ds$$

from which we deduce the theorem by a trivial change of variable. \qed

By combining (4.42) with the equality $\tilde{\Lambda}^{(\lambda, \eta)}(\rho_{\infty}) = 0$ (Theorem 4.6 (ii)), the density matrix $\rho_D(\alpha)$ must converge to $P_D(\rho_{\infty})$ and its derivative must vanish in the limit $\alpha \to \infty$. By (4.47) and Lebesgue’s dominated convergence theorem, the limit $\rho_{\infty} = P_D(\rho_{\infty})$ must solve the balance condition (4.45), as already proven in Theorem 4.7.

**Remark 4.3 (Moderate optical pump)**
The balance condition shows that the contribution of the pump to the final atomic state is of order $\eta^2 / \lambda^4$ whereas the contribution of the atom–reservoir interaction is of order one (in the parameter $\eta^2 / \lambda^4$). As explained in Section 4.7, this justifies Assumption 4, that is, $|\eta| \leq C \lambda^2$. In particular, this regime follows Observation (b) given at the beginning of Section 2.
Because of Theorem 4.7, we can interpret \( \lambda^2 A_R \), with
\[
A_R := P_\square \mathcal{L}_R P_\square ,
\]
as spontaneous transition rates and \( \frac{\partial^2}{\partial x^2} \mathcal{B}_{p,R} \) as effective stimulated rates between atomic energy levels. In order to make this precise it is natural to impose that \( A_R \) and \( \mathcal{B}_{p,R} \) generate Markov semigroups on \( \mathcal{D} \) which preserves positivity.

The operator \( A_R \) has this property for any choice of parameters because \( \mathcal{L}_R \) generates a Markov CP semigroup, which preserves the subspace \( \mathcal{D} \). Note however that this feature is in general not satisfied by the operator \( \mathcal{B}_{p,R} \). A simple counter-example with \( N = 2 \) and \( d = 3 \), where one energy level is two-fold degenerated, is given in Section 6.4. This fact is not very surprising. Indeed, as discussed in Section 4.5, the balance condition comes from a Markov approximation of the restriction of a CP dynamics. It is well-known that this kind of construction can destroy positivity [3, Section III.1].

Hence, we will assume in this section the following:

**Assumption 3 (\( \mathcal{B}_{p,R} \) as transition rates)**

The operator \( \mathcal{B}_{p,R} \) defined by (4.44) is the restriction on \( \mathcal{D} \) of the generator of a Markov CP semigroup on \( \mathcal{B}(\mathbb{C}^d) \) with an invariant space \( \mathcal{D} \subseteq \mathcal{B}(\mathbb{C}^d) \). In particular, \( \mathcal{B}_{p,R} \) generates a Markov semigroup on \( \mathcal{D} \) which preserves positivity.

A sufficient condition on the Lindbladian \( \mathcal{L}_R \) to satisfy Assumption 3 is given by Theorem 6.11 in Section 6.4. It is always satisfied if the 1st and the \( N \)th atomic energy levels are non-degenerate. Indeed, the condition stated in Theorem 6.11 physically corresponds to the following:

- The pump is uniformly resonant, i.e., the reservoir–impurity interaction does not split the spectral line corresponding to the \( N–1 \) atomic transition. This atomic spectral line may however move as a whole under the influence of the reservoir (uniform Lamb shift).
- The decoherence time is uniform for the \( N–1 \) correlations, i.e., the reservoir does not induce a splitting of \( \text{ran}(P_{N,1}) \) in smaller independent coherence subspaces.

Assuming from now Assumption 3 we are in position to define in Section 5.1 what we call **generalized Einstein coefficients**. These coefficients yield the Pauli master equation and Einstein’s relations, respectively described in Sections 5.2 and 5.3.

### 5.1 Generalized Einstein coefficients

Recall that the left and right multiplications are defined by (2.3), and the orthogonal projections \( P_{j,k} \) are defined, for all \( j, k \in \{1, \ldots, N\} \), by (4.41). Clearly,
\[
1_{\mathcal{D}_m} = \sum_{j,k=1}^{N} P_{j,k} \quad \text{and} \quad A_R = \sum_{j,k \in \{1, \ldots, N\}} A_{j,k} ,
\]
where, for all \( j, k \in \{1, \ldots, N\} \), \( A_{j,k} := P_{j,k} \mathcal{L}_R P_{k,j} \). Since \( A_R \) generates a semigroup which always preserves positivity, for any \( j, k \in \{1, \ldots, N\} \) such that \( j \neq k \), the operator \( A_{j,k} := \lambda^2 A_{j,k} \) defines a map from \( \mathcal{B}^+(\mathcal{H}_j) \) to \( \mathcal{B}^+(\mathcal{H}_j) \) and hence can be interpreted as the spontaneous transition rate from the \( k \)th to the \( j \)th atomic energy level. Here, \( \mathcal{B}^+(\mathfrak{h}) \) denotes the set of positive operators on the Hilbert space \( \mathfrak{h} \). The operator \( A_{j,j} := \lambda^2 A_{j,j} \) for any \( j \in \{1, \ldots, N\} \) is then responsible for the trace preservation of the total dynamics generated by the operator \( \lambda^2 A_R \).

The spontaneous transition rates \( A_{j,k} \) can explicitly be computed from the quantities defining the microscopic model. By (3.4), (3.6), (3.11), (3.12) and (3.13), we have
\[
A_{j,k} = 2(1-\delta_{j,k}) \sum_{\ell=1}^{m} c_{j,k}^{(\ell)} V_{j,k} V_{j,k}^{(\ell)*} + \delta_{j,k} \sum_{\ell=1}^{m} 2c_{j,k}^{(\ell)} V_{j,k} V_{j,k}^{(\ell)*} - \sum_{\ell=1}^{N} c_{j,k}^{(\ell)} (V_{j,k} V_{j,k}^{(\ell)*}) + V_{j,k}^{(\ell)*} V_{j,k}^{(\ell)}
\]
for all \( j, k \in \{1, \ldots, N\} \) and with \( \delta_{j,k} \) being the Kronecker delta. With this expression, and the corresponding ones (5.2) for \( B_{j,k} \) below, we see what role is played by the dissipative effects due to the atom–reservoir interaction for the behavior of (quasi–) steady populations of optically pumped atomic energy levels. This is already mentioned in Observation (a) at the beginning of Section 2.

Analogously, define

\[
B_{N,1} := -P_{N,N} h_p (P_{N,1})^{-1} P_{N,1} h_p P_{1,1} - P_{N,N} h_p (P_{1,N})^{-1} P_{1,N} h_p P_{1,1} \quad \text{and} \quad B_{1,N} := -P_{1,1} h_p (P_{1,N})^{-1} P_{1,N} h_p P_{N,N} - P_{1,1} h_p (P_{N,1})^{-1} P_{N,1} h_p P_{N,N} ,
\]

\[
B_{N,N,N} := P_{N,N} h_p (P_{N,1})^{-1} P_{N,1} h_p P_{N,N} + P_{N,N} h_p (P_{1,N})^{-1} P_{1,N} h_p P_{N,N} \quad \text{and} \quad B_{1,1} := P_{1,1} h_p (P_{1,N})^{-1} P_{1,N} h_p P_{1,1} + P_{1,1} h_p (P_{N,1})^{-1} P_{N,1} h_p P_{1,1} .
\]

Recall that \( h_p \in \mathcal{B}(\mathbb{C}^d) \) maps \( \mathcal{H}_1 \) to \( \mathcal{H}_N \) and its kernel equals \( \ker (h_p) = \mathcal{H}_1^+ \), whereas \( h_p^* \) maps \( \mathcal{H}_N \) to \( \mathcal{H}_1 \) with \( \ker (h_p^*) = \mathcal{H}_N^+ \), see Section 2.3. Observe meanwhile that \( \text{ran} (P_{1,N}) \), \( \text{ran} (P_{N,1}) \subset \text{ran} (P_{\mathcal{B}}) \) are invariant spaces of the Lindbladian \( \mathcal{L}_\mathcal{R} \). As a consequence, from (2.11), (5.5) and (4.44),

\[
\mathcal{B}_{p,R} = B_{N,1} + B_{1,N} + B_{N,N} + B_{1,1} .
\]

For any \( j, k \in \{1, N\} \) such that \( j \neq k \), the operator

\[
B_{j,k} := \frac{\eta^2}{4\lambda^2} B_{j,k}
\]

maps \( \mathcal{B}^+(\mathcal{H}_k) \) to \( \mathcal{B}^+(\mathcal{H}_j) \) and is associated with the stimulated transition rate from the \( k \)th to the \( j \)th atomic energy level. Similar to the spontaneous transition rates, the operators \( B_{j,j} := \frac{\eta^2}{4\lambda^2} B_{j,j} \) are such that the full dynamics generated by \( \frac{\eta^2}{4\lambda^2} B_{p,R} \) preserves traces.

The objects \( A_{j,k}, B_{j,k} \) seen as maps \( K_k \to K_k \) between cones \( K_k \subset \mathcal{B}^+(\mathcal{H}_k) \), \( K_j \subset \mathcal{B}^+(\mathcal{H}_j) \) are called here \textit{generalized Einstein coefficients}. They satisfy strong constraints, named here \textit{generalized Einstein relations}, which have consequences for the structure of the (quasi–) steady populations \( \rho_\infty \) through the corresponding balance condition satisfied by \( \rho_\infty \). These relations are discussed in Section 5.3 below, after introducing the Pauli master equation.

### 5.2 The Pauli master equation

By Theorem 4.7 we can see the unique (quasi–) steady populations \( \rho_\infty \) as the stationary state of the \textit{phenomenological (quantum) Pauli master equation}

\[
\forall t \geq 0 : \quad \frac{d}{dt} \varrho(t) = \lambda^2 \mathcal{B}_{pR} (\varrho(t)) + \frac{\eta^2}{4\lambda^2} \mathcal{B}_{p,R} (\varrho(t)) , \quad \varrho(0) = P_\mathcal{D}(\rho_{\text{at}}) ,
\]

which generalizes the usual (classical) Pauli equation for populations found in standard textbooks on laser physics.

From Assumption 3 and Theorem 6.10 the unique solution \( \varrho(t) = P_\mathcal{D} (\varrho(t)) \) of the Pauli master equation converges to the unique density matrix \( \rho_\infty \) solution of
the balance condition (4.45) as \( t \to \infty \). By Theorems 5.3 and 4.6 (ii), one thus extracts from the Pauli master equation the correct asymptotic behavior of the atomic dynamics at small couplings and large times:

\[
\limsup_{t \to \infty} \| P_D (\rho_{at} (t)) - \varrho (t) \| \leq C_{\varpi, \varepsilon} | \lambda |^{2(1 - \varepsilon)}
\]

for some finite constant \( C_{\varpi, \varepsilon} \in (0, \infty) \) depending on \( \varpi \) and \( \varepsilon \in (0, 1) \) but not on \( \rho_{at} \), \( \lambda \), and \( \eta \).

Observe also that the Pauli master equation given the true effective dynamics when \( \eta = 0 \), i.e., when there is no optical pump. In this case, the master, pre–master and Pauli equations are the same. However, for \( \eta \neq 0 \), the dynamics governed by (5.4) is generically quite different from the time evolution of the density matrix \( \{ \rho_D (\alpha) \}_{\alpha \geq 0} \), which corresponds – up to small errors – to the real atomic dynamics \( \{ P_D (\rho_{at} (t)) \}_{t \geq 0} \), cf. Theorem 3.3 and 4.6 (ii). See, for instance, the numerical examples in Section 4.1.

Furthermore, the invariance of the family \( \{ \rho_D (\alpha) \}_{\alpha \geq 0} \) is governed by a (pre–master) integro–differential equation (4.49), the Pauli master equation being its Markov approximation. Thus, the smaller the decoherence rates of the \( N-1 \) correlations as compared to the spontaneous transition rates \( A_{j,k} \) of the atom, the less accurate is the dynamics given by the Pauli master equation compared to the microscopic atomic dynamics. (The decoherence rate and Lamb shift of the \( N-1 \) correlations correspond in Theorem 6.11 to \( | \text{Re} \{ \xi_{N,1} \} | \) and \( | \text{Im} \{ \xi_{N,1} \} | \), respectively.)

