FROM BLOCH MODEL TO THE RATE EQUATIONS II: THE CASE OF ALMOST DEGENERATE ENERGY LEVELS

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Bloch equations give a quantum description of the coupling between atoms and a driving electric force. It is commonly used in optics to describe the interaction of a laser beam with a sample of atoms. In this article, we address the asymptotics of these equations for a high frequency electric field, in a weak coupling regime. The electric forcing is taken quasiperiodic in time.

We prove the convergence towards a rate equation, *i.e.* a linear Boltzmann equation, recovering in this way the physically relevant asymptotic model. It describes the transitions amongst the various energy levels of the atoms, governed by the resonances between the electric forcing and the atoms' eigenfrequencies. We also give the explicit value for the transition rates.

The present task has already been addressed in [BFCD03] in the case when the energy levels are *fixed*, and for different classes of electric fields. Here, we extend the study in two directions. First, we consider *almost degenerate* energy levels, a natural situation in practice. In this case, almost resonances might occur. Technically, this implies that the small divisor estimates needed in [BFCD03] are *false*, due to the fact that the Diophantine condition is unstable with respect to small perturbations. We use an appropriate ultraviolet cutoff to restore the analysis and to sort out the asymptotically relevant frequencies. Second, since the asymptotic rate equation may be singular in time, we completely analyze the initial time-layer, as well as the associated convergence towards an equilibrium state.

Keywords: density matrix, Bloch equations, rate equations, linear Boltzmann equation, averaging theory, small divisor estimates, degenerate energy levels.

AMS Subject Classification: 34C27, 81V80

1. Introduction

Bloch equations are a basic model used to describe the coupling between light and matter at the quantum level (see [Boy92], [NM92], [RV02]). Here, matter is described through the associated density matrix, a quantum object. In the dipole approximation, the electromagnetic wave enters through the electric field only. Bloch equations are commonly used in optics when modelling the interaction between a laser beam and a sample of atoms, whose optical properties are under study. This is the typical example we have in mind. In this context, the atoms may be in gaseous state (dry air, He, H_2, water vapor, ...), in liquid state (CS_2, CCl_4, ethanol, water, ...), or in solid state (Silica, Lucite, ...). Another case of interest in practice is a sample of independent, decoupled atoms with N energy levels (for some $N = 2$, 3, or more). In all these situations, a standard value for the laser's frequency is about $10^{14} - 10^{15}\mathrm{s}^{-1}$, to be compared with a typical unit time of the order of several ms: this is a high-frequency regime.

The present paper analyzes the asymptotic behaviour of the Bloch equations in the case of a high frequency electromagnetic forcing, when the coupling is weak, and the energy levels are discrete (see below for the precise scalings): the resonances between the field and the eigenvalues of the quantum mechanical system enforce transitions between the various energy levels of the atoms. We prove that the latter are asymptotically described by a rate equation, *i.e.* a linear Boltzmann equation. We recover in this way the rate equations formally derived in the physics literature (see e.g. [Lou91]). Mathematically, our analysis requires a precise understanding of the resonances, so that small divisor estimates and averaging techniques for Ordinary Differential Equations naturally play a key role.

A similar study has already been performed in a [BFCD03], for various high frequency forcings, when the eigenfrequencies of the atomic system are *fixed*. In the present paper, we extend in two ways the work done in [BFCD03].

First, the atoms' energy levels are here allowed to be *almost degenerate*. There are many examples of such almost degeneracies: this is the case of Zeemann hyperfine structures in complex molecules, or quantum dots submitted to an external magnetic field; high levels of an atom are also almost degenerate, due to the accumulation value at the ionisation energy. In this case, the wave's frequencies might resonate or "almost resonate" with the eigenfrequencies of the atomic system. Due to these "almost resonances", there appears the need for a new sorting out of the frequencies. Mathematically, the small divisor estimates of [BFCD03] simply become *false* in the almost degenerate case: the Diophantine estimates are unstable under small perturbations. The tool we develop in this paper is an ultraviolet cutoff procedure.

The second new point is the following: as in [BFCD03], the asymptotic rate equation that describes the above mentioned resonances may be *singular* in time. We completely analyze the initial time layer as well as the convergence towards an equilibrium state induced by this singularity.

We stress the fact that the present paper deals with a linear situation: the electromagnetic forcing is given. A full description of the light/matter interaction would require the analysis of the Maxwell-Bloch system, which has quadratic nonlinearity ([NM92]).

Let us come to quantitative statements.

The model and its scaling

According to the quantum theory, matter is described *via* a density matrix ρ, whose diagonal entry

$$\rho_{\rm d}(t,n) := \rho(t,n,n),$$

called the population, is –in the eigenstates basis– the occupation number of the n-th energy level at time t, and the off-diagonal entries

$$\rho_{\rm od}(t,n,m) := \rho(t,n,m)\ \mathbf{1}[n \neq m],$$

called the coherences, are linked to the transition probability from level n to level m (conditioned by the corresponding populations). Throughout this article we assume that the energy levels are discrete: we work below the ionisation energy of the atomic system, and the number of atoms is typically "low" (absence of continuous spectrum).

When sending the electromagnetic field through the matter, the evolution of the system is described by the Bloch equations. We refer the reader to [Boh79, Boy92, CTDRG88, Lou91, NM92, RP69, SSL77, Bid03] for textbooks about wave/matter interaction issues, where Bloch equations occur. They read, in scaled, dimensionless form ($\varepsilon > 0$ is the scaling parameter; the justification of the scaling follows),

$$\begin{aligned}\varepsilon^2\partial_t\rho(t,n,m) &= -i\omega_\varepsilon(n,m)\rho(t,n,m) \\ &\quad + i\varepsilon\sum_k\left[\phi\left(\frac{t}{\varepsilon^2}\right)V(n,k)\rho(t,k,m) - \phi\left(\frac{t}{\varepsilon^2}\right)V(k,m)\rho(t,n,k)\right] + Q_\varepsilon(\rho)(n,m),\end{aligned} \tag{1.1}$$

and the initial datum $\rho(0,n,m)$ satisfies

$$\rho(0,n,m) = 0,\ \text{if } n \neq m, \quad \rho(0,n,n) \geq 0 \quad \text{and} \quad \sum_n \rho(0,n,n) < \infty. \tag{1.2}$$

The first term on the right-hand-side of (1.1) is the free Hamiltonian of the atomic system, the second term acounts for the interaction with the wave, and the last term is a relaxation term. We now detail each of these.

The time dependent field $\phi(t) \in \mathbb{R}$ is the electric field (up to rescaling), which we assume to be quasi-periodic in time, i.e.

$$\phi(t) = \sum_{\alpha \in \mathbb{Z}^r} \phi_\alpha \exp(i\alpha \cdot \omega t), \text{ where } \alpha \cdot \omega := \alpha_1 \omega_1 + \cdots + \alpha_r \omega_r, \tag{1.3}$$

for some frequency vector $\omega \in \mathbb{R}^r$ (r is a given integer), and some Fourier coefficients ϕ_α such that $\sum_\alpha |\phi_\alpha| < \infty$. Such a field mimics a laser beam having a finite number – r – of independent frequencies, and all of their harmonics.

The quantity

$$\omega_\varepsilon(n,m) = \omega(n,m) + \delta_\varepsilon(n,m) = \Big(\omega(n) + \varepsilon^{p(n)}\delta(n)\Big) - \Big(\omega(m) + \varepsilon^{p(m)}\delta(m)\Big)$$

is the transition energy (or: transition frequency) between levels n and m, and for all n, $p(n) > 0$ is some given exponent. The reader may safely think of the case $p(n) = \text{const} = p$, but we keep track of the possible dependence of $p(n)$ upon n for sake of generality (see below on this point).

As in standard optics (see, e.g. [Boy92], page 149) the relaxation term reads

$$Q_\varepsilon(\rho)(n,m) = \begin{cases} -\varepsilon^\mu \gamma(n,m)\rho(t,n,m), & \text{if } n \neq m, \\ \\ \varepsilon^2 \left(\sum_k \left[W(k,n)\rho(t,k,k) - W(n,k)\rho(t,n,n)\right]\right), & \text{if } n = m, \end{cases}$$

where $0 \leq \mu < 1/2$ is some given exponent.

In all this analysis, the coefficients $V(n,m)$, $\gamma(n,m)$, $W(n,m)$, $\omega(n,m)$, and $\delta(n)$ are given. They are constants of the atomic system and satisfy definite symmetry and positivity properties. We also list at the beginning of section 2 the decay assumptions satisfied by these numbers. The interaction coefficient $V(n,m) \in \mathbb{C}$ is an entry in the dipole moment matrix, and it is Hermitian: $V(m,n) = V(n,m)^*$. It characterizes the atom's reaction to a given applied field. The $\omega(n)$'s and $\delta(n)$'s are real and describe the eigenfrequencies of the atomic system. Finally, the relaxation term acts as a linear Boltzmann term on the diagonal part, whereas on the off-diagonal part, it acts as a pure damping term. In the optics literature, it describes at a *heuristic* level the observed trend to equilibrium of the atomic samples under consideration. The so-called Pauli coefficient $W(n,m)$ is non-negative. The longitudinal coefficient $\gamma(n,m)$ is positive and symmetric: $\gamma(n,m) = \gamma(m,n)$. To simplify notations, we extend its definition to the case when $n = m$ introducing $\gamma(n,n) = 0$. On the more, the entries $W(n,m)$ and $W(m,n)$ are related by the standard relation (see *e.g.* [BBR01, Bid03])

$$W(n,m) = \exp\left(\frac{\omega(m,n)}{T}\right) W(m,n), \tag{1.4}$$

where T is a normalized temperature. This specific form is of great importance when describing the equilibrium states. Finally, integers $n \in \mathbb{N}$, $m \in \mathbb{N}$, and $k \in \mathbb{N}$ are labelling discrete energy levels, and time t belongs to $\mathbb{R}^+$. In the case of a finite number of energy levels, we add the restriction $n, m, k \leq N$ and so on. The dependence of the density matrix on the small parameter ε will always be implicit.

Let us comment on the scaling.

The electric field has high-frequency of size $1/\varepsilon^2$, and time t is rescaled by ε^2 accordingly in (1.1): the atoms' and wave's frequencies have the same order of magnitude. This is a realistic scaling, and it creates the possibility of a resonant interaction between the wave and the atom. Also, the typical amplitude of the field is taken of size ε. This is a standard weak coupling regime (see [Spo80, Spo91, vH55, vH57]): as is well-known, the total effect of the wave on time scales $1/\varepsilon^2$ is then of order 1 ($= 1/\varepsilon^2 \times \varepsilon^2$ =time scale $\times$(amplitude of the field)^2). In practice, the amplitude of the wave is tuned so as to create an effect of the order 1 on the atomic sample.

Concerning the free hamiltonian part of equation (1.1), a comment is in order. In the non-degenerate framework $\omega(n, m) = \omega(n) - \omega(m)$ is the difference between the frequencies of levels n and m respectively. It is either zero or an order one quantity. Here we wish to tackle the case of almost degenerate levels, replacing the frequency $\omega(n)$ by $\omega(n) + \lambda\delta(n)$ for some small adimensional parameter λ that measures the almost-degeneracy. In general, the two small parameters λ and ε are independent. A typical value of $\omega(n, m)$ is about $10^{14}\mathrm{s}^{-1}$ (corresponding to a gap in energy around 100 meV), whereas the levels' separation ($\lambda\delta(n)$) may be several orders of magnitude smaller: the value $10^{10}\mathrm{s}^{-1}$ is not seldom. The experiments reported in [HGV$^+$99] show that in quantum dots (obtained by epitaxial growth of InAs on a substrate of GaAs) submitted to an external magnetic field, the ground state and first excited state are separated by an energy gap about 70-80 meV ($\approx 10^{14}\mathrm{s}^{-1}$), and the separation with the next level varies with the field intensity (thus, λ depends on ε), down to values of the order 1 meV ($\approx 10^{12}\mathrm{s}^{-1}$). Furthermore, the coefficient λ may be in practice much larger, or much smaller, or comparable with ε, depending on the atomic sample under consideration. Note finally that for a given system, the ratio $\lambda\delta(n)/\omega(n)$ might actually vary by several orders of magnitude when index n changes (hence making the exponent p vary with n). For these reasons, and in order to describe all possible situations, we set

$$\omega_\varepsilon(n) = \omega(n) + \varepsilon^{p(n)}\delta(n).$$

It leads to different regimes, depending on the sequence of exponents $p(n)$. For given, physical values of the constants, the one or the other regime may be relevant.

