Markovian approximation for Pauli Fierz operators

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Abstract
The purpose of this article is to derive a Markovian approximation of the reduced time dynamics of observables for the Pauli-Fierz Hamiltonian with a precise control of the error terms. In that aim, we define a Lindblad operator associated to the corresponding quantum master equation. In a particular case, this allows to study the transition probability matrix.

Keywords: Pauli-Fierz Hamiltonian, Markovian approximation, Lindblad operator, quantum master equation, transition probabilities, transition rate matrix, quantum electrodynamics, QED, spin dynamics.

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1 Introduction.
Pauli-Fierz operators are used to describe the time evolution of charged particles with spins interacting with electric potentials and the quantized electromagnetic field (photons) and possibly with a non quantized external magnetic field [3, 17, 8]. In the Markovian approximation, the interaction between particles and the quantized field is usually assumed to be small and in Pauli-Fierz Hamiltonians, the terms for these interactions are multiplied by a parameter \( g \) (coupling constant). We then suppose in the following that \( g \) is small in the Pauli-Fierz Hamiltonian considered here and denoted by \( H(g) \).

The purpose of this work is to give an approximation of a Markov type for the time evolution of observables for small \( g \). In that aim, we shall omit some terms in the Hamiltonian that we expect would not modify our results below. The precise definition of the simplified Pauli-Fierz Hamiltonian \( H(g) \) is given in Section 2.

Let \( \mathcal{H}_{\text{tot}} \) be the Hilbert space of the model. Assuming that the Hamiltonian \( H(g) \) is acting in \( \mathcal{H}_{\text{tot}} \), we recall that the evolution \( X(t, g) \) of an observable \( X \) in \( \mathcal{H}_{\text{tot}} \) is usually given for any time \( t \) by:

\[
X(t, g) = e^{itH(g)} X e^{-itH(g)}.
\]

In the system studied here we consider moving spinless charged particles together with a spin-\( \frac{1}{2} \) fixed particle in the quantized electromagnetic field, with an external electric potential and an external constant magnetic field. The Hilbert space \( \mathcal{H}_{\text{tot}} \) is the completed tensor product of
the three Hilbert spaces $\mathcal{H}_{ph}$, $\mathcal{H}_{el}$ and $\mathcal{H}_{sp}$ respectively corresponding to the quantized electromagnetic field, the moving charged particles and the spin fixed particle. Concerning the observables, we assume that they act only on the moving charged particle and the spin fixed particle spaces and not on the photon space, that is, the observables are written as $I \otimes X$ with $I$ the identity in $\mathcal{H}_{ph}$ and $X$ an operator in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$. The photon vacuum state is denoted by $\Psi_0 \in \mathcal{H}_{ph}$.

In this article we are concerned with the operator denoted $\sigma_0 S(t, g)X$ mapping from $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$ into itself and defined by:

$$\langle \sigma_0 S(t, g)X f, g \rangle_{\mathcal{H}_{el} \otimes \mathcal{H}_{sp}} = \langle (I \otimes X)e^{-itH(g)}(\Psi_0 \otimes f), e^{-itH(g)}(\Psi_0 \otimes g) \rangle_{\mathcal{H}_{tot}},$$

for all $f$ and $g$ in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$. Thus, we consider time evolutions with initial states in the vacuum of photon. Note that in the case $g = 0$, one has:

$$\sigma_0 S(t, 0)X = e^{it(H_{el} + H_{sp})}X e^{-it(H_{el} + H_{sp})}$$

where $H_{el}$ and $H_{sp}$ respectively are the Schrödinger and the free spin Hamiltonian (see Section 2.1). That is,

$$\frac{d}{dt}\sigma_0 S(t, 0)X = i\sigma_0 S(t, 0)[(H_{el} + H_{sp}), X].$$

This means that, for $g = 0$, the observable evolution follows the Heisenberg equation (for the Schrödinger and the free spin Hamiltonians).

Our goal is to prove that for $g$ small enough, $\sigma_0 S(t, g)X$ follows a differential equation (master equation) with an explicit term (Lindblad operator) and two negligible terms. The first error term is estimated with $\mathcal{O}(g^4)$ and the second with $\mathcal{O}(g^2)$ but the latter can be negligible for large $t$. The simplified equation obtained when omitting these two error terms is the quantum master equation and constitutes the Markovian approximation. Let us be more specific concerning that result. We define a mapping $L(g)$ (see Proposition 2.3 below) giving for any suitable operator $X$ in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$ an operator $L(g)X$ also in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$ satisfying for all $t > 0$:

$$\frac{d}{dt}\sigma_0 S(t, g)X = \sigma_0 S(t, g)L(g)X + R_0(t, g)X + R(t, g)X.$$

The mapping $L(g)$ is called Lindblad operator (see [7, 11, 13, 19, 25, 26, 27, 30]). Concerning the error terms in this approximation, we prove for small $g$ the following estimates for all positive times:

$$\|R_0(t, g)X\| \leq Cg^2 \frac{g^2}{1 + t} \|X\|, \quad \|R(t, g)X\| \leq Cg^3 \ln(1 + t) \|X\|$$

and if the ultraviolet cutoff vanishes at the origin:

$$\|R_0(t, g)X\| \leq Cg^2 \frac{g^2}{1 + t^2} \|X\|, \quad \|R(t, g)X\| \leq Cg^3 \|X\|$$

where the above norm $\|X\|$ will be specified.

This result is precisely stated in Theorem 2.4 below and is the main result of the paper. In Section 2.2 we shall see the precise hypotheses and the definition of the chosen norms in order to get the control of the error terms.

There exists other types of corrections of the Heisenberg equation for the Schrödinger operator, for example, the Breit operator (see [6, 5]). There are other reduced dynamics, see e.g. [22, 31], which are different to the one that we consider. Return to equilibrium for Pauli-Fierz are
studied, e.g., in [4, 12, 14, 15, 16]. For Markovian approximation and weak coupling limit approximation, see [9, 31] and also [1, 30], and [32, 33, 9, 10, 30, 31] for the control of the error. Let us mention that the main result Theorem 2.4 is proved in view of possible applications in NMR (Nuclear Magnetic Resonance) for a system of several interacting electrons together with a fixed spin-$\frac{1}{2}$ particle subject to a constant external (non quantized) magnetic field. See [2] for the case of several fixed spins but without moving electrons and see [27, 23] in the positive temperature case.

In Section 5, we make explicit the operator $L(g)$ in a particular case. Namely, we consider the case of single electron without constant magnetic field and without the fixed spin particle. We make the hypotheses that the Schrödinger operator is globally elliptic in the sense of [20]. In that case, there exists an Hilbertian basis $(u_j)$ of $L^2(\mathbb{R}^3)$ with eigenfunctions $u_j$ of the Schrödinger operator $H_{el}$ (see Section 2.1 and Section 5). Denoting by $\pi_{u_j}$ the orthogonal projection on the $u_j$, we make explicit for all $m$ and $j$:

$$M_{mj} = g^{-2}\langle (L(g)\pi_{u_j})u_m, u_m \rangle.$$

The form of this matrix suggests that it is the infinitesimal generator of a Markov semi-group. One can address the question of the existence of this semi-group and whether it defines a good approximation for small $g$ of the transition probability matrix, that is, the probabilities for an electron initially in an excited state $u_m$ without photon to be at time $t$ in the state $u_j$. Different facts make this realistic. First, the matrix is triangular which means that up to this approximation, the probability for the electron to be at time $t > 0$ at an energy level higher than at the initial time is zero. Second, the diagonal elements of this matrix are related to the imaginary part of the resonances studied by Bach, Fröhlich and Sigal [3].

2 Statement of the results.

2.1 Description of the model.

The Hilbert space of the model is the completed tensor product of the three Hilbert spaces of the elements constituting the system:

$$\mathcal{H}_{tot} = \mathcal{H}_{ph} \otimes \mathcal{H}_{el} \otimes \mathcal{H}_{sp}$$

and the Hamiltonian of the model is the sum of the Hamiltonians $H_{ph}$, $H_{el}$ and $H_{sp}$ of the three elements when they are not interacting, together with the interaction Hamiltonian $H_{int}$.

Photon Hilbert space and Hamiltonian.

