Accuracy of the time-dependent Hartree-Fock approximation

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Abstract

This article examines the time-dependent Hartree-Fock (TDHF) approximation of single-particle dynamics in systems of interacting fermions. We find the TDHF approximation to be accurate when there are sufficiently many particles and the initial many-particle state is a Slater determinant, or any Gibbs equilibrium state for noninteracting fermions. Assuming a bounded two-particle interaction, we obtain a bound on the error of the TDHF approximation, valid for short times. We further show that the error of the the TDHF approximation vanishes at all times in the mean field limit.

1 Introduction

Dirac [10] invented the time-dependent Hartree-Fock equation in 1930. The time-dependent Hartree-Fock (TDHF) equation is a nonlinear Schrödinger equation designed to approximate the evolution of the single-electron state in an n-electron system. Dirac noted that the TDHF equation, originally written as a system of n coupled Schrödinger equations for occupied orbitals, may be written as a Liouville-von Neumann equation for the single-particle reduced density operator. We study the TDHF equation in this form, availing ourselves of trace norm techniques inspired by [12] to estimate the error of the TDHF approximation.

We will consider Hamiltonian dynamics of fermions interacting through a two body potential V. The energy operator for a solitary particle will be denoted by L, the energy of interaction

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of a single pair of particles will be denoted by V, and the total energy operator for a system of particles will be the sum of all single-particle energies and all pair energies. In this article we only consider bounded interaction potentials V; the case where V represents Coulomb repulsion between electrons will be treated in a forthcoming paper [3]. Although the number of particles does not change under the dynamics just described, we prefer to formulate the dynamics on a fermion Fock space, so that we may consider initial states of indeterminate particle number. We are particularly interested in initial states which are Gibbs equilibrium states (grand-canonical ensembles), for we are going to show that such initial states enhance the accuracy of the TDHF approximation.

Let L be a self-adjoint operator on $\mathbb{H}$, and let V be a bounded Hermitian operator on $\mathbb{H}\otimes\mathbb{H}$ that commutes with the transposition operator U defined by $U(x\otimes y)=y\otimes x$. We are going to discuss the dynamics on the fermion Fock space $\mathbb{F}_{\mathbb{H}}$ whose Hamiltonian H may be written in second quantized form as

$$H \;=\; \sum_{i,j}\langle j|L|i\rangle a_j^\dagger a_i \;+\; \sum_{i,j,k,l}\langle kl|V|ij\rangle a_k^\dagger a_l^\dagger a_j a_i \tag{1}$$

We will analyze the solutions of the Liouville-von Neumann equation

$$\begin{aligned} i\hbar\frac{d}{dt}D(t) &= [H,D(t)] \\ D(0) &= D_0, \end{aligned} \tag{2}$$

which is the evolution equation for the density operator in the Schrödinger picture of quantum dynamics.

We will see that (2) leads to the following equation for the single-particle density operator $\mathcal{N}_1(t)$:

$$\begin{aligned} i\hbar\frac{d}{dt}\mathcal{N}_1(t) &= [L,\mathcal{N}_1(t)] \;+\; [V,\mathcal{N}_2(t)]_{:1} \\ \mathcal{N}_1(0) &= \mathcal{N}_1(D_0). \end{aligned} \tag{3}$$

(The subscript $_{:1}$ denotes a partial trace; see definition (11) below.) This equation for $\mathcal{N}_1(t)$ is not "closed," for its right hand side involves the two-particle density operator $\mathcal{N}_2(t)$. The TDHF approximation to $\mathcal{N}_1(t)$ is the solution of the initial value problem

$$\begin{aligned} i\hbar\frac{d}{dt}F(t) &= [L,F(t)] \;+\; [V,\; F(t)^{\otimes 2}2A_2]_{:1} \\ F(0) &= \mathcal{N}_1(D(0)) \end{aligned} \tag{4}$$

where A_2 is the orthogonal projector of $\mathbb{H}\otimes\mathbb{H}$ onto the subspace of antisymmetric vectors. The existence and uniqueness of solutions of (4) were established in [6] for the case where V is a bounded operator, and in [9, 7] for the case where V is a Coulombic interaction.

The TDHF equation (4) is obtained by closing the single-particle equation (3) with the Ansatz

$$\mathcal{N}_2 = (\mathcal{N}_1 \otimes \mathcal{N}_1)2A_2 \tag{5}$$

at all times. The relation (5) holds for pure states corresponding to Slater determinants, and also for Gibbs densities. However, even supposing that $\mathcal{N}_2(0)$ satisfies (5), the interaction V is likely to introduce "correlations" in $\mathcal{N}_2(t)$, that is, departures from (5), and ignoring those correlations in the TDHF equation requires some justification.

