From Bloch model to the rate equations

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Abstract. We consider Bloch equations which govern the evolution of the density matrix of an atom (or: a quantum system) with a discrete set of energy levels. The system is forced by a time dependent electric potential which varies on a fast scale and we address the long time evolution of the system. We show that the diagonal part of the density matrix is asymptotically solution to a linear Boltzmann equation, in which transition rates are appropriate time averages of the potential. This study provides a mathematical justification of the approximation of Bloch equations by rate equations, as described in *e.g.* [Lou91]. The techniques used stem from manipulations on the density matrix and the averaging theory for ordinary differential equations. Diophantine estimates play a key role in the analysis.

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1 Introduction

In this article, we address an asymptotic model as $\varepsilon \to 0$ for the scaled Bloch equations

$$\begin{aligned}\varepsilon^2 \partial_t \rho(t,n,m) = &- \left(i\omega(n,m) + \gamma(n,m)\right) \rho(t,n,m) \\ &+ i\varepsilon \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k\right) \rho(t,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, m\right) \rho(t,n,k) \right] . \end{aligned} \tag{1}$$

More precisely, we wish to compute the asymptotic dynamics of the diagonal quantity

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

Here, $\rho(t,n,m)$ denotes the density matrix of an atomic system with discrete energy levels indexed by the integer n. The diagonal quantity $\rho_{\rm d}(t,n)$ is the occupation number of the n-th level, and the off-diagonal part $\rho(t,n,m)$ $(n \neq m)$ takes into account "correlations" between the various occupation numbers. Equation (1) describes the evolution of the atom (or the quantum system with a discrete number of energy levels), forced by a high frequency electromagnetic wave. In our scaling, the time evolution of the atom is considered over long times, of size $1/\varepsilon^2$, and the influence of the electromagnetic wave is both weak, of size ε, and it depends on the fast time scale t/ε^2 as well.

In equation (1), the quantity $\omega(n,m) = \omega(n) - \omega(m)$ is the difference between the energies $\omega(n)$ and $\omega(m)$ of atomic levels n and m respectively. Also, starting from (1), we readily assume that the atom has a natural tendency to relax to a given equilibrium state, *via* the relaxation coefficients $\gamma(n,m) > 0$ $(n \neq m)$ which act on the off-diagonal part of the density matrix only. The basic dynamics of the atom (forgetting the forcing by $\mathcal{V}$ for the moment) is thus given by a (damped) high-frequency oscillation, with frequency $1/\varepsilon^2$. On the other hand, the amplitude of the electromagnetic wave is assumed to be connected to the quantity $\mathcal{V}(t/\varepsilon^2,n,k)$ *via* the relation

$$\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right) = \phi\left(\frac{t}{\varepsilon^2}\right) V(n,k)\ ,$$

where $V(n,k)$ is an entry in the interaction potential matrix between the wave and the atom, and ϕ takes the time dependence of the wave into account. In the usual dipolar approximation (see [Lou91]), $V(n,k)$ is, up to a physical constant, an entry in the product operator matrix by the position vector written in the basis of the eigenfunctions of the atomic system. In the sequel we require this quantity to be given.

In the present regime, the dominant phenomenon as $\varepsilon \to 0$ is determined by the resonances between the high-frequency oscillations of the electromagnetic source (carried by $\phi(t/\varepsilon^2)$), and that of the atom (carried by the eigen-frequencies $\omega(n,m)$ in (1)): the wave/atom interaction enforces transitions of the quantum system between its various energy levels, a phenomenon in which those levels n and m such that the eigenfrequency $\omega(n,m)$ is resonant with the wave's frequencies, are coupled more strongly. Note that in (1), the cumulated effect of the forcing (of size ε) over the long time scale $1/\varepsilon^2$ seems at first glance diverging, of size $1/\varepsilon$: the very Hamiltonian nature of the dynamics (1) in fact makes the forcing play at second order only, so that the scaled equation (1) indeed has a limit as $\varepsilon \to 0$ (this is a weak coupling regime).

The aim of this article is to show that, in the limit $\varepsilon \to 0$, the populations of the atomic energy levels, given by the quantities $\rho(t,n,n) =: \rho_{\rm d}(t,n)$, tend to obey a linear Boltzmann equation which reads

$$\partial_t \rho_{\rm d}(t,n) = \sum_k \sigma(n,k)[\rho_{\rm d}(t,k) - \rho_{\rm d}(t,n)]. \tag{2}$$

The transition rates $\sigma(n,k)$ depend on the very value of the wave ϕ and on the atom's parameters $\omega(n,m)$, $\gamma(n,m)$, $V(n,m)$. They describe the above-mentioned resonance phenomenon. In all events, we of course have $\sigma(n,k) \geq 0$, as well as the symmetry $\sigma(n,k) = \sigma(k,n)$ (micro-reversibility).

Equation (2) provides an asymptotic model for (1). We point out that both the starting model (1) and the limiting equation (2) correspond to time-irreversible processes. Boltzmann-type equations as Eq. (2) have first been suggested by Einstein, on heuristic grounds, for two-level systems and are therefore sometimes called Einstein rate equations [Lou91].

In this paper, we show more precisely the following results.

Main Results.

- **1st case:** *the relaxation coefficients occurring in the Bloch equation (1) are* uniform *with respect to the small parameter ε, in that*

$$\inf_{n \neq m} \gamma(n,m) =: \gamma > 0.$$

In this case we establish the predicted convergence of Eq. (1) to Eq. (2) for different types of waves:

  (i) ϕ is a periodic or a quasi-periodic function (in both cases, the convergence rate from Eq. (1) to Eq. (2) can be estimated).

  (ii) ϕ is an almost periodic function, or a KBM-function (see [SV85]) (in this case, we obtain no better convergence rate than $o(1)$).

  (iii) ϕ has a slightly broadened frequency spectrum (here again we obtain no better convergence rate than $o(1)$).

The accurate meaning of Hypotheses (ii) and (iii) is stated further.

- **2nd case:** *the relaxation coefficients occurring in the Bloch equation (1) vanish as ε tends to zero, in that*

$$\inf_{n \neq m} \gamma(n,m) = \varepsilon^{\mu} \to 0,$$

for some power index $\mu > 0$. In this case, and with some smallness restrictions on the coefficient μ, we show in a way that the solution to the Bloch equation (1) tends to relax immediately to the equilibrium state of the Boltzmann equation (2). More precisely, the populations tend to satisfy the long time Boltzmann equation:

$$\partial_t \rho_{\mathrm{d}}(t,n) = \frac{1}{\varepsilon^{\mu}} \sum_k \sigma(n,k)[\rho_{\mathrm{d}}(t,k) - \rho_{\mathrm{d}}(t,n)].$$

This describes a fast convergence to the equilibrium of the operator induced by the transition rates σ (see [BFCDG03] for details on this point).

All these statements are detailed, respectively, in Theorem 1 (point (i)), Theorem 2 (point (ii)), Theorem 3 (point (iii)), and finally Theorem 4 (2nd case).

On the other hand, our analysis makes use of two types of techniques.

- First, we use classical arguments for the Bloch equation in the weak coupling regime, which makes possible to transform Eq. (1), as ε tends to 0, into a *closed* equation governing only the populations. We refer to [Cas99], [Cas02], [Cas01], or also [KL57], [KL58], [Kre83], [Zwa66] for this point.

- Second, we use classical techniques of the averaging theory for ordinary differential equations, see [LM88], [SV85] about this topic. In particular, Diophantine estimates naturally play a key role in the analysis.

We mention that an extension of the present analysis is performed in [BFCDG03], in the case where the energy-levels $\omega(n)$, $\omega(m)$ depend upon ε (this situation naturally comes up while considering quantum dots): here, *perturbed* Diophantine estimates are needed, that require a different procedure.

In the past years an extensive attention has been paid on the rigorous derivation of Boltzmann type equations from dynamical models of (classical or quantum) particles, or from models of interaction between waves and random media. We may cite [Cas99], [Cas02], [Cas01] for convergence results in the case of an electron in a periodic box. We mention the non-convergence result established in [CP02], [CP03] in a particular, periodic situation. We also quote [EY00], [Spo77], [Spo80], [Spo91] in the case of an electron weakly coupled to random obstacles, as well as the formal analysis performed in [KPR96], and the computation of the relevant cross-sections performed in [Nie96]. All these results treat the case of a linear Boltzmann equation. Let us also quote [BCEP04] for the nonlinear case. Besides, we refer the reader to [Boh79], [Boy92], [CTDRG88], [Lou91], [NM92], [SSL77] for physics textbooks about wave/matter interaction issues, which is the context of the specific problem we deal with here.

The article is organized as follows. In Section 2, we treat the case when the relaxation coefficients $\gamma(n,m)$ (for $n \neq m$) are assumed to be uniformly bounded below by a positive constant. Section 3 handles the case when the relaxation coefficients tend to zero with ε following a below-mentioned scaling hypothesis. The main results are Theorems 1, 2, 3, and 4. In the sequel, C denotes any number which does not depend on ε.

2 The uniform relaxation case

2.1 The model

We want to pass to the limit $\varepsilon \to 0$ in the following equations

$$\begin{aligned}\varepsilon^2 \partial_t \rho(t,n,m) = &-\left(i\omega(n,m) + \gamma(n,m)\right)\rho(t,n,m) \\ &+ i\varepsilon \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k\right)\rho(t,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, m\right)\rho(t,n,k)\right]. \end{aligned} \tag{3}$$

Here, the discrete indices n, m, and k index the energy levels. They belong to the set $\{1, 2, \ldots, N\}$ for some integer $N \geq 2$ (the number of levels) which can possibly be $N = +\infty$ (infinite number of levels).

Remark 1. Without altering the analysis below, we can add to Eq. (3) a relaxation term $\varepsilon^2 Q(\rho)(t,n,n)$ that reads

$$Q(\rho)(t,n,n) = \sum_p (W(p,n)\rho(t,p,p) - W(n,p)\rho(t,n,n)),$$

where $W(n,p)$ denotes the transition rate from level n to level p. Such a term would account for any process in the atomic system contributing to redistribute the populations of the different energy levels among themselves (like thermal processes, for example). In contrast, the $\gamma(n,p)$ relaxation coefficients model the processes that alter the coherence between the levels without any effect on the populations. The assumption that the relaxation for coherence (cf. below) are much faster than those for the populations underlies our analysis. This corresponds to the physical regimes used in practice [Lou91].

Here and in all this Section 2, we make the following assumptions.

- At the initial time $t = 0$, we assume that ρ is a density matrix, with vanishing off-diagonal terms (coherence), and non negative and summable diagonal terms (populations). More precisely, we suppose that

$$\rho(0,n,m) = 0, \quad \forall n \neq m, \qquad \rho(0,n,n) \geq 0, \quad \forall n, \\ \text{and} \ \ \sum_n \rho(0,n,n) < \infty. \tag{4}$$

- We require that $\gamma(n,m) > 0$ for all $n \neq m$, and $\gamma(n,n) = 0$ for all n. More exactly, we take

$$\inf_{(n,m), n \neq m} \gamma(n,m) =: \gamma > 0 \ . \tag{5}$$

  We also assume that the symmetry property $\gamma(n,m) = \gamma(m,n)$ holds for all n, m.

- We suppose that there exists a sequence $\omega(n) \in \mathbb{R}$ such that

$$\omega(n,m) = \omega(n) - \omega(m) \ .$$

- Last, we specify

$$\mathcal{V}(t,n,m) = \phi(t) V(n,m) \ ,$$

  where ϕ is a real-valued function which is bounded on $\mathbb{R}$. We moreover consider the case when the doubly indexed sequence $V(n,m)$ is Hermitian, that is $V(n,m) = V(m,n)^*$ (the star denotes the complex conjugate). In the case when $N = +\infty$, we suppose as well that

$$\sup_{n \in \mathbb{N}^*} \sum_{m \in \mathbb{N}^*} |V(n,m)| + \sup_{m \in \mathbb{N}^*} \sum_{n \in \mathbb{N}^*} |V(n,m)| < \infty \ , \tag{6}$$

  which we will denote by $V(n,m) \in l^1 l^\infty \cap l^\infty l^1$. This is an abuse of notation. The physical cases do correspond to such decay assumptions.

