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From Bloch model to the rate equations

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Abstract

In this article, we consider Bloch equations which govern the evolution of the density matrix of a quantum system with a discrete set of energy levels. We assume that the system is forced by a time dependent electric potential. We suppose that the potential 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 rôle in the analysis.

Keywords: density matrix, Bloch equations, rate equations, linear Boltzmann equation, averaging, Diophantine estimates.

1 Introduction

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

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

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

$$
\rho_d(t,n):=\rho(t,n,n) .
$$

Here, $\rho(t, n, m)$ denotes the density matrix of an atomic system with discrete energy levels indexed by the integer n, and $\rho_d(t, n)$ is the occupation number of the n-th level. The quantity $\omega(n,m) = \omega(n) - \omega(m)$ is the difference between the energies $\omega(n)$ and $\omega(m)$ of levels n and m respectively. This equation describes the evolution of an atom, or a quantum system with a discrete number of energy levels, forced by a high frequency electromagnetic wave. Its amplitude $\phi(t/\varepsilon^2)$ 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. 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 assume this quantity to be given.

The reaction of the atom is studied over long times of order $1/\varepsilon^2$, and the coupling between the atom and the wave is weak, of order ε . Last we assume that the atom has a tendency to relax to a given equilibrium state, *via* relaxation coefficients $\gamma(n, m) \geq 0$. In this regime, the predominant phenomenon as $\varepsilon \to 0$ is determined by the resonance of the electromagnetic source frequencies with the eigen-frequencies $\omega(n,m)$ of the system. Thus the wave/atom interaction tends to couple more stongly the wave frequencies that are resonant with the eigen-frequencies of the system.

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

$$
\partial_t \rho_d(t, n) = \sum_k \sigma(n, k) [\rho_d(t, k) - \rho_d(t, n)], \qquad (2)
$$

with transition levels $\sigma(n, k)$, that depend on the wave ϕ and on the atom parameters, and that 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). 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.

• 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.

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:

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

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], [CD99], 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 rôle in the analysis.

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. We can cite [Cas99], [Cas02], [Cas01], [CD99] 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 eleectron 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 [BCEP03] 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 mathematical problem we deal with.

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 4, 5, 6, and 8. 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

$$
\varepsilon^2 \partial_t \rho(t, n, m) = -(\mathrm{i}\omega(n, m) + \gamma(n, m)) \rho(t, n, m) \n+ i\varepsilon \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k\right) \rho(t, k, m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, m\right) \rho(t, n, k) \right].
$$
\n(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. Without altering the analysis below, we can add a relaxation term $\varepsilon^2 Q(\rho)(t,n,n)$ to Eq. (3) 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 coherences between the levels without any effect on the populations. The assumption that the relaxation for coherences (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 (coherences), and non negative and summable diagonal terms (populations). More precisely, we assume that

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

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

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

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

where ϕ is a real-valued function which is bounded on 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 assume 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 $\rho_{od}(t, n, m) := \rho(t, n, m) \mathbf{1}[n \neq m]$. The quantity ρ_{od} represents the offdiagonal part of the density matrix ρ , also called *coherences*.
- In the same way, we denote $\rho_d(t,n) := \rho(t,n,n)$. 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

$$
||v||_{l^{1}\infty \cap l^{\infty}l^{1}} := \sup_{n} \sum_{m} |v(n, m)| + \sup_{m} \sum_{n} |v(n, m)|.
$$

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

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

and for the populations

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

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 V belongs to $L^{\infty}(\mathbb{R}^+, l^1 l^{\infty} \cap l^{\infty} l^1)$. Therefore the linear operator which associates $\tilde{\rho}$ to $\rho \in L^{\infty}(\mathbb{R}^+, l^1)$ defined by fined 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 $\partial_t \rho \in L^{\infty}_{loc}(\mathbb{R}^+, l^1)$.

Besides these solutions to Eq. (3) have additionnal 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: $\rho_{od}(t, n, m) =$ $\rho_{\text{od}}(t, m, n)^*$. Next, for all $t \in \mathbb{R}$, trace is conserved: $\sum_n \rho_{\text{d}}(t, n) = \sum_n \rho_{\text{d}}(0, n) < \infty$. Last, for all $t \geq 0$, positiveness is conserved: $\rho_d(t,n) \geq 0$ (see [Lin76], [BBR01], [Cas01]). In particular, Eq. (8) which governs the diagonal part can be cast as

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

2.2 Towards an equation for populations

In this section, we transform the coupled system $(7)-(8)$ into one equation governing the populations $\rho_d(t, n)$ only. More precisely, we show the following lemma.

Lemma 1. Let us define the time dependent transition rate

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

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

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

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

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

Remark. 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 lemma 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 .

Remark. The following proof shows the result for a small enough value of ε . However, in the opposite case – which is of course not the one we are interested in – the lemma clearly holds true since ρ_d and $\rho_d^{(1)}$ $d_d^{(1)}$ are bounded in $L^{\infty}([0,T], l^1)$.

Proof. Lemma 1 is proved in three steps. First, the values of the coherences $\rho_{od}(t, n, k)$ are computed at the first order in ε in terms of the populations $\rho_d(t, n)$ only (see Lemma 2 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. (13)). 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 coherences. We first write an integral equation for the coherences (off-diagonal terms). Using Eq. (7) and since the initial data is $\rho_{od}(t=0, n, m) \equiv 0$, we of course have

$$
\rho_{od}(t, n, m) = i\varepsilon \int_0^{t/\varepsilon^2} ds \exp(\Omega(n, m)s) \mathcal{V}\left(\frac{t}{\varepsilon^2} - s, n, m\right) \left[\rho_d(t - \varepsilon^2 s, m) - \rho_d(t - \varepsilon^2 s, n)\right]
$$

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

We can obviously solve iteratively the integral equation (11) in terms of the unknown ρ_{od} , and obtain ρ_{od} as a *complete* expansion in powers of ε , in term of ρ_d . 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 ρ_{od} . To achieve this, we state the following result.

Lemma 2. Let us set

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

Then, for all given time $T \geq 0$, we have the estimate

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

for some constant C independent on ε .

Remark. 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.

Proof. Let $T \geq 0$ be given. We denote by A_{ε} the operator which associates to $u \in L^{\infty}([0, T], l^1)$ the quantity

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

We have the straightforward estimate

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

$$
\leq C||u||_{L^{\infty}([0,T],l^{1})}.
$$

Besides, thanks to the integral equation (11) governing ρ_{od} , the following equality holds.