We are going to prove that the absence of correlations is self-perpetuating in the mean field limit: if $\mathcal{N}_2(0)$ satisfies (5) then $\mathcal{N}_2(t)$ asymptotically satisfies (5) as the number of particles N tends to infinity and the interaction strength is scaled as $1/N$. This is the content of Theorem 5.2. To prove this theorem we first bound the error of the TDHF approximation in terms of the average particle number and interaction strength. This bound is presented in Theorem 5.1 without reference to the mean field scaling, and it applies to any system of fermions whose Hamiltonian has the form (1). Unfortunately, the bound is valid only at short times, i.e., up to a time inversely proportional to $\|V\|$ and the average particle number.

Let us advert to some shortcomings of our results. Firstly, we consider only bounded two-particle potentials V. Fortunately, the challenges presented by the Coulomb potential can be overcome [3, 5] and the mean field limit of Theorem 5.2 also holds for certain electronic systems. Secondly, the explicit error bound of Theorem 5.1 is valid only at short times, which are too short to be of interest in molecular-electronic problems, even if the Coulomb interaction between electrons is truncated at the Bohr radius. However, the model (1) does not only apply to molecular-electronic problems, and we hope that Theorem 5.1 may find other applications, perhaps to certain models of interactions between nucleons [4].

We first published our derivation of the TDHF equation in the mean field limit in [2]. There we assumed that the initial states are Slater determinants. The main theme of this article is that the initial states need not be Slater determinants; the TDHF approximation should work equally well (or badly) for all initial many-fermion states of Gibbs type. Also, in this article we are not only interested in the mean field limit, and we offer the error bound of Theorem 5.1 for the unscaled problem.

The next section gives the background on fermion Fock space, trace class operators, reduced density operators, and Gibbs equilibrium states. Section 3 carefully defines the many-particle dynamics generated by (1) and derives the single-particle equation (3) for $\mathcal{N}_1(t)$. Section 4 introduces the TDHF approximation of $\mathcal{N}_1(t)$ and its higher-order analogs. Section 5 presents Theorems 5.1 and 5.2. Finally, the appendix contains the proofs of several Propositions.

2 Definitions and notation

Consider a quantum particle whose Hilbert space is $\mathbb{H}$, i.e., a particle which, in isolation, would constitute a system whose (pure) quantum states are represented by the rank-one orthogonal projectors on some Hilbert space $\mathbb{H}$. The set of quantum states available to a system of n particles of this kind is determined by their "statistics," i.e., whether the particles are fermions, bosons, or distinguishable. If the particles are fermions, the pure states of a system of n of them are represented by the rank-one projectors onto vectors in the *antisymmetric subspace* $\mathbb{H}^{(n)}$ of the tensor power space $\mathbb{H}^{\otimes n}$.

The Hilbert space $\mathbb{H}^{\otimes n}$ is the closed span of the simple tensors

$$x_1 \otimes x_2 \otimes \cdots \otimes x_n \qquad\qquad x_1, x_2, \ldots, x_n \in \mathbb{H}$$

with the inner product

$$\langle y_1 \otimes y_2 \otimes \cdots \otimes y_n,\ x_1 \otimes x_2 \otimes \cdots \otimes x_n \rangle \ = \ \langle y_1, x_1 \rangle \langle y_2, x_2 \rangle \cdots \langle y_n, x_n \rangle.$$

Let Π_n denote the group of permutations of $\{1, 2, \ldots, n\}$. For each $\pi \in \Pi_n$, a unitary operator U_π on $\mathbb{H}^{\otimes n}$ may be defined by extending

$$U_\pi(x_1 \otimes x_2 \otimes \cdots \otimes x_n) \ = \ x_{\pi^{-1}(1)} \otimes x_{\pi^{-1}(2)} \otimes \cdots \otimes x_{\pi^{-1}(n)}$$

to all of $\mathbb{H}^{\otimes n}$. A vector $\psi \in \mathbb{H}^{\otimes n}$ is *antisymmetric* if $U_\pi(\psi) = \mathrm{sgn}(\pi)\psi$ for all $\pi \in \Pi_n$. The antisymmetric vectors in $\mathbb{H}^{\otimes n}$ form a closed subspace which will be denoted $\mathbb{H}^{(n)}$. The orthogonal projector with range $\mathbb{H}^{(n)}$ is