  We note that some more specific assumptions will have to be done on ϕ later on in this section, namely: Hypothesis 1 in Section 2.3 (r-chromatic wave), Hypothesis 2 in Section 2.4.1 (KBM-wave), and Hypothesis 3 in Section 2.4.2 (wave with spectrum broadening).

Before going any further, we introduce some notations which will be used in the sequel.

- We denote $\rho_{\mathrm{od}}(t,n,m) := \rho(t,n,m) \ \mathbf{1}[n \neq m]$. The quantity ρ_{od} represents the off-diagonal part of the density matrix ρ, also called *coherence*.

- In the same way, we denote $\rho_{\mathrm{d}}(t,n) := \rho(t,n,n)$. This is the diagonal part of ρ, also called *populations*.

- We set $\Omega(n,m) := -i\omega(n,m) - \gamma(n,m)$ and with this notation it must be understood that $n \neq m$ (of course, for $n = m$, $\Omega(n,m) = 0$ holds true).

- Given a sequence $u(n)$ or a sequence $v(n,m)$, simply or doubly indexed, the quantities $\|u\|_{l^1}$ or $\|v\|_{l^1}$ denote respectively $\sum_n |u(n)|$ or $\sum_{n,m} |v(n,m)|$, where the indices n and m belong to finite or infinite sets. In the same way, for a doubly indexed sequence $v(n,m)$, we set

$$\|v\|_{l^1 l^\infty \cap l^\infty l^1} := \sup_n \sum_m |v(n,m)| + \sup_m \sum_n |v(n,m)|.$$

With these notations, System (3) reads for the coherence

$$\partial_t \rho_{\mathrm{od}}(t,n,m) = \frac{\Omega(n,m)}{\varepsilon^2}\rho_{\mathrm{od}}(t,n,m) + \frac{i}{\varepsilon}\mathcal{V}\left(\frac{t}{\varepsilon^2},n,m\right)\left[\rho_{\mathrm{d}}(t,m) - \rho_{\mathrm{d}}(t,n)\right]$$
$$+ \frac{i}{\varepsilon}\sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\rho_{\mathrm{od}}(t,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2},k,m\right)\rho_{\mathrm{od}}(t,n,k)\right], \tag{7}$$

and for the populations

$$\partial_t \rho_{\mathrm{d}}(t,n) = \frac{i}{\varepsilon}\sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\rho_{\mathrm{od}}(t,k,n) - \mathcal{V}\left(\frac{t}{\varepsilon^2},k,n\right)\rho_{\mathrm{od}}(t,n,k)\right]. \tag{8}$$

Classical arguments (see *e.g.* [Cas99]) allow to state the existence and uniqueness of solutions to System (7)-(8) for initial data in l^1. We indeed have assumed that $\mathcal{V}$ belongs to $L^\infty(\mathbb{R}^+, l^1 l^\infty \cap l^\infty l^1)$. Therefore the linear operator which associates $\widetilde{\rho}$ to $\rho \in L^\infty(\mathbb{R}^+, l^1)$ defined by

$$\widetilde{\rho}(t,n,m) := \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\rho(t,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2},k,m\right)\rho(t,n,k)\right],$$

is continuous on $L^\infty(\mathbb{R}^+, l^1)$. This fact together with Eq. (3) implies in a straightforward way that these solutions have the following regularity: $\rho \in C^0(\mathbb{R}^+, l^1)$ and $\partial_t \rho \in L^\infty_{\mathrm{loc}}(\mathbb{R}^+, l^1)$.

Besides these solutions to Eq. (3) have additional properties and we state here those which will prove to be useful in the sequel. First, for all $t \in \mathbb{R}$, $\rho(t)$ is Hermitian,

$$\rho_{\mathrm{od}}(t,n,m) = \rho_{\mathrm{od}}(t,m,n)^*.$$

Next, for all $t \in \mathbb{R}$, trace is conserved:

$$\sum_n \rho_{\mathrm{d}}(t,n) = \sum_n \rho_{\mathrm{d}}(0,n) < \infty\ .$$

Last, for all $t \geq 0$, positiveness is conserved:

$$\rho_{\mathrm{d}}(t,n) \geq 0\ .$$

(see [Lin76], [BBR01], [Cas01]—this is a consequence of the fact that equation (1) has the so-called Lindblad property). In particular, Eq. (8) which governs the diagonal part can be cast as

$$\partial_t \rho_{\mathrm{d}}(t,n) = -\frac{2}{\varepsilon}\mathrm{Im}\left[\sum_k \mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\rho_{\mathrm{od}}(t,k,n)\right]. \tag{9}$$

It must be stressed that the Lindblad property, together with the conservation of trace, give a uniform bound on ρ_{d} in l^1. This is crucial in the sequel. An extensive use of this bound, in a different context where oscillations are much more difficult to handle, may be found in [Cas01].

2.2 Towards an equation for populations

In this section, we transform the coupled system (7)-(8) into a *closed* equation governing the populations $\rho_{\mathrm{d}}(t,n)$ only. This is a key preliminary task that allows for the proof of our main results below. More precisely, we show the following proposition.

Proposition 1. *Let us define the time dependent transition rate*

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

Let $\rho_{\mathrm{d}}^{(1)}$ be solution to

$$\partial_t \rho_{\mathrm{d}}^{(1)}(t,n) = \sum_k \Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right)\left[\rho_{\mathrm{d}}^{(1)}(t,k) - \rho_{\mathrm{d}}^{(1)}(t,n)\right], \tag{10}$$

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

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

Remark 2. Eq. (10) is a linear Boltzmann type equation with a time dependent transition rate. In a way, it yields the behavior of the populations at the dominant order in ε. We could likewise state here a proposition of the same kind giving an approximation of ρ_{d} at *each order* (in ε), thus providing a hierarchy of Boltzmann type equations for the successive approximations to ρ_{d}.

Proof. The proof is in three steps. First, the values of the coherence $\rho_{\mathrm{od}}(t,n,k)$ are computed at the first order in ε in terms of the populations $\rho_{\mathrm{d}}(t,n)$ *only* (see (12) and (13) below). Next, this result is plugged into Eq. (9) governing populations: a closed equation for populations is thus obtained. This equation is a linear Boltzmann equation with a time-delay term (see Eq. (15)). Then, there remains to show that this delay is small, so that populations tend to be solution to the delay-free equation (10). The present calculations are inspired by [Cas99], [Cas02], [Cas01].

First step: computation of coherence. We first write an integral equation for the coherence (off-diagonal terms). Using Eq. (7) and since the initial data is $\rho_{\mathrm{od}}(t=0,n,m)\equiv 0$, we have

$$\begin{aligned}
&\rho_{\mathrm{od}}(t,n,m) = \\
&i\varepsilon \int_0^{t/\varepsilon^2} ds \exp\left(\Omega(n,m)s\right) \mathcal{V}\left(\frac{t}{\varepsilon^2}-s,n,m\right)\left[\rho_{\mathrm{d}}(t-\varepsilon^2 s,m) - \rho_{\mathrm{d}}(t-\varepsilon^2 s,n)\right] \\
&\ + i\varepsilon \int_0^{t/\varepsilon^2} ds \exp\left(\Omega(n,m)s\right) \\
&\times \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}-s,n,k\right)\rho_{\mathrm{od}}(t-\varepsilon^2 s,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}-s,k,m\right)\rho_{\mathrm{od}}(t-\varepsilon^2 s,n,k)\right].
\end{aligned} \tag{11}$$

We can obviously solve iteratively the integral equation (11) in terms of the unknown ρ_{od}, and obtain ρ_{od} in terms of ρ_{d} as a *complete* series expansion in powers of ε. However, in the sequel we will merely use first order expansions in ε. Therefore we right away only compute the first term in the expansion of $\rho_{\rm od}$. To achieve this, we define the first order approximation of $\rho_{\rm od}$, as

$$
\begin{aligned}
&\rho_{\rm od}^{(0)}(t,n,m) := \\
&\int_0^{t/\varepsilon^2} ds\, \exp\left(\Omega(n,m)s\right) \mathcal{V}\left(\frac{t}{\varepsilon^2}-s,n,m\right)\left[\rho_{\rm d}(t-\varepsilon^2 s,m)-\rho_{\rm d}(t-\varepsilon^2 s,n)\right]. \qquad (12)
\end{aligned}
$$

We claim that for all given time $T \geq 0$, we have the estimate

$$
\left\|\rho_{\rm od} - i\varepsilon\rho_{\rm od}^{(0)}\right\|_{L^\infty([0,T],l^1)} \leq C\varepsilon^2, \qquad (13)
$$

for some constant C independent of ε. (In fact, the same error estimate is also valid for the extremal value $T = +\infty$. We will however not use this fact in the sequel).

To prove (13), we introduce for convenience the operator A_ε which associates to $u \in L^\infty\left([0,T],l^1\right)$ the quantity

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

We have the straightforward estimate

$$
\begin{aligned}
\|A_\varepsilon u\|_{L^\infty([0,T],l^1)} &\leq \frac{2}{\gamma}\|\mathcal{V}\|_{L^\infty(\mathbb{R},l^1 l^\infty \cap l^\infty l^1)}\|u\|_{L^\infty([0,T],l^1)} \\
&\leq C\|u\|_{L^\infty([0,T],l^1)}.
\end{aligned}
$$

Besides, thanks to the integral equation (11) governing $\rho_{\rm od}$, the following equality holds.

$$
\rho_{\rm od}(t,n,m) = i\varepsilon\rho_{\rm od}^{(0)}(t,n,m) + i\varepsilon\left(A_\varepsilon\rho_{\rm od}\right)(t,n,m).
$$

For this reason we easily estimate the difference

$$
\begin{aligned}
\|\rho_{\rm od} - i\varepsilon\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)} &\leq C\varepsilon\|A_\varepsilon\rho_{\rm od}\|_{L^\infty(\mathbb{R},l^1)} \\
&= C\varepsilon\|A_\varepsilon(\rho_{\rm od} - i\varepsilon\rho_{\rm od}^{(0)} + i\varepsilon\rho_{\rm od}^{(0)})\|_{L^\infty(\mathbb{R},l^1)} \\
&\leq C\varepsilon\|\rho_{\rm od} - i\varepsilon\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)} + C\varepsilon^2\|\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)},
\end{aligned}
$$

and thus we obtain for a small ε

$$
\|\rho_{\rm od} - i\varepsilon\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)} \leq C\varepsilon^2\|\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)},
$$

where the constant $C > 0$ is independent of ε. There remains to use the obvious estimate

$$
\|\rho_{\rm od}^{(0)}\|_{L^\infty(\mathbb{R},l^1)} \leq \frac{2}{\gamma}\|\mathcal{V}\|_{L^\infty(\mathbb{R};l^1 l^\infty \cap l^\infty l^1)}\|\rho_{\rm d}\|_{L^\infty([0,T],l^1)},
$$

together with the classical identity

$$
\|\rho_{\rm d}\|_{L^\infty([0,T],l^1)} = \|\rho_{\rm d}(t=0)\|_{l^1}, \qquad (14)
$$

and the estimate (13) is proved. It must be stressed that the key estimate (14) is based on the Lindblad property [Lin76] (which implies the conservation of positiveness for $\rho_{\rm d}$), and on the conservation of the l^1-norm of $\rho_{\rm d}$.