The single photon Hilbert space $\mathcal{H}_{ph}^1$ is:

$$\mathcal{H}_{ph}^1 = \{ f \in L^2(\mathbb{R}^3, \mathbb{R}^3), \sum_{j=1}^{3} k_j f_j(k) = 0, k \in \mathbb{R}^3 \}.$$
we use the standard operator $d\Gamma(T)$ in the Fock space (see [29]). In particular, if $M_\omega$ is the multiplication in $\mathcal{H}^1_{ph}$ by $\omega(k) = |k|$, the operator $H_{ph} = d\Gamma(M_\omega)$ is the usual free photon energy Hamiltonian.

**Electron Hilbert space and Hamiltonian.**

The electron Hilbert space is $\mathcal{H}_{el} = L^2(\mathbb{R}^{3Q})$ where $Q$ is the number of electrons. We assume that there is a constant external magnetic field $B_{ext} = (0,0,B)$ where $B \in \mathbb{R}$. The corresponding vector potential is denoted by:

$$A^{ext}(x) = \left(-B \frac{x_2}{2}, B \frac{x_1}{2}, 0\right).$$

Letting $x = (x^{(1)}, \ldots, x^{(Q)})$ be the variable in $\mathbb{R}^{3Q}$, we set for $\alpha = 1, \ldots, Q$ and $j = 1, 2, 3$:

$$D_j^{(\alpha)} = \frac{1}{i} \frac{\partial}{\partial x_j^{(\alpha)}}, \quad \nabla_j^{(\alpha)} = D_j^{(\alpha)} - A_j^{ext}(x^{(\alpha)}).$$

The Hamiltonian of the system of electrons is the $Q$-body Schrödinger operator with constant magnetic field ($\mathcal{H}_{el}, D(\mathcal{H}_{el})$):

$$H_{el} = \sum_{\alpha=1}^{Q} \sum_{j=1}^{3} (\nabla_j^{(\alpha)})^2 + V(x) \quad (2.1)$$

where $V$ is the electric potential supposed to be a real valued function on $\mathbb{R}^{3Q}$ with polynomial growth at infinity and identified with the multiplication operator by this function. This Hamiltonian operator is initially defined on $S(\mathbb{R}^{3Q})$. One can also omit the polynomial growth on the electric potential but replace in the following the Schwartz space by the set of $C^\infty$ functions with compact support.

**Spin Hilbert space and Hamiltonian.**

The spin Hilbert space of the fixed particle is $\mathcal{H}_{sp} = \mathbb{C}^2$. We denote by $\sigma_j$ the standard Pauli matrices ($1 \leq j \leq 3$). In the case of a constant magnetic field $(0,0,B)$, the energy of the spin-$\frac{1}{2}$ particle corresponds to the Hamiltonian:

$$H_{sp} = B\sigma_3. \quad (2.2)$$

**Interaction Hamiltonian.**

The interaction between electrons and the spin fixed particle with the quantized electromagnetic field is expressed with two types of operators. One type corresponds to the three components of the quantized electromagnetic vector potential and is used to define the interaction between the electron motions and the field. The other type, corresponding to the three components of the quantized magnetic field at the position $x_0$ of the spin-$\frac{1}{2}$ fixed particle, is used to define the interaction between the field and the spin particle. These operators are the image by the Segal field $\Phi_S$ of the elements $A_{jx}$ and $B_{jx}$ of $\mathcal{H}_1$ ($x \in \mathbb{R}^3, 1 \leq j \leq 3$) that we recall here the definitions:

$$A_{jx}(k) = \frac{\varphi(|k|)}{|k|^{1/2}} e^{-ik \cdot x} \left( e_j - (e_j \cdot k) \frac{k}{|k|^2} \right), \quad k \in \mathbb{R}^3 \setminus \{0\}, \quad (2.3)$$

$$B_{jx}(k) = \frac{i\varphi(|k|)|k|^{1/2}}{(2\pi)^{3/2}} e^{-ik \cdot x} \frac{k \times e_j}{|k|}, \quad k \in \mathbb{R}^3 \setminus \{0\}. \quad (2.4)$$
In the above equalities, \((e_1, e_2, e_3)\) stands for the canonical basis of \(\mathbb{R}^3\). The function \(\varphi\) is a smooth ultraviolet cutoff always supposed in the Schwartz space \(\mathcal{S}(\mathbb{R})\). Note that each \(A_{jx}\) and \(B_{jx}\) \((j\) and \(x\) fixed\) maps \(\mathbb{R}^3\) into \(\mathbb{R}^3\) and satisfy the equalities:

\[ k \cdot A_{jx}(k) = k \cdot B_{jx}(k) = 0, \quad k \in \mathbb{R}^3 \setminus \{0\}. \]

This means that the \(A_{jx}\) and \(B_{jx}\) are elements of the single photon Hilbert space \(\mathcal{H}_{\text{ph}}^1\) according to the decreasing property of \(\varphi\). The operators corresponding to the three components of the vector potential and of the magnetic field at point \(x\) respectively are \(\Phi_S(A_{jx})\) and \(\Phi_S(B_{jx})\).

In order to define and use for upcoming computations the operator \(H_{\text{int}}\), we temporarily identify \(\mathcal{H}_{\text{tot}}\) and \(L^2(\mathbb{R}^{3Q}, \mathcal{H}_{\text{ph}} \otimes \mathcal{H}_{\text{sp}})\) and set, for all \(f \in \mathcal{S}(\mathbb{R}^{3Q}, \mathcal{H}_{\text{ph}} \otimes \mathcal{H}_{\text{sp}})\) and any \(x \in \mathbb{R}^{3Q}\):

\[
(H_{\text{int}}f)(x) = \sum_{\alpha=1}^Q \sum_{j=1}^3 \left( \Phi_S(A_{j,x^{(\alpha)}}) \otimes I \right) \nabla_j^{(\alpha)} f(x) + \sum_{j=1}^3 \left( \Phi_S(B_{j,x^{(0)}}) \otimes \sigma_j \right) f(x).
\]

The tensor product in the above equality refers to \(\mathcal{H}_{\text{ph}} \otimes \mathcal{H}_{\text{sp}}\) and \(I\) denotes the identity in \(\mathcal{H}_{\text{sp}}\). It appears to be more convenient in the sequel to use integral expressions with creation operators \(a^*(k)\) and annihilation operators \(a(k)\) \((k \in \mathbb{R}^3)\) as in [3]. For any \(V \in \mathcal{H}_{\text{ph}}^1\):

\[
\Phi_S(V) = \frac{1}{\sqrt{2}} \int_{\mathbb{R}^3} \left( a^*(k)V(k) + a(k)\overline{V(k)} \right) dk.
\]

Then for each \(k \in \mathbb{R}^3\), we define an operator \(E(k)\) from \(\mathcal{H}_{\text{el}} \otimes \mathcal{H}_{\text{sp}}\) now identified with \(L^2(\mathbb{R}^{3Q}, \mathcal{H}_{\text{sp}})\) and taking values in \((\mathcal{H}_{\text{el}} \otimes \mathcal{H}_{\text{sp}})^3\) by:

\[
(E(k)f)(x) = \sum_{\alpha=1}^Q \sum_{j=1}^3 A_{j,x^{(\alpha)}}(k) \nabla_j^{(\alpha)} f(x) + \sum_{j=1}^3 B_{j,x^{(0)}}(k) \sigma_j f(x)
\]

for all \(f \in \mathcal{S}(\mathbb{R}^{3Q}, \mathcal{H}_{\text{sp}})\).