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

For this reason we easily estimate the difference

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

\n
$$
= C\varepsilon \|A_{\varepsilon} (\rho_{\text{od}} - i\varepsilon \rho_{\text{od}}^{(0)} + i\varepsilon \rho_{\text{od}}^{(0)})\|_{L^{\infty}(\mathbb{R}, l^{1})}
$$

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

and thus we obtain for a small ε

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

for some constant $C > 0$ independent on ε . There remains to use the obvious estimate

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

together with the classical identity

$$
\|\rho_{\mathbf{d}}\|_{L^{\infty}([0,T],l^{1})} = \|\rho_{\mathbf{d}}(t=0)\|_{l^{1}},
$$
\n(12)

and Lemma 2 is proved. It must be stressed that the key estimate (12) is based on the Lindblad property [Lin76] (which implies the conservation of positiveness for ρ_d), and on the conservation of the l^1 -norm of ρ_d . \Box Second step: towards a time delayed differential equation for the populations. Lemma 2 together with Eq. (9) governing ρ_d claim that

$$
\partial_t \rho_d(t,n) = \sum_k \int_0^{t/\varepsilon^2} ds \left[\rho_d(t - \varepsilon^2 s, k) - \rho_d(t - \varepsilon^2 s, n) \right]
$$
\n
$$
\tag{13}
$$

$$
\times 2\mathrm{Re}\left(\exp\left(\Omega(k,n)s\right)\mathcal{V}\left(\frac{t}{\varepsilon^2},n,k\right)\mathcal{V}\left(\frac{t}{\varepsilon^2}-s,k,n\right)\right)+\varepsilon r_{\varepsilon}(t,n),
$$

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

$$
||r_{\varepsilon}||_{L^{\infty}([0,T],l^{1})} \leq C.
$$
\n(14)

It is natural to introduce $\rho_d^{(0)}$ $d_d^{(0)}$, which is solution to

$$
\partial_t \rho_d^{(0)}(t,n) = \sum_k \int_0^{t/\varepsilon^2} ds \left[\rho_d^{(0)}(t-\varepsilon^2 s, k) - \rho_d^{(0)}(t-\varepsilon^2 s, n) \right]
$$
\n
$$
\times 2 \text{Re} \left(\exp \left(\Omega(k,n)s \right) \mathcal{V} \left(\frac{t}{\varepsilon^2}, n, k \right) \mathcal{V} \left(\frac{t}{\varepsilon^2} - s, k, n \right) \right), \tag{15}
$$

with initial data $\rho_d^{(0)}$ $d_{d}^{(0)}(0, n) = \rho_{d}(0, n)$. Clearly, Eqs (13) and (15) governing ρ_{d} and $\rho_{d}^{(0)}$ $\mathbf{d}^{(0)}$, together with the estimate (14) for r_{ε} , imply that, for all $T > 0$, there exists a constant C, independent on ε , such that

$$
\|\rho_{\mathbf{d}} - \rho_{\mathbf{d}}^{(0)}\|_{L^{\infty}([0,T],l^1)} \le C\varepsilon. \tag{16}
$$

Thus, there remains to estimate the difference between $\rho_d^{(1)}$ $d_d^{(1)}$, solution to the delay-free equation (10) , and $\rho_d^{(0)}$ $d_d^{(0)}$, solution to the integro-differential equation (15).

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 $T \geq 0$ be given. Moreover, the delayed terms $\rho_d^{(0)}$ $d_d^{(0)}(t-\varepsilon^2 s)$ in Eq. (15) read

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

Thus, Eq. (15) 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

$$
||r_{\varepsilon}||_{L^{\infty}([0,T],l^{1})} \leq C\gamma^{-2}||\partial_{t}\rho_{\mathrm{d}}^{(0)}||_{L^{\infty}([0,T],l^{1})}
$$
 using a Taylor expansion
\n
$$
\leq C\gamma^{-3}||\rho_{\mathrm{d}}^{(0)}||_{L^{\infty}([0,T],l^{1})}
$$
 using a Taylor expansion
\n
$$
\leq C||\rho_{\mathrm{d}}||_{L^{\infty}([0,T],l^{1})}
$$
 thanks to Eq. (15)
\nthanks to Eq. (16)
\nthanks to Eq. (12),

for some constant C independent on ε , only taking $\varepsilon \leq 1/2$. Lemma 1 follows from these estimates. \Box

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

Lemma 3. Let us define the time dependent transition rate

$$
\Psi\left(\frac{t}{\varepsilon^2}, k, n\right) = 2|V(n,k)|^2 \operatorname{Re} \int_0^{+\infty} ds \, \exp\left(\Omega(k,n)s\right) \phi\left(\frac{t}{\varepsilon^2}\right) \phi\left(\frac{t}{\varepsilon^2} - s\right). \tag{17}
$$

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

$$
\partial_t \rho_d^{(2)}(t,n) = \sum_k \Psi\left(\frac{t}{\varepsilon^2}, k, n\right) \left[\rho_d^{(2)}(t,k) - \rho_d^{(2)}(t,n)\right],\tag{18}
$$

with initial data $\rho_d^{(2)}$ $d_d^{(2)}(0,n) = \rho_d(0,n)$. Then, for all $T > 0$, there exists $C > 0$, independent on ε , such that

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

Proof. This result is straightforward since we have

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

(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). \Box

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 $\rho_d = \rho_d^0 + \varepsilon \rho_d^1 + \cdots$, 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 V.

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

Hypothesis 1.

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

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

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

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. 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

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

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 on α). In the sequel, it would be sufficient to assume that the Fourier coefficients of Φ are polynomially decreasing, that is ensure the estimate

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

for some coefficient C_1 , independent on 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. 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

$$
\phi(t) = \sum_{\alpha \in \mathbb{Z}^r} \phi_\alpha \exp(2i\pi\alpha \cdot \omega t). \tag{19}
$$

Moreover coefficients ϕ_{α} verify, for analyticity reasons,

$$
|\phi_{\alpha}| \le C_1 \exp(-C_2|\alpha|),\tag{20}
$$

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

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

Now we state the main theorem of this section.