Second step: towards a time delayed differential equation for the populations. Plugging the first order value $\rho_{\mathrm{od}}^{(0)}$ of ρ_{od} into Eq. (9) governing ρ_{d} gives

$$\begin{aligned}\partial_t \rho_{\mathrm{d}}(t,n) &= \sum_k \int_0^{t/\varepsilon^2} ds \left[\rho_{\mathrm{d}}(t-\varepsilon^2 s,k) - \rho_{\mathrm{d}}(t-\varepsilon^2 s,n)\right] \\ &\times 2\,\mathrm{Re}\Bigg(\exp\left(\Omega(k,n)s\right)\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\mathcal{V}\left(\frac{t}{\varepsilon^2}-s,k,n\right)\Bigg) + \varepsilon r_\varepsilon(t,n), \qquad (15)\end{aligned}$$

where, for all $T > 0$, there exists a constant $C > 0$ independent of ε such that the remainder $r_\varepsilon(t,n)$ satisfies

$$\|r_\varepsilon\|_{L^\infty([0,T],l^1)} \le C. \qquad (16)$$

Hence we introduce the natural first order expansion $\rho_{\mathrm{d}}^{(0)}$ of ρ_{d}, defined as the solution to

$$\begin{aligned}\partial_t \rho_{\mathrm{d}}^{(0)}(t,n) &= \sum_k \int_0^{t/\varepsilon^2} ds \left[\rho_{\mathrm{d}}^{(0)}(t-\varepsilon^2 s,k) - \rho_{\mathrm{d}}^{(0)}(t-\varepsilon^2 s,n)\right] \\ &\times 2\mathrm{Re}\Bigg(\exp\left(\Omega(k,n)s\right)\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\mathcal{V}\left(\frac{t}{\varepsilon^2}-s,k,n\right)\Bigg), \qquad (17)\end{aligned}$$

with initial data $\rho_{\mathrm{d}}^{(0)}(0,n) = \rho_{\mathrm{d}}(0,n)$. Clearly, Eqs (15) and (17) governing ρ_{d} and $\rho_{\mathrm{d}}^{(0)}$, together with the estimate (16) for r_ε, imply that, for all $T > 0$, there exists a constant C, independent of ε, such that

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(0)}\|_{L^\infty([0,T],l^1)} \le C\varepsilon. \qquad (18)$$

Thus, there remains to estimate the difference between $\rho_{\mathrm{d}}^{(1)}$, solution to the delay-free equation (10), and $\rho_{\mathrm{d}}^{(0)}$, solution to the integro-differential equation (17).

Third step: convergence to a delay-free equation. Synthetically we cast (10) as

$$\partial_t \rho_{\mathrm{d}}^{(1)}(t,n) = \left(B_\varepsilon \rho_{\mathrm{d}}^{(1)}\right)(t,n),$$

which defines a linear operator B_ε—as operating on sequences in l^1, for example. Let $T \ge 0$ be given. The delayed terms $\rho_{\mathrm{d}}^{(0)}(t-\varepsilon^2 s)$ in Eq. (17) read

$$\rho_{\mathrm{d}}^{(0)}(t-\varepsilon^2 s,n) = \rho_{\mathrm{d}}^{(0)}(t,n) + O\left(\varepsilon^2 s \|\partial_t \rho_{\mathrm{d}}^{(0)}\|_{L^\infty([0,T],l^1)}\right).$$

Thus, Eq. (17) yields $\partial_t \rho_{\mathrm{d}}^{(0)}(t,n) = \left(B_\varepsilon \rho_{\mathrm{d}}^{(0)}\right)(t,n) + \varepsilon^2 r_\varepsilon(t,n)$, where the remainder r_ε (we use the same notation r_ε as above in order not to overweight notations) can be estimated by

$$\begin{aligned}\|r_\varepsilon\|_{L^\infty([0,T],l^1)} &\le C\gamma^{-2}\|\partial_t \rho_{\mathrm{d}}^{(0)}\|_{L^\infty([0,T],l^1)} && \text{using a Taylor expansion} \\ &\le C\gamma^{-3}\|\rho_{\mathrm{d}}^{(0)}\|_{L^\infty([0,T],l^1)} && \text{thanks to Eq. (17)} \\ &\le C\|\rho_{\mathrm{d}}\|_{L^\infty([0,T],l^1)} && \text{thanks to Eq. (18)} \\ &\le C && \text{thanks to Eq. (14),}\end{aligned}$$

for some constant C independent of ε, only taking $\varepsilon \le 1/2$. Proposition 1 follows from these estimates. □

We end this section by stating a simplified version of Proposition 1. It consists in getting rid of the t and ε dependence in the time integral limit. This transformation proves useful in the sequel.

Proposition 2. *Let us define the time dependent transition rate*

$$\Psi\left(\frac{t}{\varepsilon^2}, k, n\right) = 2|V(n,k)|^2 \operatorname{Re}\int_0^{+\infty} ds \exp\left(\Omega(k,n)s\right)\phi\left(\frac{t}{\varepsilon^2}\right)\phi\left(\frac{t}{\varepsilon^2} - s\right). \tag{19}$$

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

$$\partial_t \rho_{\mathrm{d}}^{(2)}(t,n) = \sum_k \Psi\left(\frac{t}{\varepsilon^2}, k, n\right)\left[\rho_{\mathrm{d}}^{(2)}(t,k) - \rho_{\mathrm{d}}^{(2)}(t,n)\right], \tag{20}$$

with initial data $\rho_{\mathrm{d}}^{(2)}(0,n) = \rho_{\mathrm{d}}(0,n)$*. Then, for all* $T > 0$*, there exists* $C > 0$*, independent of* ε*, such that* $\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(2)}\|_{L^\infty([0,T],l^1)} \leq C\varepsilon$.

Proof. This result is straightforward since we have

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

(To obtain this result, compare the solutions to $\dot{y} = (1 - e^{-t/\varepsilon^2})y$ and $\dot{z} = z$, with same initial data: the size of the difference $z - y$ is ε^2, locally in time). $\square$

2.3 Limiting process and derivation of the Boltzmann equation: the case of a "r-chromatic" wave

Considering a classical expansion in power of ε reading $\rho_{\mathrm{d}} = \rho_{\mathrm{d}}^0 + \varepsilon\rho_{\mathrm{d}}^1 + \cdots$, we have up to now written an approximate equation for ρ_{d}^0, which is the lower order term in ε of ρ_{d}. This led us to Eq. (10). The above analysis may be easily extended to an expansion with all the orders in ε.

The limiting process in Eq. (10) does no longer come from a simple series expansion, but rather from averaging techniques, and, in particular, strongly depends on the precise time dependence of $\mathcal{V}$ (through $\mathcal{V}(t,n,m) = \phi(t)\, V(n,m)$).

To that aim, we specify in this section the time dependence of $\mathcal{V}$. We study the "r-chromatic" case, that is we assume the following.

Hypothesis 1. *We take* $\mathcal{V}(t,n,m) = \phi(t)\ V(n,m)$*, and specify the following time-dependence of* ϕ*,*

$$\phi(t) = \Phi(\omega_1 t, \ldots, \omega_r t),$$

where Φ *is a real-valued function,* 1*-periodic in its arguments, and we assume it is analytic on a strip* $|\operatorname{Im} z_1| \leq \sigma$*, …,* $|\operatorname{Im} z_r| \leq \sigma$*. Moreover, there is a finite number* r *of frequencies* ω_i *(*$i = 1, \cdots, r$*) which satisfy the Diophantine condition*

$$\exists C > 0, \quad \exists \kappa > 0, \quad \forall \alpha = (\alpha_1, \ldots, \alpha_r) \in \mathbb{Z}^r, \quad |\alpha\cdot\omega| \geq \frac{C}{|\alpha|^\kappa},$$

whenever $\alpha \cdot \omega \neq 0$.

We denoted $\alpha\cdot\omega = \alpha_1\omega_1 + \cdots \alpha_r\omega_r$. These assumptions fundamentally mean that we assume ϕ to be quasi-periodic and analytic.

Remark 3. Given once and for all a fixed $\delta > 0$, we can classically claim (see [Arn89]) that, for almost all value of the frequency vector $\omega = (\omega_1, \dots, \omega_r)$, there exists a constant $C(\omega) > 0$, depending on ω (and on δ), such that

$$\forall \alpha \in \mathbb{Z}^r \setminus \{0\}, \quad |\alpha \cdot \omega| \geq \frac{C(\omega)}{|\alpha|^{r-1+\delta}}.$$

Therefore, at least if $\kappa = r - 1 + \delta$ with $\delta > 0$, the above Diophantine condition holds true for almost all frequency vector ω. Hence the Diophantine condition is not much restrictive.

Also, on this basis, one could release the analytic assumption for Φ as follows: because Φ is an analytic function, the Fourier coefficients Φ_α ($\alpha \in \mathbb{Z}^r$) of Φ are exponentially decreasing ($|\Phi_\alpha| \leq C_1 \exp(-C_2|\alpha|)$ for some coefficients C_1, $C_2 > 0$ independent of α). In the sequel, it would be sufficient to assume that the Fourier coefficients of Φ are polynomially decreasing, that is

$$|\Phi_\alpha| \leq \frac{C_1}{|\alpha|^{N(r,\kappa)}},$$

for some coefficient C_1, independent of α and some exponent $N(r, \kappa) > 0$ which only depends on κ and the dimension r of the frequency vector ω. We do not continue this technical point.

Remark 4. The results we present here extend in other situations, i.e. for other types of functions ϕ, but with less accurate error estimates. We detail this point further.

First of all, let us recall a few results which are valid for a function ϕ as above. First, according to Hypothesis 1, the function ϕ has a convergent series expansion

$$\phi(t) = \sum_{\alpha \in \mathbb{Z}^r} \phi_\alpha \exp(2i\pi\alpha \cdot \omega t). \tag{21}$$

Moreover coefficients ϕ_α verify, for analyticity reasons,

$$|\phi_\alpha| \leq C_1 \exp(-C_2|\alpha|), \tag{22}$$

for some constants $C_1 > 0$ and $C_2 > 0$, independent of the multi-index α. Thus we define the time average of ϕ, denoted by $\langle \phi \rangle$, as

$$\langle \phi \rangle := \lim_{T \to \infty} \frac{1}{T} \int_0^T dt\ \phi(t). \qquad (= \phi_{(0,\dots,0)})$$

Now we state the main theorem of this section.

Theorem 1. *(i) Under the r-chromatic hypothesis 1, we introduce the asymptotic transition rate*

$$\langle \Psi \rangle(k, n) := \lim_{T \to \infty} \frac{1}{T} \int_0^T dt\ \Psi(t, k, n),$$

where Ψ is defined by (19). Let also $\rho_{\mathrm{d}}^{(3)}$ be solution to

$$\partial_t \rho_{\mathrm{d}}^{(3)}(t, n) = \sum_k \langle \Psi \rangle(k, n) \left[\rho_{\mathrm{d}}^{(3)}(t, k) - \rho_{\mathrm{d}}^{(3)}(t, n) \right], \tag{23}$$

with initial data $\rho_{\mathrm{d}}^{(3)}(0,n) = \rho_{\mathrm{d}}(0,n)$*. Then, for all* $T > 0$*, there exists a constant* $C > 0$ *independent of* ε*, such that*

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(3)}\|_{L^\infty([0,T],l^1)} \leq C\varepsilon.$$

(ii) In the specific monochromatic case when $\phi(t) = \cos(2\pi\omega t)$ *for some frequency* ω*, we may compute explicitly*

$$\langle\Psi\rangle(k,n) = \frac{1}{2}\sum_{\beta=\pm1} \frac{\gamma(k,n)\,|V(n,k)|^2}{\gamma(k,n)^2 + (2\pi\beta\,\omega + \omega(k,n))^2}. \tag{24}$$

(iii) In the r-chromatic case, formula (24) may be extended to

$$\langle\Psi\rangle(k,n) = 2\sum_{\beta\in\mathbb{Z}^r} \frac{\gamma(k,n)\;|V(n,k)|^2\;|\phi_\beta|^2}{\gamma(k,n)^2 + [2\pi\beta\cdot\omega + \omega(k,n)]^2}. \tag{25}$$

Remark 5. According to Proposition 2, Eq. (20) governing $\rho_{\mathrm{d}}^{(2)}$ is cast as

$$\partial_t\rho_{\mathrm{d}}^{(2)}(t,n) = \sum_k \Psi\left(\frac{t}{\varepsilon^2},k,n\right)\left[\rho_{\mathrm{d}}^{(2)}(t,k) - \rho_{\mathrm{d}}^{(2)}(t,n)\right].$$

On the other hand, Theorem 1 states that $\rho_{\mathrm{d}}^{(3)}$ is solution to

$$\partial_t\rho_{\mathrm{d}}^{(3)}(t,n) = \sum_k \langle\Psi\rangle(k,n)\left[\rho_{\mathrm{d}}^{(3)}(t,k) - \rho_{\mathrm{d}}^{(3)}(t,n)\right].$$

This type of convergence result is classical when averaging ordinary differential equations and we transpose here techniques described in [SV85].