The operator \(E^*(k)\) denotes the formal adjoint of \(E(k)\). Let us emphasize that each \(E(k)\) takes values in \((\mathcal{H}_{\text{el}} \otimes \mathcal{H}_{\text{sp}})^3\) and verifies:

\[
k \cdot E(k) = 0, \quad k \in \mathbb{R}^3.
\]

Therefore, for each \(f \in \mathcal{S}(\mathbb{R}^{3Q}, \mathcal{H}_{\text{sp}})\) and for each \(x \in \mathbb{R}^{3Q}\), the function \(k \rightarrow E_x(k) = (E(k)f)(x)\) belongs to \(\mathcal{H}_{\text{ph}}^1 \otimes \mathcal{H}_{\text{sp}}\) where \(\mathcal{H}_{\text{ph}}^1\) is the single photon Hilbert space. Thus, extending the Segal field \(\Phi_S\) to the Hilbert space \(\mathcal{H}_{\text{ph}}^1 \otimes \mathcal{H}_{\text{sp}}\) as in [11], we have:

\[
(H_{\text{int}}f)(x) = \Phi_S(E_x).
\]

This operator is initially defined in \(\mathcal{H}_{\text{tot}}^{\text{reg}} = \mathcal{H}_{\text{ph}}^{\text{fin}} \otimes \mathcal{S}(\mathbb{R}^{3Q}) \otimes \mathcal{H}_{\text{sp}}\). This definition may be written in a somewhat formal way but useful in the following sections as:

\[
H_{\text{int}} = \frac{1}{\sqrt{2}} \int_{\mathbb{R}^3} \left( a(k) \otimes E^*(k) + a^*(k) \otimes E(k) \right) dk.
\]

The above tensor product corresponds to the tensor product of \(\mathcal{H}_{\text{ph}}\) with \(\mathcal{H}_{\text{el}} \otimes \mathcal{H}_{\text{sp}}\).

*Full Hamiltonian.*
The following operators are initially defined in $\mathcal{H}_{tot}^{reg}$:

$$H(0) = H_{ph} + H_{el} + H_{sp}, \quad (2.7)$$

$$H(g) = H_{ph} + H_{el} + H_{sp} + gH_{int} = H(0) + gH_{int}$$

**Domain and unitary group.**

The assumption on the Hamiltonian $H_{el}$ is hypothesis $(H)$ below. We use the following notations. For any $(\alpha, J)$ with $\alpha = (\alpha_1, \ldots, \alpha_m)$ and $J = (j_1, \ldots, j_m)$ where the $\alpha_\mu = 1, \ldots, Q$ and the $j_\mu = 1, 2, 3$, we set:

$$D_{\alpha, J} = \nabla_{j_1}^{[\alpha_1]} \cdots \nabla_{j_m}^{[\alpha_m]}$$

and set $|((\alpha, J)| = m$. We make the following hypothesis $(H)$.

**Hypothesis $(H)$.** The operator $H_{el}$ defined in $(2.7)$ is essentially self-adjoint and we denote by $H_{el}$ its self-adjoint extension. We assume that there exists $C > 0$ satisfying $H_{el} + C > 0$ and we denote by $W_m^el$ the domain of $(C + H_{el})^{m/2}$. For all integers $m$, $W_m^el$ is the set of all $u \in L^2(\mathbb{R}^{3Q})$ satisfying $D_{\alpha, J}u \in L^2(\mathbb{R}^{3Q})$ if $|((\alpha, J)| \leq m$ with the norm:

$$\|u\|^2_{W_m^el} = \|u\|^2 + \sum_{|((\alpha, J)| \leq m} \|D_{\alpha, J}u\|^2$$

(2.8)

where $\| \cdot \|$ is the $L^2(\mathbb{R}^{3Q})$ norm.

This hypothesis is fulfilled for some potentials $V$. In particular, we consider in Section 5 the case of globally elliptic Schrödinger operator (20) where this hypothesis is satisfied. Our main result could be applied with other potentials.

Since we consider $H_{el} + H_{sp}$ with $H_{sp}$ bounded we shall still denote by $W_m^{el}$ the spaces associated to $H_{el} + H_{sp}$ instead of $H_{el}$ in the aim to avoid additional notations such as $W_m^{el-sp}$. For example, $W_0^{el}$ now refers to $H_{el} \otimes H_{sp}$.

**Theorem 2.1.** Under hypothesis $(H)$, the operator $H(0)$ initially defined on $\mathcal{H}_{tot}^{reg}$ is essentially self-adjoint. Let $C > 0$ be such that $H(0) + C > 0$ and let $W_m^{tot}$ be the domain of $(H(0) + C)^{m/2}$. Then, for $g$ small enough, the operator $H(g)$ with domain $W_2^{tot}$ is self-adjoint and the domain of $(H(g) + C)^{m/2}$ is $W_m^{tot}$.

Theorem 2.1 is proved in Section 3. By this theorem, the operators $e^{itH(0)}$ and $e^{itH(g)}$ make sense for $g$ small enough. See also [21] for self-adjointness of Pauli-Fierz without the condition $g$ small.

### 2.2 Observable time evolutions.

**Full, free and reduced time evolutions of electron-spin observable.**

For every operator $X$ in $H_{el} \otimes H_{sp}$, the time full evolution of $X$ is defined by:

$$S(t, g)X = e^{itH(g)}(I_{ph} \otimes X)e^{-itH(g)}.$$  

For any operator $A$ in $\mathcal{H}_{tot}$, we denote by $\sigma_0 A$ the operator defined in $H_{el} \otimes H_{sp}$ by:

$$\langle \sigma_0 Af, g \rangle_{H_{el} \otimes H_{sp}} = \langle A(\Psi_0 \otimes f), (\Psi_0 \otimes g) \rangle_{\mathcal{H}_{tot}}, \quad (2.9)$$

for all $f$ and $g$ in $H_{el} \otimes H_{sp}$.

In particular, we have for $S(t, g)X$, $t > 0$:  


Definition 2.2. We call reduced time evolution the mapping $X \to \sigma_0 S(t, g) X$ from $H_{el} \otimes H_{sp}$ into $H_{el} \otimes H_{sp}$ defined for every $t > 0$ and any operator $X$ in $H_{el} \otimes H_{sp}$ by:

$$
\langle \sigma_0 S(t, g) X f, g \rangle_{H_{el} \otimes H_{sp}} = \langle (I_{ph} \otimes X) e^{-itH(g)} (\Psi_0 \otimes f), e^{-itH(g)} (\Psi_0 \otimes g) \rangle_{H_{tot}},
$$

for all $f$ and $g$ in $H_{el} \otimes H_{sp}$.

In rest of the paper, we often omit the Hilbert space as a subscript in $\| \cdot \|$ and $\langle \cdot, \cdot \rangle$ when it is $H_{tot}$, $H_{ph}$, $H_{el}$ or $H_{el} \otimes H_{sp}$. Similarly we omit the Hilbert subscript in the notation $I$ for the identity since there is no ambiguity. Also $\mathcal{L}(H_1, H_2)$ denotes the set of bounded linear maps from $H_1$ to $H_2$ for any Hilbert spaces $H_1$ and $H_2$.

The free evolution of an operator $A$ in $H_{tot}$ is defined by:

$$
A^{free}(t) = e^{itH(0)} A e^{-itH(0)}
$$

(2.10)

and for an operator $A$ in $H_{el} \otimes H_{sp}$, we set (using the same notation since no confusion will appear):

$$
A^{free}(t) = e^{it(H_{el}+H_{sp})} A e^{-it(H_{el}+H_{sp})}.
$$

In particular, with $E(k)$ the function used to give an expression of $H_{int}$ in (2.6), we set:

$$
E^{free}(k, t) = e^{it(H_{el}+H_{sp})} E(k) e^{-it(H_{el}+H_{sp})}
$$

(2.11)

and $E^{free, *}(k, t)$ is similarly defined.

Definition of the Lindblad operator.

The Lindblad operator is defined by the following Proposition proved in Section 4.

Let us recall that the operator $E(k)$ defined in (2.5) is an operator from $H_{el} \otimes H_{sp}$ to $(H_{el} \otimes H_{sp})^3$. Therefore, we can write $E(k) = (E_1(k), E_2(k), E_3(k))$. In this subsection, we agree in order to avoid running indices in the sums to write $E^*(k) \Delta E(k)$ instead of $\sum_j E^*_j(k) \Delta E_j(k)$ for any operator $A$ in all the integrals.