$$A_n \ = \ \frac{1}{n!} \sum_{\pi \in \Pi_n} \mathrm{sgn}(\pi) U_\pi. \tag{6}$$

If $x_1, x_2, \ldots, x_n$ is an orthonormal system in a Hilbert space $\mathbb{H}$, then the vector

$$\sqrt{n!}\ A_n(x_1 \otimes x_2 \otimes \cdots \otimes x_n), \tag{7}$$

is a unit vector in $\mathbb{H}^{(n)}$. A vector of the form (7) is called a *Slater determinant*. If $\{e_\alpha\}$ is a complete orthonormal system in $\mathbb{H}$, then a basis of $\mathbb{H}^{(n)}$ may be chosen from the set of all Slater determinants of the form (7) where $\{x_1, \ldots, x_n\}$ a subset of the single particle basis of cardinality n.

If the number of particles in the system is not fixed, the appropriate Hilbert space is the direct sum of the n-particle spaces $\mathbb{H}^{(n)}$. This is the (fermion) Fock space

$$\mathbb{F}_\mathbb{H} \ = \ \mathbb{H}^{(0)} \oplus \mathbb{H}^{(1)} \oplus \mathbb{H}^{(2)} \oplus \mathbb{H}^{(3)} \oplus \cdots \qquad . \tag{8}$$

The possibility of a zero-particle state is accommodated by $\mathbb{H}^{(0)} \approx \mathbb{C}$, a one-dimensional space spanned by the *vacuum vector* Ω. The *number operator* N on $\mathbb{F}_{\mathbb{H}}$ is the self-adjoint operator whose restriction to $\mathbb{H}^{(n)}$ equals multiplication by n. Annihilation and creation operators a_x and $a_x^\dagger$ are explicitly represented on $\mathbb{F}_{\mathbb{H}}$ as discussed in [8]. The statistical state of a system of fermions with single-particle space $\mathbb{H}$ determines a bounded positive continuous functional ω on the bounded operators on $\mathbb{F}_{\mathbb{H}}$ with $\omega(I) = 1$. We are only going to consider *normal states*, i.e., those ω that satisfy

$$\omega(A) \;=\; \mathrm{Tr}(D_\omega A)$$

for some nonnegative trace class operator D_ω of unit trace, which may be called the *(statistical) density operator*.

Density operators on any Hilbert space $\mathbb{K}$ are trace class operators in particular. Let $\mathcal{T}(\mathbb{K})$ denote the real Banach space of Hermitian trace class operators T with the norm $\|T\|_1 = \mathrm{Tr}(|T|)$. We often use the inequality that, for bounded operators B on $\mathbb{K}$,

$$|\mathrm{Tr}(TB)| \;\leq\; \|T\|_1 \|B\|. \tag{9}$$

A linear functional defined on the space of compact Hermitian operators by

$$\Phi_T(K) \;=\; \mathrm{Tr}(TK) \tag{10}$$

is continuous by (9), and indeed the space $\mathcal{T}(\mathbb{K})$ is isomorphic to the Banach dual of the space of compact Hermitian operators on $\mathbb{K}$, via the isomorphism $T \longmapsto \Phi_T$.

2.1 Reduced density operators

We now restrict our attention to density operators D on $\mathbb{F}_{\mathbb{H}}$ that commute with the number operator and such that $N^m D$ is trace class for all $m \in \mathbb{N}$. For such D we will define the m^{th} *order reduced number density operators* $\mathcal{N}_m(D)$ and explain their physical significance.

We begin by defining the reduction of an n-particle statistical density operator, i.e., a positive operator of unit trace D on $\mathbb{H}^{(n)}$, to an m-particle density operator denoted $D_{:m}$. If A is an operator on $\mathbb{H}^{(n)}$, let $\overline{A}$ denote the extension of A to $\mathbb{H}^{\otimes n}$ defined by first projecting onto $\mathbb{H}^{(n)}$ and then applying A, i.e.,

$$\overline{A} \;=\; A \oplus \mathbf{0}_{\mathbb{H}^{\otimes n} \ominus \mathbb{H}^{(n)}}.$$