Proof. The proof of Theorem 1 is split in several steps.

First step: preliminary remarks and problem reduction. The proof of (24) and (25) follows from a direct calculation (see (30) below).

Therefore we only prove item (i). We choose some time $T \geq 0$. In order to show that $\rho_{\mathrm{d}}^{(2)}$ converges to $\rho_{\mathrm{d}}^{(3)}$, we now introduce, as in [SV85], an auxiliary variable $\rho_{\mathrm{d}}^{(4)}$, solution to

$$\partial_t\rho_{\mathrm{d}}^{(4)}(t,n) = \sum_k \left[\rho_{\mathrm{d}}^{(4)}(t,k) - \rho_{\mathrm{d}}^{(4)}(t,n)\right] \times \varepsilon\int_0^{1/\varepsilon} ds\,\Psi\left(\frac{t}{\varepsilon^2} + s,k,n\right), \tag{26}$$

with initial data $\rho_{\mathrm{d}}^{(4)}(0,n) = \rho_{\mathrm{d}}(0,n)$. In what follows, we successively prove

$$\|\rho_{\mathrm{d}}^{(2)} - \rho_{\mathrm{d}}^{(4)}\|_{L^\infty([0,T],l^1)} \leq C\varepsilon, \tag{27}$$

and

$$\|\rho_{\mathrm{d}}^{(4)} - \rho_{\mathrm{d}}^{(3)}\|_{L^\infty([0,T],l^1)} \leq C\varepsilon. \tag{28}$$

These two estimates end the proof of Theorem 1.

Second step: proof of (28). The proof of (28) is based on the estimate

$$\left\| \langle \Psi \rangle - \varepsilon \int_0^{1/\varepsilon} ds\ \Psi\left(\frac{t}{\varepsilon^2} + s\right) \right\|_{L^\infty([0,T], l^1 l^\infty \cap l^\infty l^1)} \leq C\varepsilon, \tag{29}$$

and Gronwall lemma.

The function $\Psi(t,k,n)$ satisfies the analytic and quasi-periodic hypothesis 1, and therefore admits a convergent series expansion like (21)-(22). More precisely, from (21) we deduce easily the formula

$$\Psi(t,k,n) = 2|V(n,k)|^2 \ \mathrm{Re} \sum_{\alpha,\beta\in\mathbb{Z}^r} \frac{\phi_\alpha \phi_\beta \exp\left(2i\pi[\alpha+\beta]\cdot\omega t\right)}{-\Omega(k,n) + 2i\pi\beta\cdot\omega}. \tag{30}$$

This developed form for Ψ allows to estimate the left hand-side in (29) by

$$\begin{aligned} 2 \sum_{\beta\in\mathbb{Z}^r} |\phi_\beta| & \left\| \frac{|V(n,k)|^2}{-\Omega(k,n) + 2i\pi\beta\cdot\omega} \right\|_{l^1 l^\infty \cap l^\infty l^1} \\ & \times \left| \phi_{-\beta} - \varepsilon \sum_{\alpha\in\mathbb{Z}^r} \int_0^{1/\varepsilon} ds\ \phi_\alpha \exp\left(2i\pi[\alpha+\beta]\cdot\omega\ [\frac{t}{\varepsilon^2} - s]\right) \right|. \end{aligned} \tag{31}$$

Thanks to the exponentially decreasing estimate given by (22), we do have the right to integrate each term separately. According to the Diophantine condition on ω, we then notice that

$$\begin{aligned} & \left| \phi_{-\beta} - \varepsilon \sum_{\alpha\in\mathbb{Z}^r} \int_0^{1/\varepsilon} ds\ \phi_\alpha \exp\left(2i\pi[\alpha+\beta]\cdot\omega(\frac{t}{\varepsilon^2} - s)\right) \right| \\ & \quad = \varepsilon \left| \sum_{\alpha+\beta\neq 0} \int_0^{1/\varepsilon} ds\ \phi_\alpha \exp\left(2i\pi[\alpha+\beta]\cdot\omega(\frac{t}{\varepsilon^2} - s)\right) \right| \\ & \quad \leq C\varepsilon \sum_{\alpha\neq-\beta} \frac{|\phi_\alpha|}{|(\alpha+\beta)\cdot\omega|} \leq C\varepsilon \sum_{\alpha\neq-\beta} |\phi_\alpha|\ |\alpha+\beta|^\kappa. \end{aligned}$$

On the other hand, we use the estimate $|-\Omega(k,n)+2i\pi\beta\cdot\omega| \geq \gamma$. This allows to estimate (31) by

$$C\varepsilon \sum_{\alpha,\beta\in\mathbb{Z}^r} |\phi_\beta|\ |\phi_\alpha|\ |\alpha+\beta|^\kappa \leq C\varepsilon,$$

and (29) is established.

Third step: proof of (27). The proof of (27) is a little more delicate. The difference $\Delta = \rho_{\mathrm{d}}^{(2)} - \rho_{\mathrm{d}}^{(4)}$ is solution to

$$\begin{aligned} \partial_t \Delta(t,n) = & \sum_k \Psi(\frac{t}{\varepsilon^2},k,n)\,[\Delta(t,k) - \Delta(t,n)] \\ & + \sum_k \left(\Psi(\frac{t}{\varepsilon^2},k,n) - \varepsilon \int_0^{1/\varepsilon} ds\, \Psi\left(\frac{t}{\varepsilon^2} + s, k, n\right) \right) \left[\rho_{\mathrm{d}}^{(4)}(t,k) - \rho_{\mathrm{d}}^{(4)}(t,n) \right], \end{aligned}$$

with vanishing initial data. Therefore we can write

$$\Delta(t,n) = \int_0^t du \sum_k \Psi(\frac{u}{\varepsilon^2},k,n)\,[\Delta(u,k)-\Delta(u,n)] \tag{32}$$
$$+\int_0^t du \sum_k \left(\Psi(\frac{u}{\varepsilon^2},k,n) - \varepsilon\int_0^{1/\varepsilon} ds\;\Psi\left(\frac{u}{\varepsilon^2}+s,k,n\right)\right)\left[\rho_{\rm d}^{(4)}(u,k)-\rho_{\rm d}^{(4)}(u,n)\right].$$

The first term of the right hand-side in (32) is estimated in l^1 norm by $\int_0^t \|\Delta(u)\|_{l^1}$. Using Gronwall lemma, the proof of (27) therefore reduces to prove the following estimate for $|t|\le T$:

$$\Big\| \int_0^t du \sum_k \left(\Psi(\frac{u}{\varepsilon^2},k,n) - \varepsilon\int_0^{1/\varepsilon} ds\,\Psi\left(\frac{u}{\varepsilon^2}+s,k,n\right)\right)$$
$$\times\left[\rho_{\rm d}^{(4)}(u,k)-\rho_{\rm d}^{(4)}(u,n)\right]\Big\|_{l^1} \le C\varepsilon. \tag{33}$$

We therefore conclude by proving (33). To this aim, we write for the contribution due to $\rho_{\rm d}^{(4)}(u,k)$ (we handle the contribution due to $\rho_{\rm d}^{(4)}(u,n)$ in a similar way),

$$\begin{aligned}
&\int_0^t du \sum_k \varepsilon\int_0^{1/\varepsilon} ds\;\Psi\left(\frac{u}{\varepsilon^2}+s,k,n\right)\rho_{\rm d}^{(4)}(u,k)\\
&\quad=\sum_k\int_0^1 ds\int_0^t du\;\Psi\left(\frac{u+\varepsilon s}{\varepsilon^2},k,n\right)\rho_{\rm d}^{(4)}(u,k)\\
&\quad=\sum_k\int_0^1 ds\int_0^t du\;\Psi\left(\frac{u+\varepsilon s}{\varepsilon^2},k,n\right)\rho_{\rm d}^{(4)}(u+\varepsilon s,k)+O_{L^\infty([0,T],l^1)}(\varepsilon),
\end{aligned}\tag{34}$$

where the $O_{L^\infty([0,T],l^1)}(\varepsilon)$ term means that the difference is estimated by $C\varepsilon$ in the norm $L^\infty([0,T],l^1)$, for some constant C independent of ε. Of course, this last estimate is obtained thanks to the definition of Ψ, as well as Eq. (26) governing $\rho_{\rm d}^{(4)}$, which implies that $\|\partial_t\rho_{\rm d}^{(4)}(t,n)\|_{L^\infty([0,T],l^1)}\le C$.
We now estimate the first term in (34) which can be rewritten as

$$\sum_k\int_0^1 ds\int_{\varepsilon s}^{t+\varepsilon s} du\;\Psi\left(\frac{u}{\varepsilon^2},k,n\right)\rho_{\rm d}^{(4)}(u,k).$$

Noticing that the quantity

$$\partial_s\left(\int_{\varepsilon s}^{t+\varepsilon s} du\;\Psi\left(\frac{u}{\varepsilon^2},k,n\right)\rho_{\rm d}^{(4)}(u,k)\right)$$

is of order ε in the $L^\infty([0,T],l^1)$ space, we estimate this term by

$$\sum_k\int_0^t du\;\Psi\left(\frac{u}{\varepsilon^2},k,n\right)\rho_{\rm d}^{(4)}(u,k)+O_{L^\infty([0,T],l^1)}(\varepsilon).$$

All these computations lead to (33), which ends the proof of Theorem 1. $\square$

Remark 6. A remark has to be made on the above proof of Theorem 1. We followed [SV85] since this proof may be extended to other types of function ϕ than the quasi-periodic functions. We refer to Section 2.4.1 for this point. Nevertheless, in the quasi-periodic case, a simpler and more straightforward proof is possible, based on integration by parts. (see Section 3.3).

2.4 Generalizing the former analysis to more general wave profiles

2.4.1 Case of a "KBM" wave

Following [LM88], [SV85], we may replace the r-chromatic hypothesis 1 by the following "KBM" (Krylov-Bogolioubov-Mitropolski) type hypothesis.

Hypothesis 2. *We take $\mathcal{V}(t,n,m) = \phi(t)V(n,m)$, and require that ϕ is a bounded C^1 function, such that, for all $t \geq 0$,*

$$\langle \Psi \rangle (n,m) := \lim_{T\to\infty} \frac{1}{T} \int_0^T du \; \Psi(t+u,n,m) \text{ exists in } l^1 l^\infty \cap l^\infty l^1.$$

Here Ψ is defined by (19) as above, that is

$$\Psi(t,n,m) = 2|V(n,m)|^2 \operatorname{Re} \int_0^{+\infty} ds \; \exp(\Omega(n,m)s)\phi(t)\phi(t-s).$$

(This limit, whenever it exists, does not depend on t.)

Under Hypothesis 2, we introduce the convergence rate

$$\delta(\varepsilon) := \sup_{0\leq T\leq 1/\varepsilon^2} \left\| \varepsilon^2 \int_0^T du \; \left[\Psi(u) - \langle\Psi\rangle\right] \right\|_{l^1 l^\infty \cap l^\infty l^1}.$$

Hypothesis 2 implies that $\delta(\varepsilon)$ tends to 0 as ε tends to 0.

We now state the main theorem of this section.

