

## *Setting and analysis of the multi-configuration time-dependent Hartree–Fock equations*

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### **Abstract**

In this paper we formulate and analyze the Multi-Configuration Time-Dependent Hartree-Fock (MCTDHF) equations for molecular systems with pairwise interaction. This set of coupled nonlinear PDEs and ODEs is an approximation of the  $N$ -particle time-dependent Schrödinger equation based on (time-dependent) linear combinations of (time-dependent) Slater determinants. The “one-electron” wave-functions satisfy nonlinear Schrödinger-type equations coupled to a linear system of ordinary differential equations for the expansion coefficients. The invertibility of the one-body density matrix (full-rank hypothesis) plays a crucial rôle in the analysis. Under the full-rank assumption a fiber bundle structure emerges and produces unitary equivalence between different useful representations of the MCTDHF approximation. For a large class of interactions (including Coulomb potential), we establish existence and uniqueness of maximal solutions to the Cauchy problem in the energy space as long as the density matrix is not singular. A sufficient condition in terms of the energy of the initial data ensuring the global-in-time invertibility is provided (first result in this direction). Regularizing the density matrix violates energy conservation. However, global well-posedness for this system in  $L^2$  is obtained with Strichartz estimates. Eventually solutions to this regularized system are shown to converge to the original one on the time interval when the density matrix is invertible.

**Key words.** Multi-configuration methods – Nonlinear Schrödinger equations, Hartree-Fock equations – Dirac-Frenkel variational principle – Strichartz estimates

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## 1. Introduction

The purpose of the present paper is to lay out the mathematical analysis of the *multi-configuration time-dependent Hartree-Fock* (MCTDHF) approximation which is used in quantum physics/chemistry for the dynamics of few electron problems, like the interaction of an atom with a strong short laser-pulse [7, 40, 41] and [23]. The MCTDHF models are natural generalizations of the time-dependent Hartree-Fock (TDHF) approximation, yielding a hierarchy of models that, in principle, should converge to the exact model.

We consider a quantum system composed of a finite number  $M$  of *fixed* nuclei of masses  $m_1, \dots, m_M > 0$  with charge  $z_1, \dots, z_M > 0$  and a finite number  $N$  of electrons. Using atomic units, the  $N$ -body Hamiltonian of the electronic system submitted to the external potential due to the nuclei is then the self-adjoint operator

$$\mathcal{H}_N = \sum_{1 \leq i \leq N} \left( -\frac{1}{2} \Delta_{x_i} + U(x_i) \right) + V(x_1, \dots, x_N) \quad (1.1)$$

acting on the Hilbert space  $L^2(\Omega^N; \mathbb{C})$  with pairwise interaction between the electrons of the form

$$V(x_1, \dots, x_N) = \sum_{1 \leq i < j \leq N} v(|x_i - x_j|),$$

with  $v$  real-valued and  $v \geq 0$ . Here and below  $\Omega$  is either the whole space  $\mathbb{R}^3$  or a bounded domain in  $\mathbb{R}^3$  with boundary conditions. The  $N$  electrons state is defined by a *wave-function*  $\Psi = \Psi(x_1, \dots, x_N)$  in  $L^2(\Omega^N)$  that is normalized by  $\|\Psi\|_{L^2(\Omega^N)} = 1$ . To account for the Pauli exclusion principle which features the fermionic nature of the electrons, the antisymmetry condition

$$\Psi(x_1, \dots, x_N) = \varepsilon(\sigma) \Psi(x_{\sigma(1)}, \dots, x_{\sigma(N)}),$$

for every permutation  $\sigma$  of  $\{1, \dots, N\}$  is imposed to the wave-function  $\Psi$ . The space of antisymmetric wave-functions will be denoted by  $\bigwedge_{i=1}^N L^2(\Omega)$ . In (1.1) and throughout the paper, the subscript  $x_i$  of  $-\Delta_{x_i}$  means derivation with respect to the  $i^{\text{th}}$  variable of the function  $\Psi$ . Next,

$$U(x) := - \sum_{m=1}^M \frac{z_m}{|x - R_m|}$$

is the Coulomb potential created by  $M$  nuclei of respective charge  $z_1, \dots, z_M > 0$  located at points  $R_1, \dots, R_M \in \mathbb{R}^3$  and  $v(x) = \frac{1}{|x|}$  is the Coulomb repulsive potential between the electrons. Actually our whole analysis carries through to more general Hamiltonians (possibly time-dependent) as explained in Section 7 below.

For nearly all applications, even with only two interacting electrons, the numerical treatment of the time-dependent Schrödinger equation (TDSE)

$$i \frac{\partial \Psi}{\partial t} = \mathcal{H}_N \Psi, \quad \Psi(0) = \Psi^0, \quad (1.2)$$

is out of the reach of even the most powerful computers, and approximations are needed.

Simplest elements of  $\bigwedge_{i=1}^N L^2(\Omega)$  are the so-called *Slater determinants*

$$\Psi(x_1, \dots, x_N) = \frac{1}{\sqrt{N!}} \det(\phi_i(x_j))_{1 \leq i, j \leq N} \quad (1.3)$$

constructed with any orthonormal family  $\phi_i$  in  $L^2(\Omega)$ . The factor  $\frac{1}{\sqrt{N!}}$  ensures the normalization condition on the wave-function. Such a Slater determinant will be denoted by  $\phi_1 \wedge \dots \wedge \phi_N$ . The family of all Slater determinants built from a complete orthonormal set of  $L^2(\Omega)$  is a complete orthonormal set of  $\bigwedge_{i=1}^N L^2(\Omega)$ . Algorithms based on the restriction to a single Slater determinant are called Hartree-Fock approximation (HF). On the other hand the basic idea of the multi-configuration methods is to use a finite linear combinations of such determinants constructed from a family of  $K (\geq N)$  orthonormal mono-electronic wave-functions.

One observes (this computation is done in Subsection 3.5) that in the absence of pairwise interacting potentials any Slater determinant constructed with orthonormal solutions  $\phi_i(x, t)$  to the single-particle time-dependent Schrödinger equation gives an exact solution of the  $N$ -particle non interacting time-dependent Schrödinger equation. Such  $\phi_i(x, t)$  are called *orbitals* in quantum chemistry. The same is true for any linear combination of Slater determinants with *constant* coefficients. Of course, the situation turns out to be completely different when pairwise interactions are added : a solution to TDSE starting with an initial data composed of one or a finite number of Slater determinants will not remain so for any time  $t \neq 0$ . Such behavior (called “explosion of rank”) is part of the common belief, but is not shown rigorously as a property of the equations, to the best of our knowledge. In the MCTDHF approach one introduces time-dependent coefficients and time-dependent orbitals to take into account pairwise interactions and to preserve the finite linear combination structure of Slater determinants in time. Using time-independent orbitals as it corresponds to a Galerkin-type approximation would save the effort for the nonlinear equations, but requires a much larger number of relevant orbitals and hence the numerical cost is much higher. The motion of the electrons in the MCTDHF framework is governed by a coupled system of  $K$  nonlinear partial differential equations for the orbitals and  $\binom{K}{N}$  ordinary differential equations for the expansion coefficients (see for instance System (3.27)).

Although MCTDHF is known for decades, the mathematical analysis has been tackled only recently. For a mathematical theory of the use of the time-independent *multi-configuration Hartree-Fock* (MCHF) ansatz in the computation of so-called ground- and bound states we refer to [27, 17, 28]. A preliminary contribution was given by Lubich [30] and Koch and Lubich [26] for the time-dependent multi-configuration Hartree (MCTDH) equations for bosons, for the simplified case of a regular and bounded interaction potential  $v$  between the electrons and a Hamiltonian without exterior potential  $U$ . The MCTDH equations (see e.g. [19] for a recent overview) are similar to MCTDHF from the functional analysis point of view, although more complicated from the algebraic point of view, since more density-matrices have to be considered in the absence of *a priori* antisymmetry

requirements on the  $N$ -particle wave-function (see also [25] for an extension to MCTDHF equations and [32] for a recent overview). Using a full-rank (*i.e.* invertibility) assumption on the one-body density matrices, the authors proved short-time existence and uniqueness of solutions in the functions space  $H^2(\mathbb{R}^3)$  for the orbitals with the help of Lie commutators techniques. Numerical algorithms are also proposed and analyzed by the groups around Scrinzi (*e.g.* [40]) and Lubich, the proof of their convergence generally requires the  $H^2$ -type regularity assumptions (see *e.g.* [31]).

We present here well-posedness results for the MCTDHF Cauchy problem in  $H^1$ ,  $H^2$  and  $L^2$ , under the full-rank assumption on the first-order density matrix and for the physically most relevant and mathematically most demanding case of Coulomb interaction. We also give sufficient conditions for global-in-time full-rank in terms of the energy of the initial data. Eventually solutions to a perturbed system with regularized density matrix are shown to converge to the original one on the time interval when the density matrix is invertible.

This paper is organized as follows. In Section 2 we give a complete analysis of the *ansatz*  $\Psi$  associated to the multi-configuration Hartree-Fock approximation. Essentially, this *ansatz* corresponds to a linear combination of Slater determinants built from a vector of complex coefficients  $C$  and a set of orthonormal, square integrable functions represented by a vector  $\Phi = (\phi_1, \phi_2, \dots, \phi_K)$ , for  $K \geq N$ . The *first-order density matrix* is introduced and represented by a complex-valued matrix  $\mathbf{\Gamma}$  which corresponds to the representation of the kernel of the first-order density matrix in the orthonormal basis  $\{\phi_1, \phi_2, \dots, \phi_K\}$ . By abuse of language this matrix  $\mathbf{\Gamma}$  depending only on the expansion coefficients  $C$  is also called *density matrix*. Its invertibility is a crucial hypothesis which will be referred to as the *full-rank hypothesis*. Under this hypothesis, the corresponding set of pairs  $(C, \Phi)$  is endowed with a structure of a *fiber bundle*. In Section 3, two set of equivalent systems are presented. The first one,  $\mathcal{S}_0$ , called *variational system* is inspired by a variational principle. The second one,  $\mathcal{S}_{\mathbf{H}}$ , will be referred to as *working equations*. In Section 4, the system  $\mathcal{S}_0$  is used to prove the propagation of the normalization constraints, the conservation of the total energy and an *a posteriori* error estimate for smooth solutions (if they exist). The system  $\mathcal{S}_{\mathbf{H}}$  is used to prove local existence, uniqueness and stability with initial data in  $H^m$  for  $m \in \mathbb{N}$ ,  $m \geq 1$ . In particular the space  $H^1$  is used to balance the singularity of the potentials (of Coulomb type) and we prove the local well-posedness using the Duhamel formula. Next, the conservation of the total energy allows to extend the local-in-time solution until the associated density matrix  $\mathbf{\Gamma}$  becomes singular. Therefore, Section 5 is devoted to a criteria based on the conservation of the energy that guarantees the global-in-time invertibility of the density matrix  $\mathbf{\Gamma}$ . To handle the possible degeneracy of this matrix, a regularized problem is considered in Section 6. For this problem the conservation of the energy does not hold anymore. Therefore we present an alternative proof based on mere mass conservation, that uses Strichartz estimates and that is also valid for singular potentials. Eventually, one expects that the solution of the regularized problem converges towards the solution of the original one as long as the unperturbed density matrix is invertible. The proof is a  $H^1$  version of the classical

“shadowing lemma” for ordinary differential equations. Finally, in the last section we list some extensions to time-dependent Hamiltonians including a laser field and/or a time-dependent external potential. The case of discrete systems is also worked out there.

Some of the results presented here have been announced in [37] and [3] and the details of the  $L^2$  theory are worked out in [34].

**Notation.**  $\langle \cdot, \cdot \rangle$  and  $\langle \cdot | \cdot \rangle$  respectively denote the usual scalar products in  $L^2(\Omega)$  and in  $L^2(\Omega^N)$ ,  $(\cdot, \cdot)$  the scalar product in  $L^2(\Omega)^K$  and  $a \cdot b$  the complex scalar product of two complex vectors  $a$  and  $b$ . The bar denotes complex conjugation. We set  $L^2_{\wedge}(\Omega^N) := \bigwedge_{k=1}^N L^2(\Omega)$  where the symbol  $\wedge$  denotes the skew-symmetric tensorial product. Throughout the paper bold face letters like  $\mathbf{P}$  correspond to one-particle operators on  $L^2(\Omega)$ , calligraphic bold face letters to operators on  $L^2(\Omega^N)$ , whereas “black board” bold face letters are reserved to matrices.  $\mathcal{L}(E; F)$  denotes the set of continuous linear mappings from  $E$  to  $F$  (as usual  $\mathcal{L}(E) = \mathcal{L}(E; E)$ ).

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## 2. Fiber Bundle Structure of the Multi-Configuration Hartree-Fock Ansatz

### 2.1. The MCHF ansatz.

For positive integers  $N \leq K$ , let  $\Sigma_{N,K}$  denote the set of increasing mappings  $\sigma : \{1, \dots, N\} \longrightarrow \{1, \dots, K\}$

$$\Sigma_{N,K} = \left\{ \sigma = \{ \sigma(1) < \dots < \sigma(N) \} \subset \{1, \dots, K\} \right\}, \quad \#\Sigma_{N,K} = \binom{K}{N} := r.$$

For simplicity the same notation is used for the mapping  $\sigma$  and its range  $\{ \sigma(1) < \dots < \sigma(N) \}$ . We introduce an open domain  $\Omega$  in  $\mathbb{R}^3$  that is either bounded or  $\mathbb{R}^3$  itself. Actually as in [17] we could as well take into account the spin coordinates and replace everywhere below  $\Omega$  by  $\Omega \times \mathbb{Z}^q$  with  $q \in \mathbb{N}$  being the number of spin states (physically  $q = 2$ ). In that case the space coordinate  $x \in \Omega$  is to be replaced by the spin-space coordinate  $x = (r; s) \in \Omega \times \mathbb{Z}^q$  with  $\int dx = \sum_{s \in \mathbb{Z}^q} \int_{\Omega} dr$ <sup>1</sup>. We define

$$\mathcal{F}_{N,K} := S^{r-1} \times \mathcal{O}_{L^2(\Omega)^K}$$

where

$$\mathcal{O}_{L^2(\Omega)^K} = \left\{ \Phi = (\phi_1, \dots, \phi_K) \in L^2(\Omega)^K : \int_{\Omega} \phi_i \bar{\phi}_j dx = \delta_{i,j} \right\}, \quad (2.1)$$

with  $\delta_{i,j}$  being the Kronecker delta and with  $S^{r-1}$  being the unit sphere in  $\mathbb{C}^r$  endowed with the complex euclidean distance

$$S^{r-1} = \left\{ C = (c_{\sigma})_{\sigma \in \Sigma_{N,K}} \in \mathbb{C}^r : \|C\|^2 = \sum_{\sigma} |c_{\sigma}|^2 = 1 \right\}. \quad (2.2)$$

Here and below we use the shorthand notation  $\sum_{\sigma}$  for  $\sum_{\sigma \in \Sigma_{N,K}}$ . To any  $\sigma \in \Sigma_{N,K}$  and  $\Phi$  in  $\mathcal{O}_{L^2(\Omega)^K}$ , we associate the Slater determinant

$$\Phi_{\sigma}(x_1, \dots, x_N) = \phi_{\sigma(1)} \wedge \dots \wedge \phi_{\sigma(N)} = \frac{1}{\sqrt{N!}} \begin{vmatrix} \phi_{\sigma(1)}(x_1) & \dots & \phi_{\sigma(1)}(x_N) \\ \vdots & & \vdots \\ \phi_{\sigma(N)}(x_1) & \dots & \phi_{\sigma(N)}(x_N) \end{vmatrix}.$$

The mapping

$$(C, \Phi) \longmapsto \Psi = \pi_{N,K}(C, \Phi) = \sum_{\sigma} c_{\sigma} \Phi_{\sigma}. \quad (2.3)$$

is multi-linear, continuous and even infinitely differentiable from  $\mathcal{F}_{N,K}$  equipped with the natural topology of  $\mathbb{C}^r \times L^2(\Omega)^K$  into  $L^2_{\lambda}(\Omega^N)$ . Its range is denoted by

$$\mathcal{B}_{N,K} = \pi(\mathcal{F}_{N,K}) = \left\{ \Psi = \sum_{\sigma} c_{\sigma} \Phi_{\sigma} : (C, \Phi) \in \mathcal{F}_{N,K} \right\}.$$

<sup>1</sup> The authors thank G. Friesecke for this observation

When there is no ambiguity, we simply denote  $\pi = \pi_{N,K}$ . The set  $\mathcal{B}_{N,N}$  is the set of single determinants or *Hartree–Fock states*. Of course  $\mathcal{B}_{N,K} \subset \mathcal{B}_{N,K'}$  when  $K' \geq K$  and actually

$$\lim_{K \rightarrow +\infty} \mathcal{B}_{N,K} = \left\{ \Psi \in L^2_\wedge(\Omega^N) : \|\Psi\| = 1 \right\},$$

in the sense of an increasing sequence of sets, since Slater determinants form an Hilbert basis of  $L^2_\wedge(\Omega^N)$  (see [29]). In particular, for  $\sigma, \tau \in \Sigma_{N,K}$ , we have

$$\langle \Phi_\sigma | \Phi_\tau \rangle = \delta_{\sigma,\tau}. \quad (2.4)$$

Observe that without orthonormality condition the formula (2.4) becomes

$$\langle \phi_1 \wedge \dots \wedge \phi_N | \xi_1 \wedge \dots \wedge \xi_N \rangle = \det(\langle \phi_i | \xi_j \rangle)_{1 \leq i,j \leq N} \quad (2.5)$$

for  $\Phi, \Xi \in L^2(\Omega)^N$  which will be used below (see [29]).

The set corresponding to the multi-configuration ansatz  $\mathcal{B}_{N,K}$  is characterized in Proposition 2.2 in Subsection 2.2 in terms of the so-called first-order density matrix, and its geometric structure is analyzed in Subsection 2.3.

## 2.2. Density Operators

For  $n = 1, \dots, N$  and for  $\Psi \in L^2_\wedge(\Omega^N)$  with  $\|\Psi\| = 1$ , a trace-class self-adjoint operator  $[\Psi \otimes \Psi]_{:n}$ , called  $n^{\text{th}}$  order density operator, is defined on  $L^2_\wedge(\Omega^n)$  through its kernel  $[\Psi \otimes \Psi]_{:n}$

$$[\Psi \otimes \Psi]_{:n}(X_n, Y_n) = \binom{N}{n} \int_{\Omega^{N-n}} \Psi(X_n, Z_n^N) \overline{\Psi}(Y_n, Z_n^N) dZ_n^N, \quad (2.6)$$

for  $1 \leq n \leq N-1$  and

$$[\Psi \otimes \Psi]_{:N}(X_N, Y_N) = \Psi(X_N) \overline{\Psi}(Y_N),$$

with the notation

$$\begin{aligned} X_n &= (x_1, \dots, x_n), & Y_n &= (y_1, \dots, y_n), \\ Z_n^N &= (z_{n+1}, \dots, z_N), & dZ_n^N &= dz_{n+1} \dots dz_N, \end{aligned}$$

and similarly for other capital letters. Our normalization follows Löwdin's [29]. A simple calculation shows that, for  $1 \leq n \leq N-1$ ,

$$[\Psi \otimes \Psi]_{:n}(X_n, Y_n) = \frac{n+1}{N-n} \int_{\Omega} [\Psi \otimes \Psi]_{:n+1}(X_n, z, Y_n, z) dz. \quad (2.7)$$

In particular, given  $1 \leq n \leq p \leq N-1$ , one can deduce the expression of  $[\Psi \otimes \Psi]_{:n}$  from the one of  $[\Psi \otimes \Psi]_{:p}$ . These operators satisfy:

**Proposition 2.1** [1,13,14,29]. *For every integer  $1 \leq n \leq N$ , the  $n$ -th order density matrix is a trace-class self-adjoint operator on  $L^2_\lambda(\Omega^n)$  such that*

$$0 \leq [\Psi \otimes \Psi]_{:n} \leq 1, \quad (2.8)$$

in the sense of operators, and

$$\mathrm{Tr}_{L^2(\Omega^n)} [\Psi \otimes \Psi]_{:n} = \binom{N}{n}.$$

Actually, the multi-configuration ansatz correspond to first-order density matrices with *finite rank*, and we have the following

**Proposition 2.2** (Löwdin's expansion theorem, Lemma 1.1 in [17] and [29]).

Let  $K \geq N$ , then

$$\mathcal{B}_{N,K} = \pi(\mathcal{F}_{N,K}) = \{ \Psi \in L^2_\lambda(\Omega^N) : \|\Psi\| = 1 \text{ and } \mathrm{rank}[\Psi \otimes \Psi]_{:1} \leq K \}.$$

More precisely, if  $\Psi = \pi(C, \Phi)$  with  $(C, \Phi) \in \mathcal{F}_{N,K}$ , then  $\mathrm{rank}[\Psi \otimes \Psi]_{:1} \leq K$  and  $\mathrm{Ran}[\Psi \otimes \Psi]_{:1} \subset \mathrm{Span}\{\phi_1, \dots, \phi_K\}$ . If  $\Psi \in \mathcal{B}_{N,K}$  and if  $\mathrm{rank}[\Psi \otimes \Psi]_{:1} = K'$  with  $N \leq K' \leq K$  and with  $\{\phi_1, \dots, \phi_{K'}\}$  being an orthonormal basis of  $\mathrm{Ran}[\Psi \otimes \Psi]_{:1}$ , then  $\Psi$  can be expanded as a linear combination of Slater determinants built from  $\{\phi_1, \dots, \phi_{K'}\}$ . The first-order (or one-particle) density matrix  $[\Psi \otimes \Psi]_{:1}$  is often denoted by  $\gamma_\Psi$  in the literature and in the course of this paper. According to Proposition 2.1 above it is a non-negative self-adjoint trace-class operator on  $L^2(\Omega)$ , with trace  $N$  and with operator norm less or equal to 1. Therefore its sequence of eigenvalues  $\{\gamma_i\}_{i \geq 1}$  satisfies  $0 \leq \gamma_i \leq 1$ , for all  $i \geq 1$ , and  $\sum_{i \geq 1} \gamma_i = N$ . In particular, at least  $N$  of the  $\gamma_i$ 's are not zero, and therefore  $\mathrm{rank} \gamma_\Psi \geq N$ , for any  $\Psi \in L^2_\lambda(\Omega^N)$ .

Similarly, if  $\Psi = \pi(C, \Phi) \in \mathcal{B}_{N,K}$ , the range of the operator  $[\Psi \otimes \Psi]_{:n}$  is  $\otimes_n \mathrm{Span}\{\Phi\}$  and its kernel is  $\left(\otimes_n \mathrm{Span}\{\Phi\}\right)^\perp$  with

$$\mathrm{Span}\{\Phi\} := \mathrm{Span}\{\phi_1, \dots, \phi_K\}.$$

Therefore the operator is represented by an Hermitian matrix in  $\otimes_n \mathrm{Span}\{\Phi\}$  whose entries turn out to depend only on the coefficients  $C$  and the dependence is quadratic. For the first- and second-order density operators we have the explicit expressions

**Proposition 2.3** ([17], Appendix 1). *Let  $\Psi = \pi(C, \Phi)$  in  $\mathcal{B}_{N,K}$ , then the operator kernel of the second-order density matrix is given by*

$$[\Psi \otimes \Psi]_{:2}(x, y, x', y') = \sum_{i,j,k,l=1}^K \gamma_{ijkl} \phi_i(x) \phi_j(y) \bar{\phi}_k(x') \bar{\phi}_l(y') \quad (2.9)$$

with

$$\gamma_{ijkl} = \frac{1}{2} (1 - \delta_{i,j})(1 - \delta_{k,l}) \sum_{\substack{\sigma, \tau | i, j \in \sigma, k, l \in \tau \\ \sigma \setminus \{i, j\} = \tau \setminus \{k, l\}}} (-1)_{i,j}^\sigma (-1)_{k,l}^\tau c_\sigma \bar{c}_\tau, \quad (2.10)$$

where for  $i \neq j$ ,

$$(-1)_{i,j}^{\sigma} = \frac{i-j}{|i-j|} (-1)^{\sigma^{-1}(i)+\sigma^{-1}(j)}.$$

Similarly, the kernel of the first-order density matrix is given by the formula

$$[\Psi \otimes \Psi]_{:1}(x, y) = \sum_{i,j=1}^K \gamma_{ij} \phi_i(x) \bar{\phi}_j(y)$$

with

$$\gamma_{ij} = \frac{2}{N-1} \sum_{k=1}^K \gamma_{ikjk} = \sum_{\substack{\sigma, \tau | i \in \sigma, j \in \tau \\ \sigma \setminus \{i\} = \tau \setminus \{j\}}} (-1)^{\sigma^{-1}(i)+\tau^{-1}(j)} c_{\sigma} \bar{c}_{\tau} \quad (2.11)$$

and

$$\gamma_{ii} = \sum_{\sigma | i \in \sigma} |c_{\sigma}|^2. \quad (2.12)$$

The first-order density matrix allows to characterize the set  $\mathcal{B}_{N,K}$  (see Proposition 2.2 above) whereas the second-order density matrix is needed to express expectation values of the energy Hamiltonian as soon as two-body interactions are involved.