For each $0 \leq m \leq n$ there exists a positive contraction from $\mathcal{T}(\mathbb{H}^{(n)})$ onto $\mathcal{T}(\mathbb{H}^{(m)})$ known as the *partial trace*. This partial trace map, which we denote by $T \mapsto T_{:m}$, is defined implicitly through the duality (10) by the requirement that

$$\mathrm{Tr}((\overline{K} \otimes I \otimes \cdots \otimes I)\overline{T}) \;=\; \mathrm{Tr}(KT_{:m})$$

for all compact Hermitian operators K on $\mathbb{H}^{(m)}$. It follows that, for any orthonormal basis $\mathcal{O}$ of $\mathbb{H}^{\otimes n-m}$,

$$\langle \xi, T_{:m}\psi\rangle \;=\; \sum_{\phi\in\mathcal{O}} \langle (\xi\oplus 0)\otimes\phi,\; \overline{T}((\psi\oplus 0)\otimes\phi)\rangle \;. \tag{11}$$

In case D is a density operator on $\mathbb{H}^{(n)}$, and if $m \leq n$, the operator $D_{:m}$ is known as the *m-particle reduced density operator* [11]. It is used to determine the expected values of m-particle observables when the n-particle system is in the statistical state D. The m-particle reduced density operators obtained from P_ψ will be denoted $(P_\psi)_{:m}$. If ψ is an n-particle Slater determinant, then the reduced density operators $(P_\psi)_{:m}$ satisfy

$$(P_\psi)_{:1} \;=\; \frac{1}{n}\sum_{j=1}^{n} P_{x_j} \tag{12}$$

$$(P_\psi)_{:m} \;=\; \frac{n^m}{\binom{n}{m}}\,(P_\psi)_{:1}^{\otimes m}\,A_m. \tag{13}$$

Now we can define the reduced number densities $\mathcal{N}_m$, which serve to describe the m-particle correlations in a system of many particles. Let D be a density operator that commutes with N. Then

$$D \;=\; \bigoplus_{n=0}^{\infty} D_n \tag{14}$$

where each D_n is a nonnegative trace class operator on $\mathbb{H}^{(n)}$. Assume that

$$\sum_{n=0}^{\infty} n^m \mathrm{Tr}(D_n) \;<\; \infty. \tag{15}$$

For such D, define the m^{th} order reduced number density

$$\mathcal{N}_m(D) \;=\; \sum_{n=m}^{\infty} \frac{n!}{(n-m)!} D_{n:m}. \tag{16}$$

The operators $\mathcal{N}_m(D)$ are called *reduced density operators* in Section 6.3.3 of [8], but we prefer to call them reduced *number* density operators because the trace of $\mathcal{N}_1$ is the average particle number. They are trace class by (15). An n-particle density operator D_n extends to a density operator

$$\widehat{D_n} \;=\; \mathbf{0}\oplus\cdots\oplus\mathbf{0}\otimes D_n\oplus\mathbf{0}\oplus\cdots \tag{17}$$

on all of Fock space. It is clear that $\mathcal{N}_m(\widehat{D_n})$ equals $\mathcal{N}_m(\widehat{D_n}) = n(n-1)\cdots(n-m+1)D_{n:m}$ if $m \leq n$ but it equals the zero operator if $m > n$.

Formula (19) below may clarify the sense in which $\mathcal{N}_1$ determines expected values of single-particle observables. Following [8], we define the second quantization $\mathrm{d}\Gamma(H)$ of a self-adjoint

operator H on $\mathbb{H}$ to be the closure of the essentially self-adjoint operator $\oplus \mathrm{d}\Gamma_n(H)$ on $\mathbb{F}_\mathbb{H}$, where $\mathrm{d}\Gamma_n(H)$ denotes the restriction to $\mathbb{H}^{(n)}$ of the operator

$$\mathrm{d}\overline{\Gamma}_n(H) \;=\; \sum_{j=1}^{n} \overset{j-1 \;\; times}{I \otimes \cdots \otimes I} \otimes H \otimes \overset{n-j \;\; times}{I \otimes \cdots \otimes I} \tag{18}$$

on $\mathbb{H}^{\otimes n}$. The operator $\mathrm{d}\Gamma(H)$ is called the *second quantization of* H; we might also call it a *single-particle observable*. Suppose that ω is a normal state on $\mathbb{F}_\mathbb{H}$ whose density D_ω satisfies (14) and (15). From (16) it then follows that

$$\omega(\mathrm{d}\Gamma(B)) \;=\; \mathrm{Tr}(\mathcal{N}_1(D_\omega)B) \tag{19}$$

for any bounded Hermitian operator B on $\mathbb{H}$. For example, the number operator N on $\mathbb{F}_\mathbb{H}$ is the second quantization of the identity operator, i.e., $N = \mathrm{d}\Gamma(I)$. From (19) we see that the trace of $\mathcal{N}_1(D_\omega)$ is the average number of particles in the system when it is in the state ω. Another example: if x is a unit vector in $\mathbb{H}$, the operator $a_x^\dagger a_x$ on $\mathbb{F}_\mathbb{H}$ is the second quantization $\mathrm{d}\Gamma(P_x)$ of the rank-one projector P_x, and (19) tells us that $\omega(a_x^\dagger a_x) = \mathrm{Tr}(\mathcal{N}_1(D_\omega)P_x)$.