Theorem 4. (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 (17). Let also $\rho_d^{(3)}$ $\int_{d}^{(3)}$ be solution to

$$
\partial_t \rho_d^{(3)}(t,n) = \sum_k \langle \Psi \rangle(k,n) \left[\rho_d^{(3)}(t,k) - \rho_d^{(3)}(t,n) \right],\tag{21}
$$

with initial data $\rho_d^{(3)}$ $d_d^{(3)}(0,n) = \rho_d(0,n)$. Then, for all $T > 0$, there exists a constant $C > 0$ independent on ε , such that

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

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

$$
\langle \Psi \rangle (k,n) = \frac{|V(n,k)|^2 \gamma(k,n)}{2} \left(\frac{1}{\gamma(k,n)^2 + (2\pi\omega - \omega(k,n))^2} + \frac{1}{\gamma(k,n)^2 + (2\pi\omega + \omega(k,n))^2} \right).
$$
\n(22)

(iii) In the r-chromatic case, formula (22) may be extended to

$$
\langle \Psi \rangle (k, n) = 2 \sum_{\beta \in \mathbb{Z}^r} \frac{\gamma(k, n) |V(n, k)|^2 |\phi_{\beta}|^2}{\gamma(k, n)^2 + [2\pi\beta \cdot \omega + \omega(k, n)]^2}.
$$
\n(23)

Remark. According to Lemma 3, Eq. (18) governing $\rho_d^{(2)}$ $\int_{d}^{(2)}$ is cast as

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

On the other hand, Theorem 4 states that $\rho_d^{(3)}$ $\int_{d}^{(3)}$ 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 4 is splitted in several steps.

First step: preliminary remarks and problem reduction. The proof of (22) and (23) follows from a direct calculation (see (28) below).

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

$$
\partial_t \rho_\mathbf{d}^{(4)}(t,n) = \sum_k \left[\rho_\mathbf{d}^{(4)}(t,k) - \rho_\mathbf{d}^{(4)}(t,n) \right] \times \varepsilon \int_0^{1/\varepsilon} ds \, \Psi\left(\frac{t}{\varepsilon^2} + s, k, n\right),\tag{24}
$$

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

$$
\|\rho_{\mathbf{d}}^{(2)} - \rho_{\mathbf{d}}^{(4)}\|_{L^{\infty}([0,T],l^{1})} \le C\varepsilon,
$$
\n(25)

and

$$
\|\rho_{\mathbf{d}}^{(4)} - \rho_{\mathbf{d}}^{(3)}\|_{L^{\infty}([0,T],l^{1})} \leq C\varepsilon.
$$
\n(26)

These two estimates of course end the proof of Theorem 4.

Second step: proof of (26) . The proof of (26) is based on the estimate

$$
\left\| \langle \Psi \rangle - \varepsilon \int_0^{1/\varepsilon} ds \, \Psi \left(\frac{t}{\varepsilon^2} + s \right) \right\|_{L^\infty([0,T], l^1/\infty \cap l^\infty l^1)} \le C\varepsilon,\tag{27}
$$

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 (19)-(20). More precisely, from (19) we deduce easily the formula

$$
\Psi(t,k,n) = 2|V(n,k)|^2 \operatorname{Re} \sum_{\alpha,\beta \in \mathbb{Z}^r} \frac{\phi_\alpha \phi_\beta \exp(2i\pi[\alpha+\beta] \cdot \omega t)}{-\Omega(k,n) + 2i\pi\beta \cdot \omega}.
$$
\n(28)

This developed form for Ψ allows to estimate the left hand-side in (27) by

$$
2\sum_{\beta \in \mathbb{Z}^r} |\phi_{\beta}| \left\| \frac{|V(n,k)|^2}{-\Omega(k,n) + 2i\pi\beta \cdot \omega} \right\|_{l^1 l^{\infty} \cap l^{\infty} l^1} \times \left| \left| \frac{|V(n,k)|^2}{\phi_{-\beta} - \sum_{\alpha \in \mathbb{Z}^r} \varepsilon \int_0^{1/\varepsilon} ds \phi_{\alpha} \exp\left(2i\pi[\alpha + \beta] \cdot \omega \left[\frac{t}{\varepsilon^2} - s\right] \right) \right|.
$$
\n(29)

Thanks to the exponentially decreasing estimate given by (20), we do have the right to integrate

each term separately. According to the Diophantine condition on ω , we then notice that

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

On the other hand, we use the estimate

$$
|-\Omega(k,n)+2i\pi\beta\cdot\omega|\geq\gamma.
$$

This allows to estimate (29) by

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

and (27) is established.

J.

Let us by the way notice that a term by term integration on the expansion (28) for Ψ leads immediately to formula (23).

Third step: proof of (25). The proof of (25) is a little more delicate. The difference $\Delta =$ $\rho_\mathrm{d}^{(2)}-\rho_\mathrm{d}^{(4)}$ $\frac{d^{(4)}}{d}$ is solution to

$$
\partial_t \Delta(t,n) = \sum_k \Psi(\frac{t}{\varepsilon^2}, k,n) \left[\Delta(t,k) - \Delta(t,n) \right] + \sum_k \left(\Psi(\frac{t}{\varepsilon^2}, k,n) - \varepsilon \int_0^{1/\varepsilon} ds \ \Psi\left(\frac{t}{\varepsilon^2} + s, k,n \right) \right) \left[\rho_d^{(4)}(t,k) - \rho_d^{(4)}(t,n) \right],
$$

with vanishing initial data. Therefore we can write

$$
\Delta(t,n) = \int_0^t du \sum_k \Psi(\frac{u}{\varepsilon^2}, k, n) \left[\Delta(u, k) - \Delta(u, n) \right]
$$
\n
$$
+ \int_0^t du \sum_k \left(\Psi(\frac{u}{\varepsilon^2}, k, n) - \varepsilon \int_0^{1/\varepsilon} ds \ \Psi\left(\frac{u}{\varepsilon^2} + s, k, n \right) \right) \left[\rho_d^{(4)}(u, k) - \rho_d^{(4)}(u, n) \right].
$$
\n(30)