Since the coefficients  $\gamma_{ij}$  only depend on  $C$ , we denote by  $\mathbf{\Gamma}(C)$  the  $K \times K$  Hermitian matrix with entries  $\bar{\gamma}_{ij}$ ,  $1 \leq i, j \leq K$  (the adjoint of the matrix representation of the first-order density operator in  $\text{Span}\{\Phi\}$ ). The matrix  $\mathbf{\Gamma}(C)$  is positive, hermitian and of trace  $N$  with the same eigenvalues as  $\gamma_{\Psi}$  and the same rank, and there exists a unitary  $K \times K$  matrix  $U$  such that  $U \mathbf{\Gamma}(C) U^* = \text{diag}(\gamma_1, \dots, \gamma_K)$  with  $0 \leq \gamma_k \leq 1$  and  $\sum_{k=1}^K \gamma_k = N$ . Hence,  $\gamma_{\Psi}$  can also be expanded as follows

$$\gamma_{\Psi}(x, y) = \sum_{i=1}^K \gamma_i \phi'_i(x) \bar{\phi}'_i(y), \quad (2.13)$$

where  $\Phi' = U \cdot \Phi$  with obvious notation and with  $\{\phi'_1, \dots, \phi'_K\}$  being an eigenbasis of  $\gamma_{\Psi}$ . Note that it is easily recovered from (2.12) that  $0 \leq \gamma_i \leq 1$  for  $C \in S^{r-1}$ .

**Remark 1.** When  $K = N$  (Hartree-Fock case),  $\gamma_{\Psi}$  being of trace  $N$  must be the projector on  $\text{Span}\{\Phi\}$ ; that is

$$\gamma_{\Psi}(x, y) = \sum_{i=1}^N \phi_i(x) \bar{\phi}_i(y) := \mathbf{P}_{\Phi}(x, y),$$

with  $\mathbf{P}_{\Phi}$  denoting the projector on  $\text{Span}\{\Phi\}$ . In this case, (2.11) and (2.10) simply reduce to  $\gamma_{ij} = \delta_{i,j}$  and  $\gamma_{ijkl} = \frac{1}{2}(\delta_{i,k}\delta_{j,l} - \delta_{i,l}\delta_{j,k})$ ; that is  $\mathbf{\Gamma}(C) = \mathbb{I}_N$ .

The representation of a wave-function  $\Psi \in \mathcal{B}_{N,K}$  in terms of expansion coefficients  $C$  and orbitals  $\Phi$  is obviously not unique as it is already seen on the Hartree-Fock ansatz. Indeed, if  $\Psi^{HF} = \phi_1 \wedge \dots \wedge \phi_N = \psi_1 \wedge \dots \wedge \psi_N$ , there exists a unique  $N \times N$  unitary transform  $U$  such that  $(\phi_1, \dots, \phi_N) = (\psi_1, \dots, \psi_N) \cdot U$ . The pre-image of  $\Psi^{HF}$  by  $\pi$  in  $\mathcal{F}_{N,N}$  is the orbit of  $(\phi_1, \dots, \phi_N)$  under the action of  $\mathcal{O}_N$ , with  $\mathcal{O}_{\ell}$  being the set of  $\ell \times \ell$  unitary matrices. In the general case under the full-rank assumption the set  $\mathcal{B}_{N,K}$  has a similar orbit-like structure as explained now.

### 2.3. Full-rank and fibration

We introduce

$$d\mathcal{B}_{N,K} := \{\Psi \in \mathcal{B}_{N,K} : \text{rank } \gamma_\Psi = K\}$$

and, by analogy,

$$d\mathcal{F}_{N,K} = \pi_{N,K}^{-1}(d\mathcal{B}_{N,K}) := \{(C, \Phi) \in \mathcal{F}_{N,K} : \text{rank } \Gamma(C) = K\}.$$

$d\mathcal{F}_{N,K}$  is the open subset of  $\mathcal{F}_{N,K}$  corresponding to invertible  $\Gamma(C)$ 's (*full-rank assumption*).

Clearly  $d\mathcal{B}_{N,N} = \mathcal{B}_{N,N}$  and  $d\mathcal{F}_{N,N} = \mathcal{F}_{N,N}$ ; that is, the full-rank assumption is automatically satisfied in the Hartree–Fock setting (see Remark 1).

On the other hand, it may happen that  $d\mathcal{B}_{N,K}$  is empty (in that case  $\mathcal{B}_{N,K} = \mathcal{B}_{N,K-1}$ ). Indeed, for  $K \geq N$  the admissible ranks of first-order density matrices must satisfy the relations [17, 28]

$$K \begin{cases} = 1 & N = 1 \\ \geq 2, \text{ even} & N = 2 \\ \geq N, \neq N+1, N \geq 3. \end{cases}$$

From now on, we only deal with pairs  $(N, K)$  with  $K$  admissible. We recall from [29] the following

**Proposition 2.4.** *Let  $(C, \Phi)$  and  $(C', \Phi')$  in  $d\mathcal{F}_{N,K}$  such that  $\pi(C, \Phi) = \pi(C', \Phi')$ . Then, there exists a unique unitary matrix  $U \in \mathcal{O}_K$  and a unique unitary matrix  $d(U) = \bar{\mathbb{U}} \in \mathcal{O}_r$  such that*

$$\Phi' = U \cdot \Phi, \quad C' = d(U) \cdot C$$

where, for every  $\sigma \in \Sigma_{N,K}$ ,

$$\Phi'_\sigma = \sum_{\tau} \mathbb{U}_{\sigma,\tau} \Phi_\tau.$$

Moreover,

$$\Gamma(C') = U \Gamma(C) U^*. \quad (2.14)$$

**Proof.** Let  $(C, \Phi)$  and  $(C', \Phi')$  in  $d\mathcal{F}_{N,K}$  such that  $\pi(C, \Phi) = \pi(C', \Phi') = \Psi \in d\mathcal{B}_{N,K}$ . From Proposition 2.2,  $\text{Span}\{\Phi\} = \text{Span}\{\Phi'\} = \text{Ran}(\gamma_\Psi)$  with  $\Phi$  and  $\Phi'$  in  $\mathcal{O}_{L^2(\Omega)^K}$ . Therefore, there exists a unique unitary matrix  $U \in \mathcal{O}_K$  such that  $\Phi' = U \cdot \Phi$ . Eqn. (2.14) follows by definition of  $\Gamma(C)$ . Accordingly, there exists a unique unitary matrix  $\mathbb{U}$  in  $\mathcal{O}_r$  that maps the family  $\{\Phi_\tau\}_{\tau \in \Sigma_{N,K}}$  to  $\{\Phi'_\sigma\}_{\sigma \in \Sigma_{N,K}}$ . More precisely, being given  $\sigma \in \Sigma_{N,K}$ , we have by a direct calculation (see also [29])

$$\Phi'_\sigma = \sum_{\tau} \mathbb{U}_{\sigma,\tau} \Phi_\tau \quad (2.15)$$

where, for all  $\sigma, \tau \in \Sigma_{N,K}$ ,

$$\begin{aligned} \mathbb{U}_{\sigma,\tau} &= \begin{vmatrix} U_{\sigma(1),\tau(1)} & \cdots & U_{\sigma(N),\tau(1)} \\ \vdots & & \vdots \\ U_{\sigma(1),\tau(N)} & \cdots & U_{\sigma(N),\tau(N)} \end{vmatrix} = \det(U_{\sigma(j),\tau(i)})_{1 \leq i,j \leq N} \\ &= \det(\langle \phi'_{\sigma(j)}; \phi_{\tau(i)} \rangle)_{1 \leq i,j \leq N}. \end{aligned} \quad (2.16)$$

By construction the  $r \times r$  matrix  $\mathbb{U}$  with matrix elements  $\mathbb{U}_{\sigma,\tau}$  is unitary. By the orthonormality of the determinants, we have

$$c'_\sigma = \langle \pi(C, \Phi) | \Phi'_\sigma \rangle = \sum_\tau c_\tau \langle \Phi_\tau | \Phi'_\sigma \rangle = \sum_\tau \overline{\mathbb{U}_{\sigma,\tau}} c_\tau, \quad (2.17)$$

hence the lemma holds with  $d(U) = \overline{\mathbb{U}}$ .

Under the full-rank assumption and given  $(N, K)$  admissible, the set  $d\mathcal{B}_{N,K}$  is a principal fiber bundle. In the terminology of differential geometry,  $d\mathcal{B}_{N,K}$  is called *the base*, and, for any  $\Psi \in d\mathcal{B}_{N,K}$ , the pre-image  $\pi^{-1}(\Psi)$  is *the fiber over*  $\Psi$ . Proposition 2.4 defines a transitive group action on  $d\mathcal{F}_{N,K}$  according to

$$\begin{aligned} (C', \Phi') &= \mathcal{U} \cdot (C, \Phi) \iff C' = d(U) \cdot C \quad \text{and} \quad \Phi' = U \cdot \Phi, \\ \mathcal{U} &:= (d(U), U) \in \mathcal{O}_r \times \mathcal{O}_K. \end{aligned} \quad (2.18)$$

Indeed on the one hand, it is clear from the expression for the matrix elements of  $d(U)$  that  $d(\mathbb{I}_K) = \mathbb{I}_r$ . On the other hand from (2.15) and (2.17) it is easily checked that  $d(UV) = d(U)d(V)$ . Therefore couples of the form  $(d(U), U)$  form a subgroup of  $\mathcal{O}_r \times \mathcal{O}_K$  that we denote by  $\mathcal{O}_K^r$ . The action of  $\mathcal{O}_K^r$  is not free on  $\mathcal{F}_{N,K}$  itself — this is illustrated in Remark 2 below on the examples of Slater determinants in  $\mathcal{F}_{N,K}$  with  $K > N$  —, but it is free on  $d\mathcal{F}_{N,K}$  and transitive over any fiber  $\pi^{-1}(\Psi)$  for every  $\Psi \in d\mathcal{B}_{N,K}$ . Therefore, the mapping  $\pi$  defines a principal bundle with fiber given by the group  $\mathcal{O}_K^r$ . We can define local (cross-)sections as continuous maps  $s : \Psi \mapsto (C, \Phi)$  from  $d\mathcal{B}_{N,K}$  to  $d\mathcal{F}_{N,K}$  such that  $\pi \circ s$  is the identity. In particular,  $d\mathcal{F}_{N,K} / \mathcal{O}_K^r$  is homeomorphic to  $d\mathcal{B}_{N,K}$ . Since the map  $\pi$  is  $C^\infty$ , one concludes from the inverse mapping theorem that the above isomorphism is also topological. In the Hartree–Fock case  $K = N$  where the full-rank assumption is automatically fulfilled,  $\pi_{N,N}^{-1}(\mathcal{B}_{N,N})$  is a so-called Stiefel manifold.

**Remark 2.** The following example illustrates the necessity of the full-rank assumption. As

$$K \leq K' \implies \mathcal{B}_{N,K} \subset \mathcal{B}_{N,K'},$$

any Slater determinant  $\Psi^{HF} = \phi_1 \wedge \cdots \wedge \phi_N$  can also be seen as an element of  $\mathcal{B}_{N,K}$  for all  $K \geq N$ . If  $K > N$ , the pre-image of  $\Psi^{HF}$  by  $\pi$  in  $\mathcal{F}_{N,K}$  does not have a similar orbit structure as shown on the following example. Let  $C = (1, 0, \dots, 0) \in S^{r-1}$  where all coordinates but the first one are 0 and let  $\Phi' = (\phi_1, \dots, \phi_N, \phi_{N+1}, \dots, \phi_K) \in \mathcal{O}_{L^2(\Omega)^K}$  with  $\phi_i \in \text{Span}\{\phi_1, \dots, \phi_N\}^\perp$  for every  $N+1 \leq i \leq K$ , then  $(C, \Phi') \in \mathcal{F}_{N,K}$

and  $\Psi_{HF} = \pi(C, \Phi)$ . Then the set  $\left(\text{Span}\{\phi_1, \dots, \phi_N\}^\perp\right)^{K-N}$  has no group-orbit structure.

Having equipped  $d\mathcal{B}_{N,K}$  with a manifold structure we turn to the study of its tangent space.

Being multi-linear with respect to the variables  $C$  and  $\Phi$ , the map  $\pi$  is clearly infinitely differentiable. Its gradients

$$\begin{aligned} \nabla \pi : \mathbb{C}^r \times L^2(\Omega)^K &\longrightarrow \mathcal{L}(\mathbb{C}^r; L^2(\Omega^N)) \times \mathcal{L}(L^2(\Omega)^K; L^2(\Omega^N)) \\ \Psi = \pi(C, \Phi) &\longmapsto \nabla \Psi = (\nabla_C \Psi, \nabla_\Phi \Psi) \end{aligned}$$

are computed as follows for every  $(C, \Phi) \in \mathcal{F}_{N,K}$ :

i) for any  $\delta C$  in  $\mathbb{C}^r$ ,

$$\nabla_C \Psi[\delta C] = \sum_{k=1}^r \delta c_k \frac{\partial \Psi}{\partial c_{\sigma_k}} = \sum_{k=1}^r \delta c_k \Phi_{\sigma_k}, \quad (2.19)$$

with  $\Sigma_{N,K} = \{\sigma_1, \dots, \sigma_r\}$ ,

ii) for any  $\zeta = (\zeta_1, \dots, \zeta_K) \in L^2(\Omega)^K$ ,

$$\nabla_\Phi \Psi[\zeta] = \sum_{k=1}^K \frac{\partial \Psi}{\partial \phi_k}[\zeta_k] = \sum_{\sigma \in \Sigma_{N,K}} c_\sigma \sum_{k=1}^K \frac{\partial \Phi_\sigma}{\partial \phi_k}[\zeta_k], \quad (2.20)$$

with

$$\frac{\partial \Psi}{\partial \phi_k}[\zeta_j] = \sum_{i=1}^N \zeta_j(x_i) \int_\Omega \Psi(x_1, \dots, x_N) \bar{\phi}_k(x_i) dx_i. \quad (2.21)$$

**Remark 3.** For every  $\sigma \in \Sigma_{N,K}$  and every  $1 \leq k \leq K$ , we have

$$\frac{\partial \Phi_\sigma}{\partial \phi_k}[\zeta] = \begin{cases} \phi_{\sigma(1)} \wedge \dots \wedge \phi_{\sigma(j-1)} \wedge \zeta \wedge \phi_{\sigma(j+1)} \wedge \dots \wedge \phi_{\sigma(N)} & \text{if } \sigma^{-1}(k) = j, \\ 0 & \text{if } k \notin \sigma. \end{cases} \quad (2.22)$$

**Remark 4.** From (2.22) we recover the Euler Formula for homogeneous functions, that reads here

$$\Psi = \frac{1}{N} \sum_{k=1}^K \frac{\partial \Psi}{\partial \phi_k}[\phi_k] := \frac{1}{N} \nabla_\Phi \Psi[\Phi]. \quad (2.23)$$

From the definition of the adjoint  $\nabla_\Phi \Psi^* \in \mathcal{L}(L^2_\lambda(\Omega^N); L^2(\Omega)^K)$  of the operator  $\nabla_\Phi \Psi$  one has for all  $\zeta \in L^2(\Omega)^K$  and  $\Xi \in L^2_\lambda(\Omega^N)$

$$\langle \nabla_\Phi \Psi^*[\Xi]; \zeta \rangle_{L^2(\Omega)^K} = \left\langle \Xi \mid \nabla_\Phi \Psi[\zeta] \right\rangle_{L^2(\Omega^N)}, \quad (2.24)$$

with

$$\frac{\partial \Psi^*}{\partial \phi_k}[\Xi](x) = N \int_\Omega \phi_k(y) \left( \int_{\Omega^{N-1}} \Xi(x, x_2, \dots, x_N) \bar{\Psi}(y, x_2, \dots, x_N) dx_2 \dots dx_N \right) dy$$

for all  $1 \leq k \leq K$ , for any function  $\Xi$  in  $L^2_\lambda(\Omega^N)$ .

It is also worth emphasizing the fact that changing  $(C, \Phi)$  to  $(C', \Phi')$  following the group action (2.18), involves a straightforward change of “variable” in the derivation of  $\Psi$ ; namely, with a straightforward chain rule,

$$\nabla_C \Psi = \mathbb{U}^* \cdot \nabla_{C'} \Psi = \nabla_{C'} \Psi \cdot d(U), \quad \nabla_\Phi \Psi = \nabla_{\Phi'} \Psi \cdot U \quad (2.25)$$

and

$$[\nabla_{\Phi'} \Psi]^* = U \cdot [\nabla_\Phi \Psi]^*. \quad (2.26)$$

The following further properties of the functional derivatives of  $\Psi$  will help to link the full-rank assumption with the possibility for  $\pi$  to be a local diffeomorphism in a neighborhood of  $\Psi_0 = \pi(C_0, \Phi_0) \in d\mathcal{B}_{N,K}$ .

**Lemma 2.5.** *Let  $(C, \Phi) \in \mathcal{F}_{N,K}$  with  $\Psi = \pi(C, \Phi)$ . Then, for all  $\zeta \in \text{Span}\{\Phi\}^\perp$ ,  $\xi \in L^2(\Omega)$  and  $\sigma, \tau \in \Sigma_{N,K}$ , we have*

$$\left\langle \frac{\partial \Phi_\tau}{\partial \phi_k} [\zeta] \middle| \Phi_\sigma \right\rangle = 0, \quad (2.27)$$

and

$$\left\langle \frac{\partial \Psi}{\partial \phi_k} [\zeta] \middle| \frac{\partial \Psi}{\partial \phi_l} [\xi] \right\rangle = \mathbf{\Gamma}_{lk} \langle \zeta, \xi \rangle, \quad (2.28)$$

for any  $1 \leq k, l \leq K$ .

**Proof.** The first claim follows immediately in virtue of (2.22) and (2.5). For the second claim we proceed as follows. Thanks to (2.22) again

$$\begin{aligned} \left\langle \frac{\partial \Psi}{\partial \phi_k} [\zeta] \middle| \frac{\partial \Psi}{\partial \phi_l} [\xi] \right\rangle &= \sum_{\sigma, \tau \mid k \in \sigma, l \in \tau} c_\sigma \bar{c}_\tau \left\langle \frac{\partial \Phi_\sigma}{\partial \phi_k} [\zeta] \middle| \frac{\partial \Phi_\tau}{\partial \phi_l} [\xi] \right\rangle \\ &= \sum_{\substack{\sigma, \tau \mid k \in \sigma, l \in \tau \\ \sigma \setminus \{k\} = \tau \setminus \{l\}}} (-1)^{\sigma^{-1}(k)} (-1)^{\tau^{-1}(l)} c_\sigma \bar{c}_\tau \langle \zeta, \xi \rangle \\ &= \mathbf{\Gamma}_{lk} \langle \zeta, \xi \rangle. \end{aligned}$$

We conclude with the help of (2.11).

From (2.23), (2.19), (2.20) and (2.21), the tangent space of  $d\mathcal{B}_{N,K}$  at  $\Psi = \pi(C, \Phi)$  is given by

$$\begin{aligned} \mathbf{T}_\Psi d\mathcal{B}_{N,K} &= \left\{ \delta \Psi = \sum_\sigma \Phi_\sigma \delta c_\sigma + \frac{1}{N} \sum_\sigma \sum_{k=1}^K c_\sigma \frac{\partial \Phi_\sigma}{\partial \phi_k} [\delta \phi_k] \in L^2_\lambda(\Omega^N) : \right. \\ &\quad \left. \delta C = (\delta c_{\sigma_1}, \dots, \delta c_{\sigma_r}) \in \mathbb{C}^r, \delta \phi_k \in \text{Span}\{\Phi\}^\perp, \forall 1 \leq k \leq K \right\}. \end{aligned} \quad (2.29)$$

Note that the tangent space (2.29) only depends on the basis point  $\Psi$  and not on the choice of coordinates  $(C, \Phi)$  in the corresponding fiber. In the terminology of physics this is the space of allowed variations around  $(C, \Phi)$  in  $\mathcal{F}_{N,K}$  according

to the constraints (2.1) and (2.2) on the expansion coefficients and the orbitals respectively.

Let  $\Psi_0 = \pi(C_0, \Phi_0)$  be in  $\mathcal{B}_{N,K}$  with invertible  $\Gamma(C_0)$ . Then the local mapping theorem at  $(C_0, \Phi_0)$  allows to define a so-called *section*  $\pi^{-1} : \Psi \mapsto (C, \Phi)$  as a  $C^1$  diffeomorphism in the neighborhood of  $\Psi_0$ . According to (2.29), we have to check that  $(0, 0)$  is the only solution in  $\mathbb{C}^r \times \text{Span}\{\Phi_0\}^\perp$  to

$$d\pi_{(C_0, \Phi_0)}(\delta C, \delta \Phi) = \sum_{\sigma} \Phi_{\sigma} \delta c_{\sigma} + \frac{1}{N} \sum_{k=1}^K \frac{\partial \Psi}{\partial \phi_k} [\delta \phi_k] = 0. \quad (2.30)$$

Indeed, on the one hand, if we scalar product the above equation with  $\Phi_{\tau}$  for any  $\tau \in \Sigma_{N,K}$  we obtain  $\delta C = 0$  in virtue of the orthonormality of Slater determinants and (2.27). On the other hand, for a given  $1 \leq l \leq K$  and any  $\xi \in L^2(\Omega)$ , the scalar product of (2.30) with  $\frac{\partial \Psi}{\partial \phi_l} [\xi]$  yields

$$\sum_{k=1}^K \left\langle \frac{\partial \Psi}{\partial \phi_k} [\delta \phi_k] \mid \frac{\partial \Psi}{\partial \phi_l} [\xi] \right\rangle = \sum_{k=1}^K \Gamma_{lk} \langle \delta \phi_k, \xi \rangle = \left\langle (\Gamma \delta \Phi)_l, \xi \right\rangle = 0$$

thanks to (2.28). Since  $\xi$  is arbitrary in  $L^2$  and since  $\Gamma$  is invertible this is equivalent to  $\delta \Phi = 0$ , hence the result. The full-rank property is mandatory for lifting continuous paths  $t \mapsto \Psi(t)$  on the basis  $d\mathcal{B}_{N,K}$  to continuous paths  $t \mapsto (C(t), \Phi(t))$  on  $d\mathcal{F}_{N,K}$ .