Theorems 5.1 and 5.2 rely on the following fact, which is proved in the appendix:

Proposition 2.1 *If D is a density operator on $\mathbb{F}_\mathbb{H}$ that commutes with N and such that $Tr(ND) < \infty$, then the operator norm of $\mathcal{N}_1(D)$ is not greater than* 1.

2.2 Gibbs equilibrium states

A Gibbs equilibrium state is that of a system of noninteracting fermions in thermal and chemical equilibrium with its environment, but only in case $e^{-\beta H}$ is trace class, where H is the single-particle Hamiltonian and $1/\beta$ is Boltzmann's constant times the temperature. In this case the density operator is proportional to $\exp(-\beta\ \mathrm{d}\Gamma(H - \mu I))$, where $\mu \in \mathbb{R}$ is the chemical potential. This density operator is diagonalizable with respect to an occupation number basis of Fock space, and occupation numbers are stochastically independent. If the single-particle Hamiltonian has discrete eigenvalues $\epsilon_1 \leq \epsilon_2 \leq \cdots$, the probability of occupation of the j^{th} level equals $1/(1 + e^{\beta(\epsilon_j - \mu)})$.

When D is a Gibbs equilibrium density then $\mathcal{N}_m(D) = \mathcal{N}_1(D)^{\otimes m} m! A_m$. The same is true when D is a pure state corresponding to a Slater determinant. Indeed, this relation characterizes the "gauge-invariant quasifree states of the CAR algebra" that have a trace class single-particle operator, and both Slater densities and Gibbs equilibrium densities are of this type.

We describe these states in probabilistic language. Let $\{\phi_j\}_{j \in J}$ be a basis of $\mathbb{H}$. One basis of $\mathbb{F}_\mathbb{H}$ consists of the vacuum vector Ω and all Slater determinants made of vectors from $\{\phi_j\}$. This basis is indexed by $\mathcal{F}$, the set of finite subsets of J including the empty set. A nonempty subset $\mathbf{s} = \{j_1, \ldots, j_n\}$ of J corresponds to a Slater determinant $\Psi(\mathbf{s})$ formed from the vectors

$\phi_{j_1}, \ldots, \phi_{j_n}$. (There are in fact two such Slater determinants, opposite in sign, and we choose one of them.) The empty subset of J corresponds to the vacuum vector Ω, i.e., $P(\oslash) = \Omega$. Let $\mathbb{P}$ be a probability measure on $\mathcal{F}$ with the σ-field of all its subsets. That is, let $\mathbb{P} : \mathcal{F} \longrightarrow [0,1]$ be equal to 0 except on a countable subset of $\mathcal{F}$, on which subset $\sum \mathbb{P}(\mathbf{s}) = 1$. Define the random variables N_j on $\mathcal{F}$ by

$$N_j(\mathbf{s}) \;=\; \begin{cases} 1 & \text{if} \quad j \in \mathbf{s} \\ 0 & \text{if} \quad j \notin \mathbf{s} \end{cases}$$

and define $N = \sum N_j$. Suppose that the N_j are independent with respect to $\mathbb{P}$, and define

$$\mathbb{P}(N_j = 1) \;=\; p(j). \tag{20}$$

Then

$$\sum_{\mathbf{s} \ni j_1, \ldots, j_n} \mathbb{P}(\mathbf{s}) \;=\; \mathbb{P}((N_{j_1} = 1) \cap \cdots \cap (N_{j_n} = 1)) \;=\; \prod_{i=1}^{n} p(j_i) \tag{21}$$

when $j_1, \ldots, j_n$ are distinct. Note that $\mathbb{E}(N) \;=\; \sum_{j \in J} p(j) \;<\; \infty$ since $\mathbb{P}(N < \infty) = 1$.