By Assumption \( 3 \) the solution of (5.4) evolves in the positive cone

\[
\mathcal{D}^+ := \mathcal{B}^+ (\mathbb{C}^d) \cap \mathcal{D} = \co \left\{ \bigcup_{k=1}^N \mathcal{B}^+ (\mathcal{H}_k) \right\}.
\]

Here, \( \co (\mathcal{m}) \) stands for the convex hull of the set \( \mathcal{m} \). By using the spontaneous and stimulated atomic transition rates \( A_{j,k}, B_{j,k} : \mathcal{B}^+ (\mathcal{H}_k) \to \mathcal{B}^+ (\mathcal{H}_j) \) defined in Section 5.1 the Pauli master equation (5.4) reads, for all \( j \in \{ 1, \ldots, N \} \),

\[
\forall t \geq 0 : \quad \frac{d}{dt} \varrho_j (t) = \sum_{k=1}^N (A_{j,k} + B_{j,k}) \varrho_k (t), \quad \varrho_j (0) = \varrho_j \in \mathcal{B}^+ (\mathcal{H}_j) \tag{5.5}
\]

with \( \varrho_j (t) = P_{j,j} (\varrho (t)) \) and where \( B_{j,k} := 0 \) if \( \{ j, k \} \not\subset \{ 1, N \} \).

In many situations, for instance in presence of symmetries, \( A_{j,k} \) and \( B_{j,k} \) define maps from subcones \( K_k \subseteq \mathcal{B}^+ (\mathcal{H}_k) \) to subcones \( K_j \subseteq \mathcal{B}^+ (\mathcal{H}_j) \) and we can write the evolution equation (5.5) with initial conditions \( \varrho_j (0) = \varrho_j \in K_j \) for \( j \in \{ 1, \ldots, N \} \). The unique solution of this initial value problem satisfies in this case \( \{ \varrho_j (t) \}_{t \geq 0} \subset K_j \) for all \( j \in \{ 1, \ldots, N \} \). If the initial density matrix \( \varrho_{at} \in \mathcal{D}^+ \) is chosen such that \( P_{j,j} (\varrho_{at}) \in K_j \) for all \( j \in \{ 1, \ldots, N \} \), the \( j \)th population \( \varrho_j (t) \) then converges to \( P_{j,j} (\varrho_{\infty}) \subset K_j \). This system of \( N \) differential equations is then the Pauli master equation of the invariant family \( \{ K_k \}_{k=1}^N \) of subcones \( K_k \subseteq \mathcal{B}^+ (\mathcal{H}_k) \).

A simple, sufficient and necessary condition on subcones \( \{ K_k \}_{k=1}^N \) to ensure that \( A_{j,k} \) and \( B_{j,k} \) both map \( K_k \) to \( K_j \) is given in the following definition:

**Definition 5.1 (Invariant family of cones)**

A family \( \{ K_k \}_{k=1}^N \) of subcones \( K_k \subseteq \mathcal{B}^+ (\mathcal{H}_k) \) is an invariant family whenever

\[
K := \co \left\{ \bigcup_{k=1}^N K_k \right\}
\]

is invariant under the action of the semigroups \( \{ e^{t \mathfrak{A}_R} \}_{t \geq 0} \) and \( \{ e^{t \mathfrak{A}_p, \mathbb{R}} \}_{t \geq 0} \).

By the Trotter product formula, observe that the subset \( K \) defined in this definition is also invariant under the action of the semigroup

\[
\left\{ \exp \left( t (\lambda^2 \mathfrak{A}_R + \frac{\eta^2}{4 \lambda^2} \mathfrak{A}_p, \mathbb{R}) \right) \right\}_{t \geq 0}
\]

Furthermore, the invariance of the family \( \{ K_k \}_{k=1}^N \) yields

\[
\forall j, k \in \{ 1, \ldots, N \} : \quad A_{j,k} (K_k), B_{j,k} (K_k) \subset K_j.
\]
Conversely, if a family \( \{ K_k \}_{k=1}^N \) of subcones \( K_k \subset B^+(\mathcal{H}_k) \) is such that \( A_{j,k} \) and \( B_{j,k} \) map \( K_k \) to \( K_j \) then \( \{ K_k \}_{k=1}^N \) is clearly an invariant family.

One trivial example of an invariant family of cones is given by taking \( K_k = B^+(\mathcal{H}_k) \) for all \( k \in \{1, \ldots, N\} \). But the smaller the dimension of such cones is, the more classical is the description of the final state via the transition rates \( \{ A_{j,k} \}_{j,k=1}^N \) and \( \{ B_{j,k} \}_{j,k=1}^N \). It can even happen that the dimension of all subcones \( \{ K_k \}_{k=1}^N \) can be chosen to be one. The latter trivially happens with \( K_k = B^+(\mathcal{H}_k) \) when all atomic levels are non-degenerated, i.e., the dimension \( n_k \) of the eigenspace \( \mathcal{H}_k \) is one for all \( k \in \{1, \ldots, N\} \). Note however that the non-degeneracy of all atomic energy levels is not a necessary condition for the existence of such one-dimensional cones and under certain circumstances the fully classical picture of the (quasi-) steady populations \( \rho_\infty \) is valid. In such a case the results of this paper directly relate the coefficients corresponding to the classical Pauli equation to microscopic quantities.

### 5.3 Einstein’s relations

By (5.2)–(5.3),

\[
B_{j,k} = \eta^2 F_{j,k}(\hat{A}_{N,1}, \hat{A}_{1,1}), \quad \text{with} \quad \hat{A}_{j,k} := \lambda^2 P_{j,k} \mathcal{L}_R P_{j,k}.
\]

In other words, \( B_{j,k} \) is proportional to the intensity \( \eta^2 \) of the pumping (monochromatic) light and proportional to a fixed function \( F_{j,k}(\hat{A}_{N,1}, \hat{A}_{1,1}) \) of the spontaneous “off-diagonal” transition rates \( \hat{A}_{N,1} \) and \( \hat{A}_{1,1} \). Einstein derived similar relations, called here Einstein AB–relations, for an atom interacting with a (broad–band, i.e., non–monochromatic) black–body radiation field in his seminal paper [11]. This was performed by using phenomenological considerations about the expected final state of the atomic populations and the asymptotics of the light intensity at large wave–numbers (Maxwell distribution).

Note that \( F_{j,k} \) strongly depends on the specific setting. The function \( F_{j,k} \) appearing in the present paper cannot be compared to the one appearing in Einstein’s work. However, the fact that the stimulated, operator–value coefficients \( B_{j,k} \) only depend on light intensity and spontaneous (operator–valued) coefficients \( \hat{A}_{j,k} \) seems to be universal. We stress that this property is rigorously derived here from a microscopic quantum mechanical description of the system under consideration and not from phenomenological assumptions.

Einstein also gives in his work a relation between the stimulated transition rates \( B_{j,k} \) and \( B_{k,j} \): Denoting the degeneracy of the \( k \)th atomic level by \( n_k \), he obtained the equations

\[
\forall j, k \in \{1, \ldots, N\} : \quad n_k B_{j,k} = n_j B_{k,j},
\]

named here Einstein BB–relations.

Let \( p_k \) denote the population in the \( k \)th atomic level and define the stimulated flux from the \( k \)th to \( j \)th atomic level by \( f_{j,k} := B_{j,k} p_k \). Then the Einstein BB–relations for fluxes reads

\[
\forall j, k \in \{1, \ldots, N\} : \quad n_j f_{j,k} - p_k n_j f_{k,j} = 0.
\]

(5.6)

In our setting, a variant of BB–relations holds for fluxes, at least for density matrices in the subcone

\[
K^0 := \text{co} \left( \bigcup_{k=1}^N K_k^0 \right),
\]

where, for all \( k \in \{1, \ldots, N\} \),

\[
K_k^0 := \mathbb{R}_0^+ \cdot 1 \left[ H_{\text{at}} = E_k \right] \subset B^+(\mathcal{H}_k).
\]

In this context and for any density matrix \( \rho \in \mathcal{S}_{\text{at}} \), the population in the \( k \)th atomic level is naturally defined to be

\[
p_k(\rho) = \text{Tr}_{C^d}(P_{k,k}(\rho)) = \text{Tr}_{C^d}(1 \left[ H_{\text{at}} = E_k \right] \rho 1 \left[ H_{\text{at}} = E_k \right]) \geq 0
\]

for \( k \in \{1, \ldots, N\} \), see (2.9). Similarly, for all \( j, k \in \{1, \ldots, N\} \)

\[
F_{j,k}(\rho) := \text{Tr}_{C^d}(B_{j,k}(\rho)) = \text{Tr}_{C^d}(B_{j,k} P_{k,k}(\rho))
\]

represents the stimulated flux from the \( k \)th to the \( j \)th atomic level with respect to the density matrix \( \rho \in \mathcal{S}_{\text{at}} \). Then one proves the following:
Proposition 5.2 (Einstein BB–relations for states in $K^0$)
For any $\rho \in K^0$,\[ \forall j, k \in \{1, \ldots, N\} : \quad p_j(\rho)n_kf_{j,k}(\rho) - p_k(\rho)n_jf_{k,j}(\rho) = 0 . \]

Proof. Clearly, for any $k \in \{1, \ldots, N\}$ and $\rho \in K^0$,\[ n_kP_{k,k}(\rho) = p_k(\rho)1[H_{at} = E_k] . \]
As a consequence, it suffices to prove the equality\[ \text{Tr}_{\mathbb{C}^d}(B_{j,k}(1[H_{at} = E_k])) = \text{Tr}_{\mathbb{C}^d}(B_{k,j}(1[H_{at} = E_j])) . \]
Recall that $B_{j,k}$ equals $B_{j,k} = 0$ for all $j \in \{2, \ldots, N - 1\}$ or $k \in \{2, \ldots, N - 1\}$. So, we only need to prove (5.7) for $j, k \in \{1, N\}$ and $j \neq k$. By (5.2),
\[ B_{1,N}(1[H_{at} = E_N]) = -\frac{\eta^2}{4\lambda^2} \left( h_p^*(E_{ran(P_{\beta}^\perp)})^{-1}(h_p) + (E_{ran(P_{\beta}^\perp)})^{-1}(h_p^*)h_p \right) , \]
\[ B_{N,1}(1[H_{at} = E_1]) = -\frac{\eta^2}{4\lambda^2} \left( h_p(E_{ran(P_{\beta}^\perp)})^{-1}(h_p^*) + (E_{ran(P_{\beta}^\perp)})^{-1}(h_p)h_p^* \right) . \]
Therefore, using the cyclicity of the trace and the identity\[ (E_{ran(P_{\beta}^\perp}))^{-1}(A) = \left[ (E_{ran(P_{\beta}^\perp}))^{-1}(A)^* \right]^* , \quad A \in \text{ran}(P_{\beta}^\perp) \subset \mathcal{H}_{at} , \]
the assertion (5.7) follows for $j, k \in \{1, N\}$ and $j \neq k$. \qed

Now, without any optical pump, i.e., for $\eta = 0$, the final density matrix $\tilde{\rho}_\infty$ belongs to the subcone $K^0$. More precisely, in this case $\rho_\infty = \rho_0 \in K^0$ is the atomic Gibbs state (2.5) with same inverse temperature $\beta$ as the one of the reservoir. As a consequence, we infer from Proposition 5.2 together with Kato’s perturbation theory [20] for non–degenerated eigenvectors that approximated Einstein BB–relations hold for the (quasi–) steady populations $\rho_\infty$ for weak pumps.

Corollary 5.3 (Einstein BB–relations at weak optical pump)
For all $(\eta, \lambda) \in \mathbb{R}^2$,\[ \forall j, k \in \{1, \ldots, N\} : \quad |p_j(\rho_\infty)n_kf_{j,k}(\rho_\infty) - p_k(\rho_\infty)n_jf_{k,j}(\rho_\infty)| \leq C_{\infty} \frac{\eta^2}{\lambda^2} . \]
Here, $C_{\infty} \in (0, \infty)$ is a constant depending on $\infty$ but not on $j, k, \rho_{at}, \lambda$ and $\eta$.