Theorem 2. *(i) Under the "KBM" hypothesis 2, we introduce $\rho_{\mathrm{d}}^{(5)}$, which is solution to*

$$\partial_t \rho_{\mathrm{d}}^{(5)}(t,n) = \sum_k \langle\Psi\rangle(k,n) \left[\rho_{\mathrm{d}}^{(5)}(t,k) - \rho_{\mathrm{d}}^{(5)}(t,n)\right],$$

with initial data $\rho_{\mathrm{d}}^{(5)}(0,n) = \rho_{\mathrm{d}}(0,n)$. Then, for all $T > 0$, there exists a constant $C > 0$ independent of ε, such that

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(5)}\|_{L^\infty([0,T],l^1)} \leq C\sqrt{\delta(\varepsilon)} \underset{\varepsilon\to 0}{\longrightarrow} 0.$$

(ii) The result of item (i) applies in particular if ϕ is an almost periodic function, that is if there exists a sequence of trigonometric polynomials $(P_l(t))_{l\in\mathbb{N}}$ such that

$$\sup_{t\in\mathbb{R}} |P_l(t) - \phi(t)| \underset{l\to\infty}{\longrightarrow} 0.$$

(iii) The result of item (i) applies also in the case when ϕ has a continuous spectrum, that is when

$$\phi(t) = \int_{\mathbb{R}} d\tau \; A(\tau)\exp(i\tau t),$$

for a frequency profile $A(\tau)$ which we will suppose – in order to simplify – to be smooth enough: $A \in \mathcal{S}(\mathbb{R})$ (it goes without saying that this assumption may be relaxed). In this case, we simply have

$$\langle \Psi \rangle \equiv 0.$$

Remark 7. Theorems 1 and 2 show that, in the limit $\varepsilon \to 0$, populations $\rho_{\mathrm{d}}(t,n)$ tend to satisfy a Boltzmann equation, with a transition rate that takes into account the resonance between the eigen-frequencies of the atom and the frequencies of ϕ. In the monochromatic or the r-chromatic case, it is obvious that resonances between the frequency vector ω of the wave ϕ and the values $\omega(n,k)$ are more strongly coupled *via* the transition rate (24) or (25). The above item (ii) shows, in a less quantitative way, that this phenomenon always occurs if the wave has a discrete spectrum which does not necessarily corresponds to a finite number r of independent frequencies (the spectrum of an almost periodic function is in general more "dense" that that of a quasi-periodic function). On the other hand, in the extreme case when the wave ϕ has a continuous spectrum, these resonances are rubbed out in a way, and the asymptotic equation governing populations is trivial.

Proof. Item (iii) is a straightforward consequence of item (i). To show item (ii), it is enough to show that Hypothesis 2 holds for an almost periodic function ϕ. To see this, we choose a sequence P_l of trigonometric polynomials which tends to ϕ. We define

$$\Psi_l(t,k,n) = 2|V(k,n)|^2 \operatorname{Re} \int_0^{+\infty} ds \ \exp(\Omega(k,n)s) P_l(t) P_l(t-s),$$

and the average $\langle \Psi_l \rangle(k,n) = \lim_{T\to\infty} \frac{1}{T} \int_0^T ds \ \Psi_l(s,k,n)$. (The limit is taken in $l^1 l^\infty \cap l^\infty l^1$, and exists for all l). Using the fact that $\operatorname{Re}\Omega(k,n) \le -\gamma < 0$ for all k, n, it is easy to see that the sequences Ψ_l and $\langle \Psi_l \rangle$ are Cauchy sequences in the appropriate spaces.

To show item (i), we use the same outline as for the proof of Theorem 1. We take a long time scale $T(\varepsilon)$ to be determined (eventually we will take $T(\varepsilon) = \sqrt{\delta(\varepsilon)}/\varepsilon^2$). We introduce $\rho_{\mathrm{d}}^{(6)}$, which is solution to

$$\partial_t \rho_{\mathrm{d}}^{(6)}(t,n) = \sum_k \left(\frac{1}{T(\varepsilon)} \int_0^{T(\varepsilon)} ds \ \Psi\left(\frac{t}{\varepsilon^2} + s, k, n\right)\right) \left[\rho_{\mathrm{d}}^{(6)}(t,k) - \rho_{\mathrm{d}}^{(6)}(t,n)\right],$$

with initial data $\rho_{\mathrm{d}}^{(6)}(0,n) = \rho_{\mathrm{d}}(0,n)$. Following the proof of Theorem 1, it is easy to find the error estimate:

$$\|\rho_{\mathrm{d}}^{(2)} - \rho_{\mathrm{d}}^{(6)}\|_{L^\infty([0,T],l^1)} \le C\varepsilon^2 T(\varepsilon). \tag{35}$$

Then, coming back to the definition of $\delta(\varepsilon)$, and using the equations governing $\rho_{\mathrm{d}}^{(6)}$ and $\rho_{\mathrm{d}}^{(5)}$ respectively, we easily establish the estimate

$$\|\rho_{\mathrm{d}}^{(5)} - \rho_{\mathrm{d}}^{(6)}\|_{L^\infty([0,T],l^1)} \le C \frac{\delta(\varepsilon)}{\varepsilon^2 T(\varepsilon)}. \tag{36}$$

The optimal choice $T(\varepsilon) = \sqrt{\delta(\varepsilon)}/\varepsilon^2$ in (35) and (36) yields Theorem 2. $\square$

2.4.2 Case of a r-chromatic wave with spectrum broadening

Theorems 1 and 2 claim that populations tend to be solution to a Boltzmann equation in the case of a wave ϕ with a discrete spectrum (case of a r-chromatic or an almost periodic function), and that this equation turns out to be trivial is the case when the wave ϕ has a continuous spectrum. Consequently it is natural to consider, as it is the case in this section, the intermediate case when the wave ϕ has a *slightly broadened* spectrum around a given frequency vector ω.

To this aim, we assume in this section that

Hypothesis 3.

$$\begin{aligned}\phi(t) &= \frac{1}{\varepsilon^{qr}}\sum_{\alpha\in\mathbb{Z}^r}\int_{\mathbb{R}^r} d\eta\; A\left(\alpha, \frac{\eta-\omega}{\varepsilon^q}\right)\exp(2i\pi\alpha\cdot\eta\; t)\\ &= \sum_{\alpha\in\mathbb{Z}^r}\int_{\mathbb{R}^r} d\eta\; A(\alpha,\eta)\exp(2i\pi\alpha\cdot[\omega+\varepsilon^q\eta]\; t),\end{aligned} \tag{37}$$

for some exponent $q>0$. We also assume that the amplitude A is sufficiently decreasing and smooth with respect to α and η and we assume also that

$$A(\alpha,\eta)\in l^1(\mathbb{Z}^r, L^1(\mathbb{R}^r)). \tag{38}$$

Note that we do not make explicit the dependence of ϕ with respect to ε in (37).

The wave ϕ being given by (37), we now establish the analogue of Theorem 1 shown in the r-chromatic case.

Theorem 3. *Let ϕ such that (37) and (38) hold. We introduce $\rho_{\mathrm{d}}^{(7)}(t,n)$, which is solution to*

$$\partial_t\rho_{\mathrm{d}}^{(7)}(t,n) = \sum_k \langle\Psi\rangle(k,n)\;\left[\rho_{\mathrm{d}}^{(7)}(t,k)-\rho_{\mathrm{d}}^{(7)}(t,n)\right],$$

with initial data $\rho_{\mathrm{d}}^{(7)}(0,n)=\rho_{\mathrm{d}}(0,n)$, where $\langle\Psi\rangle(k,n)$ is given by the formula

$$\langle\Psi\rangle(k,n) := 2|V(n,k)|^2\sum_{\beta\in\mathbb{Z}^r}\frac{\gamma(k,n)\left|\int_{\mathbb{R}^r} d\eta\; A(\beta,\eta)\right|^2}{\gamma(k,n)^2+[\omega(k,n)+2\pi\beta\cdot\omega]^2}. \tag{39}$$

Then, for all $T>0$, we have the error estimate

$$\|\rho_{\mathrm{d}}-\rho_{\mathrm{d}}^{(7)}\|_{L^\infty([0,T],l^1)} \underset{\varepsilon\to 0}{\longrightarrow} 0.$$

Remark 8. Theorem 3 shows, in particular, that spectrum broadening does not play an essential role and everything works as if the wave were straight off really r-chromatic (limiting case $q=+\infty$ in (37)).

Proof. To show Theorem 3, we proceed as in the proof of Theorems 1 and 2. We have to calculate the function Ψ as defined by (19) (once more, we do not make explicit the ε-dependence of Ψ). We easily have

$$\begin{aligned}\Psi(t,k,n) = 2|V(n,k)|^2\mathrm{Re}&\sum_{\alpha,\beta\in\mathbb{Z}^r}\int_{\mathbb{R}^{2r}} d\eta\, d\eta'\; A(\alpha,\eta)A(\beta,\eta')\\ &\times\frac{\exp\left(2i\pi\left[[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right]t\right)}{\gamma(k,n)+i\left[\omega(k,n)+2\pi\beta\cdot[\omega+\varepsilon^q\eta']\right]}.\end{aligned} \tag{40}$$

Then we *define* the asymptotic quantity

$$\langle\Psi\rangle(k,n) := 2|V(n,k)|^2 \mathrm{Re} \sum_{\beta\in\mathbb{Z}^r} \int_{\mathbb{R}^{2r}} d\eta\, d\eta'\, \frac{A(-\beta,\eta)A(\beta,\eta')}{\gamma(k,n)+i\left[\omega(k,n)+2\pi\beta\cdot\omega\right]}. \tag{41}$$

Of course, since ϕ is a real function, definition (41) is equivalent to (39).

Last, as in the proof of Theorem 2, we take a time scale $T(\varepsilon)=\varepsilon^{-\delta}$ for some (small) exponent $\delta>0$. We show the two estimates below (which are analogous to (29) and (33))

$$\left\|\langle\Psi\rangle-\varepsilon^{\delta}\int_0^{1/\varepsilon^{\delta}} ds\,\Psi\left(\frac{t}{\varepsilon^2}+s\right)\right\|_{L^\infty([0,T],l^1l^\infty\cap l^\infty l^1)} \underset{\varepsilon\to 0}{\longrightarrow} 0, \tag{42}$$

$$\left\|\int_0^t du\,\left(\Psi\left(\frac{u}{\varepsilon^2}\right)-\varepsilon^{\delta}\int_0^{1/\varepsilon^{\delta}} ds\,\Psi\left(\frac{u}{\varepsilon^2}+s\right)\right)\right\|_{L^\infty([0,T],l^1l^\infty\cap l^\infty l^1)} \le C\varepsilon^{2-\delta}. \tag{43}$$

The proof of (43) is easy and follows the same outline as the above proof of (33). We therefore merely show (42). To this aim, we begin with the definitions (40) and (41) of Ψ and its asymptotic average $\langle\Psi\rangle$. Then we estimate easily the left hand-side of (42) by

$$\begin{aligned} &C\sum_{\alpha+\beta\neq 0}\int_{\mathbb{R}^{2r}} d\eta\, d\eta'\, |A(\alpha,\eta)|\,|A(\beta,\eta')| \\ &\qquad\times\left|\frac{\exp\left(2i\pi\left[[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right]\varepsilon^{-\delta}\right)-1}{2i\pi\left[[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right]\varepsilon^{-\delta}}\right| \\ &+C\sum_{\beta}\int_{\mathbb{R}^{2r}} d\eta\, d\eta'\, |A(-\beta,\eta)|\,|A(\beta,\eta')|\left|1-\frac{\exp\left(2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]\right)-1}{2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]}\right|. \end{aligned}$$

To show that each term tends to 0, we apply the dominated convergence theorem using that function $(e^{ix}-1)/x$ is globally bounded on $\mathbb{R}$. For the second term, we notice that for all β and almost all η, η', we have

$$\frac{\exp\left(2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]\right)-1}{2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]} \underset{\varepsilon\to 0}{\longrightarrow} 1,$$

provided $0<\delta<q$, which we assume to hold true. For the first term, we write that for all α and β such that $\alpha+\beta\neq 0$, and for almost all η, η', we have

$$\begin{aligned} &\left|\frac{\exp\left(2i\pi\left[[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right]\varepsilon^{-\delta}\right)-1}{2i\pi\left[[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right]\varepsilon^{-\delta}}\right| \\ &\qquad\le \frac{C\varepsilon^{\delta}}{\left|[\alpha+\beta]\cdot\omega+\varepsilon^q[\alpha\cdot\eta+\beta\cdot\eta']\right|} \underset{\varepsilon\to 0}{\sim} \frac{C\varepsilon^{\delta}}{\left|[\alpha+\beta]\cdot\omega\right|}\le C\,\varepsilon^{\delta}, \end{aligned}$$

where we of course use the Diophantine property which is satisfied by ω. This ends the proof of (42).