The first term of the right hand-side in (30) is estimated in the l^1 norm by $C \int_0^t du \|\Delta(u)\|_{l^1}$. Using Gronwall lemma, the proof of (25) therefore reduces to prove the following estimate for $|t| \leq T$:

$$
\Big\| \int_0^t du \sum_k \left(\Psi(\frac{u}{\varepsilon^2}, k, n) - \varepsilon \int_0^{1/\varepsilon} ds \, \Psi\left(\frac{u}{\varepsilon^2} + s, k, n\right) \right) \left[\rho_d^{(4)}(u, k) - \rho_d^{(4)}(u, n) \right] \Big\|_{l^1} \le C\varepsilon.
$$
\n(31)

We therefore conclude by proving (31). To this aim, we write for the contribution due to $\rho^{(4)}_{\rm d}$ $\int_{d}^{(4)}(u,k)$ (we handle the contribution due to $\rho_d^{(4)}$ $d_d^{(4)}(u,n)$ in a similar way),

$$
\int_0^t du \sum_k \varepsilon \int_0^{1/\varepsilon} ds \Psi\left(\frac{u}{\varepsilon^2} + s, k, n\right) \rho_d^{(4)}(u, k)
$$

=
$$
\sum_k \int_0^1 ds \int_0^t du \Psi\left(\frac{u + \varepsilon s}{\varepsilon^2}, k, n\right) \rho_d^{(4)}(u, k)
$$

=
$$
\sum_k \int_0^1 ds \int_0^t du \Psi\left(\frac{u + \varepsilon s}{\varepsilon^2}, k, n\right) \rho_d^{(4)}(u + \varepsilon s, k) + O_{L^{\infty}([0, T], l^1)}(\varepsilon), \quad (32)
$$

where the $O_{L^{\infty}([0,T],l^1)}(\varepsilon)$ term means that the difference is estimated by $C\varepsilon$ in the $L^{\infty}([0,T],l^1)$ norm, for some constant C independent on ε . Of course, this last estimate is obtained thanks to the definition of Ψ , as well as Eq. (24) governing $\rho_d^{(4)}$ $d_d^{(4)}$, which implies that

$$
\|\partial_t \rho_{\rm d}^{(4)}(t,n)\|_{L^\infty([0,T],l^1)} \leq C.
$$

We now estimate the first term in (32) 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

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

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 (31), which ends the proof of Theorem 4.

Remark. A remark has to be made on the above proof of Theorem 4. We followed [SV85] since it 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 and we refer to Section 3.3 for an illustration of this fact.

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.

 \Box

Hypothesis 2.

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

where ϕ is a bounded C^1 function, such that, for all $t \geq 0$,

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

where Ψ is defined by (17) 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

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

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

We now state the main theorem of this section.

Theorem 5. (i) Under the "KBM" hypothesis 2, we introduce $\rho_d^{(5)}$ $d_d^{(0)}$, which is solution to

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

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

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

(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. Theorems 4 and 5 show that, in the limit $\varepsilon \to 0$, populations $\rho_d(t,n)$ 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 rchromatic 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 (22) or (23). The above item *(ii)* shows, in a less quantitative way, that this phenomenon always occurs if the wave has a discrete stectrum 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 $\text{Re }\Omega(k,n) \leq -\gamma < 0$ 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 4.

 $\sqrt{\delta(\varepsilon)}/\varepsilon^2$). We introduce $\rho_d^{(6)}$ First step. We take a long time scale $T(\varepsilon)$ to be determined (in fine we will take $T(\varepsilon)$ = $\mathbf{d}^{(0)}$, which is solution to

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

with initial data $\rho_d^{(6)}$ $d_d^{(0)}(0,n) = \rho_d(0,n)$. Following the proof of Theorem 4, it is easy to find the error estimate:

$$
\|\rho_{\mathbf{d}}^{(2)} - \rho_{\mathbf{d}}^{(6)}\|_{L^{\infty}([0,T],l^{1})} \le C\varepsilon^{2}T(\varepsilon). \tag{33}
$$

Second step. Coming back to the definition of $\delta(\varepsilon)$, and using the equations governing $\rho_d^{(6)}$ d and $\rho_d^{(5)}$ $_{d}^{(5)}$ respectively, we easily establish the estimate

$$
\|\rho_{\mathbf{d}}^{(5)} - \rho_{\mathbf{d}}^{(6)}\|_{L^{\infty}([0,T],l^{1})} \le C \frac{\delta(\varepsilon)}{\varepsilon^{2} T(\varepsilon)}.
$$
\n(34)

The optimal choice $T(\varepsilon) = \sqrt{\delta(\varepsilon)}/\varepsilon^2$ in (33) and (34) yields Theorem 5. \Box

2.4.2 Case of a r-chromatic wave with spectrum broadening

Theorems 4 and 5 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.

$$
\phi(t) = \frac{1}{\varepsilon^{qr}} \sum_{\alpha \in \mathbb{Z}^r} \int_{\mathbb{R}^r} d\eta \, A\left(\alpha, \frac{\eta - \omega}{\varepsilon^q}\right) \exp(2i\pi\alpha \cdot \eta \, t) \n= \sum_{\alpha \in \mathbb{Z}^r} \int_{\mathbb{R}^r} d\eta \, A(\alpha, \eta) \exp(2i\pi\alpha \cdot [\omega + \varepsilon^q \eta] \, t),
$$
\n(35)

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

$$
A(\alpha, \eta) \in l^1(\mathbb{Z}^r, L^1(\mathbb{R}^r)).
$$
\n(36)

Note that we do not make explicit the dependence of ϕ with respect to ε in (35).

The wave ϕ being given by (35), we now establish the analogue of Theorem 4 shown in the r-chromatic case.

Theorem 6. Let ϕ such that (35) and (36) hold. We introduce $\rho_d^{(7)}$ $d_{\mathbf{d}}^{(t)}(t,n)$, which is solution to

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

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

$$
\langle \Psi \rangle (k, n) := 2|V(n, k)|^2 \text{Re} \sum_{\beta \in \mathbb{Z}^r} \frac{\left| \int_{\mathbb{R}^r} d\eta \, A(\beta, \eta) \right|^2}{\gamma(k, n) + i \left[\omega(k, n) + 2\pi \beta \cdot \omega \right]}.
$$
\n(37)

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

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

Remark. Theorem 6 shows, in particular, that spectrum broadening does not play an essential $r\^ole$ and everything works as if the wave were straight off really r-chromatic (limiting case $q = +\infty \, in \, (35)$.

Proof. To show Theorem 6, we proceed as in the proof of Theorems 4 and 5.