#### 2.4. Interpretation in terms of quantum physics

The wave-function  $\Psi \in L^2(\Omega^N)$  with  $\|\Psi\| = 1$  is interpreted through the square of its modulus  $|\Psi(X_N)|^2 (= [\Psi \otimes \Psi]_{:,N}(X_N, X_N))$  that represents the density of probability of positions of the  $N$  electrons in  $\Omega^N$ . More generally, for all  $1 \leq n \leq N$ , the positive function  $X_n \mapsto [\Psi \otimes \Psi]_{:,n}(X_n, X_n)$  is in  $L^1(\Omega^n)$  with  $L^1$  norm equal to  $\binom{N}{n}$ , and it is interpreted as  $\binom{N}{n}$  times the density of probability for finding  $n$  electrons located at  $X_n \in \Omega^n$ . Any set  $\{\sigma(1), \dots, \sigma(N)\}$  for  $\sigma \in \Sigma_{N,K}$  is called a *configuration* in quantum chemistry literature and this is where the terminology *multi-configuration* comes from for wave-functions in  $\mathcal{B}_{N,K}$ . When  $\{\phi_k\}_{1 \leq k \leq K}$  is an orthonormal basis of  $\text{Ran}[\Psi \otimes \Psi]_{:,1}$  each mono-electronic function  $\phi_k$  is called an *orbital* of  $\Psi$ . When the orbitals are also eigenfunctions of  $[\pi(C, \Phi) \otimes \pi(C, \Phi)]_{:,1}$  according to (2.13) they are referred to as *natural orbitals* in the literature whereas the associated eigenvalues  $\{\gamma_i\}_{1 \leq i \leq K}$  are referred to as *occupation numbers*. Under the full-rank assumption, only occupied orbitals are taken into account. The functions with  $N-1$  variables  $\int_{\Omega} \Psi(x_1, \dots, x_N) \bar{\phi}_k(x_i) dx_i$  that appear in formula (2.21) are known as a *single-hole function* (see e.g. [5, 7]). Finally, the  $K \times K$  matrix  $\Gamma(C)$  is called *the charge- and bond matrix* (see Löwdin [29]).

“Correlation” is a key concept for many-particle systems. The intrinsic correlation of a many particle wave-function as such is a rather vague concept, with several different definitions in the literature (see among others [22, 20, 21] and the references therein). The case of a single Slater determinant is usually taken as

the definition of a wave-function with zero correlation. Hence the corresponding Hartree-Fock ansatz is not able to catch “correlation effects”. The free Schrödinger equation propagates Slater determinants (see Subsection 3.5). However, the interaction of the particles would immediately create “correlations” in the time evolution even if the initial data is a single Slater determinant. The TDHF method forces the dynamics to stay on a manifold where correlation is always zero.

Improving the approximation systematically by adding determinants brings in correlation into the *multi-configuration ansatz*. Correlation effects of the many particle wave-function can be included in the initial data and the effects of dynamical “correlation - decorrelation” can be caught in the time evolution. This is a very important conceptual advantage of MCTDHF for the modeling and simulation of correlated few electron systems. Such systems, for example in “photonics” where an atom interacting with an intense laser is measured on the femto- or attosecond scale, are increasingly studied and have given a boost to MCTDHF (see e.g. [7],[2]).

### 3. Flow on the Fiber Bundle

In this section, we consider a general self-adjoint operator  $\mathcal{H}$  in  $L^2(\Omega^N)$  with domain  $\mathcal{D}(\mathcal{H})$  whenever it is unbounded. From this point onward,  $T > 0$  is fixed. Solutions  $\Psi(t)$  are meant in the strong sense :  $\Psi(t) \in C^0(0, T; \mathcal{D}(\mathcal{H})) \cap C^1(0, T; L^2(\Omega^N))$ . Therefore all calculations below can be performed rigorously. In Section 4 below physical problems will be considered and details concerning proof of existence, uniqueness of solutions and blow-up alternatives in the appropriate functional spaces will be given.

A key point of the time-dependent case is that the set corresponding to the ansatz  $\mathcal{B}_{N,K}$  is not invariant by the Schrödinger dynamics. It is even expected (but so far not proved to our knowledge) that the solution of the exact Schrödinger equation (1.2) with initial data in  $\mathcal{B}_{N,K}$  for some finite  $K \geq N$  captures an infinite rank at any positive time as long as many-body potentials are involved (see [18] for related issues on the stationary solutions and Subsection 3.5 for the picture for non-interacting electrons). We therefore have to rely on an approximation procedure that forces the solutions to stay on the set corresponding to the ansatz for all time. In the physics literature, the MCTDHF equations are usually (formally) derived from the so-called *Dirac-Frenkel variational principle* (see, among others, [15, 16, 26] and the references therein) that demands that for all  $t \in [0, T]$ ,  $\Psi = \Psi(t) \in \mathcal{B}_{N,K}$ ,  $\Psi \in C^0(0, T; \mathcal{D}(\mathcal{H})) \cap C^1(0, T; L^2(\Omega^N))$ , and

$$\left\langle i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \mid \delta \Psi \right\rangle = 0, \quad \text{for all } \delta \Psi \in \mathbf{T}_\Psi d\mathcal{B}_{N,K}, \quad (3.1)$$

where  $\mathbf{T}_\Psi d\mathcal{B}_{N,K}$  denotes the tangent space to the differentiable manifold  $d\mathcal{B}_{N,K}$  at  $\Psi$ . Equivalently, one solves

$$\Psi(t) = \operatorname{argmin} \left\{ \left\| i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \right\|_{L^2(0, T; L^2(\Omega^N))} : \Psi \in \mathcal{B}_{N,K} \right\} \quad (3.2)$$



Actually one checks that

$$\nabla_{\Phi} \Psi^* [\mathcal{H} \Psi] = \nabla_{\bar{\Phi}} \langle \mathcal{H} \Psi | \Psi \rangle.$$

Up to the Lagrange multipliers associated to (3.3) the right-hand side in the variational system corresponds to the Fréchet derivatives of the energy expectation  $\mathcal{E}(\Psi) = \langle \mathcal{H} \Psi | \Psi \rangle$  with respect to the conjugate (independent) variables  $\bar{C}$  and  $\bar{\Phi}$ .

The variational system  $\mathcal{S}_0$  is well-suited for checking energy conservation and constraints propagation over the flow as shown in Subsection 3.1 below. However it is badly adapted for proving existence of solutions for the Cauchy problem or for designing numerical codes. Equivalent representations of the MCTDHF equations over different fibrations is made rigorous in Subsection 3.3. In particular, we prove below that the variational system is unitarily (or gauge-) equivalent to System (3.26) – named *working equations* – whose mathematical analysis in the physical case is the aim of Section 4.

**Remark 5.** Since for every  $\sigma \in \Sigma_{N,K}$ ,  $\frac{\partial \Psi}{\partial c_\sigma} = \Phi_\sigma$ , the system for the  $c_\sigma$ 's can also be expressed as

$$i \frac{dc_\sigma}{dt} = \sum_{\tau} \langle \mathcal{H} \Phi_\tau | \Phi_\sigma \rangle c_\tau. \quad (3.5)$$

This equation is then obviously linear in the expansion coefficients. Furthermore, when the  $\phi_i$ 's (or equivalently the  $\Phi_\sigma$ 's) are kept constant in time, (3.5) is nothing but a Galerkin approximation to the exact Schrödinger equation (1.2). The MCTDHF approximation then reveals as a generalization to a combination of time-dependent basis functions (with extra degree of freedom in the basis functions) of the Galerkin approximation.

### 3.1. Conservation Laws

In this subsection, we assume the full-rank assumption on the time interval  $[0, T]$ ; that is  $\mathbf{\Gamma}(C(t))$  is invertible for every  $t \in [0, T]$ . We check here that the expected conservation laws (propagation of constraints, conservation of the energy) are granted by the variational system. Recall that to avoid technicalities all calculations in this section are formal but would be rigorous for strong solutions  $(C(t); \Phi(t))$  with  $C \in C^1(0, T; \mathbb{C}^r)$  and with  $\Phi$  such that  $\Psi(t) = \pi(C(t); \Phi(t)) \in C^0(0, T; \mathcal{D}(\mathcal{H})) \cap C^1(0, T; L^2(\Omega^N))$ . We start with the following

**Lemma 3.1 (The dynamics preserves  $\mathcal{F}_{N,K}$ ).** *Let  $(C_0, \Phi_0) \in \mathcal{F}_{N,K}$  being the initial data. If there exists a solution to the system  $\mathcal{S}_0$  on  $[0, T]$  such that  $\text{rank } \mathbf{\Gamma}(C(t)) = K$  for all  $t \in [0, T]$ , then*

$$\sum_{\sigma} |c_\sigma(t)|^2 = 1, \quad \int_{\mathbb{R}^3} \phi_i(t) \bar{\phi}_j(t) dx = \delta_{i,j}, \quad \text{for all } t \in [0, T].$$

**Proof.** First we prove that  $\sum_{\sigma} |c_{\sigma}(t)|^2 = \sum_{\sigma} |c_{\sigma}(0)|^2$  for all  $t \in [0, T]$ . By taking the scalar product of the differential equation satisfied by  $C$  in  $\mathcal{S}_0$  with  $C$  itself, we get

$$\frac{d}{dt} |C(t)|^2 = 2 \Re \left( \frac{d}{dt} C(t), C(t) \right) = 2 \Im \sum_{\sigma} \left\langle \mathcal{H} \Psi | c_{\sigma} \Phi_{\sigma} \right\rangle = 2 \Im \left\langle \mathcal{H} \Psi | \Psi \right\rangle = 0,$$

thanks to the self-adjointness of  $\mathcal{H}$ , where  $\Re$  and  $\Im$  denote respectively real and imaginary parts of a complex number. From the other hand, the full-rank assumption allows to reformulate the second equation in  $(\mathcal{S}_0)$  as

$$i \frac{\partial \Phi}{\partial t} = (\mathbf{I} - \mathbf{P}_{\Phi}) \Gamma(C)^{-1} \nabla_{\Phi} \Psi^* [\mathcal{H} \Psi]. \quad (3.6)$$

(Notice that  $\mathbf{P}_{\Phi}$  commutes with  $\Gamma(C)^{-1}$ .) By definition  $\mathbf{I} - \mathbf{P}_{\Phi}$  projects on the orthogonal subspace of  $\text{Span}\{\Phi\}$ , therefore  $\frac{\partial}{\partial t} \phi_i$  lives in  $\text{Span}\{\Phi\}^{\perp}$  for all  $t$ . Hence,

$$\left\langle \frac{\partial \phi_i(t)}{\partial t}, \phi_j(t) \right\rangle = 0. \quad (3.7)$$

for all  $1 \leq i, j \leq K$  and for all  $t \in [0, T]$ . This achieves the proof of the lemma.

We now check that solutions to the variational system indeed agree with the Dirac-Frenkel variational principle.

**Proposition 3.2 (Link with the Dirac-Frenkel variational principle).** *Let  $(C, \Phi) \in d\mathcal{F}_{N,K}$  be a strong solution to  $\mathcal{S}_0$  on  $[0, T]$ . Then,  $\Psi = \pi(C, \Phi)$  satisfies the Dirac-Frenkel variational principle (3.1).*

**Proof.** We start with the characterization (2.29) of the elements in  $\mathbf{T}_{\Psi} d\mathcal{B}_{N,K}$ . Since the full-rank assumption is satisfied on  $[0, T]$ , the orbitals satisfy (3.6), and therefore  $\frac{\partial \phi_k}{\partial t} \in \text{Span}\{\Phi\}^{\perp}$  for all  $t \in [0, T]$  and  $1 \leq k \leq K$ . Firstly, being given  $\sigma \in \Sigma_{N,K}$ , we have

$$\begin{aligned} \left\langle i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \middle| \frac{\partial \Psi}{\partial c_{\sigma}} \right\rangle &= i \sum_{\tau} \frac{dc_{\tau}}{dt} \left\langle \Phi_{\tau} | \Phi_{\sigma} \right\rangle - \left\langle \mathcal{H} \Psi | \Phi_{\sigma} \right\rangle + i \sum_{\tau} c_{\tau} \left\langle \frac{\partial \Phi_{\tau}}{\partial t} | \Phi_{\sigma} \right\rangle \\ &= i \frac{dc_{\sigma}}{dt} - \left\langle \mathcal{H} \Psi | \Phi_{\sigma} \right\rangle = 0, \end{aligned} \quad (3.8)$$

thanks to the equation satisfied by  $c_{\sigma}$ . Indeed,

$$\frac{\partial \Phi_{\tau}}{\partial t} = \sum_{k=1}^K \frac{\partial \Phi_{\tau}}{\partial \phi_k} \left[ \frac{\partial \phi_k}{\partial t} \right]$$

and therefore the sum in (3.8) vanishes thanks to Lemma 2.5. Secondly, for every  $1 \leq k \leq K$  and for any function  $\zeta$  in  $\text{Span}\{\Phi\}^\perp$ , we have

$$\begin{aligned} & \left\langle i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \middle| \frac{\partial \Psi}{\partial \phi_k} [\zeta] \right\rangle \\ &= i \sum_{\sigma} \frac{dc_{\sigma}}{dt} \left\langle \Phi_{\sigma} \middle| \frac{\partial \Psi}{\partial \phi_k} [\zeta] \right\rangle + i \sum_{j=1}^K \left\langle \frac{\partial \Psi}{\partial \phi_j} \left[ \frac{\partial \phi_j}{\partial t} \right] \middle| \frac{\partial \Psi}{\partial \phi_k} [\zeta] \right\rangle - \left\langle \mathcal{H} \Psi \middle| \frac{\partial \Psi}{\partial \phi_k} [\zeta] \right\rangle \end{aligned} \quad (3.9)$$

$$\begin{aligned} &= \left\langle i \left( \mathbf{\Gamma}(C(t)) \cdot \frac{\partial \Phi}{\partial t} \right)_k - \frac{\partial \Psi^*}{\partial \phi_k} [\mathcal{H} \Psi], \zeta \right\rangle \\ &= - \left\langle \mathbf{P}_{\Phi} \frac{\partial \Psi^*}{\partial \phi_k} [\mathcal{H} \Psi], \zeta \right\rangle = 0. \end{aligned} \quad (3.10)$$

Indeed, on the one hand, in virtue of Lemma 2.5, the first term in the right-hand side of (3.9) vanishes whereas the second one identifies with  $i \sum_{j=1}^K \mathbf{\Gamma}_{kj}(C) \left\langle \frac{\partial \phi_j}{\partial t}, \zeta \right\rangle = \left\langle \left( \mathbf{\Gamma}(C(t)) \cdot \frac{\partial \Phi}{\partial t} \right)_k, \zeta \right\rangle$  since  $\frac{\partial \phi_j}{\partial t}$  and  $\zeta$  both belong to  $\text{Span}\{\Phi\}^\perp$ . On the other hand, the last line (3.10) is obtained using the equation satisfied by  $\Phi$  in  $\mathcal{S}_0$  and by observing that  $\mathbf{P}_{\Phi} \zeta = 0$  since  $\zeta \in \text{Span}\{\Phi\}^\perp$ . The proof is complete.

Let us now recall the definition of the energy

$$\mathcal{E}(\Psi) = \mathcal{E}(\pi(C, \Phi)) = \left\langle \mathcal{H} \Psi \middle| \Psi \right\rangle.$$

It is clear that  $\mathcal{E}(\Psi)$  depends on time *via*  $(C(t), \Phi(t))$ . As a corollary to Proposition 3.2 we have the following

**Corollary 3.3 (Energy is conserved by the flow).** *Let  $(C, \Phi) \in d\mathcal{F}_{N,K}$  be a solution to  $\mathcal{S}_0$  on  $[0, T]$  such that  $\pi(C(t), \Phi(t))$  lies in the domain of  $\mathcal{H}$  for all  $t$  in  $[0, T]$ . Then,*

$$\mathcal{E}(\pi(C(t), \Phi(t))) = \mathcal{E}(\pi(C^0, \Phi^0)) \quad \text{on } [0, T].$$

**Proof.** Comparing with (2.29) we observe that  $\frac{\partial \Psi}{\partial t} \in \mathbf{T}_{\Psi} d\mathcal{B}_{N,K}$ , for

$$\frac{\partial \Psi}{\partial t} = \sum_{\sigma} \frac{dc_{\sigma}}{dt} \Phi_{\sigma} + \frac{1}{N} \sum_{\sigma} \sum_{k=1}^K c_{\sigma} \frac{\partial \Phi_{\sigma}}{\partial \phi_k} \left[ \frac{\partial \phi_k}{\partial t} \right],$$

with  $\frac{\partial \phi_k}{\partial t}$  in  $\text{Span}\{\Phi\}^\perp$  whenever  $\mathbf{\Gamma}(t)$  is invertible. Then, applying Proposition 3.2 to  $\delta \Psi = \frac{\partial \Psi}{\partial t}$  one obtains

$$0 = \Re \left\langle i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \middle| \frac{\partial \Psi}{\partial t} \right\rangle = - \Re \left\langle \mathcal{H} \Psi \middle| \frac{\partial \Psi}{\partial t} \right\rangle = - \frac{1}{2} \frac{d}{dt} \left\langle \mathcal{H} \Psi \middle| \Psi \right\rangle. \quad (3.11)$$

Hence the result.

### 3.2. An a posteriori error estimate

We establish an error bound in  $L^2(\Omega)^N$  for the MCTDHF approximation compared with the exact solution to the linear TDSE (1.2). Let us introduce the projection  $\mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}$  onto the tangent space  $\mathbf{T}_\Psi d\mathcal{B}_{N,K}$  to  $d\mathcal{B}_{N,K}$  at  $\Psi$ . Then, we claim

**Lemma 3.4.** *Given an initial data  $(C^0, \Phi^0) \in d\mathcal{F}_{N,K}$  and an exact solution  $\Psi_E$  to the  $N$ -particle Schrödinger equation (1.2). Then, as long as  $(C, \Phi)$  is a solution to  $\mathcal{S}_0$  in  $d\mathcal{F}_{N,K}$ , we have for  $\Psi(t) = \pi(C(t), \Phi(t))$  and  $\Psi^0 = \pi(C^0, \Phi^0)$  the estimate:*

$$\|\Psi_E - \Psi\|_{L^2(\Omega^N)} \leq \|\Psi_E(0) - \Psi^0\|_{L^2(\Omega^N)} + \int_0^t \left\| (I - \mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}) [\mathcal{H}\Psi(s)] \right\|_{L^2(\Omega^N)} ds.$$

**Proof.** First, Proposition 3.2 expresses the fact that  $\mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}(i\frac{\partial\Psi}{\partial t} - \mathcal{H}\Psi) = 0$ . Therefore the equation satisfied by the ansatz  $\Psi$  is:

$$i\frac{\partial\Psi}{\partial t} - \mathcal{H}\Psi = (I - \mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}) \left[ i\frac{\partial\Psi}{\partial t} - \mathcal{H}\Psi \right] = -(I - \mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}) [\mathcal{H}\Psi], \quad (3.12)$$

since  $i\frac{\partial\Psi}{\partial t}$  lives in the tangent space  $\mathbf{T}_\Psi d\mathcal{B}_{N,K}$ . Next, subtracting (3.12) from (1.2), we get

$$i\frac{\partial(\Psi_E - \Psi)}{\partial t} - \mathcal{H}(\Psi_E - \Psi) = -(I - \mathcal{P}_{\mathbf{T}_\Psi d\mathcal{B}_{N,K}}) [\mathcal{H}\Psi] \quad (3.13)$$

Then, we apply the above PDE to  $\Psi_E - \Psi$  and we integrate formally over  $\Omega^N$ . The result follows by taking the imaginary part of both sides and by using the self-adjointness of  $\mathcal{H}$ .

Roughly speaking, the above lemma tells us that the closer  $\mathcal{H}\Psi$  is to the tangent space  $\mathbf{T}_\Psi d\mathcal{B}_{N,K}$ , the better the MCTDHF approximation is. Intuitively, this is true for large values of  $K$ . Let us mention that this bound was already obtained in [30] and it is probably far from being accurate. However if the MCTDHF algorithm is applied to a discrete model say of dimension  $L$  then for  $K$  large enough ( $K \geq L$ ) this algorithm coincides with the original problem (see Subsection 7.3).

### 3.3. Unitary Group Action on the Flow

The variational system  $\mathcal{S}_0$  is tailored for checking energy conservation and constraints propagation over the flow. However it is badly adapted for proving existence of solutions for the Cauchy problem or for designing numerical codes. It is therefore convenient to have at our disposal several explicit and equivalent representations of the MCTDHF equations over different foliations and to understand how they are related. This is the purpose of this subsection. Proofs of technical lemma and theorems are postponed in the Appendix to facilitate straight reading.

We start with the following (straightforward) lemma on regular flows of unitary transforms :

**Lemma 3.5 (Flow of unitary transforms).** *Let  $U_0 \in \mathcal{O}_K$  and let  $t \mapsto U(t)$  be in  $C^1([0, T]; \mathcal{O}_K)$  with  $U(0) = U_0$ . Then,  $t \mapsto M(t) := -i \frac{dU^*}{dt} U$  defines a continuous family of  $K \times K$  hermitian matrices, and for all  $t > 0$ ,  $U(t)$  is the unique solution to the Cauchy problem*

$$\begin{cases} i \frac{dU}{dt} = U(t)M(t), \\ U(0) = U_0. \end{cases} \quad (3.14)$$

*Conversely, if  $t \mapsto M(t)$  is a continuous family of  $K \times K$  Hermitian matrices and if  $U_0 \in \mathcal{O}_K$  is given, then (3.14) defines a unique  $C^1$  family of  $K \times K$  unitary matrices.*

The corresponding flow for unitary transforms on expansion coefficients is as follows:

**Corollary 3.6.** *Let  $(N, K)$  be an admissible pair, let  $t \mapsto M(t)$  be a continuous family of  $K \times K$  Hermitian matrices and let  $U^0 \in \mathcal{O}_K$ . Then, if  $t \mapsto U(t)$  denotes the unique family of unitary  $K \times K$  matrices that solves (3.14), the unitary  $r \times r$  matrix  $\mathbb{U}$  given by (2.16) is the unique solution to the differential equation*

$$\begin{cases} i \frac{d\mathbb{U}}{dt} = \mathbb{U}\mathbb{M}, \\ \mathbb{U}(0) = d(U^0), \end{cases} \quad (3.15)$$

with

$$\mathbb{M}_{\sigma, \tau} = \sum_{\substack{i \in \sigma, j \in \tau \\ \sigma \setminus \{i\} = \tau \setminus \{j\}}} (-1)^{\sigma^{-1}(i) + \tau^{-1}(j)} M_{ij}. \quad (3.16)$$

The proof of this corollary is postponed to the Appendix. The main result of this section is :

**Theorem 3.7 (Flow of unitary equivalent foliations).** *Let  $U_0 \in \mathcal{O}_K$  and  $(C_0, \Phi_0) \in d\mathcal{F}_{N,K}$ .*

*(i) Let  $t \mapsto M(t)$  be a continuous family of  $K \times K$  Hermitian matrices on  $[0, T]$  and let  $U(t) \in C^1([0, T]; \mathcal{O}_K)$  be the corresponding solution to (3.14). Assume that there exists a strong solution  $(C, \Phi)$  in  $d\mathcal{F}_{N,K}$  to  $\mathcal{S}_0$  with initial data  $(C_0, \Phi_0)$ . Then, the couple  $(C', \Phi') = \mathcal{U}(t) \cdot (C, \Phi)$  with  $\mathcal{U} \in \mathcal{O}_K^r$  defined by (2.18) and (2.16) is a strong solution to the system*

$$\begin{cases} i \frac{dC'}{dt} = \langle \mathcal{H} \Psi | \nabla_{C'} \Psi \rangle - \mathbb{M}' C', \\ i \mathbf{\Gamma}(C') \frac{\partial \Phi'}{\partial t} = (\mathbf{I} - \mathbf{P}_{\Phi'}) \nabla_{\Phi'} \Psi^* [\mathcal{H} \Psi] + \mathbf{\Gamma}(C') M' \Phi' \\ (C'(0), \Phi'(0)) = \mathcal{U}_0 \cdot (C_0, \Phi_0) \end{cases} \quad (3.17)$$

with  $\Psi = \pi(C, \Phi) = \pi(C', \Phi')$ ,  $\mathcal{U}_0 = (U_0, d(U_0)) \in \mathcal{O}_K^r$  being defined by (2.18) and with

$$M' = U M U^*, \quad \overline{M}' = U M U^*,$$

where  $\mathbb{M}$  is the  $r \times r$  Hermitian matrix with entries given by (3.16).