Proposition 2.3. Let $X$ be a bounded operator in $H_{el} \otimes H_{sp}$ and bounded from $W_{el}^1$ into itself. Then, for every $g$ small enough and all $t > 0$, the following operator is well defined from $W_{el}^2$ into $W_{el}^1 = H_{el} \otimes H_{sp}$:

$$
L(t, g) X = i [H_{el} + H_{sp}, X] + \frac{g^2}{2} \int_{\mathbb{R}^3 \times (0, t)} \left( e^{-i|k|} [E^*(k), X] E^{free}(k, -s) - e^{i|k|} E^{free}(k, -s)^* [E(k), X] \right) dk ds.
$$

(2.12)

Moreover, for small $g$, the limit as $t \to +\infty$ of $L(t, g) X$ exists in $\mathcal{L}(W_{el}^2, W_{el}^1)$ and this limit is denoted by $L(g) X$. We also have for some $C > 0$:

$$
\| L(t, g) X - L(g) X \|_{\mathcal{L}(W_{el}^2, W_{el}^1)} \leq \frac{C g^2}{1 + t} \| X \|_{\mathcal{L}(W_{el}^2, W_{el}^1)},
$$

for all $t > 0$. If the ultraviolet cutoff function $\varphi$ in (2.5), (2.4) vanishes at the origin then this inequality can be replaced by:

$$
\| L(t, g) X - L(g) X \|_{\mathcal{L}(W_{el}^2, W_{el}^1)} \leq \frac{C g^2}{1 + t^2} \| X \|_{\mathcal{L}(W_{el}^2, W_{el}^1)}.
$$

The operator $L(g) : X \to L(g) X$ is called Lindblad operator.

Main result.
We have:

\[ \frac{d}{dt} \sigma_0 S(t, g) X = \sigma_0 S(t, g) L(g) X + R_0(t, g) X + R(t, g) X \]  

(2.13)

where \( R_0(t, g) X \) is an operator from \( W^1_2 \) into \( W^{1/2}_0 \) and \( R(t) X \) is an operator from \( W^4_4 \) into \( W^1_4 \) satisfying:

\[ \| R_0(t, g) X \|_{L(W^1_0, W^{1/2}_0)} \leq C \frac{g^2}{1 + t} \| X \|_{L(W^1_0, W^{1/2}_0)} \]
\[ \| R(t, g) X \|_{L(W^4_4, W^{1/2}_0)} \leq C g^3 \ln(1 + t) \sup_{m \leq 4} \| X \|_{L(W^m_0, W^m_0)}. \]

If the ultraviolet cutoff \( \varphi \) in (2.3) (2.4) vanishes at the origin then these inequalities can be improved by:

\[ \| R_0(t, g) X \|_{L(W^2_2, W^{1/2}_0)} \leq C \frac{g^2}{1 + t^2} \| X \|_{L(W^2_4, W^{1/2}_0)} \]
\[ \| R(t, g) X \|_{L(W^4_4, W^{1/2}_0)} \leq C g^3 \sup_{m \leq 4} \| X \|_{L(W^m_0, W^m_0)}. \]

Theorem 2.4 is proved in Section 4.

3 Sobolev spaces. Proof of Theorem 2.1.

Theorem 2.1 follows from Proposition 3.2 and Proposition 3.6.

3.1 Electronic Hamiltonian.

We are first concerned with studying the action of the operator \( E(k) \) defined in (2.5) in the Sobolev spaces \( W^m_n \). Spherical coordinates for \( k \), namely \( k = \rho \omega \) (\( \rho > 0 \) and \( \omega \in S^2 \) the unit sphere centered at the origin) are used in that purpose.

Proposition 3.1. Under hypothesis (H) the operator \( E(k) \) defined in (2.5) for fixed \( k \in \mathbb{R}^3 \) maps \( W^m_n \) into \( (W^m_{n-1})^3 \). Moreover, for any \( \alpha \), all \( m \) and \( p \), there is \( C_{\text{amp}} > 0 \) such that:

\[ \| \partial_\rho^\alpha (\rho^{1/2} E(\rho \omega)) \|_{L(W^m_{n-1}((W^m_{n-1})^3))} \leq C_{\text{amp}}(1 + \rho)^{-p}. \]  

(3.1)

In particular, there exists \( C_{\text{mp}} \) satisfying:

\[ \| E(k) \|_{L(W^m_{n-1}((W^m_{n-1})^3))} \leq C_{\text{mp}}(1 + |k|)^{-p}. \]

The analogous estimates holds for \( E^*(k) \).

Proof. We use hypothesis (2.8) noticing according to the expressions (2.3) and (2.4) for the functions \( A_{jx} \) and \( B_{jx} \) that the commutator:

\[ [\nabla_j^{[\alpha]}, E(k)] = ik_j E(k). \]

Since the smooth ultraviolet cutoff \( \varphi \) in (2.3) (2.4) is supposed to belong to \( S(\mathbb{R}) \), we see that for each integer \( p \):

\[ \| \partial_\rho^\alpha (\rho^{1/2} E(\rho \omega)) \|_{L(W^m_{n-1}(W^{m-1})))} \leq C_{\text{amp}}(1 + \rho)^{-p}. \]

The proposition follows. \qed
3.2 Free Hamiltonian.

We first have the following property (\cite{28}).

**Proposition 3.2.** The operator $H(0)$ defined in $\mathcal{H}^{reg}_{tot}$ by (\ref{2.7}) is essentially self-adjoint. We also denote by $H(0)$ its self-adjoint extension. If $C > 0$ satisfies $H(0) + C > 0$ then $W^m_{tot}$ stands for the domain of $(C + H(0))^{m/2}$. For all integers $m$, we have:

$$
\|f\|^2_{W^m_{tot}} = \sum_{p+q\leq m} \|H^p_{ph}H^q_{el}f\|^2.
$$

(3.2)

We then deduce the Proposition below.

**Proposition 3.3.** We have the two following properties.

(i) If the operator $A$ maps $W^m_{el}$ into $W^p_{el}$ then $I \otimes A$ maps $W^m_{tot}$ into $W^p_{tot}$.

(ii) If the operator $B$ maps $W^m_{tot}$ into $W^p_{tot}$ then $\sigma_0(B)$ maps $W^m_{el}$ into $W^p_{el}$.

**Proof.** Let us prove the point (ii). We see that $(C + H_{el})^p \sigma_0(B) = \sigma_0((I \otimes (C + H_{el}))^p) B)$. Therefore, for each $f$ in $W^m_{el}$:

$$
\|\sigma_0(B)f\|_{W^p_{el}} \leq \|(I \otimes (C + H_{el}))^p) B(\Psi_0 \otimes f)\| \leq \|B(\Psi_0 \otimes f)\|_{W^p_{tot}}
$$

$$
\leq K\|\Psi_0 \otimes f\|_{W^m_{tot}} = K\|\Psi_0 \otimes H^m_{el}f\| = \|f\|_{W^m_{el}},
$$

for some $K > 0$. We have used above that $H_{ph}\Psi_0 = 0$.

\[\square\]

We shall also use the fact that $e^{itH(0)}$ is bounded from $W^m_{tot}$ into itself for all $m$.

3.3 Full Hamiltonian.

We now study the action of the operator $H_{int}$ in the Sobolev spaces defined in Section 3.2.

**Proposition 3.4.** The operator $H_{int}$ is bounded from $W^m_{tot}$ to $W^{m-2}_{tot}$.

For the proof, we need the following Lemma which is Lemma I.6 of [3].

**Lemma 3.5.** Let $k \rightarrow F(k)$ be a function on $\mathbb{R}^3$ taking values in $\mathcal{L}(\mathcal{H}_{el} \otimes \mathcal{H}_{sp}, (\mathcal{H}_{el} \otimes \mathcal{H}_{sp})^3)$ and satisfying $k \cdot F(k) = 0$ for all $k \in \mathbb{R}^3$. We define two operators $T_F$ and $U_F$ from $W^1_{tot}$ to $W^0_{tot}$ by:

$$
T_Ff = \int_{\mathbb{R}^3} (a(k) \otimes F^*(k))f dk,
$$

$$
U_Ff = \int_{\mathbb{R}^3} (a^*(k) \otimes F(k))f dk
$$

(with the same notation convention as for $E(k)$). Then:

$$
\|T_Ff\|^2 \leq \|(H^{1/2}_{ph} \otimes I)f\|^2 \int_{\mathbb{R}^3} \|F^*(k)\|^2 \frac{dk}{|k|},
$$

$$
\|U_Ff\|^2 \leq \|(H^{1/2}_{ph} \otimes I)f\|^2 \int_{\mathbb{R}^3} \|F(k)\|^2 \frac{dk}{|k|} + \|f\|^2 \int_{\mathbb{R}^3} \|F(k)\|^2 dk.
$$

The norms of $F(k)$ and $F^*(k)$ above are the norm of $\mathcal{L}(\mathcal{H}_{el} \otimes \mathcal{H}_{sp}, (\mathcal{H}_{el} \otimes \mathcal{H}_{sp})^3)$. 