We therefore have to calculate the function Ψ as defined by (17) (once more, we do not make explicit the ε -dependence of Ψ). We easily have

$$
\Psi(t,k,n) = 2|V(n,k)|^{2} \text{Re} \sum_{\alpha,\beta \in \mathbb{Z}^{r}} \int_{\mathbb{R}^{2r}} d\eta \, d\eta' \, A(\alpha,\eta) A(\beta,\eta') \times \frac{\exp(2i\pi \left[[\alpha+\beta] \cdot \omega + \varepsilon^{q} [\alpha \cdot \eta + \beta \cdot \eta'] \right] t)}{\gamma(k,n) + i \left[\omega(k,n) + 2\pi\beta \cdot [\omega + \varepsilon^{q}\eta'] \right]}.
$$
 (38)

Then we *define* the asymptotic quantity

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

Of course, since ϕ is a real function, definition (39) is equivalent to (37).

Last, as in the proof of Theorem 5, 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 (27) and (31))

$$
\left\| \langle \Psi \rangle - \varepsilon^{\delta} \int_0^{1/\varepsilon^{\delta}} ds \, \Psi \left(\frac{t}{\varepsilon^2} + s \right) \right\|_{L^{\infty}([0,T], l^1(\infty) \cap l^{\infty} l^1)} \xrightarrow{\varepsilon \to 0} 0,
$$
\n(40)

$$
\left\| \int_0^t du \left(\Psi\left(\frac{u}{\varepsilon^2}\right) - \varepsilon^\delta \int_0^{1/\varepsilon^\delta} ds \ \Psi\left(\frac{u}{\varepsilon^2} + s\right) \right) \right\|_{L^\infty([0,T], l^1 l^\infty \cap l^\infty l^1)} \leq C\varepsilon^{2-\delta}.
$$
 (41)

The proof of (41) is easy and follows the same outline as the above proof of (31). We therefore merely show (40).

To this aim, we begin with the definitions (38) and (39) of Ψ and its asymptotic average $\langle \Psi \rangle$. Then we estimate easily the left hand-side of (40) by

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

+
$$
C \sum_{\beta} \int_{\mathbb{R}^{2r}} d\eta \ d\eta' |A(-\beta,\eta)| |A(\beta,\eta')| \left| 1 - \frac{\exp (2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]) - 1}{2i\pi\varepsilon^{q-\delta}\beta\cdot[\eta'-\eta]} \right|.
$$

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 R. For the second term, we notice that for all β and almost all η , η' , we have

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

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

$$
\left| \frac{\exp (2i\pi [[\alpha + \beta] \cdot \omega + \varepsilon^{q}[\alpha \cdot \eta + \beta \cdot \eta']] \varepsilon^{-\delta}) - 1}{2i\pi [[\alpha + \beta] \cdot \omega + \varepsilon^{q}[\alpha \cdot \eta + \beta \cdot \eta']] \varepsilon^{-\delta}} \right|
$$

$$
\leq \frac{C\varepsilon^{\delta}}{\left| [\alpha + \beta] \cdot \omega + \varepsilon^{q}[\alpha \cdot \eta + \beta \cdot \eta'] \right|} \varepsilon^{\frac{\gamma}{\gamma+1}} \frac{C\varepsilon^{\delta}}{\left| [\alpha + \beta] \cdot \omega \right|} \leq C \varepsilon^{\delta},
$$

where we of course use the Diophantine property which is satisfied by ω . This ends the proof of (40).

To conclude, estimates (40) and (41), together with the techniques developed in the proofs of Theorems 4 and 5, yield Theorem 6. \Box

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

$$
\gamma(n,m) \to \varepsilon^{\mu} \gamma(n,m)
$$

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

$$
\varepsilon^2 \partial_t \rho(t, n, m) = -\left(i\omega(n, m) + \varepsilon^{\mu} \gamma(n, m)\right) \rho(t, n, m) \n+ i\varepsilon \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k\right) \rho(t, k, m) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, m\right) \rho(t, n, k)\right].
$$
\n(42)

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

$$
\sum_{n,m} (1+|n|)^{2+2\delta} (1+|m|)^{2+2\delta} |V(n,m)|^2 < \infty \,, \tag{43}
$$

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 (43) means that we consider a situation where the relevant energy levels of the atom are "far from the continuous spectrum".

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

$$
\mu < 1/2. \tag{44}
$$

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 (21) 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

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

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

$$
\forall \alpha = (\alpha_1, ..., \alpha_r) \in \mathbb{Z}^r, \quad \forall (k, n) \in \mathbb{N}^2 ,
$$

$$
|\alpha \cdot \omega + \omega(k, n)| \geq \frac{C}{(1 + |\alpha|)^{r - 1 + \delta} (1 + |n|)^{1 + \delta} (1 + |k|)^{1 + \delta}},
$$

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

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

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

The extension to other types of waves is discussed further.

Remark. Given once and for all a fixed $\delta > 0$, 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 $\alpha \cdot \omega + \omega(k,n) \neq 0$, 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 (42). 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 lemma.

Lemma 7. Let us define the time dependent transition rate:

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

where $\Omega(k,n) = -(i\omega(k,n) + \varepsilon^{\mu}\gamma(k,n))$. Let also $\rho_d^{(8)}$ $\int_{d}^{(8)}$ be solution to

$$
\partial_t \rho_d^{(8)}(t,n) = \sum_k \Psi_\varepsilon \left(\frac{t}{\varepsilon^2}, k,n\right) \left[\rho_d^{(8)}(t,k) - \rho_d^{(8)}(t,n)\right],\tag{45}
$$

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

$$
\|\rho_{\rm d} - \rho_{\rm d}^{(8)}\|_{L^\infty([0,T];l^1)} \le C\varepsilon^{1-2\mu}.\tag{46}
$$

Remark. Lemma 7 extends immediately to the more general case when ϕ is a bounded function on R.