(ii) Conversely, assume that there exists a strong solution  $(C, \Phi)$  in  $d\mathcal{F}_{N,K}$  to  $\mathcal{S}_0$  with initial data  $(C_0, \Phi_0)$  and let  $U(t) \in C^1([0, T]; \mathcal{O}_K)$ . Then, the couple  $(C', \Phi') = \mathcal{U}(t) \cdot (C, \Phi)$  with  $\mathcal{U} \in \mathcal{O}_K^r$  defined by (2.18) and (2.16) is a strong solution to System (3.17) with  $M(t) = -i \frac{dU^*}{dt} U$ .

**Remark 6 (Link with Lagrangian interpretation).** The equations can be derived (at least formally) thanks to the Lagrangian formulation: One writes the stationarity condition for the action

$$\mathcal{A}(\Psi) = \int_0^T \left\langle i \frac{\partial \Psi}{\partial t} - \mathcal{H} \Psi \mid \Psi \right\rangle dt$$

over functions  $\Psi = \Psi(t)$  that move on  $\mathcal{F}_{N,K}$ . The associated time-dependent Euler-Lagrange equations take the form (3.17) with  $\Psi = \pi(C, \Phi)$ ,  $M$  an hermitian matrix and with  $\mathbb{M}$  be the  $r \times r$  hermitian matrix linked to  $M$  through Eqn. (3.16) above. As observed already by Cancès and Le Bris [8], even if they appear so, the Hermitian matrices  $M$  and  $\mathbb{M}$  should not be interpreted as time-dependent Lagrange multipliers associated to the constraints  $(C, \Phi) \in \mathcal{F}_{N,K}$  since the constraints on the coefficients and the orbitals are automatically propagated by the dynamics (see Lemma 3.11), but rather as degrees of freedom within the fiber at  $\Psi$ . In particular, this gauge invariance can be used to set  $M$  and  $\mathbb{M}$  to zero for all  $t$  as observed in Lemma 3.7 and Eqn. (3.23) below, so that the above system can be transformed into the simpler system ( $\mathcal{S}_0$ ) we started from.

As a first example of the change of gauge one can use the unitary transforms to diagonalize the matrix  $\mathbf{\Gamma}$  for all time and therefore derive the evolution equations for natural orbitals following [5]

**Lemma 3.8 (Diagonal density matrix).** Let  $(C, \Phi)$  satisfying  $\mathcal{S}_0$  with initial data  $(C_0, \Phi_0)$  and let  $U_0 \in \mathcal{O}_K$  that diagonalizes  $\mathbf{\Gamma}(C_0)$ . We assume that for all time the eigenvalues of  $\mathbf{\Gamma}(C)$  are simple, that is  $\gamma_i \neq \gamma_j$  for  $1 \leq i, j \leq K$  and  $i \neq j$ . Define a  $K \times K$  Hermitian matrix by

$$M_{ij} = \begin{cases} \frac{1}{\gamma_j - \gamma_i} \left[ \left\langle \mathcal{H} \Psi \mid \frac{\partial \Psi}{\partial \phi_i} [\phi_j] \right\rangle - \left\langle \frac{\partial \Psi}{\partial \phi_j} [\phi_i] \mid \mathcal{H} \Psi \right\rangle \right] & \text{if } i \neq j, \\ 0 & \text{otherwise,} \end{cases}$$

and consider the family  $t \mapsto U(t) \in \mathcal{O}_K$  that satisfies (3.14) with  $U(t=0) = U_0$ . Then  $(C', \Phi') = \mathcal{U}(t) \cdot (C, \Phi)$  is solution to

$$\begin{cases} i \frac{dC'}{dt} = \left\langle \mathcal{H} \Psi \mid \nabla_{C'} \Psi \right\rangle - \mathbb{M}' C', \\ i \gamma_i(t) \frac{\partial \phi'_i}{\partial t} = (\mathbf{I} - \mathbf{P}_{\Phi'}) \frac{\partial \Psi^*}{\partial \phi'_i} [\mathcal{H} \Psi] + \gamma_i(t) M' \Phi' \\ (C'(0), \Phi'(0)) = \mathcal{U}_0 \cdot (C_0, \Phi_0) \end{cases}$$

with the notation of Theorem 3.7. In particular,  $\mathbf{\Gamma}(C') = \text{diag}(\gamma_1(t), \dots, \gamma_K(t))$  for every  $t$ .

**Proof.** Using the equation for the coefficients in (3.17) together with (2.11), the evolution equation for the coefficients of the density matrix writes

$$\begin{aligned} i \frac{d\gamma_{ij}}{dt} &= \sum_{\substack{\sigma, \tau: i \in \sigma, j \in \tau \\ \sigma \setminus \{i\} = \tau \setminus \{j\}}} (-1)^{\sigma^{-1}(i) + \tau^{-1}(j)} [\langle \mathcal{H} \Psi | c_\sigma \Phi_\tau \rangle - \langle c_\tau \Phi_\sigma | \mathcal{H} \Psi \rangle] \\ &+ \sum_{\substack{\kappa, \sigma, \tau: i \in \sigma, j \in \tau \\ \sigma \setminus \{i\} = \tau \setminus \{j\}}} (-1)^{\sigma^{-1}(i) + \tau^{-1}(j)} [\mathbb{M}_{\sigma, \kappa} c_\kappa \bar{c}_\tau - \mathbb{M}_{\kappa, \tau} \bar{c}_\kappa c_\sigma] \\ &= \left\langle \mathcal{H} \Psi \left| \frac{\partial \Psi}{\partial \phi_i} [\phi_j] \right. \right\rangle - \left\langle \frac{\partial \Psi}{\partial \phi_j} [\phi_i] \left| \mathcal{H} \Psi \right. \right\rangle - \sum_{k=1}^K \left\{ \Gamma_{ik} M_{kj} - M_{ik} \Gamma_{kj} \right\} \end{aligned}$$

Next, we require that

$$\gamma_{ij}(t) = \gamma_i \delta_{i,j}, \quad \text{that is} \quad \frac{d\Gamma_{ij}}{dt} = 0 \quad \forall 1 \leq i \neq j \leq K.$$

Using the above equation, a sufficient condition is given by

$$M_{ij} = \frac{1}{\gamma_i - \gamma_j} \left[ \left\langle \mathcal{H} \Psi \left| \frac{\partial \Psi}{\partial \phi_i} [\phi_j] \right. \right\rangle - \left\langle \frac{\partial \Psi}{\partial \phi_j} [\phi_i] \left| \mathcal{H} \Psi \right. \right\rangle \right]$$

This achieves the proof.

As a second application of Theorem 3.7 we investigate particular (stationary) solutions or standing waves. A standing wave for the exact Schrödinger equation is of the form  $\Psi(t, x) = e^{-i\lambda t} \Psi(x)$  with  $\lambda \in \mathbb{R}$ . In the same spirit we look for solutions  $(C', \Phi')$  of System (3.17) with  $(C', \Phi') = \mathcal{U}(t) \cdot (e^{-i\lambda t} C, \Phi)$ , where  $(C, \Phi) \in d\mathcal{F}_{N,K}$  is fixed, independent of time, and  $\mathcal{U}(t) \in \mathcal{O}_K^r$ . Using the formulas (2.25) and (2.26) for the changes of variables, we arrive at

$$\begin{cases} \left( i \frac{d\bar{U}(t)}{dt} + \mathbb{M}\bar{U} + \lambda \bar{U} \right) C = \bar{U} \left\langle \mathcal{H} \Psi \left| \nabla_C \Psi \right. \right\rangle, \\ \Gamma(C) \left( i U^* \frac{dU}{dt} - U^* M U \right) \Phi = (\mathbf{I} - \mathbf{P}_\Phi) \nabla_\Phi \Psi^* [\mathcal{H} \Psi], \\ U(0) = \mathbb{I}_K. \end{cases}$$

In the above system  $\Psi = \pi(C, \Phi)$  and  $\Gamma(C)$  are independent of time and  $\Gamma(C)$  is invertible. We start with the equation satisfied by  $\Phi$ . Observing that the left-hand side lives in  $\text{Span}\{\Phi\}$  whereas the right-hand side lives in  $\text{Span}\{\Phi\}^\perp$ , we conclude that there are both equal to zero. Therefore, there exists a  $K \times K$  matrix  $\Lambda$  that is independent of  $t$  and such that

$$\nabla_\Phi \Psi^* [\mathcal{H} \Psi] = \nabla_\Phi \left\langle \mathcal{H} \Psi \left| \Psi \right. \right\rangle = \Lambda \cdot \Phi. \quad (3.18)$$

Also since the left-hand side has to be independent of  $t$  we get

$$i \frac{dU}{dt} = M U.$$

Comparing now with the equation for the coefficients we infer from Corollary 3.6 that

$$i \frac{d\bar{\mathbb{U}}(t)}{dt} = -\mathbb{M}\bar{\mathbb{U}},$$

hence

$$\nabla_{\bar{C}} \langle \mathcal{H} \Psi | \Psi \rangle = \lambda C. \quad (3.19)$$

Equations (3.18) and (3.19) are precisely the MCHF equations that are satisfied by critical points of the energy. They were derived by Lewin [28] in the Coulomb case. The real  $\lambda$  is the Lagrange multiplier corresponding to the constraint  $C \in S^{r-1}$  whereas the Hermitian matrix  $\Lambda$  is the matrix of Lagrange multipliers corresponding to the orthonormality constraints on the orbitals. Existence of such solutions in physical case is recalled in Section 4.

The proof of Theorem 3.10 is postponed in the Appendix and we rather state before some corollaries or remarks. In the physics literature the MCTDHF equations are derived from the variational principle (3.1) under the constraints  $\Psi = \pi(C, \Phi) \in \mathcal{B}_{N,K}$  along with additional constraints on the time-dependent orbitals

$$\left\langle \frac{\partial \phi_i}{\partial t}, \phi_j \right\rangle = \langle \mathbf{G} \phi_i, \phi_j \rangle \quad \text{for all } 1 \leq i, j \leq K. \quad (3.20)$$

In the above equation  $\mathbf{G}$  is an arbitrary self-adjoint operator on  $L^2(\Omega)$  possibly time-dependent named the *gauge*. In this spirit the variational system corresponds to  $\mathbf{G} = 0$ . Therefore a gauge field is chosen *a priori* and the corresponding equations are derived accordingly. Both approaches are equivalent by observing that, to every Hermitian matrix  $M$ , one can associate a self-adjoint operator  $\mathbf{G}$  in  $L^2(\Omega)$  such that  $M_{ij} = \langle \mathbf{G} \phi_i, \phi_j \rangle$  by demanding that

$$\mathbf{G} \phi_i = \sum_{j=1}^K M_{ij} \phi_j \quad \text{for all } 1 \leq i \leq K.$$

Conversely being given the family  $t \mapsto M(t)$  in Theorem 3.10 it follows immediately from the system (3.17) that for all  $1 \leq i, j \leq K$ ,

$$i \left\langle \frac{\partial \phi'_i}{\partial t}, \phi'_j \right\rangle = M'_{ij}.$$

provided  $\mathbf{\Gamma}(C') = U \mathbf{\Gamma}(C) U^*$  is invertible on  $[0, T]$ . We state below Theorem 3.7 that is the equivalent formulation of Theorem 3.10 in terms of gauge. It is based on above remarks together with the following :

**Lemma 3.9.** *Let  $t \mapsto \mathbf{G}(t)$  be a family of self-adjoint operators on  $L^2(\Omega)$  with domain  $\mathcal{D}(\mathbf{G}(t))$  and let  $\Phi = (\phi_1(t), \phi_2(t), \dots, \phi_K(t)) \in \mathcal{O}_{L^2(\Omega)^K}$  such that  $\phi_i(t) \in \mathcal{D}(\mathbf{G}(t))$  for all  $t$  and  $t \mapsto \langle \mathbf{G}(t) \phi_i(t), \phi_j(t) \rangle$  is continuous on  $[0, T]$ . Then the matrix  $M$  with entries  $M_{ij}(t) = \langle \mathbf{G}(t) \phi_i(t), \phi_j(t) \rangle$  is Hermitian and the Cauchy problem (3.14) defines a globally well-defined  $C^1$  flow on the set of unitary  $K \times K$*

matrices. In that case, the unitary transforms  $\mathbb{U} = d(U)$  solve the Cauchy problem (3.15) with  $\mathbb{M}$  in (3.16) given by

$$\mathbb{M}_{\sigma,\tau} = \sum_{i=1}^N \langle \mathbf{G}_{x_i} \Phi_\sigma | \Phi_\tau \rangle. \quad (3.21)$$

**Remark 7.** In Lemma 3.9 the functions  $t \mapsto \phi_i(t)$  are continuous with values in the domain of  $\mathbf{G}$ . When  $\mathbf{G}$  is bounded from below it is enough to assume continuity in the form-domain. When  $\mathbf{G}$  is the Laplace operator or, more generally a one-body time-independent Schrödinger operator, we simply assume that  $\phi_i \in H^1(\mathbb{R}^3)$  or  $\phi_i \in H_0^1(\Omega)$  when  $\Omega$  is a bounded domain with Dirichlet boundary conditions. (Other boundary conditions could of course be considered.)

**Theorem 3.10 (Flow in different gauge).** Let  $U_0 \in \mathcal{O}_K$ ,  $(C_0, \Phi_0) \in d\mathcal{F}_{N,K}$  and let  $t \mapsto \mathbf{G}(t)$  be a family of self-adjoint operators in  $L^2(\Omega)$ . Assume that there exists a strong solution  $(C, \Phi)$  to  $\mathcal{S}_0$  with initial data  $(C_0, \Phi_0)$  such that  $(C, \Phi) \in d\mathcal{F}_{N,K}$  for all  $t$  and  $t \mapsto \langle \mathbf{G}(t)\phi_i(t), \phi_j(t) \rangle$  is continuous on  $[0, T]$  for every  $1 \leq i, j \leq K$ . Define the family of unitary transforms  $U(t) \in C^1([0, T]; \mathcal{O}_K)$  that satisfy (3.14) with  $M_{ij} = \langle \mathbf{G}\phi_i, \phi_j \rangle$  as in Lemma 3.9. Then the couple  $(C', \Phi') = \mathcal{U}(t) \cdot (C, \Phi)$  with  $\mathcal{U}(t) = (d(U(t)); U(t))$  defined by (2.18) and (2.16) is a strong solution to

$$(\mathcal{S}_{\mathbf{G}}) \begin{cases} i \frac{dC'}{dt} = \langle \mathcal{H}\Psi | \nabla_{C'}\Psi \rangle - \left\langle \sum_{i=1}^N \mathbf{G}_{x_i} \Psi | \nabla_{C'}\Psi \right\rangle, \\ i \Gamma(C') \frac{\partial \Phi'}{\partial t} = \Gamma(C') \mathbf{G} \Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'}) \nabla_{\Phi'} \Psi^* \left[ \mathcal{H}\Psi - \sum_{i=1}^N \mathbf{G}_{x_i} \Psi \right], \\ (C'(0), \Phi'(0)) = \mathcal{U}_0 \cdot (C_0, \Phi_0), \end{cases}$$

with  $\Psi = \pi(C, \Phi) = \pi(C', \Phi')$ ,  $\mathcal{U}_0 = (U_0, d(U_0)) \in \mathcal{O}_K^r$  being defined by (2.18) and with  $\mathbb{M}$  being the  $r \times r$  Hermitian matrix given by (3.16).

**Remark 8.** Passing from  $\mathcal{S}_0$  to  $\mathcal{S}_{\mathbf{G}}$  amounts to change the operator  $\mathcal{H}$  by  $\mathcal{H} - \sum_{i=1}^N \mathbf{G}_{x_i}$  in both equations and by adding the linear term  $\Gamma(C') \mathbf{G} \Phi'$  in the equation satisfied by  $\Phi'$ . Note that whereas solutions to  $\mathcal{S}_0$  in  $d\mathcal{F}_{N,K}$  satisfy

$$i \left\langle \frac{\partial \phi_i}{\partial t}, \phi_j \right\rangle = 0,$$

for all  $1 \leq i, j \leq K$ , solutions to  $(\mathcal{S}_{\mathbf{G}})$  satisfy

$$i \left\langle \frac{\partial \phi'_i}{\partial t}, \phi'_j \right\rangle = \langle \mathbf{G} \phi'_i, \phi'_j \rangle. \quad (3.22)$$

System  $\mathcal{S}_{\mathbf{G}}$  corresponds to the choice of gauge  $\mathbf{G}$ .

This is illustrated and summarized on Figure 3.1 below.

Theorem 3.10 and Lemma 3.9 provide with the differential equation that satisfies the unitary matrix  $U(t)$  that transforms  $\mathcal{S}_0$  into  $\mathcal{S}_{\mathbf{G}}$ . A direct calculation shows that, given two self-adjoint one-particle operators  $\mathbf{G}$  and  $\mathbf{G}'$ , the solution to

$$\begin{cases} i \frac{dU}{dt} = U M_{\mathbf{G} \rightarrow \mathbf{G}'}, \\ U(t=0) = U^0 \end{cases} \quad (3.23)$$

with  $(M_{\mathbf{G} \rightarrow \mathbf{G}'} )_{ij} = \langle (\mathbf{G} - \mathbf{G}') \phi_i, \phi_j \rangle$  maps a solution to  $\mathcal{S}_{\mathbf{G}}$  to a solution to  $\mathcal{S}_{\mathbf{G}'}$ . In particular, if we prove existence of solutions for the system  $\mathcal{S}_{\mathbf{G}}$  for some operator  $\mathbf{G}$  then we have existence of solutions for any system  $\mathcal{S}_{\mathbf{G}'}$ .

Another immediate though crucial consequence of Theorem 3.10 and Theorem 3.7 is given in Corollary 3.11 below. It states that for any choice of gauge the constraints on the expansion coefficients and on the orbitals are propagated by the flow and the energy is kept constant since it is the case for the system  $\mathcal{S}_0$ . Also the rank of the first-order density matrices does not depend on the gauge.

**Corollary 3.11 (Gauge transforms and conservation properties).** *Let  $T > 0$ . Let  $\mathbf{G}$  be a self-adjoint (possibly time-dependent) operator acting on  $L^2(\Omega)$  with domain  $\mathcal{D}(\mathbf{G})$ . Assume that there exists a strong solution to the system  $\mathcal{S}_{\mathbf{G}}$  on  $[0, T]$  such that  $\text{rank} \Gamma(C(t)) = K$  and such that the matrix  $t \mapsto \langle \mathbf{G} \phi_i, \phi_j \rangle_{1 \leq i, j \leq K}$  is continuous with  $\phi_i(t) \in \mathcal{D}(\mathbf{G})$  for all  $t$ . Then, for all  $t \in [0, T]$ ,*

$$(C(t), \Phi(t)) \in d\mathcal{F}_{N,K},$$

and the energy is conserved, that is

$$\mathcal{E}(\pi(C(t), \Phi(t))) = \mathcal{E}(\pi(C(0), \Phi(0))).$$

In addition,  $\Psi = \pi(C, \Phi)$  satisfies the Dirac-Frenkel variational principle (3.1).

*Proof of Corollary 3.11.* By Theorem 3.10 and its remark, if  $(C, \Phi)$  satisfies  $\mathcal{S}_{\mathbf{G}}$  with initial data in  $d\mathcal{F}_{N,K}$ , there exists a family of unitary transforms  $U \in C^1(0, T; \mathcal{O}_K)$  such that  $(C, \Phi) = \mathcal{U} \cdot (C', \Phi')$  where  $(C', \Phi')$  satisfies  $\mathcal{S}_0$  with same initial data. Since by Lemma 3.1,  $\mathcal{S}_0$  preserves  $\mathcal{F}_{N,K}$ , so does  $\mathcal{S}_{\mathbf{G}}$  since  $U$  and  $\mathbb{U} = d(U)$  are unitary. Then, by Lemma 2.4,  $\pi(C, \Phi) = \pi(C', \Phi') = \Psi$ , and the energy is conserved by the flow since it only depends on  $\Psi$ . Eventually Eqn. (3.1) is satisfied since  $\mathbf{T}_{\Psi} d\mathcal{B}_{N,K}$  only depends on the point  $\Psi$  on the basis  $\mathcal{B}_{N,K}$  and not on the pre-images in the fiber  $\pi^{-1}(\Psi)$ .  $\square$

So far we have considered a generic Hamiltonian  $\mathcal{H}$  and we have written down an abstract coupled system of evolution equations for this operator. In the following subsection we turn to the particular physical case of  $N$ -body Schrödinger-type operators with pairwise interactions

### 3.4. $N$ -body Schrödinger type operators with pairwise interactions

At this point, we consider an Hamiltonian in  $L^2(\Omega^N)$  of the following form

$$\mathcal{H}_N \Psi = \sum_{i=1}^N \mathbf{H}_{x_i} \Psi + \sum_{1 \leq i < j \leq N} v(|x_i - x_j|) \Psi. \quad (3.24)$$

In the above definition,  $\mathbf{H}$  is a self-adjoint operator acting on  $L^2(\Omega)$  with domain  $\mathcal{D}(\mathbf{H})$ . To fix ideas we take  $\mathbf{H} = -\frac{1}{2}\Delta + U$  with  $U \in L^2(\Omega) + L^\infty(\Omega)$ .  $v$  is a real-valued potential in  $L^2(\Omega) + L^\infty(\Omega)$ , and we denote

$$V = \sum_{1 \leq i < j \leq N} v(|x_i - x_j|).$$

Under these assumptions, the domain of  $H$  is  $H^2(\mathbb{R}^3)$  when  $\Omega = \mathbb{R}^3$  and is a subspace of  $H^2(\Omega)$  accounting for boundary conditions otherwise (e.g.  $\mathcal{D}(\mathbf{H}) = H^2(\Omega) \cap H_0^1(\Omega)$  for a bounded regular domain  $\Omega$  with Dirichlet boundary conditions). Expanding the expression of  $\mathcal{H}$  in the system  $\mathcal{S}_0$  and arguing as in the proof of Theorem 3.10 we obtain

$$\mathcal{S}_0 : \begin{cases} i \frac{dC}{dt} = \left\langle \sum_{i=1}^N \mathbf{H}_{x_i} \Psi \mid \nabla_C \Psi \right\rangle + \left\langle V \Psi \mid \nabla_C \Psi \right\rangle \\ i \Gamma(C) \frac{\partial \Phi}{\partial t} = (\mathbf{I} - \mathbf{P}_\Phi) \nabla_\Phi \Psi^* \left[ V \Psi + \sum_{i=1}^N \mathbf{H}_{x_i} \Psi \right] \\ (C(0), \Phi(0)) = (C_0, \Phi_0) \in \mathcal{F}_{N,K}. \end{cases} \quad (3.25)$$

Comparing with System  $\mathcal{S}_G$  in Theorem 3.10, one observes that the choice of gauge  $\mathbf{G} = \mathbf{H}$  leads to the equivalent system

$$\mathcal{S}_H : \begin{cases} i \frac{dC}{dt} = \left\langle V \Psi \mid \nabla_C \Psi \right\rangle, \\ i \Gamma(C) \frac{\partial \Phi}{\partial t} = \Gamma(C) \mathbf{H} \Phi + (\mathbf{I} - \mathbf{P}_\Phi) \nabla_\Phi \Psi^* [V \Psi] \\ (C(0), \Phi(0)) = (C_0, \Phi_0) \in \mathcal{F}_{N,K}, \end{cases} \quad (3.26)$$

(provided  $t \mapsto \langle \mathbf{H} \phi_i, \phi_j \rangle$  makes sense). From Corollary 3.11 we know that if the initial data in (3.26) lies in  $\mathcal{F}_{N,K}$  it persists for all time. This property allows to recast System (3.26) in a more tractable way where the equations satisfied by the orbitals form a coupled system of non-linear Schrödinger-type equations. This new system that it is equivalent to System (3.26) as long as the solution lies in  $\mathcal{F}_{N,K}$  will be referred to as *working equations* following [7, 26]. It is better adapted for well-posedness analysis as will be seen in the forthcoming section. In the following, by strong solution  $(C; \Phi)$  we mean  $C \in C^1(0, T; \mathbb{C}^r)$  and  $\Phi \in C^0(0, T; \mathcal{D}(\mathbf{H}))^K \cap C^1(0, T; L^2(\Omega))^K$ .