Proof of Lemma 3.5. For the convenience of the reader we recall the proof in [3]. We have, for all $f$ and $g$:

\[
|\langle T_F f, g \rangle|^2 \leq \int_{\mathbb{R}^3} |k|\|(a(k) \otimes I)f\|^2\,dk \int_{\mathbb{R}^3} \|(I \otimes F^*(k))g\|^2\,\frac{dk}{|k|}.
\]

We know that:

\[
\int_{\mathbb{R}^3} |k|\|(a(k) \otimes I)f\|^2\,dk \leq \|(H_{ph}^{1/2} \otimes I)f\|^2.
\]

We also have:

\[
\|U_f\|^2 = \int_{\mathbb{R}^6} \langle (a(p)a^*(k) \otimes F(k))f, (I \otimes F(p)f)dkdp
\]

\[
= \int_{\mathbb{R}^3} \|(I \otimes F(k))f\|^2\,dk + \int_{\mathbb{R}^6} \langle (a^*(k)a(p) \otimes F(k))f, (I \otimes F(p)f)\rangle d\text{ke}
\]

\[
\leq \int_{\mathbb{R}^3} \|(I \otimes F(k))f\|^2\,dk + \int_{\mathbb{R}^6} \frac{|p|}{|k|}\|(a(p) \otimes F(k))f\|^2\,d\text{ke}
\]

\[
\leq \int_{\mathbb{R}^3} \|(I \otimes F(k))f\|^2\,dk + \int_{\mathbb{R}^3} \frac{1}{|k|}\|(H_{ph}^{1/2} \otimes I)g\|^2\,dk.
\]

The Lemma follows. \hfill \Box

Proof of Proposition 3.4. We use the identity (3.2) for $W_{m}^{tot}$ together with the integral expression (2.3) for $H_{int}$. We know that:

\[
H_{ph}a(k) = a(k)(H_{ph} - |k|).
\]

Therefore, for all $f$ in $W_{2m}^{tot}$, for all integers $p$ and $q$ such that $p + q \leq m - 1$,

\[
(H_{ph}^p \otimes H_{el}^q)(a(k) \otimes E^*(k))f = \sum_{j=0}^{p} (-1)^j C_j |k|^{p-j}(a(k)H_{ph}^j \otimes H_{el}^q E(k))f.
\]

We apply Lemma 3.5 with:

\[
F_j(k) = |k|^{p-j}H_{el}^q E(k)(C + H_{el})^{-q-1}, \quad g_j = (H_{ph}^j \otimes (C + H_{el})^{q+1})f.
\]

Thus:

\[
(H_{ph}^p \otimes H_{el}^q)(a(k) \otimes E^*(k))f = \sum_{j=0}^{p} (-1)^j C_j T_{F_j}g_j.
\]

By Proposition 3.1

\[
\int_{\mathbb{R}^3} \|F_j(k)\|^2\,\frac{dk}{|k|} < \infty.
\]

The norm for $\|F_j(k)\|$ in the above integral is the norm of $L(\mathcal{H}_{el} \otimes \mathcal{H}_{sp}, (\mathcal{H}_{el} \otimes \mathcal{H}_{sp})^3)$. By the Lemma:

\[
\|(H_{ph}^p \otimes H_{el}^q)(a(k) \otimes E^*(k))f\| \leq C \sum_{j=0}^{p} \|(H_{ph}^{1/2} \otimes I)g_j\|
\]

\[
\leq C \sum_{j=0}^{p} \|(H_{ph}^{1/2} \otimes (C + H_{el})^{q+1})f\| \leq C\|f\|_{W_{2m}^{tot}}.
\]

Proposition 3.4 then follows for $m$ even for other $m$ by interpolation. \hfill \Box
Proposition 3.6. For $g$ small enough, $H(g)$ is essentially self-adjoint on its initial domain. The domain of its self-adjoint extension is $W_2^{tot}$. The domain of $H(g)^m$ is $W_2^{tot}$ for any integer $m$. The operator $e^{itH(g)}$ is bounded in $W_m^{tot}$ uniformly in $t$.

Proof. By Proposition 3.4 we have for $g$ small enough:

$$\|H(g)^m f - H(0)^m f\| \leq Cg\|f\|_{W_2^{tot}}.$$ 

Since $W_2^{tot}$ is the domain of the self-adjoint extension of $H(0)^m$, the Proposition follows from Kato Rellich Theorem. The last point of the Proposition is deduced from the first point for even $m$ since $e^{itH(g)}$ maps $D(H(g)^{m/2})$ into itself and by interpolation for the other $m$. 

4 Proof of the main results: Proposition 2.3 and Theorem 2.4.

4.1 Formal identities.

We write $A \sim B$ if $\sigma_0(A - B) = 0$ for two given operators $A$ and $B$ in $\mathcal{H}_{tot}$ where $\sigma_0$ is the operator defined in (2.9).

We begin this subsection with formal equalities and the norm estimates making sense for operators will be studied in the next subsections.

For each $t > 0$, set:

$$E(t) = \{(s_1, s_2) \in \mathbb{R}_+^2, s_1 + s_2 < t\}. \quad (4.1)$$

Theorem 4.1. For each operator $X$ in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$, we have:

$$\frac{d}{dt} e^{itH(g)}(I \otimes X)e^{-itH(g)} \sim e^{itH(g)}(I \otimes L(t,g)X)e^{-itH(g)} + R_1(t,g)X + R_2(t,g)X$$

where $L(t,g)X$ is defined in (2.3) and:

$$R_1(t,g)X = \frac{i}{2}g^3 \int_{\mathbb{R}^3 \times E(t)} e^{i(t-s_1)|k|}e^{itH(g)} \left(I \otimes [E^*(k), X] \right) e^{-is_2H(g)} \quad (4.2)$$

$$R_2(t,g)X = -\frac{i}{2}g^3 \int_{\mathbb{R}^3 \times E(t)} e^{i(s_1-t)|k|}e^{i(t-s_2)H(g)} [H_{int}, I \otimes E^{free}(k,s_1 + s_2 - t)] \quad (4.3)$$

Concerning the dependance on the parameter $g$ in the notations, we make it explicit in important terms such as $H(g), S(t,g), L(t,g), L(g), R_0(t,g), R(t,g), R_1(g), R_2(t,g)$ and not for the terms used in the computations such as $f(t), I_1(t), I_2(t), L_1(t), L_2(t), \Phi(s, X)$.

The first three steps of the proof correspond to the three Propositions below.

Proposition 4.2. For all operators $X$ in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$, we have:

$$\frac{d}{dt} e^{itH(g)}(I \otimes X)e^{-itH(g)} = i\epsilon^{itH(g)} \left(I \otimes [(H_{el} + H_{sp}), X] + g[H_{int}, I \otimes X] \right) e^{-itH(g)}$$
\[ = ie^{itH(g)}(I \otimes [(H_{cl} + H_{sp}), X])e^{itH(g)} \]
\[ + \frac{ig}{\sqrt{2}} \int_{\mathbb{R}^3} e^{itH(g)} \left( a(k) \otimes [E^*(k), X] + a^*(k) \otimes [E(k), X] \right) e^{-itH(g)} \, dk. \]

The proof of this Proposition is only a combination of the Heisenberg equation for the Pauli-
Fierz Hamiltonian with the integral expression (2.6) of \( H_{\text{int}} \).