Given a probability measure $\mathbb{P}$ on $\mathcal{F}$, we define a density operator on $\mathbb{F}_{\mathbb{H}}$ by

$$G[\mathbb{P}] \;=\; \sum_{\mathbf{s} \in \mathcal{F}} \mathbb{P}(\mathbf{s}) P_{\mathbf{s}} \;, \tag{22}$$

where $P_{\mathbf{s}}$ denotes the orthogonal projector onto the span of $\Psi(\mathbf{s})$.

Proposition 2.2 *Let $\mathbb{P}$ be as in (20) and (21). Let G denote the density operator $G[\mathbb{P}]$ of (22). Then for all $n \in \mathbb{N}$*

$$\mathcal{N}_n(G) \;=\; \mathcal{N}_1(G)^{\otimes n} n! A_n. \tag{23}$$

The preceding proposition is proved in the appendix. Proposition 2.2 and the following proposition (also proved in the appendix) together imply Proposition 2.4 below:

Proposition 2.3 *If T is a Hermitian trace class operator then*

$$\left\| T^{\otimes n} n! A_n \right\|_1 \;\leq \left\| T \right\|_1^n. \tag{24}$$

Proposition 2.4 *Let G be as in Proposition 2.2. Then $\left\| \mathcal{N}_n(G) \right\|_1 \;\leq\; \left\| \mathcal{N}_1(G) \right\|_1^n$.*

3 Definition of the dynamics on Fock space

We now define the dynamics (2) more carefully, and derive the reduced dynamics (3) from (2).

Let H be a self-adjoint operator on $\mathbb{H}$, and let V be a bounded Hermitian operator on $\mathbb{H}\otimes\mathbb{H}$ that commutes with the transposition operator $U(x\otimes y)=y\otimes x$. For $1\le j\le n$, let L_j denote the operator

$$\overset{j-1\ \ times}{I\otimes\cdots\otimes I}\otimes L\otimes\overset{n-j\ \ times}{I\otimes\cdots\otimes I}$$

on $\mathbb{H}^{\otimes n}$ (the value of $n\ge j$ is not explicit in the notation L_j but it will always be clear from context). For $1\le i<j\le n$, let $U_{(ij)}$ denote the permutation operator on $\mathbb{H}^{\otimes n}$ that transposes the i^{th} and j^{th} factors of any simple tensor $x_1\otimes\cdots\otimes x_n$, and let

$$V_{ij} \;=\; U_{(1i)}U_{(2j)}\left(V\otimes I^{\otimes n-2}\right)U_{(2j)}U_{(1i)}$$

(again, the domain $\mathbb{H}^{\otimes n}$ of V_{ij} will always be clear from context). For each n, define the operators

$$\begin{aligned} L^{(n)} &= \sum_{j=1}^{n} L_j \\ H^{(n)} &= L^{(n)} + \sum_{1\le i<j\le n} V_{ij} \end{aligned}$$

on $\mathbb{H}^{(n)}$ (these operators are defined on all of $\mathbb{H}^{\otimes n}$ but we are only considering their restrictions to the invariant subspace $\mathbb{H}^{(n)}$). The Hamiltonian operator H, which we had formally represented above by (1), is the direct sum $H=\bigoplus H^{(n)}$ defined on the domain

$$\mathcal{D}(H) \;=\; \Big\{x=\oplus x_n\in\mathbb{F}_{\mathbb{H}}:\ \sum_n \left\|H^{(n)}x_n\right\|^2 \;<\; \infty\Big\}.$$

This operator is closed and self-adjoint (see Section 6.3.1 of [8]), and $-\frac{i}{\hbar}H$ is the generator of the strongly continuous group

$$W_t \;=\; \bigoplus_{n=1}^{\infty} W_t^{(n)}$$

of unitary operators on $\mathbb{F}_{\mathbb{H}}$, where $W_t^{(n)}=\exp\big(-\frac{it}{\hbar}H^{(n)}\big)$.

It is convenient to have some notation for the free part of the dynamics, and we will subsequently use

$$U_t^{(n)} \;=\; \exp\big(-\frac{it}{\hbar}L^{(n)}\big) \qquad\qquad \text{and} \qquad\qquad U_t \;=\; \bigoplus_{n=1}^{\infty} U_t^{(n)}.$$