**Proposition 3.12 (Working equations).** *Let  $(C, \Phi)$  be a strong solution to (3.26) in  $\mathcal{F}_{N,K}$ , then it is a strong solution to*

$$\begin{cases} i \frac{dC}{dt} = \mathbb{K}[\Phi] C, \\ i \Gamma(C) \frac{\partial \Phi}{\partial t} = \Gamma(C) \mathbf{H} \Phi + (\mathbf{I} - \mathbf{P}_\Phi) \mathbb{W}[C, \Phi] \Phi, \\ (C(0), \Phi(0)) = (C^0, \Phi^0) \in \mathcal{F}_{N,K}, \end{cases} \quad (3.27)$$

where  $\mathbb{K}[\Phi]$  (resp.  $\mathbb{W}[C, \Phi]$ ) is a  $r \times r$  (resp.  $K \times K$ ) Hermitian matrix with entries

$$\mathbb{K}[\Phi]_{\sigma, \tau} = \sum_{i, j \in \tau, k, l \in \sigma} \delta_{\tau \setminus \{i, j\}, \sigma \setminus \{k, l\}} (-1)_{i, j}^{\tau} (-1)_{k, l}^{\sigma} D_v(\phi_i \bar{\phi}_k, \bar{\phi}_j \phi_l) \quad (3.28)$$

and

$$\mathbb{W}[C, \Phi]_{ij}(x) = 2 \sum_{k, l=1}^K \gamma_{ijkl} (\phi_k \bar{\phi}_l \star_{\Omega} v) \quad (3.29)$$

where here and below we denote

$$D_v(f, g) = \iint_{\Omega \times \Omega} v(|x-y|) f(x) \bar{g}(y) dx dy,$$

$$f \star_{\Omega} v = \int_{\Omega} v(\cdot - y) f(y) dy$$

and with the coefficients  $\gamma_{ijkl}$  being defined by (2.10) in Proposition 2.3. Conversely, any solution to (3.27) defines a flow on  $\mathcal{F}_{N,K}$  as long as  $\Gamma(C)$  is invertible and is therefore a solution to (3.26).

**Proof.** We have to show that for  $\Psi = \pi(C, \Phi)$  in  $\mathcal{B}_{N,K}$

$$\langle V \Psi \mid \nabla_C \Psi \rangle = \nabla_{\bar{C}} \langle V \Psi \mid \Psi \rangle = \mathbb{K}[\Phi] C \quad (3.30)$$

and

$$\nabla_{\Phi} \Psi^* [V \Psi] = \nabla_{\bar{\Phi}} \langle V \Psi \mid \Psi \rangle = \mathbb{W}[C, \Phi] \Phi. \quad (3.31)$$

We start from

$$\langle V \Psi \mid \Psi \rangle = \iint_{\mathbb{R}^3 \times \mathbb{R}^3} [\Psi \otimes \Psi]_{:2}(x, y, x, y) v(|x-y|) dx dy$$

with

$$[\Psi \otimes \Psi]_{:2}(x, y, x, y) = \sum_{i, j, k, l=1}^K \gamma_{ijkl} \phi_i(x) \phi_j(y) \bar{\phi}_k(x) \bar{\phi}_l(y)$$

according to (2.9). Since only the coefficients  $\gamma_{ijkl}$  depend on  $C$  through Eqn. (2.10) we first get

$$\nabla_{\bar{C}} \langle V \Psi \mid \Psi \rangle = \sum_{i, j, k, l=1}^K \nabla_{\bar{C}} (\gamma_{ijkl}) D_v(\phi_i \bar{\phi}_k, \bar{\phi}_j \phi_l).$$

Hence (3.28) by using again Formula (2.10).

We now turn to the proof of (3.31) starting from

$$\langle V \Psi | \Psi \rangle = \sum_{i,j,k,l=1}^K \gamma_{ijkl} \iint_{\mathbb{R}^3 \times \mathbb{R}^3} \phi_i(x) \phi_j(y) \bar{\phi}_k(x) \bar{\phi}_l(y) v(|x-y|) dx dy.$$

Then, for every  $1 \leq p \leq K$

$$\begin{aligned} \frac{\partial}{\partial \bar{\phi}_p} \langle V \Psi | \Psi \rangle &= \sum_{i,j,l=1}^K \gamma_{ijpl} ((\phi_j \bar{\phi}_l) \star v) \phi_i + \sum_{i,j,k=1}^K \gamma_{ijkp} ((\phi_i \bar{\phi}_k) \star v) \phi_j \\ &= 2 \sum_{i,j,l=1}^K \gamma_{ijpl} ((\phi_i \bar{\phi}_l) \star v) \phi_j \end{aligned}$$

by interchanging the rôle played by  $i$  and  $j$  in the first sum and by using  $\gamma_{ijkp} = \gamma_{jipk}$  and renaming  $k$  as  $l$  in the second one. Comparing with (3.29) we find

$$\frac{\partial}{\partial \bar{\phi}_p} \langle V \Psi | \Psi \rangle = 2 \sum_{j=1}^K \mathbb{W}[C, \Phi]_{pj} \phi_j.$$

To achieve the proof of the proposition we now check that the system of equations in (3.27) preserves  $\mathcal{F}_{N,K}$  as long as  $\Gamma(C)$  is invertible. The claim is obvious as regards the orthonormality of the orbitals since  $\mathbf{H}$  is self-adjoint and since  $\mathbf{I} - \mathbf{P}_\Phi$  projects on  $\text{Span}\{\Phi\}^\perp$ . On the other hand, the equation on the coefficients leads to

$$\frac{d}{dt} \|C(t)\|^2 = 2 \Im \sum_{\sigma,\tau} \mathbb{K}[\Phi]_{\sigma,\tau} c_\tau \bar{c}_\sigma = 0$$

since the matrix  $\mathbb{K}[\Phi]$  is Hermitian.

We treat apart in the last two subsections the special cases of the linear *free system* with no pairwise interaction and of the *time-dependent Hartree–Fock equations* for the evolution of a single-determinant (TDHF in short) with pairwise interaction.

### 3.5. Interactionless Systems $v \equiv 0$

In this section we consider systems for which the binary interaction potential  $v$  is switched off. Then the system (3.26) becomes

$$\begin{cases} i \frac{dC}{dt} = 0, \\ i \Gamma(C) \frac{\partial \Phi}{\partial t} = \Gamma(C) \mathbf{H} \Phi. \end{cases}$$

From the first equation the coefficients  $c_\sigma$ 's are constant during the evolution. In particular the full-rank assumption is satisfied for all time whenever it is satisfied at start. In the latter case the orbitals satisfy  $K$  independent linear Schrödinger equations

$$i \frac{\partial \Phi}{\partial t} = \mathbf{H} \Phi, \quad (3.32)$$

and the  $N$ -particle wave-function  $\Psi = \pi(C, \Phi)$  solves the exact Schrödinger equation

$$\begin{cases} i \frac{\partial \Psi}{\partial t} = \sum_{i=1}^N \mathbf{H}_{x_i} \Psi, \\ \Psi(t=0) = \pi(C_0, \Phi_0). \end{cases} \quad (3.33)$$

Conversely, the unique solution to the Cauchy problem (3.33) with  $(C_0, \Phi_0) \in d\mathcal{F}_{N,K}$  coincides with  $\pi(C(t), \Phi(t)) \in \mathcal{F}_{N,K}$  where  $\Phi(t)$  is the solution to (3.32). This is a direct consequence of the fact that the linear structure of (3.33) propagates the factorization of a Slater determinant. In particular, this enlightens the fact that the propagation of the full-rank assumption is intricately related to the non-linearities created by the interaction potential  $v$  between particles.

### 3.6. MCTDHF ( $K = N$ ) contains TDHF

The TDHF equations write (up to a unitary transform)

$$i \frac{\partial \phi_i}{\partial t} = \mathbf{H} \phi_i + \mathcal{F}_\Phi \phi_i, \quad (3.34)$$

for  $1 \leq i \leq N$ , with  $\mathcal{F}_\Phi$  being the self-adjoint operator on  $L^2(\Omega)$  that is defined by

$$\mathcal{F}_\Phi w = \left( \sum_{j=1}^N \int_{\Omega} v(|\cdot - y|) |\phi_j(y)|^2 dy \right) w - \sum_{j=1}^N \left( \int_{\Omega} v(|\cdot - y|) \bar{\phi}_j(y) w(y) dy \right) \phi_j.$$

The global-in-time existence of solutions in the energy space  $H^1(\Omega)^N$  goes back to Bove, Da Prato and Fano [6] for bounded interaction potentials and to Chadam and Glassey [12] for the Coulomb potentials. They also checked by integrating the equations that the TDHF equations propagate the orthonormality of the orbitals and that the Hartree–Fock energy is preserved by the flow. Derivation of the TDHF equations from the Dirac-Frenkel variational principle may be encountered in standard Physics textbooks (see *e.g.* [33]). Let us also mention the work [8] by Cancès and Le Bris who have investigated existence of solutions to TDHF equations including time-dependent electric field and that are coupled with nuclear dynamics.

By simply setting  $K = N$  in the MCTDHF formalism one gets

$$\#\Sigma_{N,K} = 1, \quad \mathbf{\Gamma}(t) = \mathbb{I}_N \quad (3.35)$$

and

$$\Psi(t) := C(t) \phi_1(t) \wedge \dots \wedge \phi_N(t), \quad C(t) = e^{-i\theta_\Phi(t)}$$

for some  $\theta_\Phi \in \mathbb{R}$ . In addition according to Remark 1,

$$\gamma_{jkil} = \frac{1}{2} (\delta_{i,j} \delta_{k,l} - \delta_{i,k} \delta_{j,l}). \quad (3.36)$$

Therefore with the definitions (3.28) and (3.29)

$$\begin{aligned}\mathbb{K}[\Phi] &= \langle V \phi_1 \wedge \dots \wedge \phi_N \mid \phi_1 \wedge \dots \wedge \phi_N \rangle \\ &= \sum_{i,j,k,l: \{i,j\}=\{k,l\}} (-1)^{i+p_i(j)+k+p_k(l)} D_V(\phi_i \bar{\phi}_k; \bar{\phi}_j \phi_l) \\ &= \sum_{i=1}^N \langle \mathcal{F}_\Phi \phi_i, \phi_i \rangle\end{aligned}$$

and

$$\sum_{j=1}^N \mathbb{W}[C, \Phi]_{ij} \phi_j = \mathcal{F}_\Phi \phi_i.$$

Eventually for  $N = K$ , according to (3.27), the MCTDHF system in the working form turns out to be

$$\mathcal{S}_{\mathbf{H}}(N = K) \begin{cases} \frac{d\theta_\Phi(t)}{dt} = \sum_{i=1}^N \langle \mathcal{F}_\Phi \phi_i, \phi_i \rangle, \\ i \frac{\partial \phi_i}{\partial t} = \mathbf{H} \phi_i + (\mathbf{I} - \mathbf{P}_\Phi) \mathcal{F}_\Phi \phi_i \\ \quad = \mathbf{H} \phi_i + \mathcal{F}_\Phi \phi_i - \sum_{j=1}^N \langle \mathcal{F}_\Phi \phi_i, \phi_j \rangle \phi_j \end{cases}$$

with  $\theta_\Phi(0) = 0$  and  $\Phi(0) \in \mathcal{O}_{L^2(\Omega)^N}$ . Comparing with (3.17), we introduce the  $N \times N$  Hermitian matrix  $M$  with entries  $M_{ij} = -\langle \mathcal{F}_\Phi \phi_i, \phi_j \rangle$ . According to Lemma 3.5 there exists a unique unitary matrix  $U(t)$  such that

$$\begin{cases} i \frac{dU}{dt} = -UM, \\ U(t=0) = \mathbb{I}_N. \end{cases}$$

In virtue of (2.16) the unitary matrix  $\mathbb{U}$  that transforms  $\phi_1 \wedge \dots \wedge \phi_N$  into  $(U\phi_1) \wedge \dots \wedge (U\phi_N)$  is then simply a complex number of modulus 1 ( $\mathbb{U} = \det(U)$ ) that satisfies

$$\begin{cases} i \frac{d\mathbb{U}}{dt} = -\text{tr}(M) \mathbb{U}, \\ \mathbb{U}(t=0) = 1. \end{cases} \quad (3.37)$$

Comparing (3.37) with the equation satisfied by  $\theta_\Phi(t)$  in  $\mathcal{S}_{\mathbf{H}}(N = K)$  it turns out that  $\mathbb{U} = e^{i\theta_\Phi(t)}$ . In that special case a change of gauge is simply a multiplication by a global phase factor. Applying Theorem 3.7, the functions  $\phi'_i$ ,  $1 \leq i \leq N$ , defined by  $\Phi' = U\Phi$  satisfy the standard Hartree–Fock equations (3.34) and  $C'(t) = \overline{\mathbb{U}}C(t) = 1$  for all time; that is  $\Psi' = \phi'_1 \wedge \dots \wedge \phi'_N$ . Being a special case of the MCTDHF setting we then recover “for free” that the TDHF equations propagate the orthonormality of the initial data, that they satisfy the Dirac-Frenkel variational principle and that the flow keeps the energy constant.

#### 4. Mathematical analysis of the MCDTHF Cauchy Problem

This section is devoted to the mathematical analysis of the Cauchy problem for the  $N$ -body Schrödinger operator with “physical interactions”

$$U(x) = - \sum_{m=1}^M \frac{z_m}{|x - R_m|}, \quad v(x) = \frac{1}{|x|}, \quad (4.1)$$

that is given by (3.26):

$$\mathcal{S}_{\mathbf{H}} : \begin{cases} i \frac{dC}{dt} = \langle V \Psi | \nabla_C \Psi \rangle, \\ i \mathbf{\Gamma}(C) \frac{\partial \Phi}{\partial t} = \mathbf{\Gamma}(C) \mathbf{H} \Phi + (\mathbf{I} - \mathbf{P}_{\Phi}) \nabla_{\Phi} \Psi^* [V \Psi] \\ (C(0), \Phi(0)) = (C_0, \Phi_0) \in \mathcal{F}_{N,K}. \end{cases}$$

In this section  $\Omega = \mathbb{R}^3$ . According to Proposition (3.27) solutions to  $(\mathcal{S}_{\mathbf{H}})$  lie in  $\mathcal{F}_{N,K}$  and they are therefore solutions to

$$\begin{cases} i \frac{dC}{dt} = \mathbb{K}[\Phi] C, \\ i \mathbf{\Gamma}(C) \frac{\partial \Phi}{\partial t} = \mathbf{\Gamma}(C) \mathbf{H} \Phi + (\mathbf{I} - \mathbf{P}_{\Phi}) \mathbb{W}[C, \Phi] \Phi, \\ (C(0), \Phi(0)) = (C^0, \Phi^0) \in \mathcal{F}_{N,K} \end{cases} \quad (4.2)$$

with

$$\begin{aligned} \mathbb{K}[\Phi]_{\sigma, \tau} &= \sum_{i,j \in \tau, k,l \in \sigma} \delta_{\tau \setminus \{i,j\}, \sigma \setminus \{k,l\}} (-1)_{i,j}^{\tau} (-1)_{k,l}^{\sigma} D(\phi_i \bar{\phi}_k, \bar{\phi}_j \phi_l), \\ \mathbb{W}[C, \Phi]_{ij}(x) &= 2 \sum_{k,l=1}^K \gamma_{jkil} (\phi_k \bar{\phi}_l \star \frac{1}{|x|}) \\ D(f, g) &= \iint_{\mathbb{R}^3 \times \mathbb{R}^3} \frac{1}{|x-y|} f(x) \bar{g}(y) dx dy. \end{aligned}$$

The above system is referred to as the “strong form” of the working equations. Let us emphasize again that it is equivalent to  $(\mathcal{S}_{\mathbf{H}})$  provided  $(C, \Phi) \in \mathcal{F}_{N,K}$ . The main sources of difficulties arise from the fact that the matrix  $\mathbf{\Gamma}(C)$  may degenerate and from the Coulomb singularities of the interaction potentials. Our strategy of proof works for more general potentials  $U$  and  $v$ . This is discussed in Section 7 below.

The spaces  $\mathbb{C}^r$  and  $W^{m,p}(\mathbb{R}^3)^K$  are equipped with the Euclidian norms for the vectors  $C$  and  $\Phi$  respectively

$$\|C\|^2 := \sum_{\sigma \in \Sigma_{N,K}} |c_{\sigma}|^2, \quad \|\Phi\|_{W^{m,p}}^2 := \sum_{i=1}^K \|\phi_i\|_{W^{m,p}(\mathbb{R}^3)}^2.$$

Moreover, for a  $p \times p$  matrix  $M$  we use the Frobenius norm

$$\|M\| = \sqrt{\sum_{i,j=1}^p |M_{ij}|^2}.$$

For any integer  $m$ , we introduce the spaces  $X_m := \mathbb{C}^r \times H^m(\mathbb{R}^3)^K$  endowed with the norms

$$\|(C, \Phi)\|_{X_m} = \|C\| + \|\Phi\|_{H^m}.$$

The main result in this section is the following

**Theorem 4.1 (The MCTDHF equations are well-posed).** *Let  $(C^0, \Phi^0) \in d\mathcal{F}_{N,K}$  with  $\Phi^0$  in  $H^m(\mathbb{R}^3)^K$  with  $m \in \mathbb{N}$ ,  $m \geq 1$ . Then, there exists a maximal existence time  $T^* > 0$  (possibly  $+\infty$  but independent of  $m$ ) such that:*

(i) *The MCTDHF system (3.27) admits a unique solution  $(C, \Phi)$  with*

$$C \in C^1([0, T^*]; \mathbb{C}^r), \quad \Phi \in C^0([0, T^*]; H^m(\mathbb{R}^3)^K) \cap C^1([0, T^*]; H^{m-2}(\mathbb{R}^3)^K).$$

*This solution depends continuously on the initial data  $(C^0, \Phi^0)$  in  $X_m$ . For every  $0 \leq t < T^*$ ,*

(ii)  *$(C(t), \Phi(t)) \in d\mathcal{F}_{N,K}$  and  $\mathbf{\Gamma}(C(t))$  is invertible.*

(iii) *The energy is conserved :*

$$\langle \mathcal{H}_N \Psi(t) | \Psi(t) \rangle = \langle \mathcal{H}_N \Psi^0 | \Psi^0 \rangle \quad \text{with } \Psi = \pi(C, \Phi) \text{ and } \Psi^0 = \pi(C^0, \Phi^0).$$

(iv) *The Dirac–Frenkel variational principle (3.1) is satisfied.*

(v) *When  $T^* < +\infty$  one has*

$$\limsup_{t \nearrow T^*} \|\mathbf{\Gamma}(C(t))^{-1}\| = +\infty$$

*and more precisely:*

$$\int_0^{T^*} \|\mathbf{\Gamma}(C(t))^{-1}\|^{3/2} dt = +\infty.$$

The global well-posedness in  $H^1$  and  $H^2$  of the TDHF equations goes back to Chadam and Glassey [12]. Recently Koch and Lubich [26] proved local well-posedness in  $H^2$  of the MCTDH and MCTDHF equations for regular pairwise interaction potential  $v$  and with  $U \equiv 0$  by using Lie commutators techniques. Our result extends both works. The rest of the section is devoted to the proof of this theorem. The above system with the same notation is rewritten in the “mild form” which makes sense as long as the matrix  $\mathbf{\Gamma}(C(t))$  is not singular:

$$U(t) = e^{-it\mathcal{L}} U_0 - i \int_0^t e^{-i(t-s)\mathcal{L}} \mathcal{L}(U(s)) ds \quad (4.3)$$

with

$$U = \begin{pmatrix} C \\ \Phi \end{pmatrix}, \mathcal{A} = \begin{pmatrix} 0 \\ \mathbf{H} \otimes \mathbb{I}_K \end{pmatrix}, \mathcal{L}(U) = \begin{pmatrix} \mathbb{K}[\Phi]C \\ \mathbf{\Gamma}(C)^{-1}(\mathbf{I} - \mathbf{P}_\Phi)\mathbb{W}[C, \Phi]\Phi \end{pmatrix}. \quad (4.4)$$

The strategy of proof is as follows.

In Subsection 4.1 we show that the operator  $\mathcal{L}$  is locally Lipschitz continuous on  $X_m$  for  $m \geq 1$  in the neighborhood of any point  $(C_0, \Phi_0)$  such that  $\mathbf{\Gamma}(C_0)$  is invertible. Observe in particular that  $\mathbf{\Gamma}(C)$  is a second-order homogeneous function of the coefficients  $C$  and therefore the invertibility of this matrix is a local property. Standard theory of evolution equations with locally Lipschitz non-linearities then guarantees local-in-time existence and uniqueness of a mild solution in these spaces that is continuous with respect to the initial data as long as the matrix  $\mathbf{\Gamma}(C)$  remains invertible (see e.g [36, 35]). Next for initial data in  $X_m$  with  $m \geq 2$ , the corresponding mild solution in this space is regular enough to be a strong solution to (4.2) (see [35, 11]). As shown in the previous section (Proposition 3.12), the strong solution then remains on the constraints fiber bundle  $\partial \mathcal{F}_{N,K}$  and it is therefore a solution to (3.26). Furthermore using the gauge equivalence and Corollary 3.11 one deduces that the energy of the solution is conserved and that the Dirac-Frenkel variational principle is satisfied. Recall for further use that the energy may be recasted in the following equivalent forms [17, 28]

$$\begin{aligned} \mathcal{E}(\Psi) &= \mathcal{E}(\pi(C, \Phi)) \\ &= \left( \left( \mathbf{H}\mathbf{\Gamma} + \frac{1}{2}\mathbb{W}[C, \Phi] \right) \Phi, \Phi \right)_{L^2(\Omega)^K} \\ &= \sum_{i,j=1}^K \gamma_{ij} \int_{\mathbb{R}^3} \left[ \frac{1}{2} \nabla \phi_i \cdot \nabla \bar{\phi}_j + U \phi_i \bar{\phi}_j \right] dx + \sum_{i,j,k,l=1}^K \gamma_{ijkl} D(\phi_l \bar{\phi}_i; \phi_k \bar{\phi}_j). \end{aligned} \quad (4.5)$$

In consequence for initial data in  $X_m$ ,  $m \geq 2$ , the norm of the vector  $\Phi(t)$  remains locally bounded in  $H^1$  (independently of the  $H^2$  norm). Therefore it is also a strong solution in  $H^1$  defined on the same time interval which depends only on the  $H^1$  norm and on  $\mathbf{\Gamma}(C_0)$ . Eventually using the density of  $X_2 \cap \partial \mathcal{F}_{N,K}$  in  $X_1 \cap \partial \mathcal{F}_{N,K}$  and the continuous dependence with the initial data one obtains the local-in-time existence of a strong solution in  $X_1 \cap \partial \mathcal{F}_{N,K}$  with constant energy.

In Subsection 4.2, relying on the conservation of the energy we prove the existence of the solution over a maximal time interval beyond which the density matrix degenerates. The equations themselves imply the further regularity  $C(t) \in C^1([0, T^*), \mathbb{C}^r)$  and  $\Phi(t) \in C^0([0, T^*), H^m(\mathbb{R}^3)^K) \cap C^1([0, T^*), H^{m-2}(\mathbb{R}^3)^K)$ .

#### 4.1. Properties of the one-parameter group and local Lipschitz properties of the non-linearities

As in Chadam and Glassey [12] for example, one checks that  $\{e^{it\mathcal{A}}\}_{t \in \mathbb{R}}$  is a one-parameter group of linear operators, unitary in  $X_0$  and uniformly bounded in time for  $0 \leq t \leq T$  in  $X_1$  and  $X_2$ .