To conclude, estimates (42) and (43), together with the techniques developed in the proofs of Theorems 1 and 2, yield Theorem 3. □

3 Case when relaxations tend to 0 with ε

3.1 The model

The whole analysis performed in Section 2 relies strongly on the existence of a uniform relaxation, since we assumed that

$$\gamma := \inf_{(n,m), n \neq m} \gamma(n,m) > 0.$$

In this section, we address the case when γ depends on ε, and tends to 0 with ε. We consider more specifically the case when $\gamma \sim \varepsilon^\mu$ for some exponent $\mu > 0$. Precisely, we reproduce the former analysis in the case when we perform the substitution

$$\gamma(n,m) \rightarrow \varepsilon^\mu \gamma(n,m)$$

in the original Bloch equations (3). The value of the exponent μ is specified below.

Summarizing, we now perform the asymptotic analysis as $\varepsilon \to 0$ in

$$\begin{aligned} \varepsilon^2 \partial_t \rho(t,n,m) = & -\left(i\omega(n,m) + \varepsilon^\mu \gamma(n,m)\right) \rho(t,n,m) \\ & + i\varepsilon \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k\right) \rho(t,k,m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, m\right) \rho(t,n,k) \right]. \end{aligned} \tag{44}$$

The initial data, as well as coefficients $\gamma(n,m)$, $\omega(n,m)$ and wave profile $\mathcal{V}(t,n,m)$ are chosen as in the case of uniform relaxations (see Section 2, and the assumptions listed in Section 2.1).

Unfortunately, the analysis presented in this Section 3 needs the following three restrictions.

First, we need the following strong decay assumption on the coefficients $V(n,m)$. Indeed, we assume in the whole Section 3 that

$$\sum_{n,m} (1+|n|)^{2+2\delta} \, (1+|m|)^{2+2\delta} |V(n,m)|^2 < \infty \,, \tag{45}$$

where $\delta > 0$ is as in Hypothesis 4 below, and may be arbitrarily small. This restriction stems from "small denominator" considerations (Diophantine estimates). The present decay should be compared with the milder assumption (6) made in the previous section. Though the mild decay (6) is physically relevant in most situations, the stronger assumption (45) means that we consider a situation where the relevant energy levels of the atom are "far from the continuous spectrum". Obviously, the present restriction includes the case of an atom with a finite number of energy levels, a case which is often considered in practice.

Second, we restrict in the sequel of this section to the case when

$$\mu < 1/2. \tag{46}$$

We do not know whether this exponent is optimal or not. A comment is necessary. In the case when $\gamma > 0$ (i.e. $\mu = 0$), the initial Bloch equation (3) is time-irreversible and the asymptotic equation (23) is also time-irreversible. On the other hand, in the opposite case when every coefficient $\gamma(n,m)$ is identically zero, the initial Bloch equation is time-reversible and the nature of the problem has changed. It is therefore no wonder that the following analysis displays a threshold value for the exponent μ.

Last, our analysis is restricted to the case of an r-chromatic wave and we need the

Hypothesis 4. *The function ϕ is assumed r-chromatic, in the sense that*

$$\mathcal{V}(t,n,m) = \phi(t)\ V(n,m), \qquad \text{with}\ \ \phi(t) = \Phi(\omega_1 t,\dots,\omega_r t),$$

and Φ is a real-valued function, 1-periodic in its arguments, analytic on a strip $|\mathrm{Im} z_1| \le \sigma$, ..., $|\mathrm{Im} z_r| \le \sigma$. Moreover, there is a finite number r of frequencies ω_i $(i = 1,\dots,r)$ which satisfy the following Diophantine condition: There exists a constant $C > 0$ and a number $\delta > 0$ such that

$$\begin{aligned} &\forall \alpha = (\alpha_1,\dots,\alpha_r) \in \mathbb{Z}^r, \quad \forall (k,n) \in \mathbb{N}^2\ , \\ &\qquad |\alpha\cdot\omega + \omega(k,n)| \ge \frac{C}{(1+|\alpha|)^{r-1+\delta}\ (1+|n|)^{1+\delta}\ (1+|k|)^{1+\delta}}\ , \end{aligned} \tag{47}$$

whenever $\alpha\cdot\omega + \omega(k,n) \ne 0$, and

$$\forall \alpha = (\alpha_1,\dots,\alpha_r) \in \mathbb{Z}^r, \qquad |\alpha\cdot\omega| \ge \frac{C}{(1+|\alpha|)^{r-1+\delta}}\ ,$$

whenever $\alpha\cdot\omega \ne 0$.

The extension to other types of waves is discussed later.

Remark 9. When $\alpha = 0$ in (47), the assumed lower bound corresponds to an *assumption* on the repartition of the energy levels $\omega(n,m)$: the levels are not allowed to accumulate too quickly (more than polynomially fast). Such a (mild) assumption is seen for those levels that accumulate towards the ionisation energy (the others are anyhow well separated).

On the other hand, once (47) is ensured for $\alpha = 0$, and a fixed $\delta > 0$ being chosen, it is easily proved (see [Arn89]) that, for almost all value of the frequency vector $\omega = (\omega_1,\dots,\omega_r)$, there exists a constant $C(\omega) > 0$, depending on ω (and on δ), such that

$$\forall \alpha \in \mathbb{Z}^r\ , \forall (k,n) \in \mathbb{N}^2\ , |\alpha\cdot\omega + \omega(k,n)| \ge \frac{C(\omega)}{(1+|\alpha|)^{r-1+\delta}\ (1+|n|)^{1+\delta}(1+|k|)^{1+\delta}}\ ,$$

whenever $\alpha\cdot\omega + \omega(k,n) \ne 0$, and the analogous estimate holds for $\alpha\cdot\omega$. This is an easy consequence of the fact that

$$\sum_{\alpha}\sum_{k,n}(1+|\alpha|)^{-(r-1+\delta)}\ (1+|n|)^{-(1+\delta)}(1+|k|)^{-(1+\delta)} < \infty\ .$$

Therefore, the above Diophantine condition holds true for almost all frequency vector ω, and δ may be taken arbitrarily small. Hence the Diophantine condition is not much restrictive. Also, it is needless to say that one could release the analytic assumption for Φ in the analysis performed below, and we do not continue this technical point.

Let us now return to the asymptotic analysis of (44). It follows the main steps of the case $\gamma > 0$ (i.e. $\mu = 0$).

3.2 Towards an equation for populations

In this section, we establish the following Proposition, which is parallel to Proposition 1.

Proposition 3. *Let us define the time dependent transition rate:*

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

where $\Omega(k,n) = -(i\omega(k,n)+\varepsilon^\mu\gamma(k,n))$. We assume that $\mu<1/2$. Then ρ_{d} satisfies

$$\partial_t\rho_{\mathrm{d}}(t,n) = \sum_k \Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right)\left[\rho_{\mathrm{d}}(t,k)-\rho_{\mathrm{d}}(t,n)\right] + O_{L^\infty_{\mathrm{loc}}(\mathbb{R},l^1)}\left(\varepsilon^{1-2\mu}\right), \tag{48}$$

where the symbol $O_{L^\infty_{\mathrm{loc}}(\mathbb{R},l^1)}\left(\varepsilon^{1-2\mu}\right)$ means that for all $T>0$, there exists C such that the following estimate holds

$$\left\|O_{L^\infty_{\mathrm{loc}}(\mathbb{R},l^1)}\left(\varepsilon^{1-2\mu}\right)\right\|_{L^\infty([0,T];l^1)} \le C\varepsilon^{1-2\mu}. \tag{49}$$

Remark 10. Proposition 3 extends immediately to the more general case when ϕ is a bounded function on $\mathbb{R}$.

Proof. We follow the main steps of the proof of Proposition 1.

Let $T>0$ be given. To begin with, we compute the coherence in terms of the populations, at the lowest order in ε. Therefore we write, as in Eq. (11),

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

which defines operators A_ε and $\widetilde{A}_\varepsilon$. Thus, we obtain for a given value of $T>0$:

$$\begin{aligned}
\|\rho_{\mathrm{od}}-i\varepsilon(A_\varepsilon\rho_{\mathrm{d}})\|_{L^\infty([0,T],l^1)} &= \varepsilon\|\widetilde{A}_\varepsilon\rho_{\mathrm{od}}\|_{L^\infty([0,T],l^1)}\\
&\le \varepsilon\left\|\widetilde{A}_\varepsilon\Big[\rho_{\mathrm{od}}-i\varepsilon(A_\varepsilon\rho_{\mathrm{d}})\Big]\right\|_{L^\infty([0,T],l^1)} + \varepsilon^2\|\widetilde{A}_\varepsilon A_\varepsilon\rho_{\mathrm{d}}\|_{L^\infty([0,T],l^1)}.
\end{aligned} \tag{50}$$

Besides, using the definition $\Omega(n,m) = -i\omega(n,m)-\varepsilon^\mu\gamma(n,m)$ in which $\gamma(n,m)\ge\gamma>0$, we of course have the estimates

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

Thus, we obtain in (50), for ε small enough,

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

Thereby, Eq. (8) for the populations reads

$$\begin{aligned}\partial_t \rho_{\rm d}(t,n) &= \frac{i}{\varepsilon}\sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\rho_{\rm od}(t,k,n) - \mathcal{V}\left(\frac{t}{\varepsilon^2},k,n\right)\rho_{\rm od}(t,n,k)\right]\\ &= -\sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)(A_\varepsilon\rho_{\rm d})(t,k,n) - \mathcal{V}\left(\frac{t}{\varepsilon^2},k,n\right)(A_\varepsilon\rho_{\rm d})(t,n,k)\right]\\ &\quad + O_{L^\infty([0,T];l^1)}(\varepsilon^{1-2\mu}). \qquad (51)\end{aligned}$$

Here, the symbol $O_{L^\infty([0,T];l^1)}(\varepsilon^{1-2\mu})$ means that the corresponding remaining term is estimated by $C\varepsilon^{1-2\mu}$ in the $L^\infty([0,T],l^1)$ norm, for a constant $C>0$ independent of ε. Making Eq. (51) explicit, we obtain the equation:

$$\begin{aligned}\partial_t \rho_{\rm d}(t,n) = \sum_k \int_0^{t/\varepsilon^2} ds\,\left[\rho_{\rm d}(t-\varepsilon^2 s,k) - \rho_{\rm d}(t-\varepsilon^2 s,n)\right] \qquad (52)\\ \times 2{\rm Re}\left(\exp\left(\Omega(k,n)s\right)\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\mathcal{V}\left(\frac{t}{\varepsilon^2}-s,k,n\right)\right) + O_{L^\infty([0,T];l^1)}(\varepsilon^{1-2\mu}).\end{aligned}$$

We notice here that the condition $\mu<1/2$ ensures that the remainder $O(\varepsilon^{1-2\mu})$ in the above equation is a remainder indeed.