We finally comment the scaling of the relaxation term. In our scaling, we assume that the diagonal relaxation has an influence on the same time scale as the free Hamiltonian (due to the prefactor ε^2). In practice, this term typically acts on comparable or much larger time scales than the free Hamiltonian, so that a prefactor ε^q for some $q \geq 2$ would be natural. We choose the value $q = 2$ to fix the ideas, but our analysis anyhow extends straightforwardly for any value of $q \geq 2$. More importantly, our scaling makes the off-diagonal relaxation act on a much shorter time scale than both the diagonal relaxation and the free Hamiltonian term. Again, this is a standard polarization process in optics: we model this phenomenon upon measuring the corresponding time scale by the parameter ε^μ for some $0 \leq \mu < 2$, and the constraint $\mu < 2$ is physically relevant. As before, there is no theoretical basis linking the off-diagonal relaxation with the parameter ε, and this is the reason why we do not fix the value of the parameter μ. Technically, our analysis turns out to need the strengthened restriction $0 \leq \mu < 1/2$. This threshold value for μ arises in the estimates and we do not know whether it is optimal or not. However it is no wonder that there is a threshold value. Indeed, in the case when $\mu = 0$, the initial Bloch equation (1.1) is time-irreversible and the asymptotic equation that we derive in this paper is also time-irreversible. On the other hand, in the opposite case when every coefficient $\gamma(n, m)$ is identically zero (which can also be interpreted as $\mu = \infty$), the initial Bloch equation is time-reversible and the nature of the problem has changed.

The vanishing of the off-diagonal entries means that we are dealing with "well prepared" initial data. This is a standard assumption in the field (see *e.g.* [KL57, LK58, Zwa66]).

Convergence results

Since rate equations play a central role in this paper, we introduce the following useful

Notation. *To coefficients $A(n, k)$, $n \neq k$ (which may possibly be time dependent: $A(t, n, k)$) we associate the linear Boltzmann operator $A_\sharp$ defined as*

$$A_\sharp\ \rho_{\mathrm{d}}(n) = \sum_k \left[A(k, n)\rho_{\mathrm{d}}(k) - A(n, k)\rho_{\mathrm{d}}(n)\right]. \tag{1.5}$$

If $A(n, k) \in l^\infty_n l^1_k \cap l^\infty_k l^1_n$, $A_\sharp$ is a bounded operator on l^p, $1 \leq p \leq \infty$ (see Appendix 7.1).

With this notation, our first main result asserts that equation (1.1) behaves asymptotically like a rate equation acting on the diagonal part of ρ.

Theorem 1.1. ***(Averaging of the oscillations).*** *We assume the Diophantine condition Hypothesis 1 holds true, and the ϕ_α's are decaying enough (Hypothesis 3).*
(i) First approximate dynamics. *Define the transition rate*

$$\langle\Psi_\varepsilon\rangle(k, n) = 2|V(k, n)|^2 \sum_{\beta \in \mathbb{Z}^r} \frac{\varepsilon^\mu \gamma(k, n)}{\varepsilon^{2\mu}\gamma(k, n)^2 + |\omega(k, n) + \beta\cdot\omega + \delta_\varepsilon(k, n)|^2} |\phi_\beta|^2. \tag{1.6}$$

Define $\rho^{(1)}_{\mathrm{d}} = \rho^{(1)}_{\mathrm{d}}(t, n)$ as the solution to the rate equation

$$\partial_t \rho^{(1)}_{\mathrm{d}}(t, n) = (W + \langle\Psi_\varepsilon\rangle)_\sharp\ \rho^{(1)}_{\mathrm{d}}(t, n) \tag{1.7}$$

with initial data $\rho_{\mathrm{d}}^{(1)}(0,n) = \rho_{\mathrm{d}}(0,n)$*. Then, for all* $T > 0$*, there exists* $C > 0$ *such that*

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(1)}\|_{L^\infty([0,T],l^2)} \le C\varepsilon^{1-2\mu}, \qquad \text{and } \|\rho_{\mathrm{od}}\|_{L^\infty([0,T],l^1)} \le C\varepsilon^{1-\mu}.$$

(ii) Refined approximate dynamics*. Define the* dominant *transition rate*

$$\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}(k,n) := 2|V(k,n)|^2 \frac{\varepsilon^\mu \gamma(k,n)}{\varepsilon^{2\mu}\gamma(k,n)^2 + \delta_\varepsilon(k,n)^2} \sum_{\beta\in\mathbb{Z}^r;\ \omega(k,n)+\beta\cdot\omega=0} |\phi_\beta|^2. \quad (1.8)$$

Let also $\rho_{\mathrm{d}}^{(2)}$ *be the solution to*

$$\partial_t \rho_{\mathrm{d}}^{(2)}(t,n) = \left(W + \langle\Psi_\varepsilon\rangle^{\mathrm{dom}}\right)_\sharp\ \rho_{\mathrm{d}}^{(2)}(t,n) \qquad (1.9)$$

with initial data $\rho_{\mathrm{d}}^{(2)}(0,n) = \rho_{\mathrm{d}}(0,n)$*. Then, under the additional decay Hypotheses 2, 4 and 5, for all* $T > 0$*, there exists* $C > 0$ *such that*

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(2)}\|_{L^\infty([0,T],l^2)} \le C(\varepsilon^\mu + \varepsilon^{1-2\mu}).$$

Part (i) of the Theorem is essentially contained in [BFCD03]. It states that the off-diagonal part of ρ vanishes asymptotically, while the diagonal part tends to satisfy a rate equation. The original transition rates W of equation (1.1) become $W + \langle\Psi_\varepsilon\rangle$ in (1.9) after the averaging procedure. As in [BFCD03], this Theorem is obtained in two steps: first, using manipulations on the density matrix, we prove that ρ_{d} tends to satisfy a *closed* differential equation with rapidly oscillating coefficients; second, we average the oscillations appropriately.

Now, part (ii) of the Theorem is the main part, both from the point of view of modelisation and analysis. In the transition rate $\langle\Psi_\varepsilon\rangle(k,n)$ obtained in part (i), the main contribution comes *both* from the exact resonances (when $\omega(k,n) + \beta\cdot\omega = 0$ and $\delta_\varepsilon(k,n) = 0$), and from the "almost-resonances" (when $\omega(k,n) + \beta\cdot\omega = 0$ and $\delta_\varepsilon(k,n) \neq 0$) between the wave and the atomic system: almost resonances correspond, in other words, to the case when $\omega_\varepsilon(k,n)+\beta\cdot\omega = \omega(k,n)+\beta\cdot\omega+\delta_\varepsilon(k,n)$ is small, but non zero. On the other hand, in the transition rate $\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}$ obtained in part (ii), the system only retains those Fourier modes ϕ_β for which $\omega(k,n)+\beta\cdot\omega = 0$: the almost resonances have been filtered out. To state the result in a different way, part (ii) somehow establishes that the averaging procedure does not "see" the almost resonance condition.

Analytically, the tool to prove (ii) is the ultraviolet cutoff procedure prepared in Lemma 4.1 and developed in section 4.2 below. The idea is the following. The Diophantine condition, which is the basic tool to estimate the small denominators in (1.6) or (1.8), is *not* stable under the perturbation $\delta_\varepsilon(k,n)$. There is no hope to restore its validity in any uniform sense with respect to the perturbation. Lemma 4.1 provides an alternative way for the treatment of small denominators: it asserts that those frequencies that violate the Diophantine condition are typically associated with very large Fourier modes β (or large indices n, k). We use this to balance their contribution thanks to the decay properties of the ϕ_β coefficients (or of the $V(n,k)$

coefficients). Despite its easy proof, we stress that this Lemma allows to circumvent hard analytical difficulties related to small denominators.

Note that when $\mu = 0$, part (ii) of the Theorem does not give a good approximation of $\rho_{\rm d}$, and we do not have a better description than the one from part (i), where all (resonant and non-resonant) frequencies have a contribution to the transition rates.

Our second result is the following.

Clearly, the coefficients $\langle \Psi_\varepsilon \rangle^{\rm dom}(k,n)$ may contain singular terms of size $\varepsilon^{-\mu}$ or $\varepsilon^{\mu - 2p(n)}$, and so on, depending on the relative values of μ, $p(k)$ and $p(n)$. Using the fact that the linear Boltzmann operator $(W + \langle \Psi_\varepsilon \rangle^{\rm dom})_\sharp$ has a non-positive spectrum, we completely analyze the underlying initial time layer, and behaviour outside the layer, in the case of a finite number of levels.

Theorem 1.2. ***(True limit in ε, and equilibrium states)****. Assume $N < \infty$. Then, the asymptotic evolution of $\rho_{\rm d}$ when $\varepsilon \to 0$ can be summarized as follows.*
(i) First case: $\mu = 0$. Then, for any $0 \le t \le T$, we have $\|\rho_{\rm d}(t) - \rho_{\rm d}^{(3)}(t)\|_{l^2} \le C\varepsilon$, where, denoting $\langle \Psi_0 \rangle = \lim_{\varepsilon \to 0} \langle \Psi_\varepsilon \rangle$, we define $\rho_{\rm d}^{(3)}$ as the solution to

$$\partial_t \rho_{\rm d}^{(3)}(t) = (W + \langle \Psi_0 \rangle)_\sharp \ \rho_{\rm d}^{(3)}(t) \ , \qquad \rho_{\rm d}^{(3)}(0) = \rho_{\rm d}(0) \ .$$

(ii) Second case: $\mu > 0$. Then, there exists an explicit decomposition of the entries $\langle \Psi_\varepsilon \rangle^{\rm dom}(n,m)$ of the form

$$\langle \Psi_\varepsilon \rangle^{\rm dom}(n,m) = \sum_{j=0}^{I} \varepsilon^{-\nu_j} B_j^\varepsilon(n,m) + B_{I+1}^\varepsilon(n,m) + \mathcal{O}(\varepsilon^{\nu'}) \ ,$$

such that $\nu' > 0$, the exponents ν_j are positive, decreasing (with $\nu_0 = \mu$), and each B_j^ε is an ε-dependent matrix that satisfies

$$B_j^\varepsilon \underset{\varepsilon \to 0}{\longrightarrow} B_j^0 \ , \qquad \forall \varepsilon \ge 0 \ , \ {\rm Ker} B_j^\varepsilon = {\rm Ker} B_j^0 \ .$$

On the more, all linear Boltzmann operators $(B_j^\varepsilon)_\sharp$ define non-positive operators. Let Π be the orthogonal projection (in l^2) onto $\cap_{j \le I} {\rm Ker}(B_j^\varepsilon)_\sharp$, and set $\nu = \nu_I$. Then the following estimates hold true, for any $0 \le t \le T$

$$\begin{aligned} &\left\|(1-\Pi)\rho_{\rm d}^{(2)}(t)\right\|_{l^2} \le C \ \left(\varepsilon^\nu + \exp(-ct\varepsilon^{-\nu})\right) \ , \\ &\left\|\Pi\rho_{\rm d}^{(2)}(t) - \rho_{\rm d}^{(4)}(t)\right\|_{l^2} \le C \ \left(\varepsilon^\nu + \exp(-ct\varepsilon^{-\nu}) + \|B_{I+1}^\varepsilon - B_{I+1}^0\| + \varepsilon^{\nu'}\right) \ , \end{aligned}$$

for some constants $C > 0$ and $c > 0$. Here, $\rho_{\rm d}^{(4)}$ is defined as the solution to

$$\partial_t \rho_{\rm d}^{(4)}(t) = \Pi \ (W + B_{I+1}^0)_\sharp \ \Pi \rho_{\rm d}^{(4)}(t) \ , \qquad \rho_{\rm d}^{(4)}(0) = \Pi \rho_{\rm d}^{(2)}(0) \ .$$

The precise value of matrices B_j^ε is given in section 5.

Theorem 1.2 completely describes the asymptotic dynamics satisfied by $\rho_{\rm d}^{(2)}$: the density matrix $\rho_{\rm d}^{(2)}$ immediately relaxes to some equilibrium space (after a time of size ε^ν). This is a polarization process. Past this time, there only remains a "shadow dynamics" on the relevant space. It is described by the equations of part (i) or (ii), depending on the regime. The latter are linear Boltzmann equations.

In this second part of our analysis, the perturbation $\delta(n, m)$ has a true effect, as is seen in Table 1 below (where q is the minimum value of the exponent $q(n, m)$ such that $\delta_\varepsilon(n, m) \sim \overline{\delta}(n, m)\varepsilon^{q(n,m)}$ with $\overline{\delta}(n, m) \neq 0$; see also the formulae we give in section 5): the perturbation does affect the exponent ν, as well as the asymptotic value of the transition rates.

For an infinite number of levels, Theorem 1.2 cannot be proved. However, we give two results that go in direction of extending Theorem 1.2 to this case. First we prove that the dynamics of infinitely many levels ($N = +\infty$) can be approximated by the dynamics of the conveniently truncated N-level system ($< \infty$), uniformly in time, upon chosing N large enough. Second, for *some* values of the exponents μ and ν, an equivalent to Table 1 can be given. We refer to Section 6.

μ/q	ν	difference with the unperturbed case $\delta_\varepsilon(n, m) \equiv 0$
$0 \leq \mu/q < 1$	μ	none
$\mu/q = 1$	μ	value of the transition rates
$1 < \mu/q < 2$	$2q - \mu$	value of the transition rates, and size of the time-layer
$2 \leq \mu/q < \infty$	$0 < \nu \leq \mu$	value of the transition rates, size of the time-layer, value of the projector and limit rates

Table 1. Consequences of the perturbation for a finite number of levels.