6. Appendix

We give in Section 6.1 a heuristic derivation of the time–dependent Lindbladian $L^{(\lambda, \eta)}$ (3.3) driving the effective atomic master equation (3.15) as well as the proof of Theorem 4.4 in Section 6.2. Section 6.3 is a short review on completely positive (CP) semigroups by focussing on results which are relevant for our analysis in order to facilitate the reading of the paper. Finally, in Section 6.4 we discuss positivity questions related to the interpretation as transitions rates of the coefficients of the balance condition (4.45) characterizing uniquely the (quasi–) steady populations.

6.1 Heuristic derivation of the time–dependent Lindbladian
1. It is natural to use the GNS representation $(\mathcal{H}_0, \pi, \Omega_0)$ of the initial state $\omega_0 := \omega_{at} \otimes \omega_{\mathcal{R}}$ (2.15) of the system because it provides a Hilbert space structure which simplifies the analysis [10]. Note that $\mathcal{H}_0 := \mathcal{H}_{at} \otimes \mathcal{H}_{\mathcal{R}}$, $\pi := \pi_{at} \otimes \pi_{\mathcal{R}}$, and $\Omega_0 := \Omega_{at} \otimes \Omega_{\mathcal{R}}$, where $(\mathcal{H}_{at}, \pi_{at}, \Omega_{at})$ and $(\mathcal{H}_{\mathcal{R}}, \pi_{\mathcal{R}}, \Omega_{\mathcal{R}})$ are the GNS representations of $\omega_{at}$ and $\omega_{\mathcal{R}}$. As $\omega_{at}$ is assumed to be faithful, the initial state $\omega_0$ is also faithful. In particular, $\pi$ is injective, so, for simplicity, $\pi(A)$ and $\pi(V)$ are denoted by $A$ and $V$. Moreover, the cyclic vector $\Omega_0$ of the GNS representation
is in this case separating for $\mathcal{M}$, i.e., $\mathcal{A}\Omega_0 = 0$ implies $A = 0$. Here, the von Neumann algebra $\mathcal{M} := \mathcal{V}'$ is the weak closure of the $C^*$-algebra $\pi(\mathcal{V})$. The state $\omega_0$ on $\mathcal{V}$ extends uniquely to a normal state on $\mathcal{M}$. This extension is again denoted by $\omega_0$.

If $\omega_{\text{at}} = g_{\text{at}}$ is the Gibbs state (cf. (2.5)) then $\omega_0$ is a $(\beta, \tau)$–KMS state, where $\tau := \{\tau_t\}_{t \in \mathbb{R}}$ is the strongly continuous one–parameter group of automorphisms of $\mathcal{V}$ defined by (2.14). As a consequence, by [22, Corollary 5.3.4], $\tau$ also extends uniquely to a $\sigma$–weakly continuous $\ast$–automorphism group on $\mathcal{M}$, which is also denoted by $\tau := \{\tau_t\}_{t \in \mathbb{R}}$. Moreover, this extension has a unique unitary representation $\{U_t\}_{t \in \mathbb{R}}$, i.e.,

$$\forall t \in \mathbb{R}, \ A \in \mathcal{M}: \quad \tau_t(A) = U_t A U_t^*, \tag{6.1}$$

such that $U_t \Omega_0 = \Omega_0$. As $t \mapsto \tau_t$ is $\sigma$–weakly continuous, the map $t \mapsto U_t$ is strongly continuous. Therefore, the unitary group $\{U_t\}_{t \in \mathbb{R}}$ has an anti–self–adjoint operator $iL$ as generator, i.e., $U_t = e^{itL}$. In particular, $\Omega_0 \in \text{Dom}(L)$ and $L$ annihilates $\Omega_0$, i.e., $L\Omega_0 = 0$.

Now, in the case where $\omega_{\text{at}}$ is any faithful state of the atom, observe that the space $\mathcal{H}_{\text{at}} := \mathcal{H}_{\text{at}} \otimes \mathcal{H}_R$ as well as the map $\pi := \pi_{\text{at}} \otimes \pi_R$ do not depend on $\omega_{\text{at}}$. Only the cyclic vector $\Omega_0 := \Omega_{\text{at}} \otimes \Omega_R$ of the GNS representation of $\omega_0$ changes. In particular, the von Neumann algebra $\mathcal{M}$ and the corresponding extension of the $\ast$–automorphism group $\tau$ do not depend on the initial state $\omega_{\text{at}}$ of the atom. Therefore, we can take any faithful state $\omega_{\text{at}}$ and use the operators $\{\tau_t\}_{t \in \mathbb{R}}$ acting on $\mathcal{M}$, and the operators $\{U_t\}_{t \in \mathbb{R}}$ and $L$ acting on $\mathcal{H}$ defined above for $\omega_{\text{at}} = g_{\text{at}}$. Note however that the equality $L\Omega_0 = 0$ does not hold, in general.

**2.** We then use the (Tomita–Takesaki) modular objects $\Delta$, $J$ of the pair $(\mathcal{M}, \Omega_0)$ to generalize [40], where the notion of $C$–Liouvilleans has been first introduced to investigate non-equilibrium stationary states, to the case of time–dependent $C$–Liouvilleans. For the detailed proofs of the results stated above we refer to the paper [1]. See also [2] for further applications of time-dependent $C$–Liouvilleans.

The pump term and the atom–reservoir interaction are taken into account by defining a time–dependent $C$–Liouvillean

$$\mathcal{L}_t = \mathcal{L}_t^{(\lambda, \eta)} := iL + iH_{\text{at}} \otimes 1_{\mathcal{H}_R} - iJ \Delta^{1/2} (H_{\text{at}} \otimes 1_{\mathcal{H}_R}) \Delta^{-1/2} J + iW_t - iJ \Delta^{1/2} W_t \Delta^{-1/2} J \tag{6.1}$$

with

$$W_t := \eta \cos(\varpi t) H_{\text{p}} + \lambda \sum_{\ell = 1}^m Q_{\ell} \otimes \Phi(\ell).$$

Indeed, we can prove that the time–dependent $C$–Liouvillean $\mathcal{L}_t$ generates an evolution family $\{U_{t,s}^{(\lambda, \eta)}\}_{t \geq s}$ representing in the GNS space $\mathcal{H}$ the strongly continuous two–parameter family $\{\tau^{(\lambda, \eta)}_{t,s}\}_{t \geq s}$ generated by the symmetric derivation $\delta_t^{(\lambda, \eta)}$ (2.17). More precisely, we show that the non–autonomous evolution equations

$$\forall t > s: \quad \partial_t U_{t,s}^{(\lambda, \eta)} = \mathcal{L}_t U_{t,s}^{(\lambda, \eta)}, \quad \partial_s U_{t,s}^{(\lambda, \eta)} = -U_{t,s}^{(\lambda, \eta)} \mathcal{L}_s, \quad U_{s,s} := 1 \tag{6.2}$$

on $\text{Dom}(L)$ have a unique solution $\{U_{t,s}^{(\lambda, \eta)}\}_{t \geq s} \subset \mathcal{B}(\mathcal{H})$. The operator $U_{t,s}^{(\lambda, \eta)}$ possesses a bounded inverse $(U_{t,s}^{(\lambda, \eta)})^{-1}$ for any $t \geq s$ and is $2\pi \varpi^{-1}$–periodic in the following sense:

$$\forall t \geq s: \quad U_{t,s}^{(\lambda, \eta)} = U_{t+2\pi \varpi^{-1}, s+2\pi \varpi^{-1}}^{(\lambda, \eta)}.$$

Then, one finally proves that $U_{t,s}^{(\lambda, \eta)} \Omega_0 = \Omega_0$ and

$$\forall t \geq s, \ A \in \mathcal{V}: \quad \tau^{(\eta, \lambda)}_{t,s}(A) = U_{t,s}^{(\lambda, \eta)} A (U_{t,s}^{(\lambda, \eta)})^{-1}. \tag{6.3}$$

**3.** To exploit these facts and obtain the atomic time–dependent Lindbladian described in Section 3.1 we use an explicit GNS representation of the initial state under consideration. As explained above, this GNS representation includes of course the GNS representation of $\omega_{\text{at}}$ already described in Section 2.2 and so, it remains to give
the an explicit GNS representation \((\Omega_R, \pi_R, \Omega_R)\) of the (faithful) quasi–free equilibrium state \(\omega_R\) of the fermionic reservoir at inverse temperature \(\beta \in (0, \infty)\).

The usual explicit form of the GNS representation of fermion quasi–free states is the Araki–Wyss representation \([39]\). Here we use however the (unitarily equivalent) representation introduced by Jakšić and Pillet \([40]\), the so–called Jakšić–Pillet glued representation, because it is well–adapted to spectral deformation methods.

So, consider the Hilbert space

\[
b_2 := L^2(\mathbb{R} \times S^2, \mathbb{C}),
\]

where \(S^2 \subset \mathbb{R}^3\) is the two–dimensional unit sphere centered at the origin and \(\mathbb{R} \times S^2\) (spherical coordinates of \(\mathbb{R}^3 \times \mathbb{R}^3\)) and is equipped with the measure \(d\lambda \otimes d^2s\). Here, \(d^2s\) is the usual rotation invariant measure of \(S^2\) induced by the Euclidean norm of \(\mathbb{R}^3\) and \(d\lambda\) is the Lebesgue measure. In the Jakšić–Pillet glued representation, the corresponding Hilbert space is the antisymmetric Fock space

\[
\mathcal{F}_R := \mathcal{F}_-(b_2).
\]

The cyclic vector \(\Omega_R\) is the vacuum of \(\mathcal{F}_-(b_2)\). The representation map \(\pi_R\) of the fermionic \(C^*\)–algebra \(\mathcal{V}_R\) is the \(C^*\)–homomorphism uniquely defined by

\[
\forall f \in b_1 : \quad \pi_R(a(f)) = a(g_f),
\]

where \(g_f \in b_2\) is given by

\[
\forall (p, \vartheta) \in \mathbb{R} \times S^2 : \quad g_f(p, \vartheta) = |p| (1 + e^{-\beta p})^{-1/2} \begin{cases} f(p \vartheta), & p \geq 0, \\ \overline{f(-p \vartheta)}, & p < 0. \end{cases}
\]

See also \((2.16)\).

4. We can now explicitly give the time–dependent \(C^*\)–Liouvillean \(\mathcal{L}_t\). By Lemma \(2.1\)

\[
L = L_{\text{at}} \otimes 1_\mathcal{R} + 1_\mathcal{R} \otimes L_R = (H_\text{at} - H_\text{at}) \otimes 1_\mathcal{R} + 1_\mathcal{R} \otimes d\Gamma(p).
\]

See also \((2.8)\).

The time–dependent \(C^*\)–Liouvillean \(\mathcal{L}_t\) then equals \((0.1)\) with

\[
W_t = \eta \cos(\varpi t) H_\text{at} \otimes 1_\mathcal{R} + \lambda \sum_{\ell=1}^m Q_\ell \otimes \frac{1}{\sqrt{2}}(a(g_\ell) + a^+(g_\ell))
\]

and

\[
J \Delta^{1/2} W_t \Delta^{-1/2} J = \eta \cos(\varpi t) \rho_{\text{at}}^{-1/2} H_\text{at} \rho_{\text{at}}^{1/2} \otimes 1_\mathcal{R}
\]

\[
+ i \lambda \sum_{\ell=1}^m \rho_{\text{at}}^{-1/2} Q_\ell \rho_{\text{at}}^{1/2} \otimes \frac{(-1)^{R(\text{tr} + 2)}}{\sqrt{2}} (a(e^{-\beta p \tilde{g}_\ell}) + a^+(i \tilde{g}_\ell))
\]

\(\Delta\)
Here, \( g_t(p, \partial) := g_t(p) \) (see 2.16) and \( d\Gamma(1_{h_2}) \) is the second quantization of \( 1_{h_2} \), i.e., the particle number operator acting on the antisymmetric Fock space \( \mathcal{F}_R := \mathcal{F}_-(h_2) \).