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

To begin with, we compute the coherences in terms of the populations, at the lowest order in ε . Therefore we write, as in Eq. (11),

$$
\rho_{od}(t, n, m) = i\varepsilon \int_0^{t/\varepsilon^2} ds \exp(\Omega(n, m)s) \mathcal{V}\left(\frac{t}{\varepsilon^2} - s, n, m\right) \left[\rho_d(t - \varepsilon^2 s, m) - \rho_d(t - \varepsilon^2 s, n)\right]
$$

+ $i\varepsilon \int_0^{t/\varepsilon^2} ds \exp(\Omega(n, m)s) \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2} - s, n, k\right) \rho_{od}(t - \varepsilon^2 s, k, m) - \mathcal{V}\left(\frac{t}{\varepsilon^2} - s, k, m\right) \rho_{od}(t - \varepsilon^2 s, n, k)\right]$
=: $i\varepsilon \left(A_\varepsilon \rho_d\right)(t, n, m) + i\varepsilon \left(\widetilde{A}_\varepsilon \rho_{od}\right)(t, n, m),$

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

$$
\|\rho_{\text{od}} - i\varepsilon (A_{\varepsilon}\rho_{\text{d}})\|_{L^{\infty}([0,T],l^1)} = \varepsilon \|\widetilde{A}_{\varepsilon}\rho_{\text{od}}\|_{L^{\infty}([0,T],l^1)} \tag{47}
$$

$$
\leq \varepsilon \left\|\widetilde{A}_{\varepsilon}\left[\rho_{\text{od}} - i\varepsilon (A_{\varepsilon}\rho_{\text{d}})\right]\right\|_{L^{\infty}([0,T],l^1)} + \varepsilon^2 \|\widetilde{A}_{\varepsilon}A_{\varepsilon}\rho_{\text{d}}\|_{L^{\infty}([0,T],l^1)}.
$$

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

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

Thus, we obtain in (47), for ε small enough,

$$
\|\rho_{\text{od}} - i\varepsilon (A_{\varepsilon}\rho_{\text{d}})\|_{L^{\infty}([0,T],l^1)} \leq C\varepsilon^2 \|(\widetilde{A}_{\varepsilon}A_{\varepsilon}\rho_{\text{d}})\|_{L^{\infty}([0,T],l^1)} \leq C\varepsilon^{2-2\mu} \|\rho_{\text{d}}\|_{L^{\infty}([0,T],l^1)} \leq C\varepsilon^{2-2\mu}.
$$

Thereby, Eq. (8) for the populations reads

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

$$
= - \sum_k \left[\mathcal{V}\left(\frac{t}{\varepsilon^2}, n, k \right) (A_\varepsilon \rho_d)(t, k, n) - \mathcal{V}\left(\frac{t}{\varepsilon^2}, k, n \right) (A_\varepsilon \rho_d)(t, n, k) \right]
$$
(48)

$$
+ O_{L^\infty([0,T];l^1)}(\varepsilon^{1-2\mu}).
$$

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 on ε . Making Eq. (48) explicit, we obtain the equation:

$$
\partial_t \rho_d(t,n) = \sum_k \int_0^{t/\varepsilon^2} ds \left[\rho_d(t-\varepsilon^2 s, k) - \rho_d(t-\varepsilon^2 s, n) \right] \times
$$

$$
\times 2 \text{Re} \left(\exp \left(\Omega(k,n)s \right) \mathcal{V} \left(\frac{t}{\varepsilon^2}, n, k \right) \mathcal{V} \left(\frac{t}{\varepsilon^2} - s, k, n \right) \right) \qquad (49)
$$

+
$$
O_{L^\infty([0,T];l^1)}(\varepsilon^{1-2\mu}).
$$

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 (49) by a delay-free equation. To this aim, we first estimate thanks to (49):

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

On this account, substituting the delayed terms $\rho_d(t - \varepsilon^2 s)$ by their asymptotic values $\rho_d(t)$ in the integral term in (49), we introduce an error in the $L^{\infty}([0,T], l^1)$ norm, which can be estimated by

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

This estimate together with (49) yields

$$
\partial_t \rho_d(t,n) = O_{L^{\infty}([0,T],l^1)}(\varepsilon^{1-2\mu}) + O_{L^{\infty}([0,T],l^1)}(\varepsilon^{2-3\mu})
$$

$$
+ 2\text{Re} \sum_k \left[\rho_d(t,k) - \rho_d(t,n) \right] \Psi_{\varepsilon} \left(\frac{t}{\varepsilon^2},k,n \right).
$$

Gronwall lemma ends the proof.

3.3 Asymptotic analysis of the equation for populations

As in the case of uniform relaxations, Lemma 7 reduces the problem to the asymptotic analysis of (45). 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:

$$
\Psi_{\varepsilon}\left(\frac{t}{\varepsilon^{2}},k,n\right) = 2|V(n,k)|^{2}\text{Re}\sum_{\alpha,\beta\in\mathbb{Z}^{r}}\phi_{\alpha}\phi_{\beta}\exp\left(i(\alpha+\beta)\cdot\omega\frac{t}{\varepsilon^{2}}\right) \times \frac{1-\exp\left([-\varepsilon^{\mu}\gamma(k,n)-i(\omega(k,n)+\beta\cdot\omega)]t/\varepsilon^{2}\right)}{\left[\varepsilon^{\mu}\gamma(k,n)+i(\omega(k,n)+\beta\cdot\omega)\right]}.
$$
\n(50)

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 rôle in the analysis. In the sequel we show the following theorem.

Theorem 8. Let us define the transition rate (dominant term)

$$
\Psi^{\text{dom}}\left(k,n\right) := 2\frac{|V(n,k)|^2}{\gamma(k,n)} \sum_{\beta \in \mathbb{Z}^r / \omega(k,n)+\beta \cdot \omega = 0} |\phi_{\beta}|^2. \tag{51}
$$

Let also $\rho_{\rm d}^{(9)}$ $\frac{d^{(9)}}{d}$ be solution to

$$
\partial_t \rho_d^{(9)}(t,n) = \frac{1}{\varepsilon^{\mu}} \sum_k \Psi^{\text{dom}}(k,n) \left[\rho_d^{(9)}(t,k) - \rho_d^{(9)}(t,n) \right],\tag{52}
$$

with initial data $\rho_d^{(9)}$ $d_d^{(9)}(0,n) = \rho_d(0,n)$. We assume that $\mu < 1/2$. Then for all $T > 0$, there exists $C > 0$ such that the following error estimate holds:

$$
\|\rho_{\rm d} - \rho_{\rm d}^{(9)}\|_{L^{\infty}([0,T], l^2)} \le C\varepsilon^{\mu}.
$$

 \Box

Remark. Theorem 8 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 $\partial_t \rho(t,n) =$ $\sum_k \Psi^{\text{dom}}(k,n)[\rho(t,k)-\rho(t,n)]$, 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 $\Psi^{\text{dom}} \equiv 0$ 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 8 is the following.