Bibliographical remarks

Apart from [BFCD03], in the past few years an extensive attention has been paid on the rigourous derivation of Boltzmann type equations from dynamical models of (classical or quantum) particles or models for the interaction of waves with random media. Convergence results in the case of an electron in a periodic box may be found in [Cas99, Cas02, Cas01]. We also mention the non-convergence result established in [CP02, CP03] in a particular, periodic situation. For the case when an electron is weakly coupled to random obstacles, the reader may refer to [EY00, Spo77, Spo80, Spo91] and [KPR96]. The computation of the relevant cross-sections is performed in [Nie96]. All these results address the case of a linear Boltzmann equation. A nonlinear case is studied in [BCEP04].

Outline

The article is organized as follows:

• Section 2 is devoted to the introduction of the precise notations and assumptions needed in the sequel.

• In Section 3, we completely prove Theorem 1.1-(i). Our proof uses, in a first step, classical arguments for the Bloch equation in the weak coupling regime (see [Cas99, Cas02, Cas01] and also [KL57, LK58, Kre83, Zwa66] for this point). Then, to perform the averaging procedure that leads to Theorem 1.1-(i), we use techniques of the averaging theory for ordinary differential equations (see [LM88, SV85]).

• Section 4 is devoted to the proof of Theorem 1.1-(ii). First, we state and prove Lemma 4.1: it ensures that although the Diophantine condition is not stable with respect to small perturbations, violations of the condition only occur for large values of the indices. Then, we use this fact to compensate small denominators by extra smoothness assumptions.

• Section 5 is devoted to the proof of Theorem 1.2, i.e. to the analysis of the limit process $\varepsilon \to 0$ in equation (1.9), in the case of finitely many energy levels.

• Section 6 is devoted to partial extensions of Theorem 1.2 in the case of an infinite number of levels.

• Finally, we give in Section 7 the proofs of several lemmas, concerning continuity and non-positiveness of the relaxation operators first, then about existence and uniqueness of the associated equilibrium state (this is a light version of the Perron-Frobenius Theorem). We also show the genericity of the Diophantine condition 1.

2. Functional setting

In this section, we list the assumptions needed in our analysis, together with some basic properties of the solution to the Bloch equations. The main information is Lemma 2.1 below.

Basic decay assumptions on the various coefficients

The energy levels are assumed bounded:

$$(\omega(n))_{n\in\mathbb{N}} \in l^\infty \text{ and } (\delta(n))_{n\in\mathbb{N}} \in l^\infty$$

(which is natural, since these energies are bounded by the ionisation energy). Since $\delta_\varepsilon(n) = \varepsilon^{p(n)}\delta(n)$ is supposed to be a perturbation of $\omega(n)$, we assume

$$\inf_n p(n) =: p > 0.$$

In the same way, we suppose the relaxation coefficients satisfy

$$(\gamma(n,m))_{n,m\in\mathbb{N}} \in l^\infty, \qquad \inf_{n\neq m} \gamma(n,m) =: \gamma > 0.$$

Finally, the Pauli relaxation coefficients possess the following summability condition

$$\|W\|_{l^\infty_k l^1_n \cap l^\infty_n l^1_k} := \sup_k \sum_n |W(n,k)| + \sup_n \sum_k |W(n,k)| < \infty,$$

as does the (Hermitian) matrix $V(n,m)$:

$$\|V\|_{l^\infty l^1} := \sup_k \sum_n |V(n,k)| < \infty.$$

These last two assumptions ensure the corresponding terms in (1.1) have a bounded effect on the density matrix (this is a consequence of Schur's Lemma 7.1).

Boundedness and positivity of the density matrix
Our assumptions on V, W, and ϕ ensure that the operators involved in the right-hand-side of Eq. (1.1) are continuous on $L^\infty(\mathbb{R}^+, l^1)$. Classical ordinary differential equation arguments (see *e.g.* [Cas99]) then allow to state the existence and uniqueness of a solution ρ to System (1.1) for any initial data in l^1. It satisfies $\rho \in C^0(\mathbb{R}^+, l^1)$ and $\partial_t \rho \in L^\infty(\mathbb{R}^+, l^1)$.

The key point is that summation and positiveness of ρ are preserved through the time evolution. More precisely we have the following Lemma (points (*ii*), (*iii*) are addressed in [Lin76, BBR01, Cas01]).

Lemma 2.1. *Let $\rho(t=0)$ satisfy conditions* (1.2)*. Then, under the above assumptions, there exists a unique solution $\rho \in \mathcal{C}^0(\mathbb{R}^+, l^1)$ to Eq.* (1.1)*. It satisfies, for all $t \geq 0$,*

- *(i) $\rho(t)$ is Hermitian: $\rho(t,n,m) = \rho(t,m,n)^*$,*
- *(ii) the trace of ρ is conserved: $\sum_n \rho(t,n,n) = \sum_n \rho(0,n,n) < \infty$,*
- *(iii) positiveness of populations is conserved: $\rho(t,n,n) \geq 0$.*

We stress the importance of items *(ii)* and *(iii)*, first established in [Lin76]. They give a nontrivial l^1 estimate for the diagonal part ρ_{d}. This proves to be crucial in proving Theorem 1.1 (see also [Cas01] for a situation where the oscillations are much more difficult to handle).

Technical assumptions needed in the course of the proofs
Most importantly, and as it is usual in the field of oscillations in ordinary differential equations and averaging techniques (see [Arn89, SV85, LM88]), we introduce a Diophantine condition on the frequency vector ω.

Hypothesis 1 (Diophantine condition). There exists a (small) number $\eta > 0$, and a constant $C_\eta > 0$, such that

$$\begin{aligned} &\forall \alpha = (\alpha_1, \ldots, \alpha_r) \in \mathbb{Z}^r \setminus \{0\}, \quad \forall (n,k) \in \mathbb{N}^2 \text{ such that } \alpha\cdot\omega + \omega(n,k) \neq 0, \\ &\qquad |\alpha\cdot\omega + \omega(n,k)| \geq \frac{C_\eta}{(1+|\alpha|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}}, \end{aligned} \tag{2.10}$$

and similarly

$$\forall \alpha \in \mathbb{Z}^r \setminus \{0\}, \qquad |\alpha\cdot\omega| \geq \frac{C_\eta}{(1+|\alpha|)^{r-1+\eta}}. \tag{2.11}$$

Remark. *Given once and for all a fixed $\eta > 0$, we can classically claim (see [Arn89]) that there exists a constant $C_\eta > 0$, depending on $\omega(n,m)$ and on η, such that for* *almost all value of the frequency vector $\omega = (\omega_1, \dots, \omega_r)$ Hypothesis 1 is satisfied. This is proved in Appendix 7.2. This condition is therefore not much restrictive.*

The same kind of Diophantine estimate is also needed in the case when $\alpha = 0$. It means that the energies do not converge too fast towards the ionisation energy.

Hypothesis 2 (Convergence towards the ionisation energy). There exists $C_\eta > 0$, such that

$$\forall (n,k) \in \mathbb{N}^2, \text{ either } \omega(n,k) = 0, \text{ or } \quad |\omega(n,k)| \geq \frac{C_\eta}{(1+n)^{1+\eta}(1+k)^{1+\eta}},$$

where η is the number occurring in the Diophantine assumption (Hypothesis 1).

For more technical reasons, averaging procedures require a number of decay assumptions on the Fourier coefficients ϕ_α, as well as on $V(n,m)$ and $W(n,m)$. We list them below.

From Section 3 on, the summability condition $\sum_\alpha |\phi_\alpha| < \infty$ needs to be strengthened into

Hypothesis 3 (Smoothness assumption). The Fourier coefficients ϕ_α satisfy

$$\sum_\alpha (1 + |\alpha|)^{r-1+\eta} |\phi_\alpha|^2 < \infty,$$

where η is the number occurring in the Diophantine assumption (Hypothesis 1).

From Section 4 on, we need the following stronger decay. It is used to justify the sorting out of resonant and non resonant contributions.

Hypothesis 4 (Reinforced smoothness assumption). There exists $N_\eta > 2\mu/p$ such that the Fourier coefficients ϕ_α satisfy

$$\sum_\alpha (1 + |\alpha|)^{(r-1+\eta)N_\eta} |\phi_\alpha|^2 < \infty,$$

where η is the number occurring in the Diophantine assumption (Hypothesis 1).

Hypothesis 5 ("Far from continuous spectrum" assumption). The interaction coefficients satisfy

$$\sup_n \sum_m \left((1+n)^{1+\eta}(1+m)^{1+\eta} \right)^{N_\eta} |V(n,m)|^2 < \infty,$$

where η is the number occurring in the Diophantine assumption (Hypothesis 1) and N_η is given by Hypothesis 4.

The last hypothesis means that only low levels (*i.e.* levels which are far from the continuous spectrum or ionisation threshold) really interact with the wave, with a significant contribution.

Finally, in Section 4 we also use an assumption on the interaction of low and high levels *via* the relaxation operator.

Hypothesis 6 (Weak interaction of low and high energy levels). The longitudinal relaxation coefficients satisfy, for some $K > 0$,

$$\sup_n \sum_m (1+n)^K (1+m)^K |W(n,m)| < \infty.$$

3. Proof of Theorem 1.1, part (i)

In this section, we prove part (i) of Theorem 1.1. The proof is in two main steps, and follows [BFCD03]. First, we prove that the populations ρ_{d} tend to satisfy a closed equation, which is an ODE with oscillating coefficients. Second, we average out the coefficients.

We start with a basic observation. To simplify the writing, we set

$$\Omega^\varepsilon(n,m) := -i\omega(n,m) - i\delta_\varepsilon(n,m) - \varepsilon^\mu \gamma(n,m),$$

where we stress that $\Omega^\varepsilon(n,n) = 0$. With this notation, Eq. (1.1) reads for the coherences:

$$\begin{aligned} \partial_t \rho_{\mathrm{od}}(t,n,m) &= \frac{1}{\varepsilon^2} \Omega^\varepsilon(n,m) \rho_{\mathrm{od}}(t,n,m) \\ &+ \frac{i}{\varepsilon} \phi\left(\frac{t}{\varepsilon^2}\right) V(n,m) \left[\rho_{\mathrm{d}}(t,m) - \rho_{\mathrm{d}}(t,n)\right] \\ &+ \frac{i}{\varepsilon} \phi\left(\frac{t}{\varepsilon^2}\right) \sum_k \left[V(n,k)\, \rho_{\mathrm{od}}(t,k,m) - V(k,m)\, \rho_{\mathrm{od}}(t,n,k)\right], \end{aligned} \tag{3.12}$$

and, for the populations:

$$\begin{aligned} \partial_t \rho_{\mathrm{d}}(t,n) &= \frac{i}{\varepsilon} \phi\left(\frac{t}{\varepsilon^2}\right) \sum_k \left[V(n,k)\, \rho_{\mathrm{od}}(t,k,n) - V(k,n)\, \rho_{\mathrm{od}}(t,n,k)\right] \\ &+ \sum_k [W(k,n)\rho_{\mathrm{d}}(t,k) - W(n,k)\rho_{\mathrm{d}}(t,n)]. \end{aligned} \tag{3.13}$$

As a consequence of the Hermitian properties of Eq. (1.1) recalled in Lemma 2.1, Eq. (3.13) can also be cast as

$$\begin{aligned} \partial_t \rho_{\mathrm{d}}(t,n) &= -\frac{2}{\varepsilon} \mathrm{Im} \left[\sum_k \phi\left(\frac{t}{\varepsilon^2}\right) V(n,k)\, \rho_{\mathrm{od}}(t,k,n)\right] \\ &+ \sum_k [W(k,n)\rho_{\mathrm{d}}(t,k) - W(n,k)\rho_{\mathrm{d}}(t,n)]. \end{aligned} \tag{3.14}$$

We pass to the limit on this form of the equations.

3.1. *The populations satisfy an ODE with oscillating coefficients*

In this section, we prove the

Proposition 3.1. *Define the time dependent transition rate*

$$\Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right) := 2|V(n,k)|^2\mathrm{Re}\int_0^{t/\varepsilon^2} ds\ \exp\left(\Omega^\varepsilon(k,n)s\right)\phi\left(\frac{t}{\varepsilon^2}\right)\phi\left(\frac{t}{\varepsilon^2}-s\right).$$

Then, for all $T>0$, the vector ρ_{d} satisfies

$$\partial_t\rho_{\mathrm{d}}(t) = \left(\Psi_\varepsilon\left(\frac{t}{\varepsilon^2}\right)+W\right)_\sharp \rho_{\mathrm{d}}(t) + O_{L^\infty([0,T],l^1)}\left(\varepsilon^{1-2\mu}\right). \tag{3.15}$$

Remark. *Eq.* (3.15) *is a closed linear Boltzmann equation on the populations only, with a time dependent transition rate.*

Remark. *For $s\geq 0$, and $q\geq 1$, the symbol $O_{L^\infty_{\mathrm{loc}}(\mathbb{R},l^q)}\left(\varepsilon^s\right)$ means that for all $T>0$, there exists a constant $C>0$, that does not depend on ε, such that the corresponding term is bounded:*

$$\|O_{L^\infty([0,T],l^q)}(\varepsilon^s)\|_{L^\infty([0,T],l^q)} \leq C\varepsilon^s.$$

In the whole article C denotes constants which do not depend on the (small) parameter ε. It however possibly depends on all the coefficients of the problem and on the initial data, but we will never make this dependence explicit.