5. We identify the Hilbert spaces \( \mathcal{H}_{\text{at}} \otimes \{ \Omega_R \} \subset \mathcal{H} \) and \( \mathcal{H}_{\text{at}} \) and define the orthogonal projector \( P_{\text{at}} \in \mathcal{B}(\mathcal{H}) \) on \( \mathcal{H}_{\text{at}} \equiv \mathcal{H}_{\text{at}} \otimes \{ \Omega_R \} \). Next introduce the operator \( L^{(c)}_R(t) \in \mathcal{B}(\mathcal{H}_{\text{at}}) \) defined, for each \( t \in \mathbb{R} \) and any \( \epsilon > 0 \), by

\[
L^{(c)}_R := -i \sum_{\epsilon \in \sigma(L_{\text{at}})} P^{(c)}_{\text{at}} \left( \frac{\partial}{\partial \lambda} V_t \right) (L - \epsilon + i\epsilon)^{-1} \left[ \frac{\partial}{\partial \lambda} V_t \right] P^{(c)}_{\text{at}}
\]

with

\[
V_t := W_t - J \Delta^{1/2} W_t \Delta^{-1/2} J.
\]

Here, \( P^{(c)}_{\text{at}} \) is the Riesz projector related to the eigenvalue \( \epsilon \in \sigma(L_{\text{at}}) = \sigma([H_{\text{at}}, \cdot]) \) of the free \( C \)-Liouvillean

\[
\hat{L}_{\text{at}} : \mathcal{H}_{\text{at}} \otimes \{ \Omega_R \} \rightarrow \mathcal{H}_{\text{at}} \otimes \{ \Omega_R \}
\]

defined by

\[
\hat{L}_{\text{at}} := i H_{\text{at}} \otimes 1_{\mathcal{H}_R} - i J \Delta^{1/2} (H_{\text{at}} \otimes 1_{\mathcal{H}_R}) \Delta^{-1/2} J.
\]

Observe that

\[
\hat{L}_{\text{at}}(A \Omega_0) = (L_{\text{at}} A) \Omega_0
\]

for all \( A \in \mathcal{B}(\mathbb{C}^d) \) and thus \( \sigma(\hat{L}_{\text{at}}) = \sigma(L_{\text{at}}) \). Note also that \( L^{(c)}_R \) does not depend on the time \( t \).

It is easy to check that the following limit exists

\[
L_R := \lim_{\epsilon \to 0} L^{(c)}_R.
\]

This limit operator \( L_R \) is called the level-shift operator of the atom–reservoir interaction at time \( t \in \mathbb{R} \). See, e.g., [27, Eq. (3)].

By replacing the terms in \( L_t \) containing the coupling constant \( \lambda \) by the corresponding level–shift operator \( \lambda^2 L_R \) we obtain a new generator \( L^{(\text{eff})}_t \) having \( \mathcal{H}_{\text{at}} \equiv \mathcal{H}_{\text{at}} \otimes \{ \Omega_R \} \) as an invariant space. Note further that also the subspace

\[
\mathcal{H}_{\text{at}} := \{ A \Omega_0 \in \mathcal{H}_{\text{at}} : A \in \mathcal{B}(\mathbb{C}^d), L_{\text{at}}(A) = \epsilon A, \ \epsilon \in \{ E_1 - E_N, 0, E_N - E_1 \} \}
\]

is an invariant space of \( L_R \) and \( L^{(\text{eff})}_t \) for all \( t \in \mathbb{R} \).

Denote by \( \{ U^{(\text{eff})}_{t,s} \}_{t \geq s} \) the two–parameter family of operators on \( \mathcal{H}_{\text{at}} \) generated by \( L^{(\text{eff})}_t \). From direct calculations

\[
L^{(\text{eff})}_t (A \Omega_0) = (L^{(\lambda,\eta)*}_t A) \Omega_0
\]

for all \( A \Omega_0 \in \mathcal{H}_{\text{at}} \), where \( L^{(\lambda,\eta)*}_t \) is the adjoint of the Lindbladian \( L^{(\lambda,\eta)}_t \). Thus, for all \( A \in \mathcal{D} \),

\[
U^{(\text{eff})}_{t,0} (A \Omega_0) = (\hat{\tau}^{(\lambda,\eta)}(A)) \Omega_0,
\]

where \( \{ \hat{\tau}^{(\lambda,\eta)} \}_{t \geq s} \) is the two–parameter family of operators on \( \mathcal{B}(\mathbb{C}^d) \) generated by \( L^{(\lambda,\eta)*}_t \).

Let \( \rho(t) := (\hat{\tau}^{(\lambda,\eta)*}_{0,-t})(\rho_{\text{at}}) \). Then

\[
\langle \rho(t), A \rangle_{\mathcal{H}_{\text{at}}} = \langle (\hat{\tau}^{(\lambda,\eta)*}_{0,-t})(\rho_{\text{at}}), A \rangle_{\mathcal{H}_{\text{at}}} = \langle \rho_{\text{at}}, \hat{\tau}^{(\lambda,\eta)}(A) \rangle_{\mathcal{H}_{\text{at}}}
\]

for all \( A \in \mathcal{B}(\mathbb{C}^d) \) and thus

\[
\left\langle \frac{d}{dt} \rho(t), A \right\rangle_{\mathcal{H}_{\text{at}}} = \left\langle \rho_{\text{at}}, \hat{\tau}^{(\lambda,\eta)}(L^{(\lambda,\eta)*}_t(A)) \right\rangle_{\mathcal{H}_{\text{at}}} = \left\langle L^{(\lambda,\eta)}_t(\rho(t)), A \right\rangle_{\mathcal{H}_{\text{at}}}.
\]

We used above that \( L^{(\lambda,\eta)}_t = L^{(\lambda,\eta)}_t \). Hence, the time–dependent density matrix \( \rho(t) \) satisfies the master equation

\[
\forall t \geq 0: \quad \frac{d}{dt} \rho(t) = L^{(\lambda,\eta)}_t \rho(t), \quad \rho(0) = \rho_{\text{at}}.
\]
6. In order to justify this procedure, note that for all $A \in \mathcal{D}$,
\[
\langle \rho(t), A \rangle_{\mathcal{D}_{\text{at}}} = \left\langle \Omega_0, U_0^{(\text{eff})}(t) A \Omega_0 \right\rangle,
\]
and that in \( \mathcal{P} \) we show, for $\rho_{\text{at}} \in \mathcal{D}$,
\[
\left| \left\langle \Omega_0, (U_0^{(\text{eff})} - U_{t,0}) A \Omega_0 \right\rangle \right|_{\mathcal{D}_{\text{at}}} \leq C_{\varpi} \| A \| |\lambda|
\]
for some constant $C_{\varpi} \in (0, \infty)$ depending on $\varpi$ but not on the initial state $\omega_{\text{at}}$ of the atom, the observable $A \in \mathcal{D}$, and the parameters $t, \lambda$, and $\eta$ with $|\lambda|$ sufficiently small.

6.2 Proof of Theorem 4.4

Before starting the proof, we first extend the definitions of $\Lambda^{(\lambda, \eta)}$ \(4.32\), $\tilde{\Sigma}^{(0,0)}_0$ \(4.35\), $U_0$ \(4.36\), and $\tilde{\Lambda}^{(\lambda, \eta)}$ \(4.37\) to all eigenvalues $\epsilon \in \sigma([H_{\text{at}}, \cdot])$ as follows:
\[
\tilde{\Sigma}^{(0,0)}_\epsilon := \text{span}\{W^{(k', n')}_{(k, n)} \mid (k, k') \in \bigcup_{m \in \{-2, -1, 0, 1, 2\}} t_{m+2m\varpi}, n \in \{1, \ldots, n_k\}, n' \in \{1, \ldots, n_{k'}\}\}.
\]

The unitary operator $U_\epsilon$ is defined by
\[
U_\epsilon \left( e^{i\epsilon (\omega - E_k) t + E_k t} W^{(k', n')}_{(k, n)} \right) := W^{(k', n')}_{(k, n)} \in \tilde{\Sigma}^{(0,0)}_\epsilon,
\]
whereas
\[
\Lambda^{(\lambda, \eta)} := P^{(0,0)}_\epsilon \Sigma^{(\lambda, \eta)} P^{(0,0)}_\epsilon
\quad \text{and} \quad
\tilde{\Lambda}^{(\lambda, \eta)} := U_\epsilon \Lambda^{(\lambda, \eta)} U_\epsilon^*.
\]

See, e.g., \(3.7\), \(3.8\) and \(4.33\). Observe that
\[
\tilde{\Sigma}^{(0,0)}_\epsilon \subset \mathcal{D}_{\text{at}}, \quad \tilde{\Sigma}^{(0,0)}_{-\varpi} = \tilde{\Sigma}^{(0,0)}_0 = \tilde{\Sigma}^{(0,0)}_{\varpi},
\]
whereas
\[
\forall \epsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varpi, 0, \varpi\}; \quad \tilde{\Sigma}^{(0,0)}_0 \perp \tilde{\Sigma}^{(0,0)}_{\epsilon}.
\] (6.5)

Using these observations we can deduce the spectral structure of the operators $\Lambda^{(\lambda, \eta)}_\epsilon$:

Lemma 6.1 (Spectral properties of operators $\Lambda^{(\lambda, \eta)}_\epsilon$)

For all $\epsilon \in \sigma([H_{\text{at}}, \cdot])$,
\[
\sigma(\Lambda^{(\lambda, \eta)}_\epsilon) \setminus \{-i\varpi, 0, i\varpi\} \subset i\mathbb{R} - i\mathbb{R}^+
\]
and $-i\varpi$, $0$, $i\varpi$ are simple eigenvalues of $\Lambda^{(\lambda, \eta)}_\varpi$, $\Lambda^{(\lambda, \eta)}_0$ and $\Lambda^{(\lambda, \eta)}_{-\varpi}$, respectively. Moreover, for all $\epsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varpi, 0, \varpi\}$,
\[
\sigma(\Lambda^{(\lambda, \eta)}_\epsilon) \subset i\mathbb{R} - i\mathbb{R}^+.
\]

I.e., any eigenvalue of $\Lambda^{(\lambda, \eta)}_\epsilon$ has a strictly negative real part for every $\epsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varpi, 0, \varpi\}$.

Proof. Similar to the proof of Theorem 4.6 (i), we compute that
\[
\tilde{\Lambda}^{(\lambda, \eta)} = \left( -i\epsilon - \frac{\eta}{2} \Sigma_p + \lambda^2 \Sigma_R \right) |\tilde{\Sigma}^{(0,0)}_0|.
\] (6.6)

Indeed, if $\epsilon \in \{-\varpi, 0, \varpi\}$ then $\tilde{\Sigma}^{(0,0)}_{\pm\varpi} = \tilde{\Sigma}^{(0,0)}_0$ and the computations for this case are exactly those given in the proof of Theorem 4.6. For $\epsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varpi, 0, \varpi\}$ the properties \(2.10\), \(2.12\) of the optical pump yield $\Sigma_p |\tilde{\Sigma}^{(0,0)}_0| = 0$ whereas one can readily check that the Lindbladian $\Sigma_R$ conserves the atomic subspace $\tilde{\Sigma}^{(0,0)}_0$. These properties lead to \(6.6\) for $\epsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varpi, 0, \varpi\}$. 
As a consequence,

\[
\sigma(\Lambda^{(\lambda,\eta)}_\varepsilon) = \sigma(\tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon) = -i\varepsilon + \sigma\left( \tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon \right)_{\tilde{\delta}_\varepsilon^{(0,0)}} ,
\]

where \(\tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon\) is seen as an operator acting on \(\tilde{\mathcal{D}}_{\text{at}}\), see again proof of Theorem 4.6. Moreover, \(\tilde{\delta}_\varepsilon^{(0,0)}\) is an invariant space of \(\tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon\) in \(\mathcal{B}(\mathcal{D}_{\text{at}})\). As explained in the proof of Theorem 4.6, the operator \(\tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon\) is the generator of a relaxing, Markov CP semigroup, see Definition 6.9. In particular, 0 is a non–degenerate eigenvalue of \(\Lambda^{(\lambda,\eta)}_\varepsilon\) and the corresponding eigenvector is an element of \(\tilde{\mathcal{D}}_{\pm\varepsilon}^{(0,0)} = \tilde{\mathcal{D}}_{\varepsilon}^{(0,0)}\). As a consequence, from (6.7), \(-i\varepsilon\), 0 and \(i\varepsilon\) must be non–degenerated eigenvalues of \(\Lambda^{(\lambda,\eta)}_\varepsilon\), \(\Lambda^{(\lambda,\eta)}_0\) and \(\Lambda^{(\lambda,\eta)}_{-\varepsilon}\), respectively.