The Liouville-von Neumann dynamics corresponding to (2) are given by the group

$$\mathcal{W}_t(D) \;=\; W_t D W_{-t} \tag{25}$$

of isometries of $\mathcal{T}(\mathbb{F}_{\mathbb{H}})$. (See Proposition 3.4 of [6] for a proof that groups of isometries defined in this way are strongly continuous.) Define the subspaces $\mathcal{T}_n \subset \mathcal{T}(\mathbb{H})$ consisting of all n-particle trace class operators:

$$\mathcal{T}_n \;=\; \left\{ \widehat{T} \text{ as in (17)} \;:\; T_n \in \mathcal{T}(\mathbb{H}^{(n)}) \right\}.$$

These subspaces are invariant under $\mathcal{W}_t$, and the restriction of $\mathcal{W}_t$ to $\mathcal{T}_n$ is

$$\mathcal{W}_t^{(n)}(T) \;=\; W_t^{(n)} T W_{-t}^{(n)}.$$

The generator of this group of isometries is

$$\mathcal{L}_n \;+\; \sum_{1 \le i < j \le n} [V_{ij},\, \cdot\,], \tag{26}$$

where $\mathcal{L}^{(n)}$ is the the generator of the group

$$\mathcal{U}_t^{(n)}(T) \;=\; U_t^{(n)} T U_{-t}^{(n)}, \tag{27}$$

which may be written formally as $-\frac{i}{\hbar}\sum^{n}[L_j,\, \cdot\,]$. (See [6] and references therein for a proof.) Since (26) is a bounded perturbation of $\mathcal{L}^{(n)}$, it follows that $\mathcal{W}_t(\widehat{T_n})$ equals $\widehat{T_n(t)}$, where

$$T_n(t) \;=\; \mathcal{U}_t^{(n)} T_n(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(n)} \sum_{1 \le i < j \le n} [V_{ij}, T_n(s)]\, ds \tag{28}$$

when $T_n \in \mathcal{T}_n$. Taking the m^{th} partial trace of both sides of (28) yields

$$\begin{aligned} T_{n:m}(t) \;&=\; \mathcal{U}_t^{(m)} T_{n:m}(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{1 \le i < j \le m} [V_{ij}, T_{n:m}(s)] \\ &-\; (n-m)\frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{j=1}^{m} [V_{j,m+1}, T_{n:m+1}(s)]_{:m}\, ds \end{aligned}$$

because of the symmetry properties of T and V. Multiplying both sides of the last equation by $n!/(n-m)!$ we obtain

$$\begin{aligned} \frac{n!}{(n-m)!} T_{n:m}(t) \;&=\; \mathcal{U}_t^{(m)} \frac{n!}{(n-m)!} T_{n:m}(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{1 \le i < j \le m} \left[V_{ij}, \frac{n!}{(n-m)!} T_{n:m}(s) \right] \\ &-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{j=1}^{m} \left[V_{j,m+1}, \frac{n!}{(n-m-1)!} T_{n:m+1}(s) \right]_{:m} ds. \end{aligned} \tag{29}$$

Now let $D = \oplus D_n$ be a density operator on $\mathbb{F}_{\mathbb{H}}$. Then $\mathcal{W}_t D = \oplus D_n(t)$, where $D_n(t)$ is the solution of (28) with initial condition $D_n(0) = D_n$. If D satisfies moment condition (15) then

$$\mathcal{N}_m(D(t)) \;=\; \sum_{n=m}^{\infty} \frac{n!}{(n-m)!} D(t)_{n:m}$$

is trace class. We abbreviate $\mathcal{N}_m(D(t))$ by $\mathcal{N}_m(t)$. Summing the right-hand sides of (29) with D in place of T yields

$$\begin{aligned} \mathcal{N}_m(t) \;&=\; \mathcal{U}_t^{(m)} \mathcal{N}_m(0) \;-\; \frac{i}{\hbar} \int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{1 \le i < j \le m} [V_{ij}, \mathcal{N}_m(s)] \\ &-\; \frac{i}{\hbar} \int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{j=1}^{m} [V_{j,m+1}, \mathcal{N}_{m+1}(s)]_{:m} \, ds \;, \end{aligned} \tag{30}$$

for (15) permits the interchange of the sum and the integral. This is equation (3) in integral form when $m = 1$. To summarize:

Proposition 3.1 *Let $\mathcal{U}_t$ and $\mathcal{W}_t$ be as defined in (27) and (25).*

Suppose that D is a density operator on $\mathbb{F}_{\mathbb{H}}$ of the form $D = \oplus D_n$ such that moment condition (15) holds for some $m \in \mathbb{N}$. Let $\mathcal{N}_m(t)$ denote $\mathcal{N}_m(\mathcal{W}_t(D))$. Then $\mathcal{N}_m(t)$ satisfies equation (30).