Remark. *Using Lemma 7.1 of the Appendix, and since $\mathrm{Re}\Omega^\varepsilon(n,m)\leq -\varepsilon^\mu\gamma$, uniformly in n and m, $n\neq m$, the operator Ψ_ε is a priori of order $\varepsilon^{-\mu}$ on l^2 or l^1. More precisely we have*

$$\left\|\Psi_\varepsilon(t/\varepsilon^2)_\sharp\rho_{\mathrm{d}}\right\|_{l^1} \leq C\varepsilon^{-\mu}\|V(n,k)\|^2_{l^\infty_n l^1_k\cap l^\infty_k l^1_n}\|\phi\|^2_{L^\infty(\mathbb{R})}\|\rho_{\mathrm{d}}\|_{l^1}, \tag{3.16}$$

$$\left\|\Psi_\varepsilon(t/\varepsilon^2)_\sharp\rho_{\mathrm{d}}\right\|_{l^2} \leq C\varepsilon^{-\mu}\|V(n,k)\|^2_{l^\infty_n l^1_k\cap l^\infty_k l^1_n}\|\phi\|^2_{L^\infty(\mathbb{R})}\|\rho_{\mathrm{d}}\|_{l^2}. \tag{3.17}$$

Proof. The proof is given in two steps that follow [BFCD03].
First step: computation of coherences
Since the initial data for coherences is $\rho_{\mathrm{od}}(t=0,n,m)\equiv 0$, the integral form for Eq. (3.12) reads

$$\begin{aligned}\rho_{\mathrm{od}}(t,n,m) &= i\varepsilon\int_0^{t/\varepsilon^2}\exp\left(\Omega^\varepsilon(n,m)s\right)\phi\left(\frac{t}{\varepsilon^2}-s\right)V(n,m)\\ &\quad\times\left[\rho_{\mathrm{d}}(t-\varepsilon^2 s,m)-\rho_{\mathrm{d}}(t-\varepsilon^2 s,n)\right]ds + (A_\varepsilon\rho_{\mathrm{od}})(t,n,m)\\ &=: \rho^{(0)}_{\mathrm{od}}(t,n,m) + (A_\varepsilon\rho_{\mathrm{od}})(t,n,m).\end{aligned} \tag{3.18}$$

This serves as a definition for $\rho^{(0)}_{\mathrm{od}}$. We have here defined the remainder A_ε as

$$\begin{aligned}(A_\varepsilon\rho_{\mathrm{od}})(t,n,m) &:= i\varepsilon\int_0^{t/\varepsilon^2}\exp\left(\Omega^\varepsilon(n,m)s\right)\\ &\times\sum_k\phi\left(\frac{t}{\varepsilon^2}-s\right)\left[V(n,k)\rho_{\mathrm{od}}(t-\varepsilon^2 s,k,m)-V(k,m)\rho_{\mathrm{od}}(t-\varepsilon^2 s,n,k)\right]ds.\end{aligned}$$

We now claim that for any given time $T \geq 0$, there exists a constant C, such that

$$\left\|\rho_{\rm od} - \rho_{\rm od}^{(0)}\right\|_{L^\infty([0,T],l^1)} \leq C\,\varepsilon^{2(1-\mu)}, \quad \left\|\rho_{\rm od}\right\|_{L^\infty([0,T],l^1)} \leq \qquad C\varepsilon^{1-\mu}. \tag{3.19}$$

Indeed, since $\mathrm{Re}\Omega^\varepsilon(n,m) \leq -\varepsilon^\mu \gamma < 0$, uniformly in n and m, $n \neq m$, we have

$$\begin{aligned}
&\|A_\varepsilon \rho_{\rm od}\|_{L^\infty([0,T],l^1)} \\
&\leq 2\varepsilon \left\| \int_0^{+\infty} ds \ \left|\exp\left(\Omega^\varepsilon(n,m)s\right)\right| \right\|_{l^\infty_{n,m}} \|\phi\|_{L^\infty} \|V\|_{l^\infty_n l^1_m \cap l^\infty_m l^1_n} \|\rho_{\rm od}\|_{L^\infty([0,T],l^1)} \\
&\leq C\varepsilon^{1-\mu} \|\rho_{\rm od}\|_{L^\infty([0,T],l^1)}.
\end{aligned}$$

Hence using Eq. (3.18), we recover

$$\begin{aligned}
\|\rho_{\rm od} - \rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)} &\leq C\varepsilon^{1-\mu} \|\rho_{\rm od}\|_{L^\infty([0,T],l^1)} \\
&\leq C\varepsilon^{1-\mu}\ \|\rho_{\rm od} - \rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)} + C\varepsilon^{1-\mu} \|\rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)}.
\end{aligned}$$

For ε small enough, it implies

$$\|\rho_{\rm od} - \rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)} \leq C\varepsilon^{1-\mu} \|\rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)}.$$

Then using the definition of $\rho_{\rm od}^{(0)}(t,n,m)$, we obtain

$$\|\rho_{\rm od}^{(0)}\|_{L^\infty([0,T],l^1)} \leq C\,\varepsilon^{1-\mu} \times \|\phi\|_{L^\infty}\ \|V\|_{l^\infty_n l^1_k \cap l^\infty_k l^1_n} \times \|\rho_{\rm d}\|_{L^\infty([0,T],l^1)}.$$

The crucial estimate now stems from the trace conservation property of Lemma 2.1, which reads $\|\rho_{\rm d}\|_{L^\infty([0,T],l^1)} = \|\rho_{\rm d}(t=0)\|_{l^1}$. This l^1-estimate on the diagonal terms of the density matrix is therefore sufficient to control all the off-diagonal terms in turn, and the claim (3.19) follows.

Second step: convergence to a delay-free equation

Estimate (3.19) together with Eq. (3.14) governing $\rho_{\rm d}$ imply that

$$\begin{aligned}
\partial_t \rho_{\rm d}(t,n) &= \sum_k [W(k,n)\rho_{\rm d}(t,k) - W(n,k)\rho_{\rm d}(t,n)] \\
&+ 2\sum_k \int_0^{t/\varepsilon^2} [\rho_{\rm d}(t-\varepsilon^2 s,k) - \rho_{\rm d}(t-\varepsilon^2 s,n)] \\
&\times \mathrm{Re}\left\{\exp(\Omega^\varepsilon(k,n)s)\,\phi\left(\frac{t}{\varepsilon^2}\right)\,\phi\left(\frac{t}{\varepsilon^2}-s\right)\,|V(k,n)|^2\right\} ds \\
&+ O_{L^\infty([0,T],l^1)}\left(\varepsilon^{1-2\mu}\right).
\end{aligned} \tag{3.20}$$

Now, the delayed term $\rho_{\rm d}(t-\varepsilon^2 s)$ reads

$$\rho_{\rm d}(t-\varepsilon^2 s,n) = \rho_{\rm d}(t,n) + O\left(\varepsilon^2 s\,\|\partial_t \rho_{\rm d}(\cdot,n)\|_{L^\infty([0,T])}\right).$$

Thus, Eq. (3.20) yields, using the shorter expressions defined in Notation 1.5,

$$\begin{aligned}
\partial_t \rho_{\rm d}(t,n) &= \left(\Psi_\varepsilon(t/\varepsilon^2)_\sharp\ \rho_{\rm d}\right)(t,n) \\
&+ (W_\sharp\ \rho_{\rm d})(t,n) + O_{L^\infty([0,T],l^1)}\left(\varepsilon^{1-2\mu}\right) + r_\varepsilon(t,n),
\end{aligned} \tag{3.21}$$

where the remainder r_ε can be estimated by

$$\begin{aligned}\|r_\varepsilon\|_{L^\infty([0,T],l^1)} &\le C\|\partial_t\rho_{\mathrm{d}}\|_{L^\infty([0,T],l^1)}\left\|\varepsilon^2\int_0^{+\infty} s\ \exp(\Omega^\varepsilon(n,m)s)\,ds\right\|_{l^\infty_{n,m}}\\ &\le C\varepsilon^{2-2\mu}\|\partial_t\rho_{\mathrm{d}}\|_{L^\infty([0,T],l^1)}\\ &\le C\varepsilon^{2-2\mu}\varepsilon^{-\mu}\|\rho_{\mathrm{d}}\|_{L^\infty([0,T],l^1)} \quad \text{thanks to Eqs (3.21) and (3.16)}\\ &\le C\varepsilon^{2-3\mu}.\end{aligned}$$

Including this new estimate in Eq. (3.21), Proposition 3.1 follows from

$$\partial_t\rho_{\mathrm{d}}(t,n) = \left((\Psi_\varepsilon)_\sharp\ \rho_{\mathrm{d}}\right)(t,n) + (W_\sharp\ \rho_{\mathrm{d}})(t,n) + O_{L^\infty([0,T],l^1)}\left(\varepsilon^{1-2\mu}+\varepsilon^{2-3\mu}\right). \quad \square$$

3.2. *Time averaging of transition rates*

Proposition 3.1 reduces the problem to the analysis of an ODE with fast oscillating coefficients. In this section we integrate out the oscillations of $\Psi_\varepsilon(t/\varepsilon^2)$, using averaging techniques (see *e.g.* [SV85]). As in [BFCD03], a crude averaging procedure in Eq. (3.15) would actually destroy the specific structure of the equations (rate equations), on which *all* our a priori estimates are based. This is why the simple but crucial remark that leads from Proposition 3.1 to Theorem 1.1 is the fact that the entries of $\langle\Psi_\varepsilon\rangle$ are *non-negative*. As a counterpart, this remark strongly relies on the specific form of the wave, and this is the reason why we restrict to the case of a quasiperiodic wave in all this paper (the result might be false otherwise).

The proof goes as follows. First, we observe the following

Lemma 3.1. *For any $\varepsilon > 0$, operator $\langle\Psi_\varepsilon\rangle_\sharp$ is a bounded non-positive operator on the Hilbert space l^2. In particular, the exponential $\exp(t\langle\Psi_\varepsilon\rangle_\sharp)$ is well defined as an operator on l^2 for $t \ge 0$, and its norm is less than 1, for all $t \ge 0$.*

(See Appendix 7.1, Lemma 7.1 for the proof).

Second, using the explicit value of the wave $\phi(t)$, we compute from Eq. (3.15) the time-dependent transition rate Ψ_ε:

$$\begin{aligned}\Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right) &= 2|V(n,k)|^2\mathrm{Re}\sum_{\alpha,\beta\in\mathbb{Z}^r}\phi_\alpha\phi_\beta\exp\left(i(\alpha+\beta)\cdot\omega\frac{t}{\varepsilon^2}\right)\\ &\times\frac{1-\exp\left([-\varepsilon^\mu\gamma(k,n)-i(\omega(k,n)+\beta\cdot\omega+\delta_\varepsilon(k,n))]t/\varepsilon^2\right)}{\varepsilon^\mu\gamma(k,n)+i(\omega(k,n)+\beta\cdot\omega+\delta_\varepsilon(k,n))}.\end{aligned} \tag{3.22}$$

We are now in position to perform the averaging procedure, which leads to replacing the transition rate $\Psi_\varepsilon(t/\varepsilon^2)$ by its mean value $\langle\Psi_\varepsilon\rangle(k,n) := \lim_{T\to+\infty}\dfrac{1}{T}\displaystyle\int_0^T ds\ \Psi_\varepsilon(s)$.