**Proposition 4.3.** For each \( k \) in \( \mathbb{R}^3 \), we have:
\[ e^{itH(g)}(a(k) \otimes I)e^{-itH(g)} = e^{-it|k|}(a(k) \otimes I) - \frac{ig}{\sqrt{2}} \int_0^t e^{i(s-t)|k|} e^{isH(g)}(I \otimes E(k))e^{-isH(g)} \, ds, \]
\[ e^{itH(g)}(a^*(k) \otimes I)e^{-itH(g)} = e^{it|k|}(a^*(k) \otimes I) + \frac{ig}{\sqrt{2}} \int_0^t e^{i(t-s)|k|} e^{isH(g)}(I \otimes E^*(k))e^{-isH(g)} \, ds. \]

**Proof.** One knows that:
\[ e^{itH(0)}(a(k) \otimes I)e^{-itH(0)} = e^{-it|k|}(a(k) \otimes I). \]
Let:
\[ f(t) = e^{it|k|}e^{itH(g)}(a(k) \otimes I)e^{-itH(g)} = e^{itH(g)}e^{-itH(0)}(a(k) \otimes I)e^{itH(0)}e^{-itH(g)}. \]
We see that:
\[ f'(t) = ig e^{it|k|}e^{itH(g)}[H_{\text{int}}, (a(k) \otimes I)]e^{itH(g)}. \]
According to the integral expression (2.6) of \( H_{\text{int}} \), since the operators \( a(k') \) and \( a(k) \) are commuting, and using \([a(k), a^*(k')] = \delta(k-k')\), we get:
\[ [H_{\text{int}}, (a(k) \otimes I)] = -\frac{1}{\sqrt{2}}(I \otimes E(k)). \]
Consequently:
\[ f'(t) = -\frac{ig}{\sqrt{2}} e^{it|k|}e^{itH(g)}(I \otimes E(k))e^{-itH(g)}. \]
The Proposition then follows. \( \square \)

The next Proposition is of the type of Proposition 4.2.

**Proposition 4.4.** For all operators \( Y \) in \( \mathcal{H}_{cl} \otimes \mathcal{H}_{sp} \), we have:
\[ e^{-itH(g)}(I \otimes Y)e^{itH(g)} = I \otimes Y^{\text{free}}(-t) - ig \int_0^t e^{-isH(g)}[H_{\text{int}}, (I \otimes Y^{\text{free}}(s-t))]e^{isH(g)} \, ds. \]

**Proof.** For any operator \( Z \), set:
\[ f(t) = e^{-itH(g)}e^{itH(0)}(I \otimes Z)e^{-itH(0)}e^{itH(g)}. \]
We have:
\[ f'(t) = -ige^{-itH(g)}[H_{\text{int}}, (I \otimes Z^{\text{free}}(t))]e^{itH(g)}. \]
Thus:
\[ e^{-itH(g)}(I \otimes Z^{\text{free}}(t))e^{itH(g)} = (I \otimes Z) - ig \int_0^t e^{-isH(g)}[H_{\text{int}}, (I \otimes Z^{\text{free}}(s))]e^{isH(g)} \, ds. \]
The proof is completed applying this equality to \( Z = Y^\text{free}(t) \).

\[ \square \]

End of the proof of Theorem 4.1. By Proposition 4.2, we have:

\[
\frac{d}{dt}e^{itH(g)}(I \otimes X)e^{-itH(g)} = ie^{itH(g)}\left(I \otimes [(H_{el} + H_{sp}), X]\right)e^{-itH(g)} + I_1(t)X + I_2(t)X,
\]

with:

\[
I_1(t)X = \frac{ig}{\sqrt{2}} \int_{\mathbb{R}^3} e^{itH(g)}(a(k) \otimes [E^*(k), X])e^{-itH(g)}dk, \\
I_2(t)X = \frac{ig}{\sqrt{2}} \int_{\mathbb{R}^3} e^{itH(g)}(a^*(k) \otimes [E(k), X])e^{-itH(g)}dk.
\]

Proposition 4.3 is used to rewrite \( I_1(t)X \). This gives:

\[
I_1(t)X = \frac{ig}{\sqrt{2}} \int_{\mathbb{R}^3} e^{itH(g)}(I \otimes [E^*(k), X])(a(k) \otimes I)e^{-itH(g)} = I'_1(t)X + I''_1(t)X,
\]

with:

\[
I'_1(t)X = \frac{ig}{\sqrt{2}} \int_{\mathbb{R}^3} e^{itH(g)}(I \otimes [E^*(k), X])e^{-itH(g)}e^{i\lambda(k)}(a(k) \otimes I)dk, \\
I''_1(t)X = \frac{g^2}{2} \int_{\mathbb{R}^3 \times (0,t)} e^{itH(g)}(I \otimes [E^*(k), X])e^{i(s-t)\lambda(k)}e^{i(s-t)H(g)}(I \otimes E(k))e^{-isH(g)}dkds.
\]

Notice that \( I'_1(t)X \sim 0 \). To rewrite \( I''_1(t)X \), we use Proposition 4.4 with \( t \) replaced by \( t-s \) and \( Y \) by \( E(k) \). Thus, we obtain:

\[
e^{i(s-t)H(g)}(I \otimes E(k))e^{i(t-s)H(g)} = I \otimes E^\text{free}(k, s-t)
\]

\[-ig \int_0^{t-s} e^{-is_2H}(H_{\text{int}}, (I \otimes E^\text{free}(k, s + s_2 - t)))e^{is_2H(g)}ds_2.
\]

Consequently:

\[
I_1(t)X \sim I''_1(t)X = e^{itH(g)}(I \otimes L_1(t)X)e^{-itH(g)} + R_1(t, g)X
\]

where \( R_1(t, g)X \) is defined in (4.2) and:

\[
L_1(t)X = \frac{g^2}{2} \int_{\mathbb{R}^3 \times (0,t)} e^{i(s-t)\lambda(k)}[E^*(k), X]E^\text{free}(k, s-t)dkds.
\]

Next, we similarly consider \( I_2(t)X \) and obtain:

\[
I_2(t)X \sim e^{itH(g)}(I \otimes L_2(t)X)e^{-itH(g)} + R_2(t, g)X,
\]

\[
L_2(t)X = -\frac{g^2}{2} \int_{\mathbb{R}^3 \times (0,t)} e^{is\lambda(k)}[E^\text{free}(k, -s), E(k), X]dkds.
\]

where \( R_2(t, g)X \) is defined in (4.3). We see that the operator \( L(t, g)X \) defined in (2.3) satisfies:

\[
L(t, g)X = i[(H_{el} + H_{sp}), X] + L_1(t)X + L_2(t)X.
\]

The proof is completed. \( \square \)
4.2 Norm estimates of the Lindblad operator.

We study here the main term $L(t, g)X$ and its limit as $t \to +\infty$. To this end, we need the next Proposition.

**Proposition 4.5.** Let $X$ be a bounded operator in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$ which is also bounded from $W_{1}^{el}$ to itself. Then for all $t > 0$, the following operators are well defined from $W_{2}^{el}$ into $W_{0}^{el}$:

\[
\Phi(s, X) = \int_{\mathbb{R}^{3}} e^{is|k|} [E(k)^{\star}, X] E^{\text{free}}(k, -s) dk
\]

\[
\Phi^{\star}(s, X) = \int_{\mathbb{R}^{3}} e^{-is|k|} E^{\text{free}}(k, -s)^{\star} [X, E(k)] dk.
\]

Moreover, under the above hypotheses, there exists $C > 0$ such that:

\[
\|\Phi(s, X)\|_{L(W_{2}^{el}, W_{0}^{el})} + \|\Phi^{\star}(s, X)\|_{L(W_{2}^{el}, W_{0}^{el})} \leq \frac{C}{1 + s^{2}} \|X\|_{L(W_{1}^{el}, W_{1}^{el})}.
\]

If in addition $X$ is bounded from $W_{0}^{el}$ to $W_{1}^{el}$ then we have:

\[
\|\Phi(s, X)\|_{L(W_{1}^{el}, W_{0}^{el})} + \|\Phi^{\star}(s, X)\|_{L(W_{1}^{el}, W_{0}^{el})} \leq \frac{C}{1 + s^{2}} \|X\|_{L(W_{1}^{el}, W_{1}^{el})}.
\]

If the function $\varphi$ in (2.3), (2.4) is vanishing at the origin then the first inequality can be replaced by:

\[
\|\Phi(s, X)\|_{L(W_{2}^{el}, W_{0}^{el})} + \|\Phi^{\star}(s, X)\|_{L(W_{2}^{el}, W_{0}^{el})} \leq \frac{C}{1 + s^{2}} \|X\|_{L(W_{1}^{el}, W_{1}^{el})}
\]

and similarly for the second inequality.