Finally, by Theorem 6.10

\[
\text{Re}\left\{ \sigma\left( \tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon \right) \right\} \setminus \{0\} \subset (-\infty, 0)
\]

which, for any \(\varepsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{-\varepsilon, 0, \varepsilon\}\), implies that

\[
\text{Re}\left\{ \sigma\left( \tilde{\Lambda}^{(\lambda,\eta)}_\varepsilon \right) \right\} = \text{Re}\left\{ \sigma\left( \tilde{\Lambda}^{(\lambda,\eta)}_0 \right) \right\} \setminus \{0\} \subset (-\infty, 0)
\]

because of (6.5). \qed

The proof of Theorem 4.4 needs further technical results. The next one concerns the stability of the irreducibility of quantum Markov chains (Assumption 2) under block localization:

**Lemma 6.2 (Stability of Assumption 2 under block localization)**

Let \(H \in \mathcal{B}(\mathbb{C}^d)\) be any self–adjoint operator and denote by \(\Pi_\varepsilon\) the orthogonal projection onto the eigenspace of \([H, \cdot]\) associated with the eigenvalue \(\varepsilon \in \sigma([H, \cdot])\). Then, there are \(\tilde{m} \in \mathbb{N}\), non–negative real numbers \(\{c_{j,k}\}_{j,k,\ell} \subset \mathbb{R}^+_0\), real numbers \(\{\tilde{d}(\ell)\}_{j,k,\ell} \subset \mathbb{R}\), and operators \(\{\tilde{V}_{j,k}\}_{j,k,\ell} \subset \mathcal{B}(\mathbb{C}^d)\) such that

\[
\tilde{\Sigma}_R(\rho) := \sum_{\varepsilon \in \sigma([H, \cdot])} 1_{\varepsilon} \Sigma_R 1_{\varepsilon} = -i[H_{\text{Lamb}}, \cdot] + \tilde{\Sigma}_d ,
\]

where

\[
H_{\text{Lamb}} := -\frac{1}{2} \sum_{\varepsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{0\}} \sum_{(j,k) \in \ell, \ell, \ell} \tilde{m} \tilde{d}(\ell) \tilde{V}_{j,k,\ell}^* \tilde{V}_{j,k,\ell}
\]

and, for any \(\rho \in \mathcal{D}_{\text{at}}\),

\[
\tilde{\Sigma}_d(\rho) := \frac{1}{2} \times \sum_{\varepsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{0\}} \sum_{(j,k) \in \ell, \ell} \sum_{\ell} \tilde{m} \tilde{c}(\ell) \left( 2\tilde{V}_{j,k,\ell}^* \rho \tilde{V}_{j,k,\ell}^* - \tilde{V}_{j,k,\ell}^* \rho \tilde{V}_{j,k,\ell}^* \right) .
\]

Moreover,

\[
\left( \bigcup_{\{(j,k,\ell) : c_{j,k,\ell} \neq 0\}} \{\tilde{V}_{j,k,\ell}^*\} \right)^\prime = \mathcal{B}(\mathbb{C}^d) .
\]

In particular, \(\tilde{\Sigma}_R\) is the generator of a relaxing, Markov, CP semigroup satisfying Assumption 2.

**Proof.** Let \(\{\tilde{E}_j\}_{j=1}^M\) be the eigenvalues of the self–adjoint operator \(H (M \leq d)\) and

\[
\tilde{i}_\varepsilon := \{(j,k) : \tilde{E}_j - \tilde{E}_k = \varepsilon\} \subset \{1, 2, \ldots M\} \times \{1, 2, \ldots M\} .
\]

For any \(V \in \mathcal{B}(\mathbb{C}^d)\) and each eigenvalue

\[
\varepsilon \in \sigma([H, \cdot]) = \{ \tilde{E}_j - \tilde{E}_k : j, k \in \{1, 2, \ldots M\} \} ,
\]
we define
\[ V_\varepsilon := 1_\varepsilon(V) = \sum_{(j,k) \in \mathfrak{t}_\varepsilon} 1[H = \tilde{E}_j] V 1[H = \tilde{E}_k] \in \mathcal{B}(\mathbb{C}^d). \]

By construction, note that
\[ \sum_{\varepsilon \in \sigma([H, \cdot])} V_\varepsilon = V. \tag{6.8} \]

In particular, one has
\[ V \in \text{span} \left\{ \bigcup_{\varepsilon \in \sigma([H, \cdot])} V_\varepsilon \right\}. \tag{6.9} \]

For \((j, k) \in \mathfrak{t}_\varepsilon, \varepsilon \in \sigma([H_{\text{at}}, \cdot]) \setminus \{0\}, \) and \(\varepsilon \in \sigma([H, \cdot])\) let
\[ \tilde{V}_{j,k}^{(\ell, \varepsilon)} := 1_\varepsilon(V_{j,k}^{(\ell)}) \tag{6.10} \]

By identifying the finite sets \(\{1, 2, \ldots, m\} \times \sigma([H, \cdot])\) and \(\{1, 2, \ldots, \tilde{m}\},\) we infer from (6.9) and Assumption 2 that the family \(\{\tilde{V}_{j,k}^{(\ell)}\}_{j,k,\tilde{\ell}} \subset \mathcal{B}(\mathbb{C}^d)\) of operators satisfies
\[ \left( \bigcup_{\{j,k,\tilde{\ell} \} : \tilde{\varepsilon}_{j,k}^{(\ell)} \neq 0} \{ \tilde{V}_{j,k}^{(\ell)} \} \right)^{''} = \mathcal{B}(\mathbb{C}^d). \tag{6.11} \]

For all eigenvectors \(A_{\varepsilon_1}, B_{\varepsilon_2} \in \mathcal{B}(\mathbb{C}^d)\) associated with eigenvalues \(\varepsilon_1, \varepsilon_2 \in \sigma([H, \cdot])\) respectively, i.e., \(1_{\varepsilon_1}(A) = A_{\varepsilon_1}\) and \(1_{\varepsilon_2}(B) = B_{\varepsilon_2},\) let
\[ D := A_{\varepsilon_1}B_{\varepsilon_2} \in \mathcal{B}(\mathbb{C}^d). \]

If \(D \neq 0\) then we observe that \((\varepsilon_1 + \varepsilon_2) \in \sigma([H, \cdot])\) and \(1_{\varepsilon_1 + \varepsilon_2}(D) = D, i.e., D = D_{\varepsilon_1 + \varepsilon_2}\) must be an eigenvector associated with the eigenvalue \(\varepsilon_1 + \varepsilon_2.\) Therefore, by using (6.8) and the equality \(1_{-\varepsilon}(V^*) = V_{\varepsilon}^*\), we deduce that
\[ \sum_{\varepsilon \in \sigma([H, \cdot])} 1_{\varepsilon}(V1_{\varepsilon}(\rho)V^*) = \sum_{\varepsilon \in \sigma([H, \cdot])} 1_{\varepsilon} \left( \sum_{\tilde{\varepsilon} \in \sigma([H, \cdot])} V_{\tilde{\varepsilon}}1_{\varepsilon}(\rho) \sum_{\tilde{\varepsilon} \in \sigma([H, \cdot])} 1_{\tilde{\varepsilon}}(V^*) \right) \]
\[ = \sum_{\varepsilon \in \sigma([H, \cdot])} \sum_{\tilde{\varepsilon} \in \sigma([H, \cdot])} V_{\tilde{\varepsilon}}1_{\varepsilon}(\rho) 1_{-\varepsilon}(V^*) \]
\[ = \sum_{\varepsilon \in \sigma([H, \cdot])} V_{\varepsilon}\rho V_{\varepsilon}^* \quad \tag{6.12} \]
for any \(\rho \in \mathfrak{h}_{\text{at}} = \mathcal{B}(\mathbb{C}^d).\) Similarly,
\[ \sum_{\varepsilon \in \sigma([H, \cdot])} 1_{\varepsilon}(V^*V1_{\varepsilon}(\rho)) = \sum_{\varepsilon \in \sigma([H, \cdot])} V_{\varepsilon}^*V_{\varepsilon}\rho, \quad \tag{6.13} \]
\[ \sum_{\varepsilon \in \sigma([H, \cdot])} 1_{\varepsilon}(1_{\varepsilon}(\rho)V^*) = \sum_{\varepsilon \in \sigma([H, \cdot])} \rho V_{\varepsilon}^*V_{\varepsilon} \quad \tag{6.14} \]
for any \(\rho \in \mathfrak{h}_{\text{at}}.\) Therefore, by using (3.6), (3.11), (3.13), (6.10) and (6.12)–(6.14) with the identification of the finite sets \(\{1, 2, \ldots, m\} \times \sigma([H, \cdot])\) and \(\{1, 2, \ldots, \tilde{m}\},\) we obtain the explicit decomposition of \(\tilde{\mathcal{L}}_R\) stated in the lemma. By Theorems 6.7 and 6.10 together with (6.11), it is then straightforward to verify that \(\tilde{\mathcal{L}}_R\) is the generator of a relaxing, Markov CP semigroup.

We need more precise information about the behavior of the spectral gap given in Lemma 6.1 with respect to the coupling constants \(\eta\) and \(\lambda.\) This is achieved by using the previous lemma:

Lemma 6.3 (Behavior of the spectral gap of \(\Lambda(\lambda, \eta)\))

For all \((\lambda, \eta) \in \mathbb{R} \times \mathbb{R},\)
\[ \min \left\{ |\text{Re} p| : p \in \sigma(\Lambda(\lambda, \eta)) \setminus \{0\} \right\} \geq C_{\varepsilon} \lambda^2 \]
with \(C_{\varepsilon} \in (0, \infty)\) being a constant depending on \(\varepsilon\) but not on \(\lambda, \eta.\)
Proof. We define the function
\[ g(\lambda, \eta) := \lambda^{-2} \min \left\{ |\text{Re} \{p\}| : p \in \sigma(\Lambda^{(\lambda, \eta)}) \setminus \{0\} \right\} \]
on the set \( \mathbb{R} \setminus \{0\} \times \mathbb{R} \). Observe that \( g(\lambda, \eta) \) only depends on the ratio \( \kappa := \eta / \lambda^2 \) and is strictly positive, by Theorems 6.7 and 6.10. Indeed, by the proof of Lemma 6.1,
\[ g(\lambda, \eta) = \min \left\{ |\text{Re} \{p\}| : p \in \sigma \left( \frac{\eta}{2\lambda^2} \mathcal{L}_p + \mathcal{L}_R \right) \setminus \{0\} \right\}. \]
Furthermore, by Remark 3.2, \( \kappa \mathcal{L}_p + \mathcal{L}_R \) is the generator of a Markov CP semigroup satisfying Assumption 2 for any \( \kappa \in \mathbb{R} \) (see also Remark 3.1) and Theorem 6.10 yields that this semigroup must be relaxing.

By Kato’s perturbation theory [26], for some constants \( C, c \in (0, \infty) \), \( g(\lambda, \eta) \geq C \) whenever \( \eta \leq c \lambda^2 \), i.e., when \( \kappa \leq c \). Using again Kato’s perturbation theory [26] and Theorem 6.10, \( \kappa \mapsto g(\kappa^{-\frac{1}{2}}, 1) \) is a strictly positive continuous function on the interval \([c, c']\) for any finite constant \( c' > c \). By compactness of the interval \([c, c']\), it follows that
\[ \min \left\{ g(\kappa^{-\frac{1}{2}}, 1) : \kappa \in [c, c'] \right\} > 0. \]
So, it remains to prove that \( g(\lambda, \eta) \geq C \) whenever \( \eta > c \lambda^2 \), i.e., when \( \kappa > c' \), for some constant \( C \in (0, \infty) \) and sufficiently large \( c' < \infty \). By (6.7) for \( \lambda, \epsilon = 0 \), note that
\[ \sigma(\Lambda^{(0, \eta)}) \subset i\mathbb{R}. \]
Thus, by Kato’s perturbation theory [26] for the spectrum of \( \kappa \mathcal{L}_p / 2 + \mathcal{L}_R \), the limit
\[ \lim_{\kappa \to \infty} g(\kappa^{-\frac{1}{2}}, 1) \in [0, \infty) \] (6.15)
exists and satisfies
\[ \lim_{\kappa \to \infty} g(\kappa^{-\frac{1}{2}}, 1) \geq \min \left\{ |\text{Re} \{p\}| : p \in \sigma(\mathcal{L}_R) \setminus \{0\} \right\}, \] (6.16)
where
\[ \mathcal{L}_R := \sum_{\xi \in \mathbb{Z} \setminus \{0\}} 1_\xi \mathcal{L}_R 1_\xi. \]
Here, \( 1_\xi \) denotes the spectral projection of \([H_p, \cdot]\) onto the eigenspace associated with an eigenvalue \( \xi \in \sigma([H_p, \cdot]) \). Using Lemma 6.2, \( \mathcal{L}_R \) is the generator of a relaxing, Markov, CP semigroup satisfying Assumption 2 and, by (6.16) and Theorem 6.10,
\[ \lim_{\kappa \to \infty} g(\kappa^{-\frac{1}{2}}, 1) \in (0, \infty). \]
In other words, for some constants \( C \in (0, \infty) \) and sufficiently large \( c' > c \), \( g(\kappa^{-\frac{1}{2}}, 1) \geq C \) for all \( \kappa > c' \). \(

We now use Lemma 6.3 to obtain norm estimates on the difference of the uniformly bounded semigroups \( \{e^{\alpha \Lambda^{(\lambda, \eta)}}\}_{\alpha \geq 0} \) and \( \{e^{\alpha P_0^{(\lambda, \eta)\epsilon}}\}_{\alpha \geq 0} \).