First step: splitting of $\boldsymbol{\Psi_\varepsilon}$ into an average, and an oscillating value

Theorem 1.1 amounts to estimating the difference $\Delta(t) := \rho_{\mathrm{d}}(t)-\rho_{\mathrm{d}}^{(1)}(t)$. Eq. (3.15) and Eq. (3.22) can be cast as

$$\partial_t \rho_{\mathrm{d}}(t) = \left(\Psi_\varepsilon \left(\frac{t}{\varepsilon^2}\right)_\sharp \, \rho_{\mathrm{d}}\right)(t) + (W_\sharp \, \rho_{\mathrm{d}})(t) + O\left(\varepsilon^{1-2\mu}\right),$$

$$\partial_t \rho_{\mathrm{d}}^{(1)}(t) = \left(\langle \Psi_\varepsilon \rangle_\sharp \, \rho_{\mathrm{d}}^{(1)}\right)(t) + \left(W_\sharp \, \rho_{\mathrm{d}}^{(1)}\right)(t).$$

Hence the difference $\Delta(t)$ satisfies the equation

$$\partial_t \Delta(t) = (\langle \Psi_\varepsilon \rangle_\sharp \, \Delta)(t) + \left(\Psi_\varepsilon^{\mathrm{osc}} \left(\frac{t}{\varepsilon^2}\right)_\sharp \, \rho_{\mathrm{d}}\right)(t) + (W_\sharp \, \Delta)(t) + O(\varepsilon^{1-2\mu}), \quad (3.23)$$

where $\Psi_\varepsilon^{\mathrm{osc}}\left(\frac{t}{\varepsilon^2}, k, n\right) := \Psi_\varepsilon\left(\frac{t}{\varepsilon^2}, k, n\right) - \langle \Psi_\varepsilon \rangle(k,n)$ contains the oscillatory contribution to the transition rate, which we want to prove to be negligible. Gathering the terms for which $\alpha + \beta = 0$, this contribution is equal to

$$\begin{aligned}
&\Psi_\varepsilon^{\mathrm{osc}}\left(\frac{t}{\varepsilon^2}, k, n\right) = \\
&2|V(n,k)|^2 \mathrm{Re}\Bigg(- \sum_{\beta \in \mathbb{Z}^r} \frac{|\phi_\beta|^2}{\varepsilon^\mu \gamma(k,n) + i(\omega(k,n) + \beta \cdot \omega + \delta_\varepsilon(k,n))} \\
&\qquad \times \exp\left([-\varepsilon^\mu \gamma(k,n) - i(\omega(k,n) + \beta \cdot \omega + \delta_\varepsilon(k,n))] \, \frac{t}{\varepsilon^2} \right) \\
&\quad + \sum_{\alpha \neq -\beta \in \mathbb{Z}^r} \frac{\phi_\alpha \phi_\beta \exp(i(\alpha+\beta)\cdot \omega t/\varepsilon^2)}{[\varepsilon^\mu \gamma(k,n) + i(\omega(k,n) + \beta \cdot \omega + \delta_\varepsilon(k,n))]} \\
&\qquad \times \left[1 - \exp\left([-\varepsilon^\mu \gamma(k,n) - i(\omega(k,n) + \beta \cdot \omega + \delta_\varepsilon(k,n))] \, \frac{t}{\varepsilon^2} \right) \right] \Bigg).
\end{aligned} \quad (3.24)$$

This expression carries "time-oscillations" (at frequency $\varepsilon^{-2+\mu}$ at least), which kill the possibly diverging factors $\varepsilon^{-\mu}$ (due to the denominators), and make them of size $\varepsilon^{2-2\mu}$, as we show now.

Second step: preliminary bounds

Since $V \in l_n^\infty l_k^1 \cap l_k^\infty l_n^1$ and $\sum_\beta |\phi_\beta|^2 < \infty$, we first find that

$$\|\langle \Psi_\varepsilon \rangle(k,n)\|_{l_k^\infty l_n^1 \cap l_n^\infty l_k^1} \leq C\varepsilon^{-\mu}, \quad \|\Psi_\varepsilon^{\mathrm{osc}}(t/\varepsilon^2, k, n)\|_{l_k^\infty l_n^1 \cap l_n^\infty l_k^1} C\varepsilon^{-\mu} \|u\|_{l^2},$$

for some $C > 0$ that does not depend on t and ε, which yields

$$\|\langle \Psi_\varepsilon \rangle_\sharp \, u\|_{l^2} \leq C\varepsilon^{-\mu} \|u\|_{l^2}, \quad \left\| \Psi_\varepsilon^{\mathrm{osc}}{}_\sharp \, u \right\|_{l^2} \leq C\varepsilon^{-\mu} \|u\|_{l^2}. \quad (3.25)$$

Now, according to Eq. (3.24), $\Psi_\varepsilon^{\mathrm{osc}}(t,k,n)$ is a sum of two different terms. We use the decay assumptions on V and ϕ_β to estimate the contribution of the first term by

$$C \left\| \sum_\beta |V(n,k)|^2 \frac{|\phi_\beta|^2}{|\varepsilon^\mu \gamma(k,n) + i(\omega(k,n) + \beta \cdot \omega + \delta_\varepsilon(k,n)|^2)} \right\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \leq C\varepsilon^{-2\mu}.$$

The second contribution is estimated by

$$
\begin{aligned}
&C\left\|\sum_{\alpha+\beta\neq 0}\frac{|V(n,k)|^2|\phi_\alpha||\phi_\beta|}{|\varepsilon^\mu\gamma(k,n)+i(\omega(k,n)+\beta\cdot\omega+\delta_\varepsilon(k,n))|}\cdot\frac{1}{|(\alpha+\beta)\cdot\omega|}\right\|_{l_n^\infty l_k^1\cap l_k^\infty l_n^1}\\
&\quad + C\left\|\sum_{\alpha+\beta\neq 0}\frac{|V(n,k)|^2|\phi_\alpha||\phi_\beta|}{|\varepsilon^\mu\gamma(k,n)+i((\alpha+\beta)\cdot\omega+\delta_\varepsilon(k,n))|^2}\right\|_{l_n^\infty l_k^1\cap l_k^\infty l_n^1}\\
&\quad \leq C\varepsilon^{-\mu}\sum_{\alpha,\beta}|\phi_\alpha||\phi_\beta||\alpha+\beta|^{r-1+\eta}+C\varepsilon^{-2\mu}\sum_{\alpha,\beta}|\phi_\alpha||\phi_\beta|\\
&\quad \leq C\varepsilon^{-2\mu},
\end{aligned}
$$

thanks to the Diophantine estimate (Hypothesis 1), together with Hypothesis 3. This yields

$$
\sup_{0\leq t\leq T}\left\|\int_0^{t/\varepsilon^2} ds\ \Psi_\varepsilon^{\rm osc}(s,k,n)\right\|_{l_n^\infty l_k^1\cap l_k^\infty l_n^1}\leq C\varepsilon^{-2\mu}. \tag{3.26}
$$

Third step: integral form of the equations, and final estimates
Since $\Delta(0)=0$, the integral form for Eq. (3.23) governing $\Delta(t)$ reads $\Delta(t):=\Delta^{(1)}(t)+\Delta^{(2)}(t)$, where

$$
\begin{aligned}
\Delta^{(1)}(t)&=\int_0^t ds\exp([t-s]\langle\Psi_\varepsilon\rangle_\sharp)\Psi_\varepsilon^{\rm osc}\left(\frac{s}{\varepsilon^2}\right)_\sharp\ \rho_{\rm d}(s),\\
\Delta^{(2)}(t)&=\int_0^t ds\exp([t-s]\langle\Psi_\varepsilon\rangle_\sharp)\left((W_\sharp\ \Delta)(s)+O(\varepsilon^{1-2\mu})\right).
\end{aligned}
$$

Lemma 3.1 gives the uniform boundedness of $\exp\left(t\langle\Psi_\varepsilon\rangle_\sharp\right)$, so that

$$
\|\Delta^{(2)}(t)\|_{l^2}\leq C\left(\varepsilon^{1-2\mu}+\int_0^t ds\ \|\Delta(s)\|_{l^2}\right). \tag{3.27}
$$

On the other hand, to take advantage of the time oscillations of the operator $\Psi_\varepsilon^{\rm osc}(t/\varepsilon^2)$, we carry out the natural integration by parts in the expression for $\Delta^{(1)}$:

$$
\begin{aligned}
\Delta^{(1)}(t)&=\varepsilon^2\left(\int_0^{t/\varepsilon^2} du\Psi_\varepsilon^{\rm osc}(u)\right)_\sharp\ \rho_{\rm d}(t)\\
&\quad+\varepsilon^2\int_0^t ds\ \exp([t-s]\langle\Psi_\varepsilon\rangle_\sharp)\langle\Psi_\varepsilon\rangle_\sharp\ \left(\int_0^{s/\varepsilon^2} du\Psi_\varepsilon^{\rm osc}(u)\right)_\sharp\ \rho_{\rm d}(s)\\
&\quad-\varepsilon^2\int_0^t ds\ \exp([t-s]\langle\Psi_\varepsilon\rangle_\sharp)\left(\int_0^{s/\varepsilon^2} du\Psi_\varepsilon^{\rm osc}(u)\right)_\sharp\\
&\qquad\times\left(\langle\Psi_\varepsilon\rangle+\Psi_\varepsilon^{\rm osc}\left(\frac{s}{\varepsilon^2}\right)+W+O(\varepsilon^{1-2\mu})\right)_\sharp\ \rho_{\rm d}(s),
\end{aligned}
$$

where we have used Eq. (3.15) to express $\partial_t \rho_{\rm d}(s)$. We deduce

$$\|\Delta^{(1)}\|_{L^\infty([0,T],l^2)} \leq C\varepsilon^{2-\mu} \sup_{0\leq t\leq T} \left\| \int_0^{t/\varepsilon^2} ds\ \Psi_\varepsilon^{\rm osc}(s) \right\|_{\mathcal{L}(l^2)} \|\rho_{\rm d}\|_{L^\infty([0,T],l^2)}.$$

Besides, from $\|\rho_{\rm d}\|_{L^\infty([0,T],l^2)} \leq \|\rho_{\rm d}\|_{L^\infty([0,T],l^1)} \leq C$, it follows that

$$\begin{aligned} \|\Delta^{(1)}\|_{L^\infty([0,T],l^2)} &\leq C\varepsilon^{2-\mu} \sup_{0\leq t\leq T} \left\| \int_0^{t/\varepsilon^2} ds\ \Psi_\varepsilon^{\rm osc}(s) \right\|_{\mathcal{L}(l^2)} \\ &\leq C\varepsilon^{2-\mu} \sup_{0\leq t\leq T} \left\| \int_0^{t/\varepsilon^2} ds\ \Psi_\varepsilon^{\rm osc}(s,k,n) \right\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \leq C\varepsilon^{2-3\mu}. \end{aligned}$$

This, together with estimate (3.27), and Gronwall lemma, yields $\|\Delta(t)\|_{L^\infty([0,T],l^2)} \leq C\varepsilon^{1-2\mu}$, and Theorem 1.1-(i) is proved.

4. Proof of Theorem 1.1, part (ii)

In this section we prove that the non-resonant contributions, which correspond to the triples (n,k,β) such that $\omega(k,n)+\beta\cdot\omega \neq 0$ in the transition rate (1.6), are negligible in the limit $\varepsilon \to 0$. We therefore replace the transition rate $\langle \Psi_\varepsilon \rangle$ by a purely resonant (or "dominant") transition rate $\langle \Psi_\varepsilon \rangle^{\rm dom}$. Due to small denominator problems, we need to reinforce the decay assumptions on the coefficients and assume Hypothesis 4 and 5 hold.

To prove Theorem 1.1-(ii), we have to understand the effect of a perturbation on Diophantine estimates. Lemma 4.1 below answers this problem. It is the key result of this section, and gives us the tool to avoid hard arithmetic problems linked to small denominators. The ultraviolet cutoff procedure attached with this Lemma is then performed in section 4.2.

4.1. *Perturbed Diophantine estimates*

Lemma 4.1. *If ω and $\omega(n,k)$ satisfy the Diophantine condition* (2.10) *and Hypothesis 2 with constants η and C_η, then the following assertion holds. Let $(n,k,\beta) \in \mathbb{N}\times\mathbb{N}\times\mathbb{Z}^r$ satisfy*

$$|\beta\cdot\omega + \omega(n,k) + \delta_\varepsilon(n,k)| \leq \frac{1}{2}\frac{C_\eta}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}},$$

then (with $p = \inf_n p(n)$)

$$(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta} \geq \frac{C_\eta \varepsilon^{-p}}{4|\delta|_{l^\infty}}.$$

Remark. *In other words, those coefficients $\omega(n,k)+\varepsilon^p\delta(n,k)$, that are capable of violating the Diophantine condition* (2.10)*, are necessarily associated with values of the triple (n,k,β) which are very large when $\varepsilon \to 0$.*

Proof.
Set $K = C_\eta/2$ and take (n,k,β) such that

$$|\beta\cdot\omega + \omega(n,k) + \delta_\varepsilon(n,k)| \le \frac{K}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}}.$$

Then

$$|\beta\cdot\omega + \omega(n,k)| - |\delta_\varepsilon(n,k)| \le \frac{K}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}},$$

and according to condition (2.10) (or Hypothesis 2, when $\beta = 0$)

$$\begin{aligned}\frac{2K}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}} &- |\delta_\varepsilon(n,k)| \\ &\le \frac{K}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}}.\end{aligned}$$

Hence

$$\frac{K}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}} \le |\delta_\varepsilon(n,k)| \le 2\varepsilon^p|\delta|_{l^\infty},$$

which ends the proof of Lemma 4.1. $\square$

4.2. *Proof of the main theorem: the ultra-violet cutoff procedure*

First step: Integral formulation
Our task is to estimate the difference $\Delta(t) = \rho_{\mathrm{d}}^{(2)}(t) - \rho_{\mathrm{d}}^{(1)}(t)$, where $\rho_{\mathrm{d}}^{(2)}(t)$ and $\rho_{\mathrm{d}}^{(1)}(t)$ are respectively solution to

$$\begin{aligned}\partial_t\rho_{\mathrm{d}}^{(2)}(t) &= \left(\langle\Psi_\varepsilon\rangle_\sharp^{\mathrm{dom}}\,\rho_{\mathrm{d}}^{(2)}\right)(t) + \left(W_\sharp\,\rho_{\mathrm{d}}^{(2)}\right)(t),\\ \partial_t\rho_{\mathrm{d}}^{(1)}(t) &= \left(\langle\Psi_\varepsilon\rangle_\sharp\,\rho_{\mathrm{d}}^{(1)}\right)(t) + \left(W_\sharp\,\rho_{\mathrm{d}}^{(1)}\right)(t),\end{aligned}$$