**Proof.** We use spherical coordinates for $k, k = \rho \omega, \rho > 0$ and $\omega \in S^{2}$. We have:

\[
\Phi(s, X) = \sum_{j=1}^{3} \int_{\mathbb{R}^{3} \times S^{2}} e^{is\rho|j|} [F_{j}(\rho, \omega), X] G_{j}(\rho, \omega, s) \rho d\rho d\omega,
\]

with:

\[
F_{j}(\rho, \omega) = \rho^{1/2} E_{j}(\rho \omega)^{\star}, \quad G_{j}(\rho, \omega, s) = \rho^{1/2} E_{j}^{\text{free}}(\rho \omega, -s).
\]

According to Proposition 3.1 we get:

\[
\|\partial^{\rho}_{\rho} F_{j}(\rho, \omega)\|_{L(W_{p}^{el}, W_{p-1}^{el})} \leq C(1 + \rho)^{-2}.
\]

Using the hypotheses on the operator $X$, we obtain:

\[
\|\partial^{\rho}_{\rho} [F_{j}(\rho, \omega), X]\|_{L(W_{p}^{el}, W_{p-1}^{el})} \leq C(1 + \rho)^{-2} \sup_{m \leq p} \|X\|_{L(W_{m}^{el}, W_{m}^{el})}.
\]

By Proposition 3.1 and since $e^{it(H_{el} + H_{sp})}$ is uniformly bounded from $W_{k}^{el}$ into itself for each $k$, we get:

\[
\|\partial^{\rho}_{\rho} G_{j}(\rho, \omega, s)\|_{L(W_{2}^{el}, W_{1}^{el})} \leq C(1 + \rho)^{-2}.
\]

We can integrate twice by parts with the variable $\rho$ in the expression of $\Phi(s, X)$. We deduce:

\[
s^{2} \Phi(s, X) = \sum_{j=1}^{3} \int_{\mathbb{R}^{3} \times S^{2}} e^{is\rho^{2}} \partial^{\rho}_{\rho} \left( \rho [F_{j}(\rho, \omega), X] G_{j}(\rho, \omega, s) \right) d\rho d\omega.
\]
To prove the last points of the Theorem, we note that if the function $\varphi$ vanishes at the origin then we can integrate by parts three times instead of two.

The proof of the Proposition is completed. □

We then deduce the next result.

**Proposition 4.6.** Let $X$ be a bounded operator in $\mathcal{H}_{el} \otimes \mathcal{H}_{sp}$ which is also bounded from $W_1^{el}$ to itself. Then the operator $L(t, g)X$ defined in (2.3) is bounded uniformly in $t$ from $W_2^{el}$ to $W_0^{el}$. Moreover, as $t$ tends to $+\infty$, this operator tends in $\mathcal{L}(W_2^{el}, W_0^{el})$ to a limit $L(g)X$ satisfying:

$$\|L(t, g)X - L(g)X\|_{\mathcal{L}(W_2^{el}, W_0^{el})} \leq \frac{Cg^2}{1+t} \|X\|_{\mathcal{L}(W_1^{el}, W_1^{el})}$$

and the above factor $1/(1+t)$ is improved to $1/(1+t^2)$ if the ultraviolet cutoff $\varphi$ vanishes at the origin.

Proposition 2.3 is then proved.

### 4.3 Estimates on the integral rests.

We now need to control the norms in some spaces of the operators $R_1(t, g)X$ and $R_2(t, g)X$ given by (4.2) and (4.3).

**Proposition 4.7.** For each integer $m$, there exists $C_m > 0$ such that the operator $R_1(t, g)$ defined in (4.2) satisfies:

$$\|R_1(t, g)X\|_{\mathcal{L}(W_1^{el}, W_m^{el})} \leq C_m g^3 \ln(1+t) \sup_{j \leq m} \|X\|_{\mathcal{L}(W_j^{el}, W_0^{el})}.$$  

We also have:

$$\|\sigma_0 R_1(t, g)X\|_{\mathcal{L}(W_1^{el}, W_0^{el})} \leq C_m g^3 \ln(1+t) \sup_{j \leq 4} \|X\|_{\mathcal{L}(W_j^{el}, W_0^{el})}.$$  

If the smooth cutoff $\varphi$ in (2.3) and (2.4) vanishes at the origin then the factor $\ln(1+t)$ can be omitted in the above inequalities.

The same estimates holds true for $R_2(t, g)$ instead of $R_1(t, g)$.

**Proof.** In the definition (4.2) of $R_1(t, g)X$, we use spherical coordinates for $k$, namely $k = \rho \omega$ with $\rho > 0$ and $\omega \in S^2$ the unit sphere centered at the origin. Thus, we can write:

$$R_1(t, g)X = g^3 \int_{E(t)} \Phi(t, s_1, s_2) ds_1 ds_2$$

with:

$$\Phi(t, s_1, s_2) = \int_{\mathbb{R}_+ \times S^2} e^{i(t-s_1)\rho} e^{itH(g)} \left( I \otimes [F(\rho, \omega, \rho, X)] e^{-is_2H(g)} \right)$$

$$[H_{int}, I \otimes G(\rho, \omega, s_1, s_2, t)] e^{i(s_2-t)H(g)} \rho d\rho d\omega$$

where:

$$F(\rho, \omega) = \rho^{1/2} E^*(\rho \omega)$$

$$G(\rho, \omega, s_1, s_2, t) = \rho^{1/2} E(\rho \omega, s_1 + s_2 - t).$$
According to Proposition 3.1, the function $F$ takes its values in $L(W^el_j, W^el_{j-1})$ and we have:

$$\|\partial^\alpha_F(\rho, \omega)\|_{L(W^el_j, W^el_{j-1})} \leq C_j(1 + \rho)^{-2}.$$ 

Under the hypothesis of Theorem 2.4 we see:

$$\|\partial^\alpha_F[X, F(\rho, \omega)]\|_{L(W^el_j, W^el_{j-1})} \leq C_j(1 + \rho)^{-2}\sup_{k \leq j} \|X\|_{L(W^el_k, W^el_j)}.$$ 

By Proposition 3.3,

$$\|\partial^\alpha(I \otimes [X, F(\rho, \omega)])\|_{L(W^tot_j, W^tot_{j-1})} \leq C_j(1 + \rho)^{-2}\sup_{k \leq j} \|X\|_{L(W^el_k, W^el_j)}.$$ 

In the same way and since $e^{it(H_{el} + H_{sp})}$ is uniformly bounded from $W^el_k$ into itself for all $k$, we deduce:

$$\|\partial^\alpha G(\rho, \omega, s_1, s_2, t)\|_{L(W^el_j, W^el_{j-1})} \leq C_j(1 + \rho)^{-2}$$ 

where $C_j$ is independent on $t, s_1, s_2$. Therefore, by Proposition 3.3,

$$\|\partial^\alpha(I \otimes G(\rho, \omega, s_1, s_2, t))\|_{L(W^tot_j, W^tot_{j-1})} \leq C_j(1 + \rho)^{-2}.$$ 

By Proposition 3.4 $H_{int}$ is bounded from $W^tot_j$ to $W^tot_{j-2}$. Thus:

$$\|\partial^\alpha[H_{int}, I \otimes G(\rho, \omega, s_1, s_2, t)]\|_{L(W^tot_j, W^tot_{j-3})} \leq C_j(1 + \rho)^{-2}.$$ 

Finally, we know that $e^{itH(g)}$, $e^{-is_2H(g)}$ and $e^{i(s_2-t)H(g)}$ are uniformly bounded from $W^tot_j$ into itself for all $j$. We can integrate twice by parts with the variable $\rho$ in (4.3). In view of the above estimates, we get:

$$(1 + |t - s_1|^2)\|\Phi(t, s_1, s_2)\|_{L(W^tot_j, W^tot_{j-4})} \leq C_j\sup_{k \leq j} \|X\|_{L(W^el_k, W^el_j)}.$$ 

If the cutoff $\varphi$ vanishes at 0, we have $F(0, \omega) = 0$ and $G(0, \omega, s_1, s_2, t) = 0$. In equality (4.3), we can integrate three times by parts and obtain:

$$(1 + |t - s_1|^3)\|\Phi(t, s_1, s_2)\|_{L(W^tot_j, W^tot_{j-4})} \leq C_j\sup_{k \leq j} \|X\|_{L(W^el_k, W^el_j)}.$$ 

If $E(t)$ is the set defined in (4.1), we see that:

$$\int_{E(t)} \frac{ds_1 ds_2}{1 + |t - s_1|^2} \leq C \ln(1 + t), \quad \int_{E(t)} \frac{ds_1 ds_2}{1 + |t - s_1|^3} \leq C.$$ 

The first point in the Theorem is then deduced. The second one follows by Proposition 3.3.