Following the former analysis, we approximate the delayed differential equation (52) by a delay-free equation. To this aim, we first estimate thanks to (52):

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

On this account, substituting the delayed terms $\rho_{\rm d}(t-\varepsilon^2 s)$ by their asymptotic values $\rho_{\rm d}(t)$ in the integral term in (52), we introduce an error in the $L^\infty([0,T],l^1)$ norm, which can be estimated by

$$C\varepsilon^2\int_0^{T/\varepsilon^2} ds\, e^{-\varepsilon^\mu s}\; s\|\partial_t\rho_{\rm d}\|_{L^\infty([0,T],l^1)} \le C\varepsilon^2\,\frac{1}{\varepsilon^{3\mu}}.$$

This estimate together with (52) yields

$$\begin{aligned}\partial_t\rho_{\rm d}(t,n) \;&=\; O_{L^\infty([0,T],l^1)}(\varepsilon^{1-2\mu}) + O_{L^\infty([0,T],l^1)}(\varepsilon^{2-3\mu})\\ &\quad +2{\rm Re}\sum_k\left[\rho_{\rm d}(t,k)-\rho_{\rm d}(t,n)\right]\Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right).\end{aligned}$$

The Proposition is proved. □

3.3 Asymptotic analysis of the equation for populations

As in the case of uniform relaxations, Proposition 3 reduces the problem to the asymptotic analysis of (48). The techniques developed in Section 2 show that this study only amounts to the study of some averages of function $\Psi_\varepsilon(t/\varepsilon^2,k,n)$. From now on, we restrict once and for all to the case when function ϕ is r-chromatic, in that Hypothesis 4 holds. In this case, function Ψ_ε has the explicit value:

$$\begin{aligned}\Psi_\varepsilon\left(\frac{t}{\varepsilon^2},k,n\right) &= 2|V(n,k)|^2{\rm Re}\sum_{\alpha,\beta\in\mathbb{Z}^r}\phi_\alpha\phi_\beta\exp\left(i(\alpha+\beta)\cdot\omega\frac{t}{\varepsilon^2}\right)\\ &\quad\times\frac{1-\exp\left([-\varepsilon^\mu\gamma(k,n)-i(\omega(k,n)+\beta\cdot\omega)]t/\varepsilon^2\right)}{[\varepsilon^\mu\gamma(k,n)+i(\omega(k,n)+\beta\cdot\omega)]}. \qquad (53)\end{aligned}$$

Clearly, resonances which correspond to contributions for values of the parameters α, β, n, k such that $\alpha = -\beta$, *and* $\omega(k, n) + \beta \cdot \omega = 0$, play a special role in the analysis. In the sequel we show the following theorem.

Theorem 4. *Let us define the transition rate (dominant term)*

$$\Psi^{\mathrm{dom}}(k, n) := 2 \frac{|V(n, k)|^2}{\gamma(k, n)} \sum_{\substack{\beta \in \mathbb{Z}^r \\ \omega(k,n)+\beta\cdot\omega=0}} |\phi_\beta|^2. \tag{54}$$

Let also $\rho_{\mathrm{d}}^{(8)}$ *be solution to*

$$\partial_t \rho_{\mathrm{d}}^{(8)}(t, n) = \frac{1}{\varepsilon^\mu} \sum_k \Psi^{\mathrm{dom}}(k, n) \, [\rho_{\mathrm{d}}^{(8)}(t, k) - \rho_{\mathrm{d}}^{(8)}(t, n)], \tag{55}$$

with initial data $\rho_{\mathrm{d}}^{(8)}(0, n) = \rho_{\mathrm{d}}(0, n)$*. We assume that* $\mu < 1/2$*. Then for all* $T > 0$*, there exists* $C > 0$ *such that the following error estimate holds:*

$$\|\rho_{\mathrm{d}} - \rho_{\mathrm{d}}^{(8)}\|_{L^\infty([0,T],l^2)} \leq C \, \left(\varepsilon^\mu + \varepsilon^{1-2\mu}\right).$$

Remark 11. Theorem 4 states in a way that in the case when relaxations tend to zero with ε (slowly enough), populations relax immediately to the equilibrium states of the equation $\partial_t \rho(t, n) = \sum_k \Psi^{\mathrm{dom}}(k, n)[\rho(t, k) - \rho(t, n)]$, with some time boundary layer of size ε^μ. This instantaneous relaxation to an equilibrium state of course only affects the energy levels that resonate exactly with the wave.

In other words, everything goes on as if it were possible to pass to the limit as ε goes to zero, keeping first the relaxations, then pass to the limit in the relaxation term ε^μ. Note that the degenerate case $\Psi^{\mathrm{dom}} \equiv 0$ is not *a priori* excluded in the analysis.

The reader should notice that the error estimate takes place in l^2 norm here.

The simple but crucial remark that leads to Theorem 4 is the following.

Lemma 1. *Let us define the operator* B^{dom} *on the Hilbert space* l^2*, which associates to a sequence* $u(n) \in l^2$ *the sequence* $(B^{\mathrm{dom}}u)(n) \in l^2$ *according the formula:*

$$(B^{\mathrm{dom}}u)(n) = \sum_k \Psi^{\mathrm{dom}}(k, n)[u(k) - u(n)].$$

Then, operator B^{dom} *is a bounded* non-positive *operator on the Hilbert space* l^2*. In particular, the exponential* $\exp(tB^{\mathrm{dom}})$ *is well defined as an operator on* l^2 *as* $t \geq 0$*, and its norm is estimated by* 1*, for all* $t \geq 0$*.*

Remark 12. In the case of an arbitrary wave ϕ, we cannot compute as easily the main contribution Ψ^{dom} in the asymptotic process $\varepsilon \to 0$. Therefore we do not have any sign property at our disposal as put forward in Lemma 1. Because this property proves to be crucial in the sequel (and the asymptotic result in Theorem 4 does certainly not hold when the operator B^{dom} has no sign), this explains why we restrict the analysis to the case of a r-chromatic wave when relaxations tend to zero with ε.

Proof. The proof of Lemma 1 is obvious, and relies on the positiveness $\Psi^{\mathrm{dom}}(k, n) \geq 0$, and the symmetry $\Psi^{\mathrm{dom}}(k, n) = \Psi^{\mathrm{dom}}(n, k)$. □

Proof. We now prove Theorem 4. Let $T > 0$ be given. We estimate the difference $\rho_{\rm d} - \rho_{\rm d}^{(8)}$ in $L^\infty([0,T], l^2)$ norm. Thanks to Proposition 3, the equation governing $\rho_{\rm d}$ reads

$$\partial_t \rho_{\rm d}(t,n) = \sum_k \Psi_\varepsilon\left(\frac{t}{\varepsilon^2}, k, n\right)[\rho_{\rm d}(t,k) - \rho_{\rm d}(t,n)] + O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right),$$

where the transition rate Ψ_ε is given by (53).

First step: splitting of Ψ_ε. We split Ψ_ε into a dominant and two residual contributions, as follows:

$$\Psi_\varepsilon\left(\frac{t}{\varepsilon^2}, k, n\right) := \frac{\Psi^{\rm dom}(k,n)}{\varepsilon^\mu} + \Psi^{\rm res}\left(\frac{t}{\varepsilon^2}, k, n\right) + \widetilde{\Psi}^{\rm res}(k,n),$$

where $\Psi^{\rm dom}$ is defined by (54). By (53), the two residual contributions are

$$\widetilde{\Psi}^{\rm res}(k,n) := 2\,|V(n,k)|^2\,\varepsilon^\mu \sum_{\substack{\beta\in\mathbb{Z}^r,\\ \omega(k,n)+\beta\cdot\omega\neq 0}} \frac{|\phi_\beta|^2}{\varepsilon^{2\mu}\gamma(k,n)^2 + (\omega(k,n)+\beta\cdot\omega)^2},$$

and, respectively, $\Psi^{\rm res}\left(\frac{t}{\varepsilon^2}, k, n\right) := 2\,|V(n,k)|^2 \times \mathrm{Re}\left(\mathrm{I}+\mathrm{II}+\mathrm{III}+\mathrm{IV}\right)$, where

$$\mathrm{I} = -\varepsilon^{-\mu} \sum_{\substack{\beta\in\mathbb{Z}^r,\\ \omega(k,n)+\beta\cdot\omega=0}} \frac{|\phi_\beta|^2}{\gamma(k,n)} \exp\left(-\varepsilon^\mu\gamma(k,n)\,\frac{t}{\varepsilon^2}\right)$$

$$\mathrm{II} = -\sum_{\substack{\beta\in\mathbb{Z}^r,\\ \omega(k,n)+\beta\cdot\omega\neq 0}} \frac{|\phi_\beta|^2}{\varepsilon^\mu\gamma(k,n) + i(\omega(k,n)+\beta\cdot\omega)} \times \exp\left([-\varepsilon^\mu\gamma(k,n) \ - i(\omega(k,n)+\beta\cdot\omega)]\frac{t}{\varepsilon^2}\right)$$

$$\mathrm{III} = \varepsilon^{-\mu} \sum_{\substack{\alpha+\beta\neq 0,\\ \omega(k,n)+\beta\cdot\omega=0}} \frac{\phi_\alpha\phi_\beta}{\gamma(k,n)} \exp(-i(\alpha+\beta)\cdot\omega\,\frac{t}{\varepsilon^2}) \left[1 - \exp\left(-\varepsilon^\mu\gamma(k,n)\,\frac{t}{\varepsilon^2}\right)\right]$$

$$\mathrm{IV} = \sum_{\substack{\alpha+\beta\neq 0,\\ \omega(k,n)+\beta\cdot\omega\neq 0}} \frac{\phi_\alpha\phi_\beta}{[\varepsilon^\mu\gamma(k,n) + i(\omega(k,n)+\beta\cdot\omega)]} \exp\left(i(\alpha+\beta)\cdot\omega\,\frac{t}{\varepsilon^2}\right) \times\left[1 - \exp\left([-\varepsilon^\mu\gamma(k,n) - i(\omega(k,n)+\beta\cdot\omega)]\,\frac{t}{\varepsilon^2}\right)\right]\Bigg).$$

Now, the point in the sequel is that the first residual contribution $\tilde{\Psi}^{\rm res}$ is small (of order ε^μ) thanks to the real part, while the second residual contribution $\Psi^{\rm res}$ carries "time-oscillations" (at frequency $\varepsilon^{-2+\mu}$ at least), which kill the diverging factors $\varepsilon^{-\mu}$ and make them of size $\varepsilon^{2-2\mu}$.

Second step: preliminary bounds. Let us readily notice that the following three estimates are straightforward:

$$\sum_{k,n} |\Psi^{\mathrm{dom}}(k,n)| \leq C, \ \sum_{k,n} |\Psi^{\mathrm{res}}(t/\varepsilon^2,k,n)| \leq C\varepsilon^{-\mu}, \ \sum_{k,n} |\widetilde{\Psi}^{\mathrm{res}}(k,n)| \leq C\ \varepsilon^{\mu}\ , \quad (56)$$

for some $C>0$ independent of t and ε. All these bounds indeed stem from the decay assumptions made on $V(k,n)$ as well as on the Fourier coefficients ϕ_β. For instance, we may prove the last (and most difficult) bound appearing in (56) by writing

$$\begin{aligned}
\sum_{k,n} |\widetilde{\Psi}^{\mathrm{res}}(k,n)| &\leq C\ \varepsilon^{\mu} \sum_{\substack{k,n,\beta,\\ \omega(k,n)+\beta\cdot\omega\neq 0}} \frac{|\phi_\beta|^2 |V(k,n)|^2}{|\omega(k,n)+\beta\cdot\omega|^2} \\
&\leq C\ \varepsilon^{\mu} \sum_{k,n,\beta} (1+|\beta|)^{2(r-1+\delta)}\ (1+|n|)^{2(1+\delta)}\ (1+|k|)^{2(1+\delta)}\ |\phi_\beta|^2\ |V(k,n)|^2 \\
&\leq C\ \varepsilon^{\mu}\ .
\end{aligned}$$

The Diophantine estimate on ω (Hypothesis 4) is used in the second inequality, while the strong decay assumption (45) is used in the last estimate.