Hence

$$\partial_t\Delta(t) = (\langle\Psi_\varepsilon\rangle_\sharp\,\Delta)(t) + \left(\langle\Psi_\varepsilon\rangle_\sharp^{\mathrm{neg}}\,\rho_{\mathrm{d}}^{(2)}\right)(t) + (W_\sharp\,\Delta)(t). \tag{4.28}$$

Here, the transition coefficient

$$\langle\Psi_\varepsilon\rangle^{\mathrm{neg}}(k,n) = \langle\Psi_\varepsilon\rangle^{\mathrm{dom}}(k,n) - \langle\Psi_\varepsilon\rangle(k,n)$$

contains the contributions to the transition rate, that we want to prove to be negligible. Since $\Delta(0) = 0$ the integral form for (4.28) reads

$$\begin{aligned}\Delta(t) = \int_0^t ds\ &\exp([t-s]\langle\Psi_\varepsilon\rangle)\left(\langle\Psi_\varepsilon\rangle_\sharp^{\mathrm{neg}}\,\rho_{\mathrm{d}}^{(2)}\right)(s)\\ &+ \int_0^t ds\ \exp([t-s]\langle\Psi_\varepsilon\rangle)(W_\sharp\,\Delta)(s).\end{aligned}$$

Second step: Estimating $\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}$

In view of Eqs (1.6) and (1.8), we have

$$\|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} = \sup_n \sum_{k,\beta;\ \omega(n,k)+\beta\cdot\omega\neq 0} \frac{2|V(n,k)|^2 \varepsilon^\mu \gamma(k,n) |\phi_\beta|^2}{\varepsilon^{2\mu}\gamma(k,n)^2 + |\omega(k,n)+\beta\cdot\omega+\delta_\varepsilon(k,n)|^2}.$$

We split this expression into two contributions according to the fact that

$$|\beta\cdot\omega + \omega(n,k) + \delta_\varepsilon(n,k)| \geq \frac{1}{2} \frac{C_\eta}{(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}},$$

or not. Using Lemma 4.1 for the second contribution, we obtain

$$\begin{aligned}
\|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \leq \sup_n \Bigg\{ & \sum_{k,\beta;\ \omega(n,k)+\beta\cdot\omega\neq 0} \frac{4|V(n,k)|^2 \varepsilon^\mu \gamma(k,n) |\phi_\beta|^2}{C_\eta} \\
& \times (1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta} \\
+ & \sum_{k,\beta;\ \omega(n,k)+\beta\cdot\omega\neq 0} \mathbf{1}\Big[(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta} \geq C\varepsilon^{-p}\Big] \\
& \times \frac{2|V(n,k)|^2 \varepsilon^{-\mu} |\phi_\beta|^2}{\gamma} \Bigg\}.
\end{aligned}$$

The first sum is estimated using Hypotheses 4, 2 and 5. The second term is first multiplied and divided by the quantity $[(1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}]^{N_\eta}$. Therefore we get

$$\begin{aligned}
&\|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \leq C\varepsilon^\mu \\
&\quad + C\varepsilon^{N_\eta p - \mu} \sum_{n,k,\beta} \left((1+|\beta|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}\right)^{N_\eta} |V(n,k)|^2 |\phi_\beta|^2.
\end{aligned}$$

Since we assumed that $N_\eta > 2\mu/p$, we finally have $\|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \leq C\varepsilon^\mu$.

Third step: Conclusion

Using Lemma 3.1 (non-positiveness of $\langle \Psi_\varepsilon \rangle$) leads to

$$\begin{aligned}
\|\Delta(t)\|_{l^2} &\leq C \|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}\|_{\mathcal{L}(l^2)} \|\rho_{\mathrm{d}}^{(2)}\|_{L^\infty([0,T],l^2)} + C\int_0^t ds\ \|\Delta(s)\|_{l^2} \\
&\leq C \|\langle \Psi_\varepsilon \rangle^{\mathrm{neg}}(n,k)\|_{l_n^\infty l_k^1 \cap l_k^\infty l_n^1} \left[\|\Delta\|_{L^\infty([0,T],l^2)} + \|\rho_{\mathrm{d}}^{(1)}\|_{L^\infty([0,T],l^2)}\right] \\
&\quad + C\int_0^t ds\ \|\Delta(s)\|_{l^2}.
\end{aligned}$$

A by-product of Theorem 1.1-(i) is that the quantity $\|\rho_{\rm d}^{(1)}\|_{L^\infty([0,T],l^2)}$ can be estimated by a constant. Therefore

$$\begin{aligned}\|\Delta(t)\|_{l^2} &\le C\|\langle\Psi_\varepsilon\rangle^{\rm neg}(n,k)\|_{l_n^\infty l_k^1\cap l_k^\infty l_n^1}(1+\|\Delta\|_{L^\infty([0,T],l^2)}) + C\int_0^t ds\ \|\Delta(s)\|_{l^2}\\ &\le C\varepsilon^\mu(1+\|\Delta\|_{L^\infty([0,T],l^2)}) + C\int_0^t ds\ \|\Delta(s)\|_{l^2}.\end{aligned}$$

Thanks to the Gronwall lemma, we recover $\|\Delta\|_{L^\infty([0,T],l^2)} \le C\varepsilon^\mu$, and Theorem 1.1-(ii) is proved. □

5. Proof of Theorem 1.2: time-layers and limiting dynamics

In all this section we assume $N<\infty$. We derived in Theorem 1.1 the rate equation (1.9) on $\rho_{\rm d}^{(2)}$, with the (modified) transition rates

$$W_\varepsilon^{\rm mod}(n,m) = \langle\Psi_\varepsilon\rangle^{\rm dom}(n,m) + W(n,m).$$

For a fixed ε, the underlying dynamics is standard (see Appendix 7.1). However, in most cases, the coefficients become singular as $\varepsilon\to 0$. Therefore the time evolution of the solution obeys two different regimes as ε goes to zero: first a time-layer, and then relaxation to a slow, asymptotic dynamics. This is what we prove now.

5.1. *Setting for the time-layer*

We split $\langle\Psi_\varepsilon\rangle^{\rm dom}(n,m)$ into different contributions according to the size of $\delta_\varepsilon(n,m)$. To simplify notations, we set

$$C(n,m) = 2|V(n,m)|^2 \sum_{\beta\in\mathbb{Z}^r;\ \omega(n,m)+\beta\cdot\omega=0} |\phi_\beta|^2.$$

Now, observe that for given indices n,m, as $\varepsilon\to 0$, either $\delta_\varepsilon(n,m)\equiv 0$, or $\delta_\varepsilon(n,m)\sim \overline{\delta}(n,m)\varepsilon^{q(n,m)}$ with $\overline{\delta}(n,m)\neq 0$ and

$$q(n,m) = \min(p(n),p(m)) \quad (q(n,m) := \infty \text{ when } \delta_\varepsilon(n,m)\equiv 0). \tag{5.29}$$

Then we have the fairly natural decomposition

$$\langle\Psi_\varepsilon\rangle^{\rm dom}(n,m) = \sum_{j=0}^J \varepsilon^{-\nu_j} B_j^\varepsilon(n,m),$$

where B_0^ε accounts for the asymptotically unperturbed rates, and the other B_j^ε's take care of the perturbed ones, with decreasing strength of the singularity $\varepsilon^{-\nu_j}$. More precisely, we first define

$$\nu_0=\mu, \quad B_0^\varepsilon(n,m) = \frac{C(n,m)\,\gamma(n,m)}{\gamma(n,m)^2+\varepsilon^{-2\mu}\delta_\varepsilon(n,m)^2}\,\mathbf{1}[q(n,m)>\mu].$$

Then we take $(q_j)_{1\leq j\leq J}$ as the distinct, decreasing ordered values of the remaining $q(n,m)$'s, and for each $j\geq 1$, we set

$$\nu_j = 2q_j - \mu, \quad B_j^\varepsilon(n,m) = \frac{C(n,m)\,\gamma(n,m)}{\varepsilon^{2(\mu-q_j)}\gamma(n,m)^2 + \varepsilon^{-2q_j}\delta_\varepsilon(n,m)^2}\,\mathbf{1}[q(n,m)=q_j]\,.$$

With the above notations, we clearly have, for all j, $B_j^\varepsilon \to B_j^0$ in $l^\infty l^1$, as $\varepsilon\to 0$ (and therefore also as an operator on l^2). Also, Eq. (1.9) governing $\rho_{\mathrm{d}}^{(2)}$ reads accordingly

$$\partial_t \rho_{\mathrm{d}}^{(2)} = \left(\sum_{j=0}^{J} \varepsilon^{-\nu_j} B_j^\varepsilon + W\right)_\sharp \rho_{\mathrm{d}}^{(2)}. \tag{5.30}$$

Using the block decomposition described in Appendix 7.1, we notice the important fact that for all j, the kernel of B_j^ε is constant:

$$\forall\varepsilon\geq 0\,, \qquad \mathrm{Ker}(B_j^\varepsilon) = \mathrm{Ker}(B_j^0).$$

In the remainder part of this section, we exploit this decomposition of $\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}$ to describe completely the limiting dynamics of $\rho_{\mathrm{d}}^{(2)}$, solution to (5.30). Since the non-singular perturbations $\varepsilon^{-\nu_j}B_j^\varepsilon$ for $\nu_j<0$ have no influence on the dynamics for times $\mathcal{O}(1)$, when $\nu_J<0$, we define I such that

$$\mu = \nu_0 > \cdots > \nu_I > 0 = \nu_{I+1} > \cdots > \nu_J.$$

5.2. *A finite dimensional lemma*

Lemma 5.1. *Let $\nu_0 > \cdots > \nu_I > 0 > \nu_{I+2} > \cdots > \nu_J$. For each j, let $B_j^\varepsilon \in \mathcal{M}_N(\mathbb{R})$ be a symmetric non-positive matrix such that $B_j^\varepsilon \to B_j^0$, and assume that* $\mathrm{Ker}(B_j^\varepsilon)$ *is constant for $\varepsilon\geq 0$. Let Π be the orthogonal projection onto $\cap_{j\leq I}\mathrm{Ker}B^0$. Then there exists a constant $c>0$ and $\varepsilon_0>0$, such that any non-zero eigenvalue λ^ε of $(1-\Pi)(\sum_j \varepsilon^{-\nu_j}B_j^\varepsilon)(1-\Pi)$ satisfies*

$$\lambda^\varepsilon \leq -c\varepsilon^{-\nu_I},\ \forall\varepsilon\in]0,\varepsilon_0].$$

Proof. Let λ_ε be such an eigenvalue, and $x^\varepsilon\in\mathrm{Range}(1-\Pi)$ an associated eigenvector of $\sum_j \varepsilon^{-\nu_j}B_j^\varepsilon$ with norm one. We simply estimate, using the nonpositivity of the B_j^ε,

$$\varepsilon^{\nu_I}\lambda^\varepsilon\|x^\varepsilon\|^2 = \left(\left(\sum_j \varepsilon^{\nu_I-\nu_j}B_j^\varepsilon\right)x^\varepsilon, x^\varepsilon\right) \leq \sum_{j=0}^{I}(B_j^\varepsilon x^\varepsilon, x^\varepsilon) + o(1).$$

On the other hand, up to extracting a subsequence of x^ε converging to x^0, we have the convergence $(B_j^\varepsilon x^\varepsilon, x^\varepsilon)\to(B_j^0 x^0, x^0)$ for all j. Hence any number c such that $c<\min_{\|x\|\leq 1,\Pi x=0}(-(\sum_{j=0}^{I}B_j^0)x,x)$ gives the upper bound $\lambda^\varepsilon\leq -c\varepsilon^{-\nu_I}$, for ε small enough. $\square$

As an immediate consequence of Lemma 5.1, we obtain

Lemma 5.2. *Let $\nu_0 > \cdots > \nu_I > \nu_{I+1} = 0 > \nu_{I+2} > \cdots > \nu_J$. For each j, let $B_j^\varepsilon \in \mathcal{M}_N(\mathbb{R})$ be a symmetric matrix such that $B_j^\varepsilon \to B_j^0$, and assume that $\mathrm{Ker}(B_j^\varepsilon)_\sharp$ is constant for $\varepsilon \geq 0$. Let Π be the orthogonal projection onto $\cap_{j\leq I}\mathrm{Ker}(B_j^\varepsilon)_\sharp$. If y is solution to*

$$\partial_t y = \left(\sum_{j=0}^{J} \varepsilon^{-\nu_j} B_j^\varepsilon + W\right)_\sharp y$$

and if z is solution to

$$\partial_t z = \Pi(B_{I+1}^0)_\sharp\, \Pi z, \qquad z(0) = \Pi y(0), \tag{5.31}$$

then we have the error estimates

$$\|(1-\Pi)y\| \leq C(\varepsilon^{\nu_I} + \exp(-ct\varepsilon^{-\nu_I})), \tag{5.32}$$

$$\|\Pi(y-z)\| \leq C(\varepsilon^{\nu_I} + \exp(-ct\varepsilon^{-\nu_I}) + \|B_{I+1}^\varepsilon - B_{I+1}^0\| + \varepsilon^{\nu_{I+2}}), \tag{5.33}$$

where C depends on the B_j^0's, $y(0)$, T and c (the constant of Lemma 5.1).