We now show that the operator  $\mathcal{L}$  in the right-hand side of (4.3) is a locally bounded and locally Lipschitz continuous mapping in a small enough neighborhood of any  $(C_0, \Phi_0)$  in  $X_m$  such that  $\Gamma(C_0)$  is invertible for every  $m \geq 1$ . The operator  $\mathcal{L}$  reveals as a composition of locally bounded and locally Lipschitz continuous mappings as now detailed. We first recall that invertible matrices form an open subset of  $\mathcal{M}_{K \times K}(\mathbb{C})$  and that the mapping  $M \mapsto M^{-1}$  is locally Lipschitz continuous since

$$\begin{aligned} \|M^{-1} - \tilde{M}^{-1}\| &= \|M^{-1}(\tilde{M} - M)\tilde{M}^{-1}\| \\ &\leq \|M^{-1}\| \|\tilde{M}^{-1}\| \|M - \tilde{M}\|. \end{aligned}$$

In addition, being quadratic, the mapping  $C \mapsto \Gamma(C)$  is for any integer  $m$  and independently of  $m$  locally Lipschitz in  $X_m$  in a small enough neighborhood of any  $(C_0, \Phi_0)$  such that  $\Gamma(C_0)$  is invertible. The same holds true for the mapping  $C \mapsto \Gamma(C)^{-1}$  by composition of locally bounded and locally Lipschitz functions.

The operator  $\mathbf{P}_\Phi$  is a sum of  $K$  terms of the form  $\langle \phi, \cdot \rangle_{L^2} \phi$  with  $\phi$  in  $H^m(\mathbb{R}^3)$ . Hence, for  $m \geq 0$ ,

$$\|\mathbf{P}_\Phi\|_{\mathcal{L}(H^m)} \lesssim \|\Phi\|_{L^2} \|\Phi\|_{H^m} \quad (4.6)$$

where here and below  $\lesssim$  is a shorthand for a bound with a universal positive constant that only depends on  $K$  and  $N$ . Therefore  $\Phi \mapsto \mathbf{P}_\Phi$  is locally Lipschitz from  $X_m$  to  $\mathcal{L}(H^m)$  since it is quadratic with respect to  $\Phi$ . To deal with the other nonlinearities we start with recalling a few properties of the Coulomb potential taken from [12, Lemma 2.3]. Their proof is a straightforward application of Cauchy–Schwarz’ and Hardy’s inequalities and we skip it. Let  $\phi, \psi \in H^1(\mathbb{R}^3)$ , then with  $r = |x|$ ,  $(\phi\psi) \star \frac{1}{r} \in W^{1,\infty}(\mathbb{R}^3)$ , and we have

$$\|(\phi\psi) \star \frac{1}{r}\|_{L^\infty(\mathbb{R}^3)} \leq 2 \|\nabla\phi\|_{L^2(\Omega)} \|\psi\|_{L^2(\Omega)} \quad (4.7)$$

and

$$\left\| \nabla \left( (\phi\psi) \star \frac{1}{r} \right) \right\|_{L^\infty(\mathbb{R}^3)} \leq 4 \|\nabla\phi\|_{L^2(\Omega)} \|\nabla\psi\|_{L^2(\Omega)}.$$

As a consequence of above inequalities and by an induction argument that is detailed in [9] for example, we have, for  $\Phi \in H^m(\mathbb{R}^3)^K$  and for every  $1 \leq i, j, k \leq K$ ,

$$\left\| \left( (\phi_i \phi_j) \star \frac{1}{r} \right) \phi_k \right\|_{H^m(\mathbb{R}^3)} \lesssim \|\Phi\|_{H^{m'}}^2 \|\Phi\|_{H^m} \lesssim \|\Phi\|_{H^m}^3 \quad (4.8)$$

with  $m' = \max(m-1, 1)$ . First, recall from (3.29), that  $\mathbb{W}[C, \Phi] \Phi$  is a sum of terms of the form  $\gamma_{jkil} \phi_j \left( \frac{1}{r} \star \phi_k \bar{\phi}_l \right)$  with the coefficients  $\gamma_{jkil}$  depending quadratically on  $C$  according to (2.10). They are therefore locally Lipschitz continuous with respect to  $C$ . Gathering with (4.8) we have

$$\|\mathbb{W}[C, \Phi] \Phi\|_{H^m} \lesssim \|C\|^2 \|\Phi\|_{H^{m'}}^2 \|\Phi\|_{H^m} \lesssim \|C\|^2 \|\Phi\|_{H^m}^3. \quad (4.9)$$

The mapping  $(C, \Phi) \mapsto \mathbb{W}[C, \Phi] \Phi$  is then locally bounded in  $X_m$  and being quadratic in  $C$  and cubic in  $\Phi$  it is locally Lipschitz continuous in  $X_m$  by a standard polarization argument. In particular, the first bounds reveals a linear dependence on the  $H^m$

norm. Eventually, for every  $1 \leq i, j, k, l \leq K$ , using (4.7) and Hölder's inequality we obtain

$$|D(\phi_j \bar{\phi}_i, \phi_k \bar{\phi}_l)| \lesssim \|\Phi\|_{L^2}^3 \|\Phi\|_{H^1} \lesssim \|\Phi\|_{H^m}^4,$$

the last line being a direct consequence of (4.8). In particular this proves

$$\|\mathbb{K}[\Phi]\| \lesssim \|\Phi\|_{L^2}^3 \|\Phi\|_{H^1}, \quad (4.10)$$

$$\|\mathbb{K}[\Phi]C\| \lesssim \|C\| \|\Phi\|_{H^m}^4 \quad (4.11)$$

and that  $(C, \Phi) \mapsto \mathbb{K}[\Phi]C$  is locally Lipschitz continuous in  $X_m$  since according to (3.28),  $\mathbb{K}[\Phi]C$  is a finite sum of terms of this kind up to some universal constant.

For any  $m \geq 1$  existence and uniqueness of a solution  $(C(t), \Phi(t))$  to the integral equation (4.3) in a neighborhood of  $(C^0, \Phi^0)$  in  $\mathcal{C}^0(0, T; X_m)$  for  $0 < T$  small enough follows by Segal's Theorem [36], which also ensures the continuity with respect to the initial data in  $X_m$ .

We now turn to the existence of a maximal solution and to the blow-up alternative in  $X_1$ .

#### 4.2. Existence of the maximal solution and blow-up alternative

To simplify notation, from now on we use the shorthand  $\mathbf{\Gamma}(t)$  for  $\mathbf{\Gamma}(C(t))$ . Existence of a global-in-time solution requires to control uniformly both the  $H^1$  norm of  $\Phi$  and the norm of  $\mathbf{\Gamma}^{-1}(t)$ . With the conservation of the energy this turns to be equivalent to control only the norm of  $\mathbf{\Gamma}^{-1}(t)$ . Let  $T^*$  denotes the maximal existence time and assume that  $T^* < +\infty$ . We first show that

$$\limsup_{t \uparrow T^*} \|\mathbf{\Gamma}(t)^{-1}\| = +\infty. \quad (4.12)$$

We argue by contradiction and assume that there exists a positive constant  $M_0$  such that for all  $t \in [0, T^*)$ ,  $\|\mathbf{\Gamma}(t)^{-1}\| \leq M_0$ . We now prove that there exists a positive constant  $K_0$  such that

$$\forall t \in [0, T^*), \quad \|\Phi(t)\|_{H^1} \leq K_0. \quad (4.13)$$

Thanks to Lemma 3.3 and Corollary 3.11, the energy is preserved by the flow, and therefore using the expression (4.5)

$$\begin{aligned} \left( \mathbf{H}\mathbf{\Gamma}(t) \Phi(t), \Phi(t) \right) &\leq \left( \mathbf{H}\mathbf{\Gamma}(t) \Phi(t), \Phi(t) \right) + \frac{1}{2} \left( \mathbb{W}[C, \Phi] \Phi(t), \Phi(t) \right) \\ &= \mathcal{E}(\pi(C, \Phi)) = \mathcal{E}(\pi(C^0, \Phi^0)) \end{aligned}$$

for all  $0 \leq t < T^*$  since, with  $\Psi = \pi(C, \Phi)$ ,

$$\left\langle V \Psi | \Psi \right\rangle = \left( \mathbb{W}[C, \Phi] \Phi(t), \Phi(t) \right) \geq 0$$

for  $v \geq 0$ . As in [28, 17], Kato's inequality then yields that

$$\|\sqrt{\mathbf{\Gamma}} \Phi\|_{H^1} \leq M_1$$

where  $M_1$  is a positive constant independent of  $t \geq 0$ . Now let  $\mu(t) \in (0, 1]$  be the smallest eigenvalue of the hermitian matrix  $\mathbf{\Gamma}(t)$ . Then according to the definition of the Frobenius norm

$$\|\mathbf{\Gamma}^{-1}(t)\| = \left( \sum_{k=1}^K \frac{1}{\mu_k(t)^2} \right)^{\frac{1}{2}},$$

with  $\{\mu_1(t), \dots, \mu_K(t)\}$  being the eigenvalues of  $\mathbf{\Gamma}(t)$ , hence

$$\frac{1}{\mu(t)} \leq \|\mathbf{\Gamma}^{-1}\| \leq \frac{\sqrt{K}}{\mu(t)} \quad \text{and} \quad \frac{1}{\sqrt{\mu(t)}} \leq \|\sqrt{\mathbf{\Gamma}^{-1}}\| \leq \frac{K^{1/4}}{\sqrt{\mu(t)}}$$

for all  $t \in [0, T^*)$ . Therefore

$$\|\Phi\|_{H^1} \leq \frac{K^{1/4}}{\sqrt{\mu(t)}} \|\sqrt{\mathbf{\Gamma}}\Phi\|_{H^1} \leq K^{1/4} M_1 \|\mathbf{\Gamma}^{-1}\|^{1/2}. \quad (4.14)$$

In particular, this shows (4.13) with  $K_0 = K^{1/4} M_1 M_0^{1/2}$ . Therefore, for any  $t \in [0, T^*)$  arguing as above, we may build a solution to the system on  $[t, t + T_0]$  for  $T_0 > 0$  that only depends on  $M_0$  and  $K_0$ . Since  $t$  is arbitrary close to  $T^*$  we reach a contradiction with the definition of  $T^*$ . Hence (4.12).

Now, taking the derivative with respect to  $t$  of both sides of  $\mathbf{\Gamma}\mathbf{\Gamma}^{-1} = \mathbb{I}_K$ , we get

$$\frac{d\mathbf{\Gamma}^{-1}}{dt} = -\mathbf{\Gamma}^{-1} \frac{d\mathbf{\Gamma}}{dt} \mathbf{\Gamma}^{-1}, \quad (4.15)$$

for all  $t \in [0, T^*)$ . From the expression of  $\mathbf{\Gamma}$  in terms of  $C$  and since  $\|C\| = 1$ , it holds

$$\left\| \frac{d\mathbf{\Gamma}}{dt} \right\| \lesssim \left\| \frac{dC}{dt} \right\| \lesssim \|\Phi\|_{H^1} \lesssim \|\mathbf{\Gamma}^{-1}\|^{1/2}$$

in virtue of the bound (4.10) on  $\mathbb{K}[\Phi]$  using the fact that  $\|\Phi\|_{L^2} = K$ . Inserting the last bound above in (4.15) and integrating over  $t$  yields

$$\|\mathbf{\Gamma}(t)^{-1}\| \leq \|\mathbf{\Gamma}(0)^{-1}\| + \text{const.} \int_0^t \|\mathbf{\Gamma}(s)^{-1}\|^{3/2} ds,$$

for all  $t \in [0, T^*)$ . Because of (4.12), this implies that  $\int_0^{T^*} \|\mathbf{\Gamma}(s)^{-1}\|^{3/2} ds = +\infty$ .

So far we have proved the local well-posedness of the MCTDHF equations in  $X_m$  for every  $m \geq 1$  and the existence of a maximal solution in  $H^1$  until time  $T^*$  when the density matrix becomes singular. We prove now that  $T^*$  is the maximal time of existence regardless of the imposed regularity on the solution. Let  $(C, \Phi)$  be a solution in  $X_2$ , then it is in particular a maximal solution in  $X_1$ . We have to show that the  $H^2$  norm of  $\Phi$  cannot explode at finite time  $0 < \tau < T^*$ . Indeed, for any  $\tau < T^*$ , we have

$$\max_{0 \leq t \leq \tau} \|\mathbf{\Gamma}(t)^{-1}\| \lesssim 1 \quad (4.16)$$

by definition of  $T^*$ , hence

$$\max_{0 \leq t \leq \tau} \|\Phi(t)\|_{H^1} \lesssim 1. \quad (4.17)$$

From the Duhamel formula for the PDEs system (4.3)–(4.4) and using the bounds (4.6) and (4.8) together with  $\|\Phi\|_{L^2} = 1$  and  $\|C\| = 1$ , we get for all  $t \in [0, \tau]$

$$\|\Phi(t)\|_{H^2} \leq \|\Phi^0\|_{H^2} + C \sup_{[0, \tau]} \int_0^t \|\Phi(s)\|_{H^2} ds$$

where  $C$  is a positive constant that only depends on the local bounds (4.16) and (4.17). By Gronwall's lemma we infer

$$\max_{0 \leq t \leq \tau} \|\Phi(t)\|_{H^2} \lesssim e^{C\tau}$$

hence the conclusion. The proof for any  $m \geq 2$  follows then by a straightforward induction argument using the corresponding bounds (4.6) and (4.8) by assuming that  $\max_{0 \leq t \leq \tau} \|\Phi(t)\|_{H^{m-1}} \lesssim 1$ .

The proof of Theorem 4.1 is now complete.

#### 4.3. Existence of Standing wave solutions

In the present case the equations for the coefficients write (3.19) while Eqn. (3.18) for the orbitals becomes:

$$\mathbf{\Gamma}(C) \mathbf{H} \Phi + \mathbb{W}[C, \Phi] \Phi = \Lambda \cdot \Phi \quad (4.18)$$

according to Proposition 3.12. In [27] Le Bris has proved the existence of ground-states - that is, minima of the energy over the set  $\mathcal{F}_{N,K}$  - for the physical Hamiltonian (1.1), on the whole space  $\mathbb{R}^3$ , and under the assumptions  $K = N + 2$  and  $\sum_{m=1}^M z_m > N - 1$ . Later on Friesecke extended this result to general admissible pairs  $(N, K)$ , under the same assumption on the nuclear charge. Finally Lewin proved the existence of infinitely many critical points of the MCHF energy for any pairs  $(N, K)$ , hence the existence of infinitely many solutions to the coupled system (4.18) – (3.19) that satisfy the full-rank assumption. All these solutions then give rise to infinitely many standing waves of the MCTDHF system and thereby to particular global-in-time solutions.

The conservation of the invertibility of the matrix  $\mathbf{\Gamma}(t)$  being an essential issue in the MCTDHF setting it is natural to give sufficient condition for such property.

### 5. Sufficient condition for global-in-time existence

In this section we focus again on the  $N$ -body Schrödinger operator (1.1) with physical interactions (4.1). For any  $K \geq N + 1$  with fixed  $N$ , we denote

$$\mathcal{I}(K) = \inf \{ \mathcal{E}(\pi(C, \Phi)) : (C, \Phi) \in \mathcal{F}_{N,K} \}$$

the “ $K$ - ground state energy”. Obviously one has

$$\forall K' \leq K \leq \infty, \quad \inf \sigma(\mathcal{H}_N) \leq \mathcal{I}(K) \leq \mathcal{I}(K'), \quad (5.1)$$

with  $\inf \sigma(\mathcal{H}_N)$  being the bottom of the spectrum of  $\mathcal{H}_N$  on  $L^2_\lambda(\Omega^N)$ . Recall that the maximal rank hypothesis corresponds to the following equivalent facts :

- (i) The rank of the operator  $[\pi(C, \Phi) \otimes \pi(C, \Phi)]_{:,1}$  is equal to  $K$ ;
- (ii) The  $K \times K$  matrix  $\mathbf{\Gamma}(C)$  is invertible;
- (iii) The smallest eigenvalue of  $\mathbf{\Gamma}(C)$  is strictly positive.

Since this is satisfied for  $K = N$  (Hartree–Fock case) and since  $K$  must be admissible, we now assume that  $K \geq N + 2$ . The main result of this section is the following:

**Theorem 5.1.** *Let  $(C^0, \Phi^0) \in \mathcal{F}_{N,K}$  be an initial data in (3.27) with  $\mathbf{\Gamma}(C^0)$  invertible. Assume that  $T^* < +\infty$  then*

$$\mathcal{E}(\pi(C^0, \Phi^0)) \geq \mathcal{I}(K - 1).$$

As an immediate by-product we get a sufficient condition ensuring the global-in-time invertibility of the matrix  $\mathbf{\Gamma}(C(t))$ .

**Corollary 5.2.** *If  $(C^0, \Phi^0) \in d\mathcal{F}_{N,K}$  satisfies*

$$\mathcal{I}(K) \leq \mathcal{E}(\pi(C^0, \Phi^0)) < \mathcal{I}(K - 1), \quad (5.2)$$

*then  $T^* = +\infty$ ; that is, the maximal solution is global-in-time.*

**Remark 9.** The hypothesis  $\sum_{m=1}^M z_m \geq N$  in (4.1) implies the relation  $\mathcal{I}(K) < \mathcal{I}(K - 2)$  [27, 18]. Therefore (5.2) can always be satisfied by changing  $K$  into  $K - 1$ .

**Remark 10.** A key difficulty in the proof of above theorem is that the energy functional  $\Psi \mapsto \mathcal{E}(\Psi)$  is not weakly lower semi-continuous in  $H^1(\mathbb{R}^{3N})$  while it is in  $H^1(\Omega^{3N})$  for any bounded domain  $\Omega$  as already observed by Friesecke [17]. When  $\Omega$  is a bounded domain of  $\mathbb{R}^3$  or when the potential  $U$  is non-negative, the proof of Theorem 5.1 is much easier thanks to the lower semi-continuity, and it is detailed in [3]. In the general case the proof is in the very spirit of Lewin’s one for the convergence of critical points of the energy functional [28].

**Proof of Theorem 5.1.** Let  $(C, \Phi)$  be the maximal solution to (3.27) on  $[0, T^*)$  with initial data  $(C^0, \Phi^0)$  given by Theorem 4.1. We assume that  $T^* < +\infty$ , then

$$\limsup_{t \nearrow T^*} \|\mathbf{\Gamma}(C(t))^{-1}\| = +\infty.$$

Equivalently, with the eigenvalues of  $\mathbf{\Gamma}(C)$  being arranged in decreasing order  $0 \leq \gamma_K \leq \gamma_{K-1} \leq \dots \leq \gamma_1 \leq 1$ , this means

$$\liminf_{t \nearrow T^*} \gamma_K(t) = 0.$$

Then there exists a sequence  $t_n$  converging to  $T^*$ , a positive number  $\beta$  and an integer  $N+1 \leq m \leq K$  such that

$$\lim_{n \rightarrow +\infty} \gamma_m(t_n) = 0 \quad \text{and} \quad 0 < \beta \leq \gamma_{m-1}(t_n). \quad (5.3)$$

Indeed, since  $\sum_{k=1}^K \gamma_k(t) = N$ , for all  $t \in [0, T^*)$ , at least  $N$  eigenvalues stay away from zero when  $t$  goes to  $T^*$ . We denote  $C^n = C(t_n)$ ,  $\Phi^n = \Phi(t_n)$ ,  $\gamma_i^n = \gamma_i(t_n)$ ,  $\mathbf{\Gamma}^n = \mathbf{\Gamma}(C^n)$  and so on for other involved quantities.

For all  $n \geq 1$ ,  $(C^n, \Phi^n) \in d\mathcal{F}_{N,K}$ . Thus according to Proposition 2.4, there exists a unique sequence of unitary transforms  $\mathcal{U}^n \in \mathcal{O}_K^r$  that map  $(C^n, \Phi^n)$  into  $(C'^n, \Phi'^n)$  with  $\Phi'^n$  being an eigenbasis for the operator  $\gamma^n := \gamma_{\Psi^n}$ . In particular the corresponding matrix  $\mathbf{\Gamma}^n := \mathbf{\Gamma}(C'^n)$  is diagonal. In other words,

$$\begin{aligned} \Psi^n &:= \pi(C^n, \Phi^n) = \sum_{\sigma} c_{\sigma}^n \Phi_{\sigma}^n = \sum_{\sigma} c'_{\sigma}{}^n \Phi'_{\sigma}{}^n = \pi(C'^n, \Phi'^n), \\ \gamma^n &= \sum_{i,j=1}^K \gamma_{ij}^n \phi_i^n \otimes \overline{\phi_j^n} = \sum_{i=1}^K \gamma_i^n \phi_i'^n \otimes \overline{\phi_i'^n}. \end{aligned}$$

Since the group of unitary transforms is compact, we may argue equivalently on the sequence  $(C'^n, \Phi'^n)$  that we keep denoting by  $(C^n, \Phi^n)$  for simplicity. From (5.3)

$$\begin{cases} \lim_{n \rightarrow +\infty} \gamma_i^n = 0 \text{ for all } m \leq i \leq K, \\ \liminf_{n \rightarrow +\infty} \gamma_i^n \geq \beta > 0 \text{ for all } 1 \leq i \leq m-1. \end{cases} \quad (5.4)$$

Then,

$$\text{for all } \sigma \in \Sigma_N^K, \quad \{m, \dots, K\} \cap \sigma \neq \emptyset \implies \lim_{n \rightarrow +\infty} c_{\sigma}^n = 0, \quad (5.5)$$

for  $\gamma_i^n = \sum_{\sigma \in \Sigma_N^K} |c_{\sigma}^n|^2$  in virtue of (2.12). In particular, the sequence  $C^n \in S^{r-1}$  being compact

$$\lim_{n \rightarrow +\infty} \sum_{\sigma \subset \{1, \dots, m-1\}} |c_{\sigma}^n|^2 = 1. \quad (5.6)$$

We decompose

$$\Psi^n = \pi(C^n, \Phi^n) = \Psi_n^+ + \Psi_n^-$$

with

$$\Psi_n^- = \sum_{\sigma \cap \{m, \dots, K\} \neq \emptyset} c_\sigma^n \Phi_\sigma^n, \quad \Psi_n^+ = \sum_{\sigma \cap \{m, \dots, K\} = \emptyset} c_\sigma^n \Phi_\sigma^n.$$

Then

$$\lim_{n \rightarrow +\infty} \|\Psi_n^-\|_{L^2(\mathbb{R}^{3N})} = 0$$

as a consequence of (5.5) and since each determinant  $\Phi_\sigma^n$  is normalized in  $L^2(\mathbb{R}^{3N})$ . Hence

$$\lim_{n \rightarrow +\infty} \|\Psi_n^- - \Psi_n^+\|_{L^2(\mathbb{R}^{3N})} = 0. \quad (5.7)$$

Since the MCTDHF flow keeps the energy constant, we have

$$\mathcal{E}(\pi(C^n, \Phi^n)) = \mathcal{E}(\pi(C^0, \Phi^0)),$$

for all  $n \geq 1$ . This property provides with additional information on the sequence  $(C^n; \Phi^n)$ . Using the fact that the  $\phi_i^n$ 's diagonalize  $\gamma^n$ , the energy (4.5) rewrites

$$\begin{aligned} \mathcal{E}(\pi(C^n, \Phi^n)) &= \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} \left[ \frac{1}{2} |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx + \sum_{i,j,k,l=1}^K \gamma_{ijkl}^n D(\phi_i^n \bar{\phi}_i^n; \phi_k^n \bar{\phi}_j^n) \\ &\geq \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} \left[ \frac{1}{2} |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx, \end{aligned} \quad (5.8)$$

where in (5.8) we used the positivity of the two-body interaction potential  $v$ . By the Kato inequality, for any  $0 < \varepsilon < 1$ , there exists  $C_\varepsilon > 0$  such that

$$|U| \leq -\varepsilon \Delta + C_\varepsilon$$

in the sense of self-adjoint operators. Then

$$\sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} U |\phi_i^n|^2 dx \geq -\varepsilon \left( \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} |\nabla \phi_i^n|^2 dx \right) - C_\varepsilon N.$$