Lemma 9. 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 $(B^{\text{dom}}u)(n) \in l^2$ according the formula:

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

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

Proof. The proof of Lemma 9 is obvious, and relies on the positiveness property $\Psi^{\text{dom}}(k, n) \geq 0$, and the symmetry $\Psi^{\text{dom}}(k, n) = \Psi^{\text{dom}}(n, k)$. \Box

Remark. 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 9. Because this property proves to be crucial in the sequel (and the asymptotic result in Theorem 8 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. We now prove Theorem 8. To this aim, we estimate the difference $\rho_d^{(8)} - \rho_d^{(9)}$ $\int_{d}^{(9)}$. The equation governing $\rho_d^{(8)}$ $\frac{1}{d}$ reads

$$
\partial_t \rho_{\rm d}^{(8)}(t,n) = \sum_k \Psi_{\varepsilon} \left(\frac{t}{\varepsilon^2}, k, n \right) \left[\rho_{\rm d}^{(8)}(t,k) - \rho_{\rm d}^{(8)}(t,n) \right],
$$

where the transition rate Ψ_{ε} is given by (50).

First step: splitting of Ψ_{ε} . We split Ψ_{ε} into a dominant and two residual contributions, as follows:

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

where Ψ^{dom} is defined by (51). From (50), the two residual contributions are defined as

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

and, respectively,

$$
\Psi^{\text{res}}\left(\frac{t}{\varepsilon^{2}},k,n\right) := 2|V(n,k)|^{2} \times \times \left(-\varepsilon^{-\mu} \sum_{\substack{\beta \in \mathbb{Z}^{r}, \\ \beta \in \mathbb{Z}^{r}, \\ \omega(k,n)+\beta \cdot \omega = 0}} \frac{|\phi_{\beta}|^{2}}{\gamma(k,n)} \exp(-\varepsilon^{\mu}\gamma(k,n) \frac{t}{\varepsilon^{2}}) - \text{Re} \sum_{\substack{\omega(k,n)+\beta \cdot \omega = 0 \\ \omega(k,n)+\beta \cdot \omega \neq 0}} \frac{|\phi_{\beta}|^{2}}{\varepsilon^{\mu}\gamma(k,n) + i(\omega(k,n)+\beta \cdot \omega)} \times \exp\left(-\varepsilon^{\mu}\gamma(k,n) - i(\omega(k,n)+\beta \cdot \omega)\right) \frac{t}{\varepsilon^{2}}) + \varepsilon^{-\mu} \text{Re} \sum_{\substack{\alpha \cdot \beta \cdot \beta \cdot \omega = 0 \\ \omega(k,n)+\beta \cdot \omega = 0}} \frac{\phi_{\alpha}\phi_{\beta}}{\gamma(k,n)} \exp(-i(\alpha+\beta)\cdot \omega \frac{t}{\varepsilon^{2}}) \left[1 - \exp(-\varepsilon^{\mu}\gamma(k,n) \frac{t}{\varepsilon^{2}})\right] + \text{Re} \sum_{\substack{\alpha \cdot \beta \cdot \beta \cdot \omega \neq 0 \\ \omega(k,n)+\beta \cdot \omega \neq 0}} \frac{\phi_{\alpha}\phi_{\beta}}{\left[\varepsilon^{\mu}\gamma(k,n) + i(\omega(k,n)+\beta \cdot \omega)\right]} \exp(i(\alpha+\beta)\cdot \omega \frac{t}{\varepsilon^{2}}) \times \times \left[1 - \exp(-\varepsilon^{\mu}\gamma(k,n) - i(\omega(k,n)+\beta \cdot \omega)\right] \frac{t}{\varepsilon^{2}})]\right).
$$

Now, the point in the sequel is that the first residual contribution Ψ^{res} is small (of order ε^{μ}) thanks to the real part, while the second residual contribution $\tilde{\Psi}^{\text{res}}$ 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:

$$
\sum_{k,n} |\Psi^{\text{dom}}(k,n)| \le C, \quad \sum_{k,n} |\Psi^{\text{res}}(t/\varepsilon^2, k,n)| \le C\varepsilon^{-\mu}, \quad \sum_{k,n} |\widetilde{\Psi}^{\text{res}}(k,n)| \le C \varepsilon^{\mu}, \tag{53}
$$

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 (53) by writing

$$
\sum_{k,n} |\widetilde{\Psi}^{\text{res}}(k,n)| \leq C \varepsilon^{\mu} \sum_{\substack{k,n,\beta, \\ \omega(k,n)+\beta \cdot \omega \neq 0}} \frac{|\phi_{\beta}|^{2} |V(k,n)|^{2}}{|\omega(k,n)+\beta \cdot \omega|^{2}}
$$

$$
\leq C \varepsilon^{\mu} \sum_{k,n,\beta} (1+|\beta|)^{2(r-1+\delta)} (1+|n|)^{2(1+\delta)} (1+|k|)^{2(1+\delta)} |\phi_{\beta}|^{2} |V(k,n)|^{2}
$$

$$
\leq C \varepsilon^{\mu}.
$$

The Diophantine estimate on ω (Hypothesis 4) is used in the second inequality, while the strong decay assumption (43) is used in the last estimate.

Third step: rewriting the equations. With the notations introduced above, Eq. (45) for $\rho_d^{(8)}$ d becomes

$$
\partial_t \rho_d^{(8)}(t,n) = \varepsilon^{-\mu} \sum_k \Psi^{\text{dom}}(k,n) [\rho_d^{(8)}(t,k) - \rho_d^{(8)}(t,n)] + \sum_k \Psi^{\text{res}} \left(\frac{t}{\varepsilon^2}, k,n \right) [\rho_d^{(8)}(t,k) - \rho_d^{(8)}(t,n)] + \sum_k \widetilde{\Psi}^{\text{res}}(k,n) [\rho_d^{(8)}(t,k) - \rho_d^{(8)}(t,n)],
$$

and this equation should be compared with

$$
\partial_t \rho_{\rm d}^{(9)}(t,n) = \varepsilon^{-\mu} \sum_k \Psi^{\rm dom}(k,n) [\rho_{\rm d}^{(9)}(t,k) - \rho_{\rm d}^{(9)}(t,n)].
$$