Proof. Simply observe that $\partial_t(1-\Pi)y = (\sum_{j=0}^{I} \varepsilon^{-\nu_j} B_j^\varepsilon)_\sharp\ (1-\Pi)y + (1-\Pi)(B_{I+1}^\varepsilon)_\sharp\, y + o(\|y\|)$, from which we deduce

$$\|(1-\Pi)y\| \leq \exp(-ct\varepsilon^{-\nu_I})\|y(0)\| + C\int_0^t ds \exp(-(t-s)c\varepsilon^{-\nu_I})\|y(s)\|.$$

This establishes (5.32). To prove (5.33), write $\partial_t \Pi(y-z) = \Pi(B_{I+1}^0)_\sharp\ \Pi(y-z) + \Pi(B_{I+1}^\varepsilon - B_{I+1}^0)_\sharp\, \Pi y + \Pi(B_{I+1}^\varepsilon)_\sharp\ (1-\Pi)y + \Pi\sum_{j\geq I+2}(B_j^\varepsilon)_\sharp\ y$. $\square$

5.3. *Proof of Theorem 1.2*

Lemmas 5.1 and 5.2, together with the splitting obtained in section 5.1, imply Theorem 1.2. We simply explain here the different cases summarized in Table 1. Note that in the framework of [BFCD03] the B_j^ε's are identically zero for $j \geq 1$ (and $q = \infty$ in the discussion below).

(i) Case when $\mu = 0$. This case plays a special role, since we have to use the transition rates $\langle\Psi_\varepsilon\rangle$ of Theorem 1.1-(i) then. We have

$$\langle\Psi_\varepsilon\rangle(n,m) = 2|V(n,m)|^2 \sum_{\beta\in\mathbb{Z}^r} \frac{\gamma(n,m)}{\gamma(n,m)^2 + |\omega(n,m) + \beta\cdot\omega + \varepsilon^p\delta(n,m)|^2}|\phi_\beta|^2.$$

Hence at leading order, we have $\rho_{\mathrm{d}} = \rho_{\mathrm{d}}^{(3)}$, where

$$\partial_t \rho_{\mathrm{d}}^{(3)} = (\langle\Psi_0\rangle + W)_\sharp\ \rho_{\mathrm{d}}^{(3)}, \qquad \rho_{\mathrm{d}}^{(3)}(0) = \rho_{\mathrm{d}}(0),$$

and $\langle\Psi_0\rangle(n,m) = 2|V(n,m)|^2 \sum_{\beta\in\mathbb{Z}^r} \dfrac{\gamma(n,m)}{\gamma(n,m)^2 + |\omega(n,m) + \beta\cdot\omega|}|\phi_\beta|^2$.

(ii) Case when $\mu > 0$: the remaining cases depend on the value of $q{:=}\inf_{n,m} q(n,m)$ (see (5.29)), which fixes the possible values of the coefficients.

• When $q > \mu$, the transition rates satisfy $\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}(n,m) \sim \varepsilon^{-\mu} C(n,m)/\gamma(n,m)$. The size of the time layer (which is ε^μ), the value of the projector Π, and the limit transition rates $W(n,m)$, are the same as in the unperturbed case $q = \infty$.

• When $q = \mu$, the value of the transition rates is modified, since

$$\text{for } q_j = \mu, \quad B_j^0(n,m) = \frac{C(n,m)\,\gamma(n,m)}{\gamma(n,m)^2 + \overline{\delta}(n,m)^2} \mathbf{1}[q(n,m) = \mu].$$

• When $q \in]\mu/2, \mu[$, Eq. (5.30) really involves at least three different time scales (ε^μ, ε^{ν_J}, and 1), and another value of the transition rates is allowed,

$$\text{for } q_j \in [\mu/2, \mu[, \quad B_j^0(n,m) = \frac{C(n,m)\,\gamma(n,m)}{\overline{\delta}(n,m)^2} \mathbf{1}[q(n,m) = \mu].$$

• When $q = \mu/2$, in addition, the contribution of B_j^0 for $j = J$ competes with that of W, since the corresponding ν_j is zero. The limit transition rates are thus modified (W is replaced by $W + B_{I+1}^0$), as well as the projector Π ($\mathrm{Ker}(B_j^0)_\sharp$ is not included then).

• When q decreases below $\mu/2$, parts of $\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}$ asymptotically vanish (and this also modifies Π): those for which $\nu_j = 2q_j - \mu < 0$. Hence for small values of $q(n,m)$ compared to μ, two almost degenerate levels n and m for which $\delta_\varepsilon(n,m) \neq 0$ are already too far apart to resonate with the wave.

6. The infinite dimensional case

In this section, we give partial extensions of Theorem 1.2 in the case $N = +\infty$. The main point is Proposition 6.1, which allows to measure the error made in the infinite dimensional system when retaining only finitely many levels. Second, we give a result analogous to Theorem 1.2 for some range of the exponants μ and p.

6.1. *Restriction to a finite number of levels*

Consider a solution ρ to Bloch equations (1.1) with initial datum $\rho(0)$, and infinitely many quantum levels. For any given $N \in \mathbb{N}$, define π^N as the projection on the space $\mathbb{C}^{\mathbb{N}^2}$ of infinite matrices onto the space of $N \times N$ matrices, by

$$(\pi^N u)(n,m) := u(n,m)\mathbf{1}[n, m < N].$$

To ρ, we then associate the N-level *truncated system* (6.34),

$$\begin{aligned} &\varepsilon^2 \partial_t \rho^N(t,n,m) = -i\omega_\varepsilon(n,m)\rho^N(t,n,m) + (\pi^N Q_\varepsilon)(\rho^N)(n,m) \\ &+ i\varepsilon \sum_{k<N} \left[\phi\left(\frac{t}{\varepsilon^2}\right) V(n,k)\rho^N(t,k,m) - \phi\left(\frac{t}{\varepsilon^2}\right) V(k,m)\rho^N(t,n,k) \right]. \qquad (6.34) \end{aligned}$$

with initial datum $\rho^N(0)$ naturally defined as

$$\rho^N(0) := \pi^N \rho(0).$$

We prove below that the dynamics of ρ (the system with infinitely many energy levels) may be approximated by that of ρ^N, uniformly in time, upon taking N large enough. As a consequence, when $\varepsilon \to 0$, the dynamics of ρ resembles that of $\rho_{\mathrm{d}}^{N,(2)}$, the solution to the truncated system obtained from Eq. (1.9):

$$\partial_t \rho_{\mathrm{d}}^{N,(2)} = (\pi^N \langle \Psi_\varepsilon \rangle^{\mathrm{dom}} + \pi^N W)_\sharp \, \rho_{\mathrm{d}}^{N,(2)}, \quad \rho_{\mathrm{d}}^{N,(2)}(0) = \pi^N \rho(0). \tag{6.35}$$

Proposition 6.1. *Under Hypotheses 1 to 6, for all $\nu, T, \varepsilon > 0$, there exists an integer N such that,*

$$\text{if} \quad \|(1-\pi^N)\rho(0)\|_{l^2} \le \nu, \ \text{then } \|\rho - \rho^N\|_{L^\infty([0,T],l^2)} \le 2\nu + C(\varepsilon^\mu + \varepsilon^{1-2\mu}),$$

where $C = C(T)$ is the constant from Theorem 1.1, and N has the form

$$N = N_0(\nu, T)\varepsilon^{-\frac{\mu}{(1+\eta)N_\eta}},$$

with η and N_η given by Hypotheses 1 and 4.

If, in addition, no resonance occurs between the wave and high energy levels (i.e. there exists $M \in \mathbb{N}$ such that, when $\min(n,k) > M$, the set $\{\beta \in \mathbb{Z}^r;\ \omega(n,k)+\beta\cdot\omega = 0\}$ is empty), then N has the form $N = N_0(\nu, T)$, uniformly with respect to ε.

Proof. Theorem 1.1 applies for both the infinite and the finite number of levels problems, therefore

$$\begin{aligned}
\|\rho - \rho_{\mathrm{d}}^{(2)}\|_{L^\infty([0,T],l^2)} &\le C(\varepsilon^\mu + \varepsilon^{1-2\mu}),\\
\|\rho^N - \rho_{\mathrm{d}}^{N,(2)}\|_{L^\infty([0,T],l^2)} &\le C(\varepsilon^\mu + \varepsilon^{1-2\mu}).
\end{aligned}$$

We only need to estimate the difference $\Delta := \rho_{\mathrm{d}}^{(2)} - \rho_{\mathrm{d}}^{N,(2)}$, which is solution to

$$\partial_t \Delta = \big(\langle \Psi_\varepsilon \rangle^{\mathrm{dom}} + W\big)_\sharp \ \Delta + \big(\langle \Psi_\varepsilon \rangle^{\mathrm{dom}} - \langle \Psi_\varepsilon \rangle^{\mathrm{dom},N}\big)_\sharp \ \rho_{\mathrm{d}}^{N,(2)} + \big(W - \pi^N W\big)_\sharp \ \rho_{\mathrm{d}}^{N,(2)}.$$

Thanks to the non-positiveness property of the operators associated with $\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}$ and W, an integral formulation leads to

$$\begin{aligned}
&\|\Delta\|_{L^\infty([0,T],l^2)}\\
&\le C\Big(\|(1-\pi^N)\rho(0)\|_{l^2} + \|\big(\langle \Psi_\varepsilon \rangle^{\mathrm{dom}} - \pi^N \langle \Psi_\varepsilon \rangle^{\mathrm{dom}}\big)_\sharp \ \rho_{\mathrm{d}}^{N,(2)}\|_{L^\infty([0,T],l^2)}\\
&\qquad\qquad + \|\big(W - \pi^N W\big)_\sharp \ \rho_{\mathrm{d}}^{N,(2)}\|_{L^\infty([0,T],l^2)}\Big)\\
&\le C\Big(\|(1-\pi^N)\rho(0)\|_{l^2} + \|\langle \Psi_\varepsilon \rangle^{\mathrm{dom}} - \pi^N \langle \Psi_\varepsilon \rangle^{\mathrm{dom}}\|_{l_k^\infty l_n^1 \cap l_n^\infty l_k^1}\\
&\qquad\qquad + \|W - \pi^N W\|_{l_k^\infty l_n^1 \cap l_n^\infty l_k^1}\Big).
\end{aligned}$$

The first term goes to zero as N goes to infinity simply because the initial datum $\rho(0)$ is in l^2. The third one reads

$$\begin{aligned}
&\sup_{n>N}\sum_k |W(n,k)| + \sup_n \sum_{k>N} |W(n,k)| + \sup_{k>N}\sum_n |W(n,k)| + \sup_k \sum_{n>N} |W(n,k)| \\
&\quad \le C\left(\sup_{n>N}\sum_k |W(n,k)| + \sup_n \sum_{k>N} |W(n,k)|\right) \\
&\quad \le CN^{-K}\left(\sup_{n>N}\sum_k (1+n)^K |W(n,k)| + \sup_n \sum_{k>N} (1+k)^K |W(n,k)|\right) \le CN^{-K},
\end{aligned}$$

thanks to Hypothesis 6. Thus, this term is also $o(1)$ uniformly with respect to ε as N goes to infinity. Finally, taking into account the fact that $\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}$ is symmetric, the second term is

$$\begin{aligned}
&2\left(\sup_{n>N}\sum_k |\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}(n,k)| + \sup_n \sum_{k>N} |\langle\Psi_\varepsilon\rangle^{\mathrm{dom}}(n,k)|\right) \\
&\qquad \le C\varepsilon^{-\mu} \sup_{n>N}\sum_k \sum_{\beta;\ \omega(n,k)+\beta\cdot\omega=0} |\phi_\beta|^2 |V(n,k)|^2 \\
&\qquad \le C\varepsilon^{-\mu} N^{-(1+\eta)N_\eta} \sup_{n>N}\sum_k (1+n)^{(1+\eta)N_\eta} \sum_{\beta;\ \omega(n,k)+\beta\cdot\omega=0} |\phi_\beta|^2 |V(n,k)|^2 \\
&\qquad \le C\varepsilon^{-\mu} N^{-(1+\eta)N_\eta},
\end{aligned}$$

which vanishes in fact when no resonance occurs between the wave and high energy levels. Else, we obtain a $o(1)$ as ε goes to zero under the condition $N \gg \varepsilon^{-\mu/N_\eta(1+\eta)}$.