Lemma 6.4 (Semigroup estimates)
For sufficiently small \( |\lambda| \), there are constants \( C_{\infty}, c_{\infty} \in (0, \infty) \) depending on \( \infty \) but not on \( \lambda, \eta \) and \( \alpha \) such that
\[ \forall \alpha \geq 0 : \quad \|e^{\alpha \Lambda^{(\lambda, \eta)}} - e^{\alpha P_0^{(\lambda, \eta)\epsilon}}\| \leq C_{\epsilon}(\lambda^2 + e^{-\alpha \lambda^2 \epsilon}) \].

Proof. Note that the continuous semigroup \( \{e^{\alpha \Lambda^{(\lambda, \eta)}}\}_{\alpha \geq 0} \) on \( \chi_{\text{st}} \) can be represented through the inverse Laplace transform of the resolvent of its generator. Indeed, by Lemma 6.1 combined with [41] Proof of Corollary 5.15,
\[ e^{\alpha \Lambda^{(\lambda, \eta)}} - E = \lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{w+iL} \int_{w-iL} e^{\alpha z} \left( (z - \Lambda^{(\lambda, \eta)})^{-1} - \frac{1}{z} \right) \text{d}z \right\} + 1 - E \] (6.17)
for any $w > 0$ and with
\[
E := \frac{1}{2\pi i} \oint_{|z| = R'} e^{\alpha z} (z - \Lambda(\lambda, \eta))^{-1} \, dz = \frac{1}{2\pi i} \oint_{|z| = R'} (z - \Lambda(\lambda, \eta))^{-1} \, dz
\]  
(6.18)
and $R' > 0$ sufficiently small. Note that 0 is a simple eigenvalue of $\Lambda(\lambda, \eta)$ and the last equality follows from the fact that the map
\[
z \mapsto (1 - e^{\alpha z})(z - \Lambda(\lambda, \eta))^{-1}
\]
is holomorphic near $z = 0$. By Lemma 6.3, the Riesz projection $E$ associated with the generator $\Lambda(\lambda, \eta)$ is well-defined for sufficiently small $R' \in (0, c\lambda^2)$ at fixed $\lambda \neq 0$. Using again the spectral properties of $\Lambda(\lambda, \eta)$ given in Lemma 6.3, we can push the integration path of the complex integral in (6.17) to sufficiently small, but a strictly negative real part $w' = -c\lambda^2 < 0$ as follows:
\[
\lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{-w+il}^{w-il} e^{\alpha z} \left( (z - \Lambda(\lambda, \eta))^{-1} - \frac{1}{z} \right) \, dz \right\}
\]
\[
= \lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{-c\lambda^2-il}^{-c\lambda^2+il} e^{\alpha z} \left( (z - \Lambda(\lambda, \eta))^{-1} - \frac{1}{z} \right) \, dz \right\}
\]
\[
+ \frac{1}{2\pi i} \oint_{|z|=R'} e^{\alpha z} (z - \Lambda(\lambda, \eta))^{-1} \, dz - \frac{1}{2\pi i} \oint_{|z|=R'} e^{\alpha z} \, dz
\]
\[
= \lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{-c\lambda^2-il}^{-c\lambda^2+il} \frac{e^{\alpha z}}{z} (z - \Lambda(\lambda, \eta))^{-1} \Lambda(\lambda, \eta) \, dz \right\} + E - 1.
\]
Note that $c \in (0, \infty)$ is a sufficiently small constant depending on $\varpi$ but not on $\lambda$, $\eta$, and $\alpha$. By (6.17), it follows that
\[
e^{\alpha \Lambda(\lambda, \eta)} - E
\]
\[
= \lim_{L \to \infty} \left\{ \lambda^{-2} \frac{1}{2\pi i} \int_{-L}^{L} e^{\alpha (ix - c) \lambda^2} \left( (ix - c) - \lambda^{-2} \Lambda(\lambda, \eta) \right)^{-1} \Lambda(\lambda, \eta) \, dx \right\}
\]  
(6.19)
for all $\alpha \in \mathbb{R}_0^+$. Observe that we have additionally used an obvious change of variable in the last equation to extract the factor $\lambda^{-2}$. Indeed, by (6.6)–(6.7) and Assumption [1] ($|\eta| \leq C\lambda^2$), we have $\|\Lambda(\lambda, \eta)\| = O(\lambda^2)$ and the limit (6.19) yields the upper bound
\[
\|e^{\alpha \Lambda(\lambda, \eta)} - E\|
\]
\[
\leq e^{-c\lambda^2} \lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{-L}^{L} \frac{1}{\|ix - c\|} \left\| (ix - c) - \lambda^{-2} \Lambda(\lambda, \eta) \right\|^{-1} \, dx \right\}
\]  
(6.20)
for all $\alpha \in \mathbb{R}_0^+$. Hence we need to bound the integrand of the last integral by some integrable function not depending on the parameters $\lambda, \eta$. To this end, first observe that
\[
\lambda^{-2} \Lambda(\lambda, \eta) = U_0^* \left( \mathcal{L}_R + \frac{\varpi}{2} \mathcal{E}_p \right) U_0
\]  
(6.21)
with the ratio $\varpi := \eta/\lambda^2 \in [-\varpi_0, \varpi_0]$ for some fixed $\varpi_0 \in (0, \infty)$. See (4.37), Theorem 4.6 (i) and Assumption [1]. Therefore, we define the map $\hat{g} : \mathbb{R} \times [-\varpi_0, \varpi_0] \to \mathbb{R}$ by
\[
\hat{g}(x, \varpi) := \left\| (ix - c) - \mathcal{L}_R - \varpi \mathcal{E}_p \right\|^{-1} < \infty.
\]  
(6.22)
This function is well-defined for sufficiently small $c > 0$ because of Lemma 6.3. Moreover, since
\[
|\hat{g}(x_1, \varpi_1) - \hat{g}(x_2, \varpi_2)|
\]
\[
\leq |\hat{g}(x_1, \varpi_1)| \, |\hat{g}(x_2, \varpi_2)| \left( \|x_1 - x_2\| + \|\varpi_1 - \varpi_2\| \|\mathcal{E}_p\| \right),
\]
the function \( \tilde{g} \) is (locally Lipschitz) continuous on its domain of definition. By compactness, for any \( L \in \mathbb{R}_0^+ \) there is a constant \( C \in (0, \infty) \) depending on \( \varpi \) but not on \( \lambda, \eta \) such that
\[
\sup_{(x, \varpi) \in [-L, L] \times [-\varpi_0, \varpi_0]} \tilde{g}(x, \varpi) \leq C . \tag{6.23}
\]

On the other hand, the Neumann series
\[
((ix - c) - \mathcal{L}_R - \varpi \mathcal{L}_p)^{-1} = \sum_{n=0}^{\infty} (ix - c)^{-(n+1)} \{ \mathcal{L}_R + \varpi \mathcal{L}_p \}^n
\]
implies that, for sufficiently large \( x \gg 1 \) and \( \varpi \in [-\varpi_0, \varpi_0] \),
\[
\tilde{g}(x, \varpi) \leq \sum_{n=0}^{\infty} \|ix - c\|^{-(n+1)} \{ \|\mathcal{L}_R\| + \|\varpi\|\|\mathcal{L}_p\| \}^n
\]
because \( \mathcal{L}_R \) and \( \mathcal{L}_p \) are bounded operators on \( \mathcal{H}_{\text{sat}} \). As a consequence, there is a sufficiently large constant \( C \in (0, \infty) \) depending on \( \varpi \), \( \lambda, \eta \) such that
\[
\forall x \in \mathbb{R}, \ \varpi \in [-\varpi_0, \varpi_0] : \quad \tilde{g}(x, \varpi) \leq \min \left\{ C, \frac{2}{\|ix - c\|} \right\} \tag{6.24}.
\]

By (6.20)–(6.24), we conclude the existence of a constant \( C \in (0, \infty) \) depending on \( \varpi \) but not on \( \lambda, \eta \) such that
\[
\forall \alpha \geq 0 : \quad \|e^{\alpha \Lambda(\lambda, \eta)} - E\| \leq Ce^{-\alpha \lambda^2} . \tag{6.25}
\]

In the same way we obtain (6.20), we derive the upper bound
\[
\left\| e^{\alpha P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} - \tilde{E}} \right\| \leq e^{-\alpha \lambda^2} \lim_{L \to \infty} \left\{ \frac{1}{2\pi i} \int_{-L}^{L} \frac{1}{\|ix - c\|} \left\| (ix - c) - \alpha^{-2} P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} - 1 \right\| \ dx \right\} . \tag{6.26}
\]

Here,
\[
\tilde{E} := \frac{1}{2\pi i} \oint_{|z| = R'} e^{\alpha z} (z - P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)})^{-1} \ dz \tag{6.27}
\]
is an operator associated with the generator \( P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} \). The latter is well–defined for sufficiently small \( R' \in (0, c\lambda^2) \) at fixed \( \lambda \neq 0 \).

Namely, for \( |\lambda| \ll 1 \) and some constant \( C \in (0, \infty) \) depending on \( \varpi \) but not on \( \lambda, \eta \), observe that
\[
\| P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} \| \leq C \lambda^2 \quad \text{and} \quad \| G^{(\lambda, \eta)} P_0 \| \leq C \lambda^2 ,
\]
using \( P_0^{(0,0)} G^{(0,0)} = 0 \). Hence, combining these last upper bounds with (4.30) and the triangle inequality we get
\[
\| P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} - \Lambda(\lambda, \eta) \| = \| P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)} - P_0^{(0,0)} G^{(\lambda, \eta)} P_0^{(0,0)} \| \leq C \lambda^4 \tag{6.28}
\]
with \( C \in (0, \infty) \) not depending on \( \lambda, \eta \) and \( \varpi \). In particular, the spectral properties of \( \Lambda(\lambda, \eta) \) given in Lemma 6.3 together with (6.28) implies that \( \tilde{E} \) is well–defined for a sufficiently small, but strictly positive \( R' \in (0, c\lambda^2) \) and satisfy
\[
\| \tilde{E} - E \| \leq C \lambda^4 \oint_{|z| = R'} \|(z - \Lambda(\lambda, \eta))^{-1} \| \| (z - P_0^{(\lambda, \eta)} G^{(\lambda, \eta)} P_0^{(\lambda, \eta)})^{-1} \| \ dz \leq C \lambda^2 \tag{6.29}
\]
for some constant \( C \in (0, \infty) \) not depending on the (sufficiently small coupling) constants \( |\lambda|, |\eta| \).
To prove the inequalities (6.26) and (6.29), note that the (non-degenerate) eigenvalue 0 is the unique element of the spectrum of $\Lambda^{(\lambda, \eta)}$ within the disc of radius $R'$, by Lemma 6.3. By (6.28) and Kato’s perturbation theory of discrete eigenvalues, there is a unique (non-degenerate) eigenvalue of $P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}$ and no other element of its spectrum within the disc of radius $R'$, provided that $|\lambda|$ is sufficiently small. In fact, this eigenvalue is also 0 because $G^{(\lambda, \eta)}$ is a closed operator and the constant function $\Omega_0 \in \mathcal{D}_{ev}$ satisfies $G^{(\lambda, \eta)^*}\Omega_0 = 0$ for the adjoint $G^{(\lambda, \eta)^*}$ of $G^{(\lambda, \eta)}$ (implying that 0 is also an eigenvalue of $G^{(\lambda, \eta)}$ and thus of $P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}$). Hence, the map

$$z \mapsto (1 - e^{\alpha z})(z - P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)})^{-1}$$

is holomorphic near $z = 0$. Consequently,

$$\tilde{E} = \frac{1}{2\pi i} \oint_{|z|=R'} (z - P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)})^{-1} dz$$

is the Riesz projection onto the eigenspace associated with the eigenvalue 0 of $P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}$ and by (6.18), the first inequality of (6.29) follows.