Therefore, inserting into (5.8),

$$\sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} |\nabla \phi_i^n|^2 dx \leq cste.$$

Thus, for all  $1 \leq i \leq K$ ,  $\sqrt{\gamma_i^n} \phi_i^n$  is bounded in  $H^1(\mathbb{R}^3)$ . Then, from (5.4) and extracting subsequences if necessary, we obtain the alternative

for all  $m \leq i \leq K$ ,  $\sqrt{\gamma_i^n} \phi_i^n$  converges to 0 weakly in  $H^1(\mathbb{R}^3)$  and strongly in  $L^2(\mathbb{R}^3)$ ,

(5.9)

and

for all  $1 \leq i \leq m-1$ ,  $\phi_i^n$  is bounded in  $H^1(\mathbb{R}^3)$ . (5.10)

Since, under the hypotheses on  $U$ , the map  $\varphi \mapsto \int_{\mathbb{R}^3} U |\varphi|^2 dx$  is weakly lower semi-continuous on  $H^1(\mathbb{R}^3)$ , we deduce from (5.9) that

$$\liminf_{n \rightarrow +\infty} \sum_{i=1}^K \gamma_i^n \int_{\mathbb{R}^3} \left[ \frac{1}{2} |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx \geq \liminf_{n \rightarrow +\infty} \sum_{i=1}^{m-1} \gamma_i^n \int_{\mathbb{R}^3} \left[ |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx. \quad (5.11)$$

We now check that

$$\liminf_{n \rightarrow +\infty} \sum_{i,j,k,l=1}^K \gamma_{ijkl}^n D(\phi_l^n \bar{\phi}_i^n; \phi_k^n \bar{\phi}_j^n) = \liminf_{n \rightarrow +\infty} \sum_{i,j,k,l=1}^{m-1} \gamma_{ijkl}^n D(\phi_l^n \bar{\phi}_i^n; \phi_k^n \bar{\phi}_j^n) \quad (5.12)$$

by showing that

$$\liminf_{n \rightarrow +\infty} \sum_{\substack{i,j,k,l=1 \\ \{i,j,k,l\} \cap \{m,\dots,K\} \neq \emptyset}}^K \gamma_{ijkl}^n D(\phi_l^n \bar{\phi}_i^n; \phi_k^n \bar{\phi}_j^n) = 0. \quad (5.13)$$

Let  $\{i,j,k,l\} \cap \{m,\dots,K\} \neq \emptyset$ . We assume without loss of completeness that  $i \geq m$ . From the expression (2.10) for  $\gamma_{ijkl}^n$ , we observe that

$$\begin{aligned} |\gamma_{ijkl}^n| &\lesssim \min(\sqrt{\gamma_i^n}; \sqrt{\gamma_j^n}) \min(\sqrt{\gamma_k^n}; \sqrt{\gamma_l^n}) \\ &\lesssim \min(\gamma_i^n; \gamma_k^n; \gamma_j^n; \gamma_l^n)^{1/2}, \end{aligned} \quad (5.14)$$

since  $0 \leq \gamma^n \leq 1$ . We thus get

$$\text{if } \{i,j,k,l\} \cap \{m,\dots,K\} \neq \emptyset, \quad \lim_{n \rightarrow +\infty} \gamma_{ijkl}^n = 0, \quad (5.15)$$

from (5.4). Then thanks to (4.7) and (5.14)

$$\begin{aligned} |\gamma_{ijkl}^n D(\phi_l^n \bar{\phi}_i^n; \phi_k^n \bar{\phi}_j^n)| &\lesssim \sqrt{\gamma_i^n} \sqrt{\gamma_k^n} \|\nabla \phi_k^n\|_{L^2} \|\phi_l^n\|_{L^2} \|\phi_j^n\|_{L^2} \|\phi_l^n\|_{L^2} \\ &\lesssim \sqrt{\gamma_i^n} \end{aligned}$$

since the  $L^2$  norms of the orbitals equal 1 and since in any case  $\sqrt{\gamma_j^n} \nabla \phi_j^n$  is bounded in  $L^2$  independently of  $n$ . Therefore each term which appears in the sum in (5.13) converges to 0 as  $n$  goes to infinity thanks to (5.9). Claim (5.13) then follows.

Gathering together (5.11) and (5.12) we have

$$\begin{aligned} &\liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(C^n; \Phi^n)) \\ &\geq \liminf_{n \rightarrow +\infty} \left[ \sum_{i=1}^{m-1} \gamma_i^n \int_{\mathbb{R}^3} \left[ |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx + \sum_{i,j,k,l=1}^{m-1} \gamma_{ijkl}^n D(\phi_i^n \bar{\phi}_l^n; \phi_k^n \bar{\phi}_j^n) \right] \end{aligned} \quad (5.16)$$

The point now consists in showing that the right-hand side in (5.16) is bounded from below by  $\liminf_{n \rightarrow +\infty} \mathcal{E}(\Psi_n^+)$ . Indeed, let us set  $\tilde{\Psi}^n = \pi(\tilde{C}^n, \tilde{\Phi}^n)$  where  $\tilde{C}^n = (c_\sigma^n)_{\sigma \in \{1,\dots,m-1\}} \in \mathbb{C}^{\binom{K}{m-1}}$  and  $\tilde{\Phi}^n = (\phi_1^n, \dots, \phi_{m-1}^n) \in \mathcal{O}_{L^2(\Omega)^{m-1}}$ . There is a slight

difficulty arising here from the fact that (with obvious notation)  $\tilde{\gamma}_{ij}^n$  is close but different from  $\gamma_{ij}^n \delta_{ij}^n$  and similarly for  $\tilde{\gamma}_{ijkl}^n$  and  $\gamma_{ijkl}^n$ . (Also  $\tilde{C}^n$  is not normalized in  $\mathbb{C}^{m-1}$  (only asymptotically) but this will be dealt with afterwards.)

First we observe that because of (2.11) for every  $i, j \in \{1, \dots, m-1\}$ ,

$$\gamma_i^n \delta_{ij}^n - \tilde{\gamma}_{ij}^n = \sum_{\substack{(\sigma \cup \tau) \cap \{m, \dots, K\} \neq \emptyset \\ i \in \sigma, j \in \tau, \sigma \setminus \{i\} = \tau \setminus \{j\}}} (-1)^{\sigma^{-1}(i) + \tau^{-1}(j)} c_\sigma^n \bar{c}_\tau^n$$

goes to 0 as  $n$  goes to infinity thanks to (5.5). In addition, each term of the form  $\int_{\mathbb{R}^3} \left[ \frac{1}{2} \nabla \phi_i^n \cdot \nabla \bar{\phi}_j^n + U \phi_i^n \cdot \bar{\phi}_j^n \right] dx$  is bounded independently of  $n$  for  $i, j \in \{1, \dots, m-1\}$ . Therefore

$$\sum_{i=1}^{m-1} \gamma_i^n \int_{\mathbb{R}^3} \left[ \frac{1}{2} |\nabla \phi_i^n|^2 + U |\phi_i^n|^2 \right] dx = \sum_{i,j=1}^{m-1} \tilde{\gamma}_{ij}^n \int_{\mathbb{R}^3} \left[ \frac{1}{2} \nabla \tilde{\phi}_i^n \cdot \nabla \tilde{\phi}_j^n + U \tilde{\phi}_i^n \tilde{\phi}_j^n \right] dx + o(1). \quad (5.17)$$

For the same reason, and with obvious notation, for all  $1 \leq i, j, k, l \leq m-1$ ,

$$\lim_{n \rightarrow +\infty} |\gamma_{ijkl}^n - \tilde{\gamma}_{ijkl}^n| = 0$$

since according to (2.10) the extra terms in these differences only involve coefficients  $c_\sigma^n$  with  $\sigma \cap \{m, \dots, K\} \neq \emptyset$ . Again each term of the form  $D(\phi_i^n \bar{\phi}_l^n; \phi_k^n \bar{\phi}_j^n)$  is bounded independently of  $n$  for  $i, j, k, l \in \{1, \dots, m-1\}$ . Therefore

$$\sum_{i,j,k,l=1}^{m-1} \gamma_{ijkl}^n D(\phi_i^n \bar{\phi}_l^n; \phi_k^n \bar{\phi}_j^n) = \sum_{i,j,k,l=1}^{m-1} \tilde{\gamma}_{ijkl}^n D(\phi_i^n \bar{\phi}_l^n; \phi_k^n \bar{\phi}_j^n) + o(1). \quad (5.18)$$

Therefore, gathering together (5.16), (5.17) and (5.18),

$$\liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(C^n; \Phi^n)) \geq \liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(\tilde{C}^n; \tilde{\Phi}^n)) = \liminf_{n \rightarrow +\infty} \mathcal{E}(\tilde{\Psi}^n). \quad (5.19)$$

Since  $\tilde{C}^n$  is not in  $\mathcal{S}^{(m-1)^{-1}}$  (it is only the case asymptotically),  $(\tilde{C}^n; \tilde{\Phi}^n)$  is not in  $\mathcal{F}_{N,m-1}$ , thus we cannot bound immediately  $\mathcal{E}(\tilde{\Psi}^n)$  from below by  $I(m-1)$ . Anyway, in virtue of (5.6),

$$\lim_{n \rightarrow +\infty} \|\tilde{\Psi}^n\|^2 = 1. \quad (5.20)$$

The energy being quadratic with respect to  $\Psi$

$$\mathcal{E}(\tilde{\Psi}^n) = \|\tilde{\Psi}^n\|^2 \mathcal{E}\left(\frac{\tilde{\Psi}^n}{\|\tilde{\Psi}^n\|}\right) \geq \|\tilde{\Psi}^n\|^2 I(m-1), \quad (5.21)$$

for  $\tilde{\Psi}^n / \|\tilde{\Psi}^n\| \in \mathcal{F}_{N,m-1}$  for all  $n \geq 1$ . Gathering together (5.19), (5.20) and (5.21) and taking the limit as  $n$  goes to infinity we deduce

$$\liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(C^n, \Phi^n)) \geq I(m-1). \quad (5.22)$$

Hence the theorem.  $\square$

**Remark 11.** When  $\Omega$  is a bounded domain of  $\mathbb{R}^3$ , any sequence in  $\mathcal{F}_{N,K}$  is relatively compact in  $\mathbb{C}^r \times L^2(\Omega)^K$  thanks to the Rellich theorem. On the other hand, the energy functional  $\Psi \mapsto \mathcal{E}(\Psi)$  is weakly lower semi-continuous in  $H^1(\Omega^{3N})$ . Therefore it is easily checked in that case that

$$\liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(C^n; \Phi^n)) \geq \liminf_{n \rightarrow +\infty} \mathcal{E}(\pi(C^*; \Phi^*)) \geq I(m-1)$$

with  $(C^*; \Phi^*) \in \mathcal{F}_{N,m-1}$  being the weak limit of the sequence  $(\tilde{C}^n; \tilde{\Phi}^n)$  introduced in the above proof.

**Remark 12 (Stability, Consistency and Invertibility of the density matrix  $\Gamma$ ).**

The main factor in the instability of the working equations or any gauge-equivalent system, is the inverse of the density matrix. In the present section, criteria for the global invertibility of  $\Gamma(C)$  have been given. These criteria do not provide with an uniform estimate for  $\|\Gamma^{-1}\|$ , and furthermore increasing the consistency of the MCTDHF approximation leads to an increase of the number  $K$  of orbitals. As usual consistency and stability are both necessary and antinomic. Indeed, the most obvious observation is that one always has

$$\|\Gamma^{-1}\| \geq \frac{K}{N},$$

for  $\Gamma$  has at most  $K$  positive eigenvalues whose sum equals  $N$ . Therefore the smallest can be at most  $N/K$ . These considerations lead either to a limitation on  $K$  or to a regularization or a ‘‘cut-off’’ of  $\Gamma^{-1}$ . In fact the ‘‘consistency’’ in the sense of numerical analysis is obtained with fixed  $N$  by letting  $K$  go to infinity. This is basically different from the idea (in spirit of statistical mechanics) of letting  $N$  go to infinity [4].

## 6. Stabilization of $\Gamma$ and existence of $L^2$ solutions

In the above analysis, both for existence of maximal solutions and for global invertibility of the density matrix, the conservation of energy plays a crucial rôle. Besides the theoretical interest, the analysis of an MCTDHF system with infinite (or non conserved) energy but finite mass is relevant. Indeed, to circumvent the possible degeneracy of the density matrix, physicists resort to *ad hoc* methods like perturbations of this matrix in order to ensure its invertibility. Typically, this is achieved as follows

$$\Gamma_\varepsilon = \Gamma + \varepsilon \text{Id} \tag{6.1}$$

(see e.g. [7]), or by taking

$$\Gamma_\varepsilon = \Gamma + \varepsilon \exp(-\Gamma/\varepsilon) \tag{6.2}$$

for small values of  $\varepsilon$  (see [5]). Note that in latter case vanishing eigenvalues are perturbed at order  $\varepsilon$  while the others are unchanged up to exponentially small

errors in terms of  $\varepsilon$ . Then for  $\varepsilon > 0$  the perturbed system reads

$$\begin{cases} i \frac{dC}{dt} = \mathbb{K}[\Phi] C, \\ i \frac{\partial \Phi}{\partial t} = \mathbf{H} \Phi + \mathbf{\Gamma}_\varepsilon(C)^{-1} (\mathbf{I} - \mathbf{P}_\Phi) \mathbb{W}[C, \Phi] \Phi, \\ C(0) = C^0, \quad \Phi(0) = \Phi^0. \end{cases} \quad (6.3)$$

On the other hand, when a laser field is turned on, the Hamiltonian of the system is then time-dependent which is a relevant configuration from the physical point of view (see [7] and Section 7 below). In such situation, the conservation of the energy fails and a recourse to alternative theories is necessary.

However in both situations the  $L^2$  norm (which corresponds to the electronic charge) is conserved and this justifies an  $L^2$  analysis of the MCTDHF outside the energy space. Therefore the Strichartz estimates turn out to be a natural tool in the same spirit as Castella [9] for the Schrödinger-Poisson equation and Zagatti [39]. for the TDHF equations. Strichartz estimates read

$$\|e^{-it\Delta} \phi\|_{L^p(0,T;L^q(\mathbb{R}^3))} \leq C(q) T^{\frac{3}{q}-\frac{1}{2}},$$

for any *Strichartz pairs*  $(p, q) \in [2, +\infty) \times [2, 6]$  with  $\frac{2}{3p} = (\frac{1}{2} - \frac{1}{q})$ . (Estimates for the endpoints  $p = 2$  and  $q = 6$  are more intricate and due to Keel and Tao [24]). In [34], existence and uniqueness of global-in-time mild solutions has been obtained for  $L^2$  initial data. The result is summarized in the following.

**Proposition 6.1.** [34] *Let  $\varepsilon > 0$ . For any initial data  $(C_0, \Phi_0)$  in  $d\mathcal{F}_{N,K}$  and for any Strichartz pairs  $(p, q)$ , the  $\varepsilon$ -regularized working equations admit a unique global mild solution*

$$C_\varepsilon(t) \in L^\infty(\mathbb{R}^+; \mathbb{C}^r), \quad \Phi_\varepsilon(t) \in L^\infty(\mathbb{R}^+; L^2(\mathbb{R}^3))^K \cap L_{\text{loc}}^p(\mathbb{R}^+; L^q(\mathbb{R}^3))^K$$

*that lives in  $\mathcal{F}_{N,K}$  for all  $t \geq 0$ . If in addition  $\Phi_0 \in H^1(\mathbb{R}^3)^K$  then  $\Phi_\varepsilon$  lies in  $C^0(\mathbb{R}^+; H^1(\mathbb{R}^3))^K$ .*

The proof is as follows. For every  $\varepsilon > 0$  existence and uniqueness of a mild solution in  $L^\infty(\mathbb{R}^+; L^2(\mathbb{R}^3))^K$  is proved on a time interval  $[0, T_\varepsilon)$  with  $T_\varepsilon > 0$  that only depends on  $\varepsilon$  and on the  $L^2$  norm of the perturbed orbitals that is preserved by the flow. Therefore solutions can be extended to  $[0; 2T_\varepsilon)$ , next  $[0; 3T_\varepsilon)$ ... and so on, thereby providing with global-in-time solutions.

Eventually one expects that whenever the original solution is well-defined (with a non-degenerate density matrix  $\mathbf{\Gamma}(t)$ ) on a time interval  $0 \leq t < T^*$  it will be on the same interval the limit for  $\varepsilon \rightarrow 0$  of the solution of the perturbed working equations. This is the object of the following

**Theorem 6.2.** *Let  $(C_0, \Phi_0) \in \mathcal{S}^{r-1} \times (H^1(\mathbb{R}^3))^K$ . Assume that the corresponding strong solution  $(C(t), \Phi(t))$  to (3.27) is well-defined on  $[0, T]$  and is such that*

$$\sup_{0 \leq t \leq T} \|\mathbf{\Gamma}(t)^{-1}\| \leq M < +\infty. \quad (6.4)$$

Then, on the same time interval it is the limit in  $\mathbb{C}^r \times H^1(\mathbb{R}^3)^K$  for  $\varepsilon \rightarrow 0$  of the solution  $(C_\varepsilon, \Phi_\varepsilon)$  to the regularized problem (6.3) with same initial data.

**Proof.** We first recall the obvious *a posteriori* bounds

$$\|C\| = \|C_\varepsilon\| = 1, \quad \|\Phi\|_{L^2} = \|\Phi_\varepsilon\|_{L^2} = 1$$

on  $[0, T]$ , and, as a consequence of (6.4) and the energy conservation,

$$\max_{0 \leq t \leq T} \|\Phi(t)\|_{H^1} \leq M'$$

with  $M' = M'(\mathcal{E}(\pi(C_0, \Phi_0)), M)$ . We can also rely on the orthonormality of the orbitals in  $\Phi$  and  $\Phi_\varepsilon$ . We introduce the notation

$$U = \begin{pmatrix} C \\ \Phi \end{pmatrix}, \quad U_\varepsilon = \begin{pmatrix} C_\varepsilon \\ \Phi_\varepsilon \end{pmatrix}, \quad \mathcal{A} = \begin{pmatrix} 0 \\ \mathbf{H} \end{pmatrix}, \quad \mathcal{B}_{(\varepsilon)}(U) = \begin{pmatrix} \mathbb{K}[\Phi]C \\ \mathbf{\Gamma}_{(\varepsilon)}^{-1} \mathbb{B}(U) \end{pmatrix}$$

where

$$\mathbb{B}(U) = (\mathbf{I} - \mathbf{P}_\Phi) \mathbb{W}[C, \Phi] \Phi$$

and where the index  $(\varepsilon)$  means that the claim holds both for the regularized system and the initial one, uniformly in  $\varepsilon$ . System (6.3) can also be written in synthetic form:

$$U(t) = e^{-it\mathcal{A}} U_0 - i \int_0^t e^{-i(t-s)\mathcal{A}} \mathcal{B}(U(s)) ds, \quad (6.5)$$

$$U_\varepsilon(t) = e^{-it\mathcal{A}} U_0 - i \int_0^t e^{-i(t-s)\mathcal{A}} \mathcal{B}_\varepsilon(U_\varepsilon(s)) ds. \quad (6.6)$$

Since the initial  $\Phi_\varepsilon(0) = \Phi_0$  is in  $H^1$  and since the regularized system propagates the regularity,  $\Phi_\varepsilon$  is in  $H^1(\mathbb{R}^3)^K$  for all time.

We fix  $\varepsilon > 0$ . We introduce a parameter  $\eta > 0$  to be made precise later and the set

$$I_\varepsilon = \{t \in [0, T] : \|U_\varepsilon(t) - U(t)\| \leq \eta\}$$

with  $\|U\| = \|C\| + \|\Phi\|_{H^1}$ . The mappings  $t \mapsto U_{(\varepsilon)}(t)$  being continuous from  $[0, T]$  to  $X_T := \mathbb{C}^r \times L^\infty(0, T; H^1(\mathbb{R}^3)^K)$ , the set  $I_\varepsilon$  is closed and since  $\|U_\varepsilon(0) - U(0)\| = 0$ , there exists a maximal time  $T_\varepsilon > 0$  in  $I_\varepsilon$  such that

$$\forall t \in [0, T_\varepsilon], \quad \|U_\varepsilon(t) - U(t)\| \leq \eta.$$

We now prove by contradiction that  $T_\varepsilon = T$ . Assume then  $T_\varepsilon < T$ .