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

$$
(B^{\text{res}}(t/\varepsilon^2)u)(n) := \sum_{k} \Psi^{\text{res}}\left(\frac{t}{\varepsilon^2}, k, n\right) [u(k) - u(n)],
$$

$$
(\widetilde{B}^{\text{res}}u)(n) := \sum_{k} \widetilde{\Psi}^{\text{res}}(k, n)[u(k) - u(n)].
$$

We know from estimates (53) that:

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

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

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

With these notations, and if functions $\rho_d^{(8)}$ $\ell_d^{(8)}$ and $\rho_d^{(9)}$ $d_d^{(9)}$ are considered as functions of the time t with values in l^2 , Eqs (45) and (52) governing $\rho_d^{(8)}$ $\ell_d^{(8)}$ and $\rho_d^{(9)}$ $\int_{d}^{(9)}$ respectively read

$$
\partial_t \rho_d^{(8)}(t) = \varepsilon^{-\mu} B^{\text{dom}} \rho_d^{(8)}(t) + B^{\text{res}} \left(\frac{t}{\varepsilon^2}\right) \rho_d^{(8)}(t) + \widetilde{B}^{\text{res}} \rho_d^{(8)}(t),
$$

$$
\partial_t \rho_d^{(9)}(t) = \varepsilon^{-\mu} B^{\text{dom}} \rho_d^{(9)}(t).
$$

From this, in order to estimate the difference

$$
\Delta(t) := \rho_{\rm d}^{(8)}(t) - \rho_{\rm d}^{(9)}(t),
$$

we write

$$
\partial_t \Delta(t) = \varepsilon^{-\mu} (B^{\text{dom}} \Delta)(t) + B^{\text{res}} \left(\frac{t}{\varepsilon^2}\right) \rho_{\text{d}}^{(8)}(t) + \widetilde{B}^{\text{res}} \left(\frac{t}{\varepsilon^2}\right) \rho_{\text{d}}^{(8)}(t),
$$

and we solve directly

$$
\Delta(t) = \int_0^t ds \exp([t - s] \varepsilon^{-\mu} B^{\text{dom}}) B^{\text{res}}\left(\frac{s}{\varepsilon^2}\right) \rho_d^{(8)}(s)
$$

$$
+ \int_0^t ds \exp([t - s] \varepsilon^{-\mu} B^{\text{dom}}) \widetilde{B}^{\text{res}} \rho_d^{(8)}(s)
$$

=:
$$
\Delta^{(1)}(t) + \Delta^{(2)}(t).
$$
 (54)

We now estimate separately each term on the right hand-side of (54).

Fourth step: estimating the first term in (54) . To take advantage of the time oscillations of operator $B^{\text{res}}(t/\varepsilon^2)$, and to display clearly that the right hand-side of (54) tends to 0 with ε , we carry out a natural integration by parts in the integral with respect to s and we obtain

$$
\Delta^{(1)}(t) = \varepsilon^2 \left(\int_0^t ds \ B^{\text{res}}\left(\frac{s}{\varepsilon^2}\right) \right) \rho_d^{(8)}(t)
$$

+ $\varepsilon^{2-\mu} \int_0^t ds \ \exp([t-s] \varepsilon^{-\mu} B^{\text{dom}}) \ B^{\text{dom}}\left(\int_0^{s/\varepsilon^2} du \ B^{\text{res}}(u)\right) \rho_d^{(8)}(s)$
- $\varepsilon^2 \int_0^t ds \ \exp([t-s] \varepsilon^{-\mu} B^{\text{dom}}) \left(\int_0^{s/\varepsilon^2} du \ B^{\text{res}}(u)\right) \left(\varepsilon^{-\mu} B^{\text{dom}} + B^{\text{res}}\left(\frac{s}{\varepsilon^2}\right)\right) \rho_d^{(8)}(s).$

Consequently, taking advantage of the bounds (53), as well as Lemma 9 (non-positiveness of B^{dom} , we obtain the estimate

$$
\|\Delta^{(1)}\|_{L^{\infty}([0,T],l^2)} \leq C\varepsilon^{2-\mu} \sup_{0\leq t\leq T} \Big\|\int_0^{t/\varepsilon^2} ds\ B^{\text{res}}(s) \Big\|_{\mathcal{L}(l^2)} \ \|\rho_{\text{d}}^{(8)}\|_{L^{\infty}([0,T],l^2)}.
$$

Besides we have

$$
\|\rho_{\mathbf{d}}^{(8)}\|_{L^{\infty}([0,T],l^2)} \leq \|\rho_{\mathbf{d}}^{(8)}\|_{L^{\infty}([0,T],l^1)}
$$

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

thanks to (46), and the fact that ρ_d is bounded. It follows that

$$
\|\Delta^{(1)}\|_{L^{\infty}([0,T],l^2)} \le C\varepsilon^{2-\mu} \sup_{0\le t\le T} \left\| \int_0^{t/\varepsilon^2} ds \, B^{\text{res}}(s) \right\|_{\mathcal{L}(l^2)}.
$$
 (55)

There remains to estimate the supremum occurring in (55).

To this aim, we proceed as in the proof of Theorem 4 (see (29) 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 (43)), we deduce the estimate

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

In short, we showed the estimate

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

and this last quantity tends to 0 with ε .

Fifth step: estimating the second term in (54). The analysis of $\Delta^{(2)}$ now uses (53) in a direct way, in that we write

$$
\|\Delta^{(2)}(t)\|_{L^{\infty}([0,T],l^2)} \leq C T \sup_{s \in [0,T]} \|\widetilde{B}^{\text{res}} \rho_{\text{d}}^{(8)}(s)\|_{l^2} \leq C \|\widetilde{B}^{\text{res}}\|_{\mathcal{L}(l^2)} \sup_{s \in [0,T]} \rho_{\text{d}}^{(8)}(s)\|_{l^2} \leq C \|\widetilde{\Psi}^{\text{res}}\|_{l^1} \leq C \varepsilon^{\mu},
$$

where we made use of the non-positiveness of B^{dom} , the boundedness of $\rho_{\text{d}}^{(8)}$ $\int_{d}^{(8)}$ (see the previous step), and the estimate (53) proved before.

Conclusion. Theorem 8 is now proved since $\mu < 2 - 2\mu$. \Box

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