Now, using (6.28), the fact that $\tilde{g}(x, \kappa)$ is uniformly bounded, and the second resolvent equation for sufficiently small $|\lambda|$, we get the upper bound

$$\left\|(ix - \pi^{-2} P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)})^{-1}\right\| \leq C \tilde{g}(x, \eta/\lambda^2)$$

for some constant $C \in (0, \infty)$ depending on $\varpi$ but not on $\lambda, \eta$. Therefore, similar to (6.25), we infer from (6.26) and properties of the function $\tilde{g}$ that there is a constant $C \in (0, \infty)$ depending on $\varpi$ but not on $\lambda, \eta$ such that

$$\forall \alpha \geq 0 : \quad \|e^{\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}} - \tilde{E}\| \leq Ce^{-c\alpha\lambda^2}.$$

Combining this with (6.25) and (6.29), one obtains the statement of the lemma. \hfill \Box

We now conclude by the proof of Theorem 4.4 that is, we show, for sufficiently small $|\lambda|$ and any $\varepsilon \in (0, 1)$, that

$$\left\|\exp\left(\alpha\Lambda^{(\lambda, \eta)}\right) - \exp\left(\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}\right)\right\| \leq C_{\varpi, \varepsilon}|\lambda|^{2(1-\varepsilon)}, \quad (6.30)$$

where the constant $C_{\varpi, \varepsilon} \in (0, \infty)$ depends on $\varpi$, $\varepsilon$ but not on $\lambda, \eta$, and $\alpha$.

Proof of Theorem 4.4. Note that the semigroups $\{e^{\alpha\Lambda^{(\lambda, \eta)}}\}_{\alpha \geq 0}$ and $\{e^{\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}}\}_{\alpha \geq 0}$ are uniformly bounded in $\lambda, \eta$ as the first one is a CP semigroup and the second one is the restriction of $\{T_\alpha\}_{\alpha \geq 0}$, see (4.7). Thus, Duhamel’s formula yields the inequality

$$\|e^{\alpha\Lambda^{(\lambda, \eta)}} - e^{\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}}\| \leq \alpha C\|P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)} - \Lambda^{(\lambda, \eta)}\| \quad (6.31)$$

for some constant $C \in (0, \infty)$ not depending on $\lambda, \eta, \varpi$, and $\alpha$. By (6.28), it follows that

$$\|e^{\alpha\Lambda^{(\lambda, \eta)}} - e^{\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}}\| \leq C_{\varpi}\alpha\lambda^4 \quad (6.32)$$

with $C_{\varpi} \in (C, \infty)$ depending on $\varpi$ but not on $\lambda, \eta$, and $\alpha$. Finally, we infer from (6.32) and Lemma 6.4 that

$$\|e^{\alpha\Lambda^{(\lambda, \eta)}} - e^{\alpha P_0^{(\lambda, \eta)}G^{(\lambda, \eta)}P_0^{(\lambda, \eta)}}\| \quad (6.33)$$

for any $\varepsilon \in (0, 1)$ and we obtain (6.30) for sufficiently small $|\lambda|$. \hfill \Box
6.3 Completely positive (CP) semigroups

In the theory of open quantum systems, one is usually interested on the restricted dynamics of some small quantum object interacting with macroscopic reservoirs. This restricted time–evolution is described by a map \( \mathcal{C} \) on the set of density matrices of the small system. See, for instance, [3 Section 1.2.1]. Properties of such maps (cf. [3 Section 1.2.2]) motivate the definition of the class of completely positive (CP) operators:

**Definition 6.5 (Completely positive (CP) maps)**

A positive map \( \mathcal{C} \in \mathcal{B}(\mathcal{B}(\mathcal{X})) \) acting on the set \( \mathcal{B}(\mathcal{X}) \) of bounded operators on a Hilbert space \( \mathcal{X} \) is called completely positive (CP) if the extended map \( \mathcal{C} \otimes 1_{\mathcal{B}(\mathcal{H})} \) remains positive for all \( n \in \mathbb{N} \). If \( \mathcal{C}(1_{\mathcal{X}}) = 1_{\mathcal{X}} \), then the operator \( \mathcal{C} \) is called a unital map.

Completely positive (CP) semigroups are defined as being the semigroups which are CP maps for all times:

**Definition 6.6 (Completely positive (CP) semigroups)**

A semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \subset \mathcal{B}(\mathcal{B}(\mathcal{X})) \), with \( \mathcal{X} \) being a Hilbert space, is CP if the map \( \mathcal{C}_t \) is CP for all \( t \in \mathbb{R}_0^+ \). If \( \mathcal{C}_t \) is unital for any \( t \in \mathbb{R}_0^+ \), then we call \( \{\mathcal{C}_t\}_{t \geq 0} \) unital.

From now on and until the end of Section 6.3, \( \mathcal{X} \) is always a \( n \)-dimensional Hilbert space. We denote by \( \mathcal{B}_2(\mathcal{X}) \equiv \mathcal{B}(\mathcal{X}) \) the Hilbert space of Hilbert–Schmidt operators with scalar product

\[
\langle A, B \rangle_{\mathcal{B}_2(\mathcal{X})} := \text{Tr}(A^* B), \quad A, B \in \mathcal{B}_2(\mathcal{X}).
\]

In the special case where a semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \subset \mathcal{B}(\mathcal{B}_2(\mathcal{X})) \) acts on \( \mathcal{B}_2(\mathcal{X}) \), we can define its (unique) adjoint semigroup \( \{\mathcal{C}^*_t\}_{t \geq 0} \subset \mathcal{B}(\mathcal{B}_2(\mathcal{X})) \) as usual via the equations

\[
\forall t \geq 0 : \quad \langle \mathcal{C}^*_t(A), B \rangle_{\mathcal{B}_2(\mathcal{X})} = \langle A, \mathcal{C}_t(B) \rangle_{\mathcal{B}_2(\mathcal{X})}, \quad A, B \in \mathcal{B}_2(\mathcal{X}).
\]

Note that if the CP semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \) is unital then \( \{\mathcal{C}^*_t\}_{t \geq 0} \) is CP and preserves the trace. A CP semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \) is called Markov CP semigroup if it preserves the trace. Generators of Markov CP semigroups \( \{\mathcal{C}_t\}_{t \geq 0} \) and of their adjoint groups \( \{\mathcal{C}^*_t\}_{t \geq 0} \) can be characterized in the finite dimensional case as follows (cf. [12 Theorem 2.2] and [13 Theorem 2.1]):

**Theorem 6.7 (Generators of Markov CP semigroups – I)**

Let \( \dim \mathcal{X} = n \in \mathbb{N} \). The operator \( L \in \mathcal{B}(\mathcal{B}_2(\mathcal{X})) \) is the generator of a Markov CP semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \) if and only if

\[
L(\rho) = -i [h, \rho] + \frac{1}{2} \sum_j \left\{ [V_j, \rho V_j^*] + [V_j^*, \rho V_j]\right\}, \quad A \in \mathcal{B}_2(\mathcal{X}),
\]

where \( h = h^* \in \mathcal{B}_2(\mathcal{X}) \) and \( \{V_j\} \subset \mathcal{B}(\mathcal{X}) \) is a family of operators with \( \sum_j V_j^* V_j \in \mathcal{B}(\mathcal{X}) \). Additionally, the adjoint semigroup \( \{\mathcal{C}^*_t\}_{t \geq 0} \) is in this case the unitary CP semigroup with generator

\[
L^*(A) = i [h, A] + \frac{1}{2} \sum_j \left\{ [V_j^*, A] V_j + V_j^* [A, V_j]\right\}, \quad A \in \mathcal{B}_2(\mathcal{X}).
\]

Generators of CP semigroups are also called Lindbladian or Lindblad (–Kossakowski) generators.

A more compact characterization of generators of Markov CP semigroups is given by [27 Sect. 4.3]:

**Theorem 6.8 (Generators of Markov CP semigroups – II)**

Let \( \dim \mathcal{X} = n \in \mathbb{N} \). The operator \( L \in \mathcal{B}(\mathcal{B}_2(\mathcal{X})) \) is the generator of a CP semigroup \( \{\mathcal{C}_t\}_{t \geq 0} \) if and only if there is a completely positive map \( \Xi \in \mathcal{B}(\mathcal{B}(\mathcal{X})) \) and an operator \( \Delta \in \mathcal{B}(\mathcal{X}) \) such that

\[
L = \Delta + \Delta^* + \Xi.
\]

Such a CP semigroup is Markov if and only if \( L^*(1_{\mathcal{X}}) = 0 \).
The relaxing property of Markov CP semigroups \( \{ C_t \}_{t \geq 0} \subset B(\mathcal{B}(X)) \), which is crucial for our analysis, is defined as follows:

**Definition 6.9 (Relaxing semigroups)**

A Markov CP semigroup \( \{ C_t \}_{t \geq 0} \subset B(\mathcal{B}(X)) \) is called relaxing if there is a unique trace–one positive \( \rho_\infty \in \mathcal{B}(\mathcal{B}(X)) \), i.e., a density matrix \( \rho_\infty \), such that, for any density matrix \( \rho \in \mathcal{B}(\mathcal{B}(X)) \),

\[
\lim_{t \to \infty} C_t(\rho) = \rho_\infty.
\]

In other words, a relaxing, Markov CP semigroup has a unique invariant equilibrium state. Moreover, this state can be approximated by the density matrix \( C_t(\rho) \) for large times and any initial state with density matrix \( \rho \).

Spohn [30, Theorem 2] gave in 1977 a characterization of relaxing semigroups which turns out to be very useful in our context:

**Theorem 6.10 (Condition for a Markov CP semigroup to be relaxing)**

Let \( \dim X = n \in \mathbb{N} \). Let \( \{ C_t \}_{t \geq 0} \subset B(\mathcal{B}(X)) \) be a Markov CP and \( C_0 \) semigroup with generator \( L \) given by Theorem 6.4. If the space spanned by the family \( \{ V_j \} \) satisfies

\[
\text{span} \{ V_j \} = \text{span} \{ V_j^* \} \subset \mathcal{B}(X)
\]

and the bicommutant

\[
\{ V_j \}'' = \mathcal{B}(X),
\]

then \( \{ C_t \}_{t \geq 0} \) is relaxing. In particular, \( 0 \) is a non–degenerated eigenvalue of \( L \) and

\[
\max \{ \text{Re} \{ w \} \mid w \in \sigma(L) \setminus \{ 0 \} \} < 0.
\]

As explained after Assumption 2, the condition (6.34) is a non–commutative version of the irreducibility of classical Markov chains.