\textit{End of the proof of Theorem 2.4.} By Theorem 4.1 we have (2.13) with:

$$R_0(t, g)X = L(t, g)X - L(g)X, \quad R(t, g)X = R_1(t, g)X + R_2(t, g)X.$$ 

The norm estimate of $R_0(t, g)X$ comes from Proposition 4.6 and the norm estimate of $R_1(t, g)X$ and $R_2(t, g)X$ from Proposition 4.7. □
5 Transition probabilities.

We shall make explicit in this section the matrix of the operator $L(g)$ in a suitable basis, and in a particular case. In this Section, there is one (spinless) electron ($Q = 1$), no fixed spin particle and no external magnetic field ($A^{\text{ext}} = 0$). Thus:

$$\mathcal{H}_{\text{tot}} = \mathcal{H}_{\text{ph}} \otimes \mathcal{H}_{\text{el}}, \quad \mathcal{H}_{\text{el}} = L^2(\mathbb{R}^3)$$

Therefore $H_{\text{el}}$ is the usual Schrödinger operator:

$$H_{\text{el}} = -\Delta + V.$$  

We make the following assumptions.

(H') The potential $V$ is a non negative real valued $C^\infty$ function on $\mathbb{R}^3$, and there exists a real $M > 0$ and a constant $C_\alpha > 0$ for each multi-index $\alpha$, such that:

$$|\partial^\alpha V(x)| \leq C_\alpha (1 + |x|)^{M-|\alpha|}.$$  

(H'') There exists $\gamma > 0$ such that:

$$V(x) \geq \gamma |x|^M.$$  

The following result is proved in [20].

**Proposition 5.1.** Under hypotheses (H') and (H'') the operator $H_{\text{el}}$ initially defined on $\mathcal{S}(\mathbb{R}^3)$ has a unique self-adjoint extension. The spectrum of $H_{\text{el}}$ is discrete. It is the set of finite multiplicity eigenvalues $\mu_j$ going to $+\infty$. There exists an Hilbertian basis $(u_j)$ of $\mathcal{H}_{\text{el}}$ satisfying:

$$H_{\text{el}}u_j = \mu_j u_j.$$  

Each $u_j$ belongs to the intersection of the spaces $W^m_{el}$. We have for all $m > 0$:

$$D(H_{el}^m) = \{ u \in H^{2m}(\mathbb{R}^3), \ V(x)^m u \in L^2(\mathbb{R}^3) \}.$$  

Consequently, Hypothesis (H) of Section 3 is satisfied if the above assumptions (H')(H'') hold. We can rearrange the eigenfunctions in such a way that the sequence $(\mu_j)$ is non decreasing.

We next define the transition probabilities. Let $\pi_{u_j}$ be the orthogonal projection in $\mathcal{H}_{\text{el}}$ on the vector space spanned by $u_j$.

**Definition 5.2.** For all unitary eigenfunctions $u_j$ and $u_m$ of $H_{\text{el}}$, and for each $t > 0$, the transition probability of $u_m$ to $u_j$ is defined by:

$$P_{mj}(t) = \langle (\sigma_0 S(t, g) \pi_{u_j}) u_m, u_m \rangle_{\mathcal{H}_{\text{el}}} = \langle (I \otimes \pi_{u_j}) e^{-itH(g)}(\Psi_0 \otimes u_m), e^{-itH(g)}(\Psi_0 \otimes u_m) \rangle_{\mathcal{H}_{\text{tot}}} = \langle \sigma_0 S(t, g) \pi_{u_j}, \pi_{u_m} \rangle_{HS},$$

where $\langle , \rangle_{HS}$ is the scalar product of two Hilbert-Schmidt operators in $\mathcal{H}_{\text{el}}$.

These identities together with our main result leads us to make explicit the following matrix:

$$M_{mj} = g^{-2} \langle (L(g) \pi_{u_j}), \pi_{u_m} \rangle_{HS} = g^{-2} \langle (L(g) \pi_{u_j}) u_m, u_m \rangle_{\mathcal{H}_{\text{el}}}. \quad (5.1)$$

We now omit the subscript $\mathcal{H}_{\text{el}}$ in the scalar product of $\mathcal{H}_{\text{el}}$.

We believe that this matrix is the generator of a semi-group of Markov matrices which perhaps will be a good approximation of the matrix $P_{mj}(t)$.  


Theorem 5.3. The $M_{mj}$ defined by (5.1) satisfy:

$$M_{mj} = \int_{\mathbb{R}^3} \left| \langle E_\alpha(k)u_m, u_j \rangle \right|^2 d\sigma(k)$$

if $\mu_j < \mu_m$

$$M_{mj} = 0$$

if $\mu_j > \mu_m$

$$M_{jj} = -\sum_{k \neq j} M_{kj}$$

$$M_{mj} = 0$$

if $\mu_j = \mu_m$ and $j \neq m$.

Therefore $M_{mj} \geq 0$ if $\mu_j < \mu_m$, $M_{mj} \leq 0$ if $\mu_j = \mu_m$ and $M_{mj} = 0$ if $\mu_j > \mu_m$. We have, for each $j$:

$$\sum_m M_{mj} = 0.$$

We remark that these properties are supposed to be verified by the infinitesimal generator of a Markov semi-group. We do not know if the matrix $M_{mj}$ is indeed the infinitesimal generator of a semi-group neither if we get a good approximation of the $P_{mj}(t)$. If this holds true then, up to this approximation, the transition probability of an initial electronic state to a higher electronic energy level state is zero.

Proof. By the definition (2.3) of $L(g)$, since $[H_{el}, \pi_{u_j}] = 0$, we have:

$$L(g)\pi_{u_j} =$$

$$\frac{g^2}{2} \lim_{t \to +\infty} \int_{\mathbb{R}^3 \times (0,t)} \left(e^{-is|k|} [E^*(k), \pi_{u_j}] E^{free}(k,-s) - e^{is|k|} E^{free}(k,-s)^* [E(k), \pi_{u_j}] \right) dk ds.$$

We have, for all integers $j$ and $m$:

$$\langle L(g)\pi_{u_j}, \pi_{u_m} \rangle_{HS} = \text{Tr} (\pi_{u_m} L(g)\pi_{u_j}).$$

By the definition (2.11), we have for the traces:

$$\text{Tr} ([E^*(k), \pi_{u_j}] E^{free}(k,-s)\pi_{u_m}) = e^{is(\mu_j - \mu_m)} \langle E(k)u_m, u_j \rangle^2$$

$$- e^{is\mu_j} \delta_{jm} \langle e^{-isH_{el}} E(k)u_j E(k)u_j \rangle$$

and

$$\text{Tr} (E^{free}(k,-s)^* [E(k), \pi_{u_j}]\pi_{u_m}) = e^{-is\mu_j} \delta_{jm} \langle e^{isH_{el}} E(k)u_j E(k)u_j \rangle$$

$$- e^{is(\mu_j - \mu_m)} \langle E(k)u_m, u_j \rangle^2.$$

Therefore:

$$g^2 A_{mj} = \langle L(g)\pi_{u_j}, \pi_{u_m} \rangle_{HS} = g^2 (A_{mj} + \delta_{jm} \lambda_j)$$

with:

$$A_{mj} = \lim_{t \to +\infty} \int_{\mathbb{R}^3 \times (0,t)} \cos(s(|k| + \mu_j - \mu_m)) \langle E(k)u_m, u_j \rangle^2 dk ds.$$

We know that, for each suitable function $F$, we have, if $\lambda < 0$:

$$\lim_{t \to +\infty} \int_{\mathbb{R}^3 \times (0,t)} \cos(s(|k| + \lambda)) F(k) dk dt = \pi \int_{|k|=\lambda} F(k) d\sigma(k).$$

This limit is zero if $\lambda \geq 0$. We then indeed obtain the expression of $M_{mj}$ for $m \neq j$. For the diagonal elements $M_{jj}$, the above computations are not used but rather $L(g)I = 0$, denoting by $I$ the identity operator on $H_{el}$. Since $I$ is the sum of the $\pi_{u_j}$, it follows that $\sum_j \langle L(g)\pi_{u_j}, \pi_{u_m} \rangle_{HS} = 0$ for each $m$. This completes the proof of Theorem 5.3. □
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