$\square$

6.2. *A version of Theorem 1.2*

In the infinite dimensional case, adopting the strategy of Section 5 above, the convergence of B^ε towards B^0 is not sufficient to conclude. In this paragraph however, we use series expansions when they only include one non-positive order in ε, i.e. when

$$\langle\Psi_\varepsilon\rangle^{\mathrm{dom}} + W = \varepsilon^{-\nu} A + o(1),$$

for some A and ν. In this case, we consider at leading order the solution $\rho_{\mathrm{d}}^{(5)}$ of

$$\partial_t \rho_{\mathrm{d}}^{(5)} = \varepsilon^{-\nu} A_\sharp\, \rho_{\mathrm{d}}^{(5)}. \tag{6.36}$$

Clearly, Eq. (6.36)) defines an approximate solution in that Gronwall's Lemma ensures

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(5)}\|_{L^\infty([0,T],l^2)} = o(1).$$

Now, we list the cases where such an homogeneous rate operator occurs, restricting the discussion to $\mu > 0$ for simplicity: all such cases anyhow require $W = 0$.

• **Case when $\mu < p$.** Series expansion yields

$$\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}(n,m) = \varepsilon^{-\mu} \frac{C(n,m)}{\gamma(n,m)} + O(\varepsilon^{2p-3\mu}),$$

and the above discussion allows to conclude if $2p - 3\mu > 0$.
• **Case when $\mu = p$.** No series expansion is needed and we have exactly

$$\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}(n,m) = \varepsilon^{-\mu} \frac{C(n,m)\gamma(n,m)}{\gamma(n,m)^2 + \delta(n,m)^2}.$$

• **Case when $\mu > p$.** Series expansion leads to

$$\begin{aligned}\langle \Psi_\varepsilon \rangle^{\mathrm{dom}}(n,m) &= \varepsilon^{-\mu} \frac{C(n,m)}{\gamma(n,m)} \mathbf{1}(\delta(n,m) = 0) \\ &\quad + \varepsilon^{-(2p-\mu)} \frac{C(n,m)\gamma(n,m)}{\delta(n,m)^2} \mathbf{1}(\delta(n,m) \neq 0) + O(\varepsilon^{3\mu-4p}).\end{aligned}$$

We can conclude in two cases: either $\delta(n,m)$ is always nonzero, $\mu > \frac{4}{3}p$. In this case $\nu = 2p - \mu$. Or $\mu > 2p$ and $\nu = \mu$.

7. Appendix

7.1. *Relaxation operators*

In this section, we give (more or less) standard lemmas concerning the general rate equation

$$\partial_t \rho_{\mathrm{d}}(t) = A_\sharp \; \rho_{\mathrm{d}}(t) \; . \tag{7.37}$$

7.1.1. *Continuity and non-positiveness*

Lemma 7.1. *Let $A(n,m) \in l^\infty_n l^1_m \cap l^\infty_m l^1_n$.*

(i) Its associated operator $A_\sharp$ is bounded on the spaces l^q, $1 \leq q \leq \infty$, and

$$\|A_\sharp \; u\|_{l^q} \leq \|A(n,m)\|_{l^\infty_n l^1_m \cap l^\infty_m l^1_n} \|u\|_{l^q}.$$

Similarly if $u \in l^q_{n,m}(\mathbb{N} \times \mathbb{N})$, we have

$$\left\| \sum_k A(n,k)u(k,m) - A(k,m)u(n,k) \right\|_{l^q_{n,m}} \leq \|A(n,m)\|_{l^\infty_n l^1_m \cap l^\infty_m l^1_n} \|u(n,m)\|_{l^q_{n,m}}.$$

(ii) If in addition $A(n,m) \geq 0$, then for all positive integer N, the spectrum of the restriction of $A_\sharp$ to $\mathbb{R}^N$ is contained in $\{\mathrm{Re}\lambda < 0\} \cup \{0\}$.

(iii) If $A(n,m) \geq 0$ is symmetric, $A_\sharp$ is non-positive on l^2, and the exponential $\exp(tA_\sharp)$ is well defined as an operator on l^2 when $t \geq 0$. Its norm is 1.

Proof.

(i) For $q = \infty$ or $q = 1$, the result is immediate. The remaining cases are obtained by interpolation.

(ii) The localization of the eigenvalues of $A_\sharp$ is obtained *via* the Hadamard-Gerschgorin method applied to $M := {}^t\overline{A_\sharp} = {}^tA_\sharp$, whose eigenvalues are the conjugates of those of $A_\sharp$: if λ is an eigenvalue of M, there exists an index n such that

$$|\lambda - M(n,n)| \leq \sum_{m \neq n} |M(n,m)|.$$

Remarking that for $m \neq n$, $M(n,m) = A_\sharp(m,n) \geq 0$, and $M(n,n) = -\sum_{m\neq n} M(n,m)$, the conclusion is straightforward.

(iii) In the symmetric case, compute for all $u \in l^2$,

$$\begin{aligned}(A_\sharp\, u, u) &= \sum_n \sum_{m \neq n} A_\sharp(n,m) u(m) u(n) + \sum_n A_\sharp(n,n) u(n)^2 \\ &\leq \frac{1}{2} \sum_n \sum_{m\neq n} A_\sharp(n,m)(|u(m)|^2 + |u(n)|^2) - \sum_n \sum_{m \neq n} A_\sharp(m,n) u(n)^2 = 0.\end{aligned}$$

The norm of $\exp(tA_\sharp)$ is 1 on l^2 for $t > 0$ because $A_\sharp$ has a non-trivial kernel (see below). $\square$

7.1.2. *Convergence to equilibrium*

We define the asymptotic state $\underline{\rho}$ associated with Eq. (7.37) and with the initial datum $\rho(0)$, the limit (in l^1), if it exists, of the solution $\rho_{\mathrm{d}}(t)$ to Eq. (7.37) with the initial value $\rho(0)$ as t goes to infinity. Such an asymptotic state necessarily belongs to the kernel of $A_\sharp$. For that reason, we study the kernel of operators $A_\sharp$ modelled on $W_\sharp$. To do so, we require that the –finite or infinite– matrix A, written in the eigenstates basis $e = (e_1, e_2, \dots)$, should have the following property $(\mathcal{P})$.

$$A(m,n) = 0 \Leftrightarrow A(n,m) = 0. \qquad (\mathcal{P})$$

Such an assumption is clearly satisfied by the coefficients W of the introduction (see (1.4)): a vanishing column in $A_\sharp$ corresponds to a vanishing line, and conversely.

For such a matrix, it is possible to split $A_\sharp$ into irreducible blocks, as follows: each block is built up upon retaining the coefficients $A_\sharp^L := \{(A_\sharp)(n,m)\}_{n,m\in L}$, where L is some subset of indices, characterized by the property

$$\begin{aligned} m \leq n \in L \quad \Leftrightarrow \quad \exists m =: m_1 \leq \cdots \leq m_s := n,\ &\text{such that} \\ m_j \in L \text{ for all } &j = 1, \dots, s \\ \text{and } A_\sharp(m_j, m_{j+1}) \neq 0 \text{ for all } &j = 1, \dots, s-1. \end{aligned}$$

(this is the irreducibility property of the Perron-Frobenius theorem). Under these circumstances, the kernel of $A_\sharp$ is simply the direct sum of the kernels of all the irreducible blocks $A_\sharp^L$. More precisely, the Perron-Frobenius theorem, together with the positivity property of Lemma 7.1, immediately imply the

Proposition 7.1. *Let $A(m,n) \in l_m^\infty l_n^1 \cap l_n^\infty l_m^1$ satisfy property $(\mathcal{P})$. Assume $A(m,n) \geq 0$ when $m \neq n$. In addition, suppose that the above decomposition of*

$A_\sharp$ only involves finite dimensional *irreducible blocks $A_\sharp^L$. Then, each irreducible block $A_\sharp^L$ has a one-dimensional kernel.*
As a corollary, for each initial datum $\rho(0) \in l^1$ satisfying Eq. (1.2)*, there is a* unique *asymptotic state $\underline{\rho}$, i.e. an element of $l^1 \cap \mathrm{Ker} A_\sharp$ that satisfies the additional trace constraint*

$$\sum_{n\in L} \underline{\rho}(n) = \sum_{n\in L} \rho(0,n), \tag{7.38}$$

whenever $A_\sharp^L := \{(A_\sharp)(m,n)\}_{m,n\in L}$ is an irreducible block of $A_\sharp$.

Proof. Simply observe that the vector $\sum_{n\in L} e_n$ belongs to the kernel of ${}^tA_\sharp^L$. Then, the positivity property of Lemma 7.1 and the Perron-Frobenius Theorem immediately imply that 0 is the largest eigenvalue of $A_\sharp^L$: it has thus multiplicity one. □

In the case when $A_\sharp$ possesses an *infinite dimensional* irreducible block, proposition 7.1 becomes false. We have the

Proposition 7.2. *Consider $A(m,n) \in l_m^\infty l_n^1 \cap l_n^\infty l_m^1$ satisfying property $(\mathcal{P})$, $A(m,n) \geq 0$ when $m \neq n$, and either symmetric, or in Pauli form (relation* (1.4)*). Suppose that there exists an irreducible block $A_\sharp^L$ that has* infinite dimension*. Then, the kernel of ${A_\sharp}^L$, as an operator on l^1, is $\{0\}$.*

As a corollary, for any initial datum $\rho(0)$ such that the sequence $\{\rho(0,n)\}_{n\in L}$ is non-identically zero, there is no asymptotic state *$\underline{\rho}$, i.e. no element $\underline{\rho} \in l^1 \cap \mathrm{Ker} A_\sharp$ that satisfies the trace constraint* (7.38)*.*

Proof. We have

$$\begin{aligned} u \in \mathrm{Ker} A_\sharp^L \Leftrightarrow\ & {}^tu \in \mathrm{Ker}\, {}^tA_\sharp^L && \text{in the symmetric case,} \\ \Leftrightarrow\ & \left(u(n) \exp\left(\frac{\omega(n)}{T}\right);\ n \in L \right) \in \mathrm{Ker}\, {}^tA_\sharp^L && \text{in the ``Pauli'' case.} \end{aligned}$$

Since we have the bounds

$$0 < \exp\left(\frac{\omega(1)}{T}\right) \leq \exp\left(\frac{\omega(n)}{T}\right) \leq \exp\left(\frac{\omega_{\text{ionisation}}}{T}\right)$$

for all n, this correspondence preserves the summability property. Finally, the proof of Proposition 7.1 shows that the kernel of ${}^tA_\sharp^E$ *in* l^1 is $\{0\}$. This gives the result□

Remark. *In the symmetric case (when $A(m,n) = A(n,m)$), according to Lemma 7.1, the l^2-norm of the solution ρ_d to Eq.* (7.37) *is decreasing in time; thus, it tends to a certain value $r \geq 0$. This means that ρ_d approaches a limit cycle in l^2 belonging the intersection of the sphere $\|\rho_\mathrm{d}\|_{l^2} = r$ and the hyperplane where the l^1-norm is one (assuming for simplicity that there is no strict $A_\sharp$-stable subspace of l^1). In this case, only weak convergence (to zero) can occur.*

7.2. *Diophantine estimates*

We show the genericity of Hypothesis 1.

Lemma 7.2. *For all $\eta > 0$ and all real sequence $\omega(n,m)$, there exists a constant $C_\eta > 0$, such that for almost all value of the frequency vector $\omega = (\omega_1, \dots, \omega_r)$,*

$$\forall \alpha = (\alpha_1, \dots, \alpha_r) \in \mathbb{Z}^r \setminus \{0\}, \quad \forall (n,k) \in \mathbb{N}^2 \text{ such that } \alpha \cdot \omega + \omega(n,k) \neq 0,$$
$$|\alpha \cdot \omega + \omega(n,k)| \geq \frac{C_\eta}{(1+|\alpha|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}}.$$

Proof. We follow the standard approach (see *e.g.* [AG91]). Restricting ω to a ball B in $\mathbb{R}^r$, we show that the measure of the set of "bad frequencies" violating the inequality for all constant C is zero.

For $\eta, c > 0$, $\alpha \in \mathbb{Z}^r \setminus \{0\}$ and $(n,k) \in \mathbb{N}^2$ fixed, set

$$B^{\eta,c}_{\alpha,n,k} := \left\{ \omega \in B;\ |\alpha \cdot \omega + \omega(n,k)| \leq \frac{c}{(1+|\alpha|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}} \right\}.$$

This limitates ω in the direction of α. Introducing a constant K which depends on the size of B only, we obtain

$$\mathrm{meas}\left(B^{\eta,c}_{\alpha,n,k}\right) \leq \frac{Kc}{(1+|\alpha|)^{r-1+\eta}(1+n)^{1+\eta}(1+k)^{1+\eta}}.$$

Now, with $\eta, c > 0$ fixed, the measure of the set of frequencies for which the inequality is false at least for some (α, n, k) is less than the sum (over α, n and k) of the ones above, and thus is $O(c)$. □

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