### 6.4 The operator \( \mathfrak{B}_{p,R} \) and the conservation of positivity

1. We first give an elementary example of an operator \( \mathfrak{B}_{p,R} \) defined by (4.44) which does not preserve positivity. Let \( d = 3 \), \( N = 2 \), \( \varpi = 1 \) and

\[
H_{\mathfrak{at}} := \begin{pmatrix}
0 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{pmatrix},
\]

\[
Q_1 := \begin{pmatrix}
0 & 1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix},
\]

\[
Q_2 := \begin{pmatrix}
0 & 0 & 1 \\
0 & 0 & 0 \\
1 & 0 & 0
\end{pmatrix},
\]

in the canonical orthonormal basis of \( \mathbb{C}^3 \). Choose the coupling functions \( f_\ell \) such that \( g_\ell(\epsilon) \geq 0 \) for \( \ell \in \{ 1, 2 \} \) and \( \epsilon \in \{-1, 0, 1\} \), see (2.16). Then, the family \( \{ V_j^{(\ell)} \}_{j, \ell} \subset B(\mathbb{C}^3) \) of operators defined by (3.9) for \( \ell \in \{ 1, 2 \} \) and \( j, k \in \{ 1, N \} \) satisfy Assumption 2. With this choice, the operators \( W_{(1,1)}^{(N,1)}, W_{(1,1)}^{(N,2)}, W_{(1,1)}^{(N,1)}, W_{(1,1)}^{(N,2)} \in \mathfrak{B}_{\mathfrak{at}} \) defined by (4.33) are eigenvectors of \( L_R \) with eigenvalues \( \gamma_1, \gamma_2, \bar{\gamma}_1, \bar{\gamma}_2 \in \mathbb{C} \setminus \{ 0 \} \), respectively. By setting

\[
H_p := \begin{pmatrix}
0 & 1 & 1 \\
1 & 0 & 0 \\
1 & 0 & 0
\end{pmatrix},
\]

we infer from (5.2) that

\[
\mathfrak{B}_{N,1} \begin{pmatrix}
1 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} = -\begin{pmatrix}
0 & 0 & 0 \\
0 & 2 \text{Re}\{\bar{\gamma}_1^{-1}\} & 0 \\
0 & 0 & 2 \text{Re}\{\gamma_2^{-1}\}
\end{pmatrix} \begin{pmatrix}
0 & 0 & 0 \\
2 \text{Re}\{\gamma_1^{-1}\} & 0 & 0 \\
0 & 0 & 2 \text{Re}\{\bar{\gamma}_2^{-1}\}
\end{pmatrix}
\]

in the canonical orthonormal basis of \( \mathbb{C}^3 \). Indeed, note that

\[
H_p \begin{pmatrix}
1 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} = W_{(1,1)}^{(N,1)} + W_{(1,1)}^{(N,2)}; \quad \begin{pmatrix}
1 & 0 & 0 \\
0 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} H_p^* = W_{(N,1)}^{(1,1)} + W_{(N,2)}^{(1,1)}.
and
\[ W_{(1,1)}^{(N,j)} h_p = W_{(N,1)}^{(N,j)} + W_{(N,2)}^{(N,j)}, \quad h_p W_{(N,j)}^{(1,1)} = W_{(N,1)}^{(N,1)} + W_{(N,j)}^{(N,1)}, \quad j \in \{1, 2\}. \]

Thus,
\[
B_{N,1} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} = - \left( \gamma_1^{-1} W_{(1,1)}^{(N,1)} + \gamma_2^{-1} W_{(1,1)}^{(N,2)} \right) h_p^* - h_p \left( \bar{\gamma}_1^{-1} W_{(N,1)}^{(1,1)} + \bar{\gamma}_2^{-1} W_{(N,1)}^{(1,1)} \right) \\
= -\gamma_1^{-1} (W_{(N,1)}^{(N,1)} + W_{(N,2)}^{(N,1)}) - \gamma_2^{-1} (W_{(N,2)}^{(N,1)} + W_{(N,1)}^{(N,2)}) \\
- \bar{\gamma}_1^{-1} (W_{(N,1)}^{(N,1)} + W_{(N,2)}^{(N,1)}) - \bar{\gamma}_2^{-1} (W_{(N,2)}^{(N,1)} + W_{(N,1)}^{(N,2)}). 
\]

Let
\[ D := \det \left( \begin{array}{cc} 2 \Re \{ \gamma_1^{-1} \} & \gamma_1^{-1} + \bar{\gamma}_2^{-1} \\ \gamma_1^{-1} + \bar{\gamma}_2^{-1} & 2 \Re \{ \gamma_2^{-1} \} \end{array} \right) \in \mathbb{R} \]
be the determinant of the lower 2 × 2 block diagonal part of the above self–adjoint matrix. One can clearly find coupling functions \( f_\ell \) for \( \ell \in \{1, 2\} \) such that \(|g_1(1)|^2 = |g_2(1)|^2\), but
\[ \mathcal{P} \mathcal{P}(g_1(1)) \neq \mathcal{P} \mathcal{P}(g_2(1)). \]

This implies that
\[ \Re \{ \gamma_1^{-1} \} = \Re \{ \gamma_2^{-1} \} \quad \text{and} \quad \Im \{ \gamma_1^{-1} \} \neq \Im \{ \gamma_2^{-1} \}. \]

However, in this case
\[ D = - (\Im \{ \gamma_1^{-1} \} - \Im \{ \gamma_2^{-1} \})^2 < 0 \]
showing that \( B_{N,1} \) is not a map from \( \mathcal{B}^+(\mathcal{H}_1) \) to \( \mathcal{B}^+(\mathcal{H}_N) \) and hence \( B_{p,R} \) is in general not the generator of a semigroup on \( \mathcal{D} \) which preserves positivity.

2. Note that under certain conditions the weighted stimulated transition rates defined via the operator
\[ B_{p,R} := \int_0^\infty e^{-s \mathcal{L}_p} \mathcal{L}_p e^{s \mathcal{L}_p} \mathcal{L}_p P_\mathcal{D} \ ds \]
are well–defined. This is the case, for instance, for strong decoherence, i.e., if
\[ \min(\Re \{ \sigma(\mathcal{L}_R P_\mathcal{D}) \}) > \max(\Re \{ \sigma(\mathcal{L}_R P_\mathcal{D}^4) \}) \]
as in this situation
\[ \| e^{-s \mathcal{L}_p} \mathcal{L}_p P_\mathcal{D} \| \| e^{s \mathcal{L}_p} \mathcal{L}_p P_\mathcal{D} \| \leq C e^{-cs} \]
for constants \( C, c \in (0, \infty) \) and all \( t \geq 0 \). Observe that the norm \( \| e^{-t \mathcal{L}_p} \| \) exponentially grows as \( t \to \infty \), generally. In this case we obtain, moreover, that
\[ e^{\mathbb{B}_{p,R} P_\mathcal{D}} = \lim_{\kappa \to 0} P_\mathcal{D} \exp(\kappa^{-2} t \mathcal{L}_R + \kappa^{-1} t \mathcal{L}_p) P_\mathcal{D}. \]

In particular, \( \mathbb{B}_{p,R} \) is the generator of a positivity preserving semi–group on \( \mathcal{D} \).

Therefore we could wonder whether or not \( \mathbb{B}_{p,R} \) also generates a positive semigroup if the system shows strong decoherence. Unfortunately, this turns out to be not the case. This can be seen in a simple variation of the example above: By introducing a third atom–reservoir interaction term in the model above with coupling satisfying \(|g_3(0)|^2 > 0\) and
\[
Q_3 := \begin{pmatrix} q_3 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad q_3 \in \mathbb{R},
\]
the diagonal part $\mathcal{L}_\mathcal{R} P_D$ is not changed whereas $\mathcal{L}_\mathcal{R} P_D^\perp$ is shifted by $-q_3^2|g_3(0)|^2 P_D^\perp$. Hence the strong decoherence regime can be always attained for sufficiently large $q_3$. As the eigenvalues $\gamma_1, \gamma_2, \tilde{\gamma}_1, \tilde{\gamma}_2$ of $W^{(N,1)}_{(1,1)}, W^{(N,2)}_{(1,1)}$, $W^{(1,1)}_{(N,1)}, W^{(1,1)}_{(N,2)}$ are simultaneously shifted by $-q_3^2|g_3(0)|^2$, $D$ remains negative for all $q_3 \in \mathbb{R}$.

3. Nevertheless, Assumption 3 can be verified for a large class of models. For instance, this condition is always satisfied if the 1st and the $N$th energy levels of the atomic part of the model are non-degenerated. This is a special case of the following theorem:

**Theorem 6.11 ($\mathcal{B}_{p,\mathcal{R}}$ as transition rates)**

Assume that $P_{N,1} \mathcal{L}_\mathcal{R} P_{N,1} = \xi_{N,1} P_{N,1}$ for some $\xi_{N,1} \in \mathbb{C}$. Then Assumption 3 holds.

**Proof.** Using (3.6) and (3.11)–(3.13), we observe that

$$
\tilde{\xi}_{N,1} P_{1,N} (\rho) = (\xi_{N,1} P_{N,1} (\rho))^* = (P_{N,1} \mathcal{L}_\mathcal{R} P_{N,1} (\rho))^* = P_{1,N} \mathcal{L}_\mathcal{R} P_{1,N} (\rho)
$$

for any self-adjoint $\rho = \rho^* \in \mathfrak{H}_{\text{at}}$. Since any element of $\mathfrak{H}_{\text{at}}$ is a sum of self-adjoint elements and $\mathcal{L}_\mathcal{R}$ as well as $P_{1,N}$ and $P_{1,N}$ are linear operators, one obtains that

$$
P_{1,N} \mathcal{L}_\mathcal{R} P_{1,N} = \tilde{\xi}_{N,1} P_{1,N}.
$$

Using the notation $\xi := \xi_{N,1}$, we get that

$$
P_{N,1} \left( \mathcal{C}_{\text{ran}(P_D^\perp)} \right)^{-1} P_{N,1} = \xi^{-1} P_{N,1}, \quad P_{1,N} \left( \mathcal{C}_{\text{ran}(P_D^\perp)} \right)^{-1} P_{1,N} = \tilde{\xi}^{-1} P_{1,N}.
$$

Thus, by (5.2),

$$
B_{N,1} = -\xi^{-1} P_{N,N} h_p^* P_{N,1} h_p P_{1,1} - \xi^{-1} P_{N,N} h_p P_{1,N} h_p^* P_{1,1}.
$$

Using the properties of $h_p$ and $A B = B A$,

$$
B_{N,1} = -\frac{2 \text{Re} (\xi) h_p h_p^*}{|\xi|^2}.
$$

Similarly, one also finds

$$
B_{1,N} = -\frac{2 \text{Re} (\xi) h_p h_p^*}{|\xi|^2}.
$$

On the other hand, we have

$$
B_{N,N} = \tilde{\xi}^{-1} h_p h_p^* P_{N,N} + \xi^{-1} P_{N,N} h_p h_p^*,
$$

$$
B_{1,1} = \xi^{-1} h_p^* h_p P_{1,1} + \tilde{\xi}^{-1} P_{1,1} h_p^* h_p.
$$

Therefore, by (5.3), we can split the operator $\mathcal{B}_{p,\mathcal{R}}$ on the subspace $D$ as follows:

$$
\mathcal{B}_{p,\mathcal{R}} = \Delta + \Delta^* + \Xi
$$

with

$$
\Delta := \text{Im} (\xi) \left( h_p h_p^* + h_p^* h_p \right) + \frac{\text{Re} (\xi)}{|\xi|^2} \left( h_p h_p^* + h_p^* h_p \right)
$$

$$
= \tilde{\xi}^{-1} \left( h_p h_p^* + h_p^* h_p \right) \in \mathcal{B}(\mathbb{C}^d)
$$

and

$$
\Xi := B_{1,N} + B_{N,1} \in \mathcal{B}(\mathfrak{H}_{\text{at}}).
$$
Since $\mathcal{L}_R$ is the generator of a relaxing Markov CP semigroup on $\mathcal{H}_{\text{at}}$ (Remark 3.2), $\text{Re}(\xi) \leq 0$ and $\Xi$ is thus a completely positive map. Clearly,

$$
\left( \Delta + \Delta^* + \Xi \right)^* (1) = \left( \Delta^* + \Delta + \Xi \right) (1) = 0.
$$

Therefore, we infer from (6.35) and Theorem 6.8 that $\mathcal{B}_{p,R}$ generates a Markov CP semigroup on $\mathcal{H}_{\text{at}}$. As $\mathcal{D}$ is an invariant subspace of this generator, $\mathcal{B}_{p,R}$ generates a Markov semigroup on $\mathcal{D}$ which preserves positivity. □

ACKNOWLEDGMENTS

This work has been supported by the grant MTM2010-16843 of the Spanish “Ministerio de Ciencia e Innovación” and a grant of the “Inneruniversitäre Forschungsförderung” of the Johannes Gutenberg University in Mainz. We thank Volker Bach for his support and the referees for pointing out a mistake in the proof of Lemma 4.1.

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