Third step: rewriting the equations. With the notations introduced above, Eq. (48) for ρ_{d} becomes

$$\begin{aligned}
\partial_t \rho_{\mathrm{d}}(t,n) \quad &= \quad \varepsilon^{-\mu} \sum_k \Psi^{\mathrm{dom}}(k,n)[\rho_{\mathrm{d}}(t,k)-\rho_{\mathrm{d}}(t,n)] \\
&\quad + \sum_k \Psi^{\mathrm{res}}\left(\frac{t}{\varepsilon^2},k,n\right)[\rho_{\mathrm{d}}(t,k)-\rho_{\mathrm{d}}(t,n)] \\
&\quad + \sum_k \widetilde{\Psi}^{\mathrm{res}}(k,n)[\rho_{\mathrm{d}}(t,k)-\rho_{\mathrm{d}}(t,n)] + O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right),
\end{aligned}$$

and this equation should be compared with

$$\partial_t \rho_{\mathrm{d}}^{(8)}(t,n) = \varepsilon^{-\mu} \sum_k \Psi^{\mathrm{dom}}(k,n)[\rho_{\mathrm{d}}^{(8)}(t,k)-\rho_{\mathrm{d}}^{(8)}(t,n)].$$

To reduce notations, we now introduce for all t the operators $B^{\mathrm{res}}(t/\varepsilon^2)$ and $\widetilde{B}^{\mathrm{res}}$ naturally associated with Ψ^{res} and $\widetilde{\Psi}^{\mathrm{res}}$, which operate on l^2:

$$\begin{aligned}
\left(B^{\mathrm{res}}(t/\varepsilon^2)u\right)(n) \quad &:= \quad \sum_k \Psi^{\mathrm{res}}\left(\frac{t}{\varepsilon^2},k,n\right)[u(k)-u(n)]\ , \\
(\widetilde{B}^{\mathrm{res}}u)(n) \quad &:= \quad \sum_k \widetilde{\Psi}^{\mathrm{res}}(k,n)[u(k)-u(n)]\ .
\end{aligned}$$

We know from estimates (56) that:

• B^{dom} is a bounded operator on l^2, and its norm is estimated by C,

• for all value of t the operator $B^{\mathrm{res}}(t)$ is likewise bounded on l^2, and its norm is estimated by $C\varepsilon^{-\mu}$,

• the norm of $\widetilde{B}^{\mathrm{res}}$ is bounded by $C\varepsilon^{\mu}$.

With these notations, and if functions $\rho_{\rm d}$ and $\rho_{\rm d}^{(8)}$ are considered as functions of the time t with values in l^2, Eqs (48) and (55) governing $\rho_{\rm d}$ and $\rho_{\rm d}^{(8)}$ respectively read

$$\begin{aligned}
&\partial_t \rho_{\rm d}(t) = \varepsilon^{-\mu} B^{\rm dom} \rho_{\rm d}(t) + B^{\rm res}\left(\frac{t}{\varepsilon^2}\right) \rho_{\rm d}(t) + \widetilde{B}^{\rm res} \rho_{\rm d}(t) + O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right),\\
&\partial_t \rho_{\rm d}^{(8)}(t) = \varepsilon^{-\mu} B^{\rm dom} \rho_{\rm d}^{(8)}(t).
\end{aligned}$$

From this, in order to estimate the difference $\Delta(t) := \rho_{\rm d}(t) - \rho_{\rm d}^{(8)}(t)$, we write

$$\begin{aligned}
\partial_t \Delta(t) = \varepsilon^{-\mu}(B^{\rm dom}\Delta)(t) + B^{\rm res}\left(\frac{t}{\varepsilon^2}\right) \rho_{\rm d}(t) + \widetilde{B}^{\rm res}\left(\frac{t}{\varepsilon^2}\right) \rho_{\rm d}(t)\\
+ O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right),
\end{aligned}$$

and we solve directly

$$\begin{aligned}
\Delta(t) &= \int_0^t ds\ \exp([t-s]\varepsilon^{-\mu} B^{\rm dom})\ B^{\rm res}\left(\frac{s}{\varepsilon^2}\right) \rho_{\rm d}(s)\\
&\quad + \int_0^t ds\ \exp([t-s]\varepsilon^{-\mu} B^{\rm dom})\ \widetilde{B}^{\rm res} \rho_{\rm d}(s) + O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right)\\
&=: \Delta^{(1)}(t) + \Delta^{(2)}(t) + O_{L^\infty([0,T],l^2)}\left(\varepsilon^{1-2\mu}\right). \qquad (57)
\end{aligned}$$

We now estimate separately each term on the right hand-side of (57).

Fourth step: estimating the first term in (57). To take advantage of the time oscillations of operator $B^{\rm res}(t/\varepsilon^2)$, and to display clearly that the right hand-side of (57) tends to 0 with ε, we carry out a natural integration by parts in the integral with respect to s and we obtain

$$\begin{aligned}
&\Delta^{(1)}(t) = \varepsilon^2 \left(\int_0^{t/\varepsilon^2} ds\ B^{\rm res}(s)\right) \rho_{\rm d}(t)\\
&+ \varepsilon^{2-\mu} \int_0^t ds\ \exp([t-s]\varepsilon^{-\mu} B^{\rm dom})\ B^{\rm dom} \left(\int_0^{s/\varepsilon^2} du\ B^{\rm res}(u)\right) \rho_{\rm d}(s)\\
&- \varepsilon^2 \int_0^t ds \exp([t-s]\varepsilon^{-\mu} B^{\rm dom}) \left(\int_0^{s/\varepsilon^2} du B^{\rm res}(u)\right) \left(\varepsilon^{-\mu} B^{\rm dom} + B^{\rm res}\left(\frac{s}{\varepsilon^2}\right)\right) \rho_{\rm d}(s).
\end{aligned}$$

Consequently, taking advantage of the bounds (56), as well as Lemma 1 (non-positiveness of $B^{\rm dom}$), we obtain the estimate

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

Besides we have

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

It follows that

$$\|\Delta^{(1)}\|_{L^\infty([0,T],l^2)} \leq C\varepsilon^{2-\mu} \sup_{0\leq t\leq T} \left\| \int_0^{t/\varepsilon^2} ds\ B^{\rm res}(s) \right\|_{\mathcal{L}(l^2)}. \qquad (58)$$

There remains to estimate the supremum occurring in (58).

To this aim, we proceed as in the proof of Theorem 1 (see (31) and the next estimates): we integrate at sight function Ψ^{res}, and taking advantage of the Diophantine condition on the frequency vector ω (Hypothesis 4), together with the strong decay of V (assumption (45)), we deduce the estimate

$$\sup_{0\le t\le T}\left\|\int_0^{t/\varepsilon^2} ds\ B^{\text{res}}(s)\right\|_{\mathcal{L}(l^2)} \le \sup_{0\le t\le T}\left\|\int_0^{t/\varepsilon^2} ds\ \Psi^{\text{res}}(s,k,n)\right\|_{l^1} \le C\varepsilon^{-\mu}.$$

In short, we showed the estimate

$$\|\Delta^{(1)}(t)\|_{L^\infty([0,T],l^2)} \le C\varepsilon^{2-2\mu},$$

and this last quantity tends to 0 with ε.

Fifth step: estimating the second term in (57). The analysis of $\Delta^{(2)}$ now uses (56) in a direct way, in that we write

$$\begin{aligned}\|\Delta^{(2)}(t)\|_{L^\infty([0,T],l^2)} &\le C\,T\sup_{s\in[0,T]}\|\widetilde{B}^{\text{res}}\rho_{\text{d}}(s)\|_{l^2}\\ &\le C\|\widetilde{B}^{\text{res}}\|_{\mathcal{L}(l^2)}\sup_{s\in[0,T]}\rho_{\text{d}}(s)\|_{l^2}\\ &\le C\|\widetilde{\Psi}^{\text{res}}\|_{l^1}\le C\,\varepsilon^\mu\,,\end{aligned}$$

where we made use of the non-positiveness of B^{dom}, the boundedness of ρ_{d} (see the previous step), and the estimate (56) proved before.

Conclusion. Theorem 4 is now proved since the total error has size $\varepsilon^\mu+\varepsilon^{1-2\mu}+\varepsilon^{2-2\mu}$. $\square$

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References

[Arn89] V.I. Arnold. *Mathematical Methods of Classical Mechanics.* Number 60 in Graduate Texts in Mathematics. Springer-Verlag, 1989.

[BBR01] B. Bidégaray, A. Bourgeade, and D. Reignier. Introducing physical relaxation terms in Bloch equations. *J. Comput. Phys.*, 170:603–613, 2001.

[BCEP04] D. Benedetto, F. Castella, R. Esposito, and M. Pulvirenti. Some considerations on the derivation of the nonlinear quantum Boltzmann equation. *to appear in J. Stat. Phys.*, 2004.

[BFCDG03] B. Bidégaray-Fesquet, F. Castella, E. Dumas, and M. Gisclon. From Bloch model to the rate equations II: the case of almost degenerate energy levels. *preprint*, 2003.

[Boh79] A. Bohm. *Quantum Mechanics*. Texts and monographs in Physics. Springer-Verlag, 1979.

[Boy92] R.W. Boyd. *Nonlinear Optics*. Academic Press, 1992.

[Cas99] F. Castella. On the derivation of a quantum Boltzmann equation from the periodic von Neumann equation. *Mod. Math. An. Num.*, 33(2):329–349, 1999.

[Cas01] F. Castella. From the von Neumann equation to the quantum Boltzmann equation in a deterministic framework. *J. Stat. Phys.*, 104(1/2):387–447, 2001.

[Cas02] F. Castella. From the von Neumann equation to the quantum Boltzmann equation II: identifying the Born series. *J. Stat. Phys.*, 106(5/6):1197–1220, 2002.

[CP02] F. Castella and A. Plagne. A distribution result for slices of sums of squares. *Math. Proc. Cambridge Philos. Soc.*, 132(1):1–22, 2002.

[CP03] F. Castella and A. Plagne. Non-derivation of the quantum Boltzmann equation from the periodic Schrödinger equation. *Indiana Univ. Math. J.*, 51(4):963–1016, 2003.

[CTDRG88] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg. *Processus d'interaction entre photons et atomes*. Savoirs actuels. Intereditions/Editions du CNRS, 1988.

[EY00] L. Erdös and H.T. Yau. Linear Boltzmann equation as the weak coupling limit of a random Schrödinger equation. *Comm. Pure Appl. Math.*, 53(6):667–735, 2000.

[KL57] W. Kohn and J.M. Luttinger. Quantum theory of electrical transport phenomena. *Phys. Rev.*, 108:590–611, 1957.

[KL58] W. Kohn and J.M. Luttinger. Quantum theory of electrical transport phenomena. II. *Phys. Rev.*, 109:1892–1909, 1958.

[KPR96] J.B. Keller, G. Papanicolaou, and L. Ryzhik. Transport equations for elastic and other waves in random media. *Wave Motion*, 24(4):327–370, 1996.

[Kre83] H.J. Kreuzer. *Nonequilibrium thermodynamics and its statistical foundations*. Monographs on Physics and Chemistry of Materials. Oxford Science Publications, 1983.

[Lin76] G. Lindblad. On the generators of quantum dynamical semigroups. *Comm. Math. Phys.*, 48:119–130, 1976.

[LM88] P. Lochak and C. Meunier. *Multiphase averaging for classical systems. With applications to adiabatic theorems*. Number 72 in Applied Mathematical Sciences. Springer-Verlag, 1988.

[Lou91] R. Loudon. *The quantum theory of light*. Clarendon Press, Oxford, 1991.

[Nie96] F. Nier. A semi-classical picture of quantum scattering. *Ann. Sci. Ec. Norm. Sup., 4. Sér.*, 29:149–183, 1996.

[NM92] A.C. Newell and J.V. Moloney. *Nonlinear Optics.* Advanced Topics in the Interdisciplinary Mathematical Sciences. Addison-Wesley Publishing Company, 1992.

[Spo77] H. Spohn. Derivation of the transport equation for electrons moving through random impurities. *J. Stat. Phys.*, 17(6):385–412, 1977.

[Spo80] H. Spohn. Kinetic equations from Hamiltonian dynamics: Markovian limits. *Rev. Mod. Phys.*, 52(3):569–615, 1980.

[Spo91] H. Spohn. *Large Scale Dynamics of interacting particles.* Springer, Berlin, 1991.

[SSL77] M. Sargent, M.O. Scully, and W.E. Lamb. *Laser Physics.* Addison-Wesley, 1977.

[SV85] J.A. Sanders and F. Verhulst. *Averaging methods in nonlinear dynamical systems*, volume 59 of *Applied Mathematical Sciences.* Springer-Verlag, 1985.

[Zwa66] R. Zwanzig. *Quantum Statistical Mechanics.* Gordon and Breach, New-York, 1966.