Subtracting (6.5) to (6.6) and taking norms first yields to

$$\begin{aligned} \|C_\varepsilon(t) - C(t)\| &\leq \int_0^t \|C_\varepsilon\| \|\mathbb{K}[\Phi_\varepsilon] - \mathbb{K}[\Phi]\| + \|C_\varepsilon - C\| \|\mathbb{K}[\Phi]\|, \\ &\leq C(\eta) \int_0^t \|U_\varepsilon - U\| ds \end{aligned} \quad (6.7)$$

for all  $0 \leq t \leq T_\varepsilon$ . Here and below  $C(\eta) = C(M, \mathcal{E}(\pi(C_0, \Phi_0)), \eta)$  denotes a positive constant that may vary from line to line but that is independent of  $\varepsilon$  and continuous and non-decreasing with respect to  $\eta$ . Indeed we use the fact that the non-linearity  $\Phi \mapsto \mathbb{K}[\Phi]$  is locally Lipschitz continuous in  $H^1$  (Subsection 4.1) together with the uniform bound

$$\max_{0 \leq t \leq T_\varepsilon} \|\Phi_\varepsilon\|_{H^1} \leq M' + \eta.$$

On the other hand, we write

$$\begin{aligned} \|\Phi_\varepsilon(t) - \Phi(t)\| &\lesssim \int_0^t \|\mathbb{B}(U_\varepsilon)\|_{H^1} \|\mathbf{\Gamma}_\varepsilon^{-1} - \mathbf{\Gamma}^{-1}\| + \|\mathbf{\Gamma}^{-1}\| \|\mathbb{B}(U_\varepsilon) - \mathbb{B}(U)\|_{H^1} \\ &\leq C(\eta) \int_0^t \left( \|\mathbf{\Gamma}_\varepsilon^{-1} - \mathbf{\Gamma}^{-1}\| + \|U_\varepsilon - U\|_{H^1} \right) ds, \end{aligned} \quad (6.8)$$

by using the local Lipschitz bounds of  $U \mapsto \mathbb{B}(U)$  given in Subsection 4.1. We now turn to the quantity  $\|\mathbf{\Gamma}_\varepsilon^{-1} - \mathbf{\Gamma}^{-1}\|$ . Both regularization (6.1) and (6.2) of the density matrix take the form :

$$\mathbf{\Gamma}_\varepsilon = \mathbf{\Gamma}(C_\varepsilon) + \varepsilon g(C_\varepsilon)$$

with  $\|g(C_\varepsilon)\| \leq \varepsilon$ . Then,

$$\begin{aligned} \|\mathbf{\Gamma}_\varepsilon - \mathbf{\Gamma}\| &\leq \|\mathbf{\Gamma}(C_\varepsilon) - \mathbf{\Gamma}(C)\| + \varepsilon \\ &\leq \kappa(\|C_\varepsilon - C\| + \varepsilon) \end{aligned} \quad (6.9)$$

by using the obvious bound  $\|\mathbf{\Gamma}(C)\| \lesssim \|C\|^2$  for  $C, C_\varepsilon \in S^{r-1}$ , where  $\kappa$  only depends on  $N$  and  $K$ . We now assume that

$$\varepsilon, \eta \leq \frac{1}{4\kappa M}, \quad (6.10)$$

where  $M$  is given in the statement of the theorem. Using

$$\mathbf{\Gamma}_\varepsilon = \left( I - (\mathbf{\Gamma} - \mathbf{\Gamma}_\varepsilon) \mathbf{\Gamma}^{-1} \right) \mathbf{\Gamma},$$

we deduce

$$\mathbf{\Gamma}_\varepsilon^{-1} = \mathbf{\Gamma}^{-1} \left( I - (\mathbf{\Gamma} - \mathbf{\Gamma}_\varepsilon) \mathbf{\Gamma}^{-1} \right)^{-1} = \mathbf{\Gamma}^{-1} \sum_{n \geq 0} \left( (\mathbf{\Gamma} - \mathbf{\Gamma}_\varepsilon) \mathbf{\Gamma}^{-1} \right)^n.$$

Therefore

$$\begin{aligned} \|\mathbf{\Gamma}_\varepsilon^{-1} - \mathbf{\Gamma}^{-1}\| &\leq \sum_{n \geq 1} \|\mathbf{\Gamma} - \mathbf{\Gamma}_\varepsilon\|^n \|\mathbf{\Gamma}^{-1}\|^{n+1} \\ &\leq \sum_{n \geq 1} M^{n+1} \kappa^n (\|C_\varepsilon - C\| + \varepsilon)^n \end{aligned}$$

by using (6.9). Hence

$$\begin{aligned} \|\mathbb{I}_\varepsilon^{-1} - \mathbb{I}^{-1}\| &\leq M^2 \kappa (\|C_\varepsilon - C\| + \varepsilon) \sum_{n \geq 0} M^n \kappa^n (\|C_\varepsilon - C\| + \varepsilon)^n \\ &\leq 2M^2 \kappa (\|C_\varepsilon - C\| + \varepsilon) \end{aligned} \quad (6.11)$$

since  $M \kappa (\|C_\varepsilon(t) - C(t)\| + \varepsilon) \leq \frac{1}{2}$  by (6.10) and for  $t$  in  $[0, T_\varepsilon]$ . Inserting (6.11) in (6.8) we get:

$$\|\Phi_\varepsilon(t) - \Phi(t)\| \leq C(\eta) \int_0^t (\|U_\varepsilon(s) - U(s)\| + \varepsilon) ds. \quad (6.12)$$

Eqn. (6.12) together with (6.7) finally leads to

$$\|U_\varepsilon(t) - U(t)\| \leq C(\eta) \int_0^t (\|U_\varepsilon(s) - U(s)\| + \varepsilon) ds, \quad (6.13)$$

for all  $t \in [0, T_\varepsilon]$ . Eventually, thanks to Gronwall's inequality,

$$\max_{0 \leq t \leq T_\varepsilon} \|U_\varepsilon(t) - U(t)\| \leq \varepsilon e^{C(\eta)T}. \quad (6.14)$$

With  $\eta$  as in (6.10), next

$$\varepsilon \leq \min\left(\frac{1}{4\kappa M}, \frac{\eta}{2} e^{-C(\eta)T}\right), \quad (6.15)$$

we get

$$\max_{0 \leq t \leq T_\varepsilon} \|U_\varepsilon(t) - U(t)\| \leq \frac{\eta}{2}.$$

By continuity of  $t \mapsto \|U_\varepsilon(t) - U(t)\|$ , we may then find  $T'_\varepsilon > T_\varepsilon$  such that  $\max_{0 \leq t \leq T'_\varepsilon} \|U_\varepsilon(t) - U(t)\| \leq \eta$ . Hence the contradiction with the definition of  $T_\varepsilon$ . Therefore,  $I_\varepsilon = [0, T]$  and, going back to (6.14) we obtain:

$$\max_{0 \leq t \leq T} \|U_\varepsilon(t) - U(t)\| \leq \varepsilon e^{C(\eta)T}, \quad (6.16)$$

for say  $\eta = \frac{1}{4\kappa M}$  and  $\varepsilon$  small enough, satisfying (6.15), whence the result.

**Remark 13.** Theorem 6.2 implies that

$$\forall 0 < \varepsilon \leq 1, \quad \sup_{0 \leq t \leq T} \|\mathbb{I}_\varepsilon(t)^{-1}\| \leq M. \quad (6.17)$$

Conversely if for a sequence of regularized solutions the bound (6.17) holds uniformly with respect to  $\varepsilon$ , then one can obviously pass to the limit  $\varepsilon$  to 0 and obtain thereby a strong solution to (3.27) on  $[0, T]$  with  $\mathbb{I}(C)$  invertible. Hence, for the limit unperturbed system, the maximal existence  $T^*$  is strictly bigger than  $T$ . Therefore, if the maximal existence time  $T^*$  is finite for the unperturbed system the bound (6.9) fails, that is  $\|\mathbb{I}_\varepsilon(t)^{-1}\|$  explodes at  $T^*$  when  $\varepsilon$  approaches 0, and this might be observed on numerical simulations.

In the forthcoming (and last) section we comment on straight extensions of the above analysis.

## 7. Extensions

The present contribution is focused on the algebraic and functional analysis properties of the MCTDHF equations for fermions. Multi-configuration approximations can also be considered for symmetric wave-functions or also for wave-functions with no symmetry (see *e.g.* [5, 26]). The mathematical analysis of the equations which play the rôle of the “working equations” of Section 3 is similar. On the other hand, the fermionic case is important by itself and leads to much better geometric structure in terms of principal fiber bundle as described in Section 2. Hence our choice. Our results could be generalized to general (symmetric)  $n$ -body interactions as well including the  $n$ -body density matrices.

### 7.1. Beyond Coulomb potentials.

Although above results and proofs are mainly detailed for Coulomb potentials they carry through more general real-valued potentials. Indeed well-posedness results in  $H^1$  and  $H^2$  are still valid for  $U$  and  $v$  in the class  $L^p(\mathbb{R}^3) + L^\infty(\mathbb{R}^3)$  with  $p > 3/2$ , and  $v \geq 0$ . These conditions ensure that  $\mathcal{H}_N$  is self-adjoint in  $L^2(\Omega^N)$ , that the one-body operator  $-\frac{1}{2}\Delta + U$  is a semi-bounded self-adjoint in  $L^2(\mathbb{R}^3)$  with domain  $H^2(\mathbb{R}^3)$  and that the Kato inequality holds for the potential  $U$ . Under these assumptions, the energy space is  $\mathbb{C}^r \times H^1(\mathbb{R}^3)^K$  (respectively  $\mathbb{C}^r \times H_0^1(\Omega)^K$  when  $\Omega$  is a bounded domain with Dirichlet boundary conditions) and the propagator  $e^{-itH}$  is a one-parameter group of unitary operators in  $H^2(\mathbb{R}^3)$  and in  $H^1(\mathbb{R}^3)$ .

For the global well-posedness sufficient condition to hold true (Theorem 5.1 and its corollary) further conditions on the potentials are required to ensure that the energy functional is weakly lower semi-continuous on the energy space. Sufficient conditions are (for example)  $U \geq 0$  or  $U_-$  (the negative part of  $U$ ) tending to 0 at infinity at least in a weak sense.

### 7.2. Extension to time-dependent potentials.

One of the basic use of the MCTDHF is the simulation of ultra-short light pulses with matter [40]. Describing this situation leads to the same type of equations but with the one-body Hamiltonian  $\mathbf{H}$  being replaced by a one-body time dependent hamiltonian

$$\mathbf{H}_{\omega,A}(t) := (i\nabla + A(t))^2 + \omega(t)U(x)$$

with  $\omega(t)$  and  $A(t)$  real,  $A(0) = 0$  and  $U$  as in the above subsection. A typical example is  $A(t) = A_0 \exp(-t/\tau)^2 \sin(\alpha t)$  for some positive real parameters  $A_0$ ,  $\alpha$  and  $\tau$  [40, 41]. This does not change neither the algebraic and geometrical structure of the equations nor the definition of the density matrix  $\mathbf{\Gamma}$  nor the notion of full-rank. The potential vector  $A$  being independent of the  $x$  variable the energy space is  $H^1$ . With convenient hypotheses (say  $\omega$  and  $A$  continuous, bounded with bounded derivatives), the results in Section 4 concerning local-in-time  $H^1$  well-posedness of the Cauchy problem remain valid. For generalization of the use of Strichartz estimates and the local  $L^2$  well-posedness one should follow for example [10]. Since

the energy is now time-dependent extra hypothesis have to be introduced for the persistence of the full-rank assumption done in Section 5.

Assume that  $\omega(t)$  and  $A(t)$  take their values in a bounded set (the set of “control”  $\mathcal{C}$ ) and that their derivatives are also bounded. The system  $\mathcal{S}_0$  (3.25) with  $\mathbf{H}$  replaced by  $\mathbf{H}_{\omega,A}(t)$  keeps on preserving the constraints since Lemma 3.1 only relies on the self-adjointness of the Hamiltonian. Similarly solutions to (3.25) satisfy the Dirac–Frenkel variational principle. The energy is no longer conserved by the flow. Indeed, following the lines of the proof of Corollary 3.3, we have

$$\frac{d}{dt} \mathcal{E}(\Psi(t)) = \frac{d}{dt} \langle \mathcal{H}(t) \Psi(t) | \Psi(t) \rangle = (\omega'(t) + 2A(t)A'(t)) \langle \Psi(t) | \Psi(t) \rangle,$$

with the prime denoting time derivatives. However

$$\mathcal{E}(\Psi(t)) = \omega(t) + 2A(t)^2 + \mathcal{E}(\Psi(0)) - \omega(0),$$

and the energy is controlled for any finite time, whence the existence of a maximal solutions in  $H^1$  as long as the matrix  $\mathbf{\Gamma}(C(t))$  remains invertible.

To adapt the result concerning the global full-rank hypothesis, we introduce the minimization problems for any real numbers  $\bar{\omega}$  and  $\bar{A}$

$$\mathcal{J}_{\bar{\omega},\bar{A}}(K) = \inf \left\{ \mathcal{E}_{\bar{\omega},\bar{A}}(\Psi) : \Psi \in \mathcal{B}_{N,K} \right\}$$

with

$$\mathcal{E}_{\bar{\omega},\bar{A}}(\Psi) = \left( \left( \mathbf{H}_{\bar{\omega},\bar{A}} \mathbf{\Gamma} + \frac{1}{2} \mathbb{W}[C, \Phi] \right) \Phi, \Phi \right)_{L^2(\Omega)^K}$$

for  $\Psi = \pi(C, \Phi)$ . The global-in-time conservation of full-rank in Theorem 5.1 remains true under the hypothesis

$$\begin{aligned} \mathcal{E}_{\omega(0),A(0)}(\Psi(0)) &< \inf \left\{ \mathcal{J}_{\bar{\omega},\bar{A}}(K-1) : |\bar{\omega}| \leq \|\omega\|_{L^\infty(\mathbb{R}^+)}, |\bar{A}| \leq \|A\|_{L^\infty(\mathbb{R}^+)} \right\} \\ &\quad - \|\omega\|_{L^\infty(\mathbb{R}^+)} - 2\|A\|_{L^\infty(\mathbb{R}^+)}^2 + \omega(0). \end{aligned}$$

There is a lot of room for improvement in the above argument. For example, if we assume that, for all time, the solution  $\Psi = \pi(C, \Phi) \in d\mathcal{B}_{N,K}$  satisfies

$$\left\langle \frac{\partial \mathcal{H}}{\partial t} \Psi(t) | \Psi(t) \right\rangle \leq h(t) \langle \mathcal{H}(t) \Psi(t) | \Psi(t) \rangle$$

for a given function  $h$ , then by the Gronwall lemma

$$\langle \mathcal{H}(t) \Psi(t) | \Psi(t) \rangle - \langle \mathcal{H}(0) \Psi_0 | \Psi_0 \rangle \leq \exp \left( \int_0^t h(s) ds \right).$$

The result of Theorem 5.1 remains true provided

$$\mathcal{E}(\Psi_0) = \langle \mathcal{H}(0) \Psi_0 | \Psi_0 \rangle \leq \mathcal{J}(K-1) - \exp \left( \int_0^{+\infty} h(s) ds \right).$$

### 7.3. Discrete systems.

The emphasis has been but in particular for the functional analysis on the case when  $\Omega = \mathbb{R}^3$  although in the first part we have described the problem in any open subset of  $\mathbb{R}^3$ . In fact all the formal and algebraic derivations can also be adapted to the case when  $\Omega$  is a discrete set equipped with a discrete Lebesgue measure and in particular when  $\Omega$  is a finite set [3]. Such situation is important for two reasons. On the one hand many models of quantum physics (the Ising model for instance) involve a discrete Hamiltonian defined on a discrete set. On the other hand the discretization of the original problem in view of any numerical algorithm leads to a discrete problem.

Up to now only a rough *a posteriori* error estimate has been proven. However if the MCTDHF algorithm is applied to a discrete model say of dimension  $L$  then one always has  $K \leq L$ . The error formula (3.13) shows that, for  $K = L$ , the MCTDHF algorithm is exact. It should be eventually observed that in general the two operations : - Discretization of the original  $N$ -particle problem and use of a MCTDHF approximation or - Use of a MCTDHF approximation and then discretization of the equations, lead to different algorithms.

## Appendix – Proofs of technical lemmas in Subsection 3.3

### Proofs of Corollary 3.6 and Lemma 3.9

For  $\sigma$  and  $\tau$  given and fixed  $1 \leq j \leq N$  it is convenient to denote by  $\mathbb{U}_{\sigma, \tau(j)}$  the column vector in  $\mathbb{C}^N$  with entries  $(\mathbb{U}_{\sigma(i), \tau(j)})_{1 \leq i \leq N}$  and by

$$[\mathbb{U}_{\tau(1)}, \mathbb{U}_{\tau(2)}, \dots, \mathbb{U}_{\tau(N)}]_{\sigma}$$

the determinant composed with these vectors. With this notation (3.14) gives

$$i \frac{d\mathbb{U}_{\sigma, \tau(j)}}{dt} = \sum_{k=1}^K M_{k, \tau(j)} \mathbb{U}_{\sigma, k} \quad (7.1)$$

Differentiating the relation

$$\mathbb{U}_{\sigma, \tau} = [U_{\tau(1)}, U_{\tau(2)}, \dots, U_{\tau(N)}]_{\sigma}$$

and using the multi-linearity with respect to the column vectors and Eqn. (7.1) one obtains:

$$i \frac{d\mathbb{U}_{\sigma, \tau}}{dt} = \sum_{\substack{1 \leq k \leq K \\ 1 \leq j \leq N}} M_{k, \tau(j)} [U_{\tau(1)}, U_{\tau(2)}, \dots, U_{\tau(j-1)}, U_k, U_{\tau(j+1)}, \dots, U_{\tau(N)}]_{\sigma}. \quad (7.2)$$

On the other hand since  $\mathbb{U}(t)$  is a flow of unitary matrices it is solution to a differential equation of the following type:

$$i \frac{d\mathbb{U}_{\sigma, \tau}}{dt} = \sum_{\tau'} [U_{\tau'(1)}, U_{\tau'(2)}, \dots, U_{\tau'(N)}]_{\sigma} \tilde{\mathbb{M}}_{\tau', \tau} \quad (7.3)$$

Identification of the coefficients of

$$[U_{\tau'(1)}, U_{\tau'(2)}, \dots, U_{\tau'(N)}]_{\sigma}$$

gives, taking in account the number of permutation needed to change

$$\tau(1), \tau(2), \dots, \tau(j-1), k, \tau(j+1), \dots, \tau(N) \text{ into } \tau'(1), \tau'(2), \dots, \tau'(N)$$

$$\tilde{M}_{\tau', \tau} = \sum_{\substack{k \in \tau', j \in \tau \\ \tau' \setminus \{k\} = \tau \setminus \{j\}}} M_{k,j} (-1)^{\tau^{-1}(j) + \tau'^{-1}(k)}.$$

Let us now prove (3.21). Let  $\sigma, \tau \in \Sigma_{N,K}$ . We first observe that

$$\sum_{i=1}^N \mathbf{G}_{x_i} \Phi_{\sigma} = \sum_{i=1}^N \phi_{\sigma(1)} \wedge \dots \wedge \mathbf{G} \phi_{\sigma(i)} \wedge \dots \wedge \phi_{\sigma(N)}. \quad (7.4)$$

Now we use (2.5) and the Laplace method to develop a determinant with respect to the row that contains the terms involving  $\mathbf{G}$  to get

$$\begin{aligned} \sum_{i=1}^N \langle \mathbf{G}_{x_i} \Phi_{\sigma} | \Phi_{\tau} \rangle &= \sum_{i=1}^N \langle \phi_{\sigma(1)} \wedge \dots \wedge \mathbf{G} \phi_{\sigma(i)} \wedge \dots \wedge \phi_{\sigma(N)} | \Phi_{\tau} \rangle \\ &= \sum_{i,j=1}^N (-1)^{i+j} \langle \mathbf{G} \phi_{\sigma(i)}, \phi_{\tau(j)} \rangle \delta_{\sigma \setminus \{\sigma(i)\}, \tau \setminus \{\tau(j)\}}, \end{aligned}$$

in virtue of (2.4). Hence (3.21) using (3.16) and the definition of  $M$ .  $\square$

#### *Proofs of Theorem 3.10 and Theorem 3.7.*

Let  $(C(t), \Phi(t))$  be a solution to  $\mathcal{S}_0$  and let  $\mathbf{G}$  be as in the statement of the theorem. With  $M_{ij} = \langle \mathbf{G} \phi_i, \phi_j \rangle$  we define the family of unitary transforms  $U(t)$  according to Lemma 3.9 and  $d(U)(t) = \bar{U}(t)$  is then given by Corollary 3.6. We set  $\mathbb{V} = \bar{U}$ ,  $C'(t) = \mathbb{V}(t) C(t)$  and  $\Phi'(t) = U(t) \Phi(t)$ . Thanks to (3.15),  $\mathbb{V}$  solves

$$\begin{cases} i \frac{d\mathbb{V}}{dt} = -\mathbb{V} \bar{M}, \\ \mathbb{V}(0) = d(U^0). \end{cases} \quad (7.5)$$

Then, for all  $\sigma \in \Sigma_{N,K}$ ,

$$\begin{aligned} i \frac{dC'}{dt} &= i \frac{d\mathbb{V}}{dt} C + \mathbb{V} i \frac{dC}{dt} = -\mathbb{V} \bar{M} \mathbb{V}^* C' + \mathbb{V} \langle \mathcal{H} \Psi | \nabla_C \Psi \rangle \\ &= -\mathbb{V} \bar{M} \mathbb{V}^* C' + \mathbb{V} \langle \mathcal{H} \Psi | \nabla_C \Psi \mathbb{V} \rangle \end{aligned}$$

thanks to (2.25) and (7.5). On the one hand, since  $\mathbb{V}$  is unitary,

$$\mathbb{V} \langle \mathcal{H} \Psi | \nabla_C \Psi \mathbb{V} \rangle = \langle \mathcal{H} \Psi | \nabla_C \Psi \rangle.$$

On the other hand, when  $M$  is obtained through  $\mathbf{G}$ , we get by a direct calculation from (3.21)

$$\left(\nabla\overline{\mathbf{M}}\nabla^*C'\right)_\sigma = \sum_\tau \left\langle \sum_{i=1}^N \mathbf{G}_{x_i} \Phi'_\tau | \Phi'_\sigma \right\rangle c'_\tau = \left\langle \sum_{i=1}^N \mathbf{G}_{x_i} \Psi | \Phi'_\sigma \right\rangle.$$

Combining these two facts we get the first equation in  $\mathcal{S}_{\mathbf{G}}$ , namely

$$i \frac{dC'}{dt} = \left\langle \mathcal{H} \Psi | \nabla_{C'} \Psi \right\rangle - \left\langle \sum_{i=1}^N \mathbf{G}_{x_i} \Psi | \nabla_{C'} \Psi \right\rangle.$$

We turn now to the equation satisfied by  $\Phi'$ . To simplify the notation we use the shorthand  $\mathbf{\Gamma}$  for  $\mathbf{\Gamma}(C)$  and  $\mathbf{\Gamma}'$  for  $\mathbf{\Gamma}(C')$  respectively. Then, using  $\mathbf{\Gamma}' = U \mathbf{\Gamma} U^*$  and (3.14), we have

$$\begin{aligned} i \mathbf{\Gamma}' \frac{\partial \Phi'}{\partial t} &= \mathbf{\Gamma}' i \frac{dU}{dt} \Phi + \mathbf{\Gamma}' U i \frac{\partial \Phi}{\partial t} \\ &= \mathbf{\Gamma}' U M U^* \Phi' + U \mathbf{\Gamma} i \frac{\partial \Phi}{\partial t} \\ &= \mathbf{\Gamma}' U M U^* \Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'}) U \nabla_{\Phi} \Psi^* [\mathcal{H} \Psi] \\ &= \mathbf{\Gamma}' U M U^* \Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'}) \nabla_{\Phi'} \Psi^* [\mathcal{H} \Psi] \end{aligned} \quad (7.6)$$

thanks to (2.26) and since clearly  $\mathbf{P}_{\Phi'} = \mathbf{P}_{\Phi}$  for  $\text{Span}\{\Phi\} = \text{Span}\{\Phi'\}$ . It is easily checked that when  $M$  is given through  $\mathbf{G}$  we have

$$(U M U^*)_{ij} = \langle \mathbf{G} \phi'_i, \phi'_j \rangle$$

and therefore

$$U M U^* \Phi' = \mathbf{P}_{\Phi'} \mathbf{G} \Phi'.$$

Hence (7.6) also writes

$$i \mathbf{\Gamma}' \frac{\partial \Phi'}{\partial t} = \mathbf{\Gamma}' \mathbf{G} \Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'}) \nabla_{\Phi'} \Psi^* [\mathcal{H} \Psi] - (\mathbf{I} - \mathbf{P}_{\Phi'}) \mathbf{\Gamma}' \mathbf{G} \Phi'.$$

We now check that, for all  $1 \leq i \leq N$ ,

$$(\mathbf{I} - \mathbf{P}_{\Phi'}) (\mathbf{\Gamma}' \mathbf{G} \Phi')_i = (\mathbf{I} - \mathbf{P}_{\Phi'}) \frac{\partial \Psi^*}{\partial \phi'_i} \left[ \sum_{j=1}^N \mathbf{G}_{x_j} \Psi \right],$$

thereby proving that

$$i \mathbf{\Gamma}' \frac{\partial \Phi'}{\partial t} = \mathbf{\Gamma}' \mathbf{G} \Phi' + (\mathbf{I} - \mathbf{P}_{\Phi'}) \nabla_{\Phi'} \Psi^* \left[ \mathcal{H} \Psi - \sum_{i=1}^N \mathbf{G}_{x_i} \Psi \right].$$

Indeed, for all  $\xi \in L^2(\Omega)$ , using (2.28) in Lemma 2.5 in (7.7) and using (7.4) in (7.8), we have

$$\begin{aligned} \langle (\mathbf{I} - \mathbf{P}_\Phi) (\mathbf{\Gamma} \mathbf{G} \Phi)_i, \xi \rangle &= \sum_{k=1}^K \Gamma_{ik} \langle \mathbf{G} \phi_k, (\mathbf{I} - \mathbf{P}_\Phi) \xi \rangle \\ &= \sum_{k=1}^K \left\langle \frac{\partial \Psi}{\partial \phi_k} [\mathbf{G} \phi_j] \mid \frac{\partial \Psi}{\partial \phi_i} [(\mathbf{I} - \mathbf{P}_\Phi) \xi] \right\rangle \end{aligned} \quad (7.7)$$

$$\begin{aligned} &= \left\langle \sum_{j=1}^N \mathbf{G}_{x_j} \Psi \mid \frac{\partial \Psi}{\partial \phi_i} [(\mathbf{I} - \mathbf{P}_\Phi) \xi] \right\rangle \quad (7.8) \\ &= \left\langle (\mathbf{I} - \mathbf{P}_\Phi) \frac{\partial \Psi^*}{\partial \phi_i} \left[ \sum_{j=1}^N \mathbf{G}_{x_j} \Psi \right], \xi \right\rangle \end{aligned}$$

by the definition (2.24) of  $\frac{\partial \Psi^*}{\partial \phi_i}$ ; whence the result since  $\xi$  is arbitrary in  $L^2(\Omega)$ .  $\square$

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