4 The TDHF hierarchy

The existence and uniqueness of mild solutions of the TDHF equation (4) is established in [6]. There it is shown that the integral equation

$$F(t) \;=\; \mathcal{U}_t^{(1)} F(0) \;-\; \frac{i}{\hbar} \int_0^t \mathcal{U}_{t-s}^{(1)} \left[V,\; F(s)^{\otimes 2} 2A_2 \right]_{:1} ds \tag{31}$$

has a unique solution $F(t)$ for any Hermitian trace class operator $F(0)$. Define $\mathcal{F}_1(t) = F(t)$ and, for $m > 1$, define

$$\mathcal{F}_m(t) \;=\; F(t)^{\otimes m} m! A_m. \tag{32}$$

We proceed to derive equations for the $\mathcal{F}_m(t)$ from (31).

First, set $G(t) = \mathcal{U}_{-t}^{(1)} F(t)$, so that

$$G(t) \;=\; F(0) \;-\; \frac{i}{\hbar} \int_0^t \mathcal{U}_{-s}^{(1)} \left[V,\; F(s)^{\otimes 2} 2A_2 \right]_{:1} ds.$$

Now apply the product rule (in integral form) to $G(t)^{\otimes m}$:

$$\begin{aligned} G(t)^{\otimes m} &= F(0)^{\otimes m} - \frac{i}{\hbar}\sum_{j=1}^{m}\int_0^t G(s)^{\otimes j-1}\otimes \mathcal{U}_{-s}^{(1)}\left[V,\ F(s)^{\otimes 2}2A_2\right]_{:1}\otimes G(s)^{\otimes n-j}ds \\ &= F(0)^{\otimes m} - \frac{i}{\hbar}\int_0^t \mathcal{U}_{-s}^{(m)}\sum_{j=1}^{m}\Big[V_{j,m+1},\ F(s)^{\otimes m+1}\big(I-U_{(j,m+1)}\big)\Big]_{:m}ds. \end{aligned}$$

Apply $\mathcal{U}_t^{(m)}$ to both sides of the preceding equation to obtain

$$F(t)^{\otimes m} = \mathcal{U}_t^{(m)}F(0)^{\otimes m} - \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)}\sum_{j=1}^{m}\Big[V_{j,m+1},\ F(s)^{\otimes m+1}\big(I-U_{(j,m+1)}\big)\Big]_{:m}ds.$$

Multiply both sides of the last equation by $m!A_m$ on the left, noting that $\mathcal{U}_s^{(m)}(X)A_m = \mathcal{U}_s^{(m)}(XA_m)$ and that A_m commutes with $\sum V_{j,m+1}$:

$$\mathcal{F}_m(t) = \mathcal{U}_t^{(m)}\mathcal{F}_m(0) - \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)}\sum_{j=1}^{m}\Big[V_{j,m+1},\ F(s)^{\otimes m+1}\big(I-U_{(j,m+1)}\big)m!A_m\Big]_{:m}ds.$$

Since $(m+1)!A_{m+1} = \big(I - U_{(1,m+1)}\ \cdots\ - U_{(m,m+1)}\big)m!A_m$, the last equation may be rewritten

$$\begin{aligned} \mathcal{F}_m(t) &= \mathcal{U}_t^{(m)}\mathcal{F}_m(0) - \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)}\sum_{j=1}^{m}\left[V_{j,m+1},\ \mathcal{F}_{m+1}(s)\right]_{:m}ds \\ &+ \sum_{1\le j\ne k\le m}\frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)}\Big[V_{j,m+1},\ F(s)^{\otimes m+1}U_{(k,m+1)}m!A_m\Big]_{:m}ds. \end{aligned} \tag{33}$$

We call these equations for the $\mathcal{F}_m(t)$ the *TDHF hierarchy*.

The trace norm of the last term in (33) is bounded by

$$m(m-1)\frac{2}{\hbar}\int_0^t \Big\|\left\{V_{m-1,m+1}U_{(m,m+1)}\big(\mathcal{F}_m(s)\otimes F(s)\big)\right\}_{:m}\Big\|_1 ds.$$

It can be verified that

$$\big(V_{m-1,m+1}U_{(m,m+1)}(\mathcal{F}_m(s)\otimes F(s))\big)_{:m} = \big(I^{\otimes m-1}\otimes F(s)\big)V_{m-1,m}\mathcal{F}_m(s),$$

whence the trace norm of the last term in (33) does not exceed

$$m(m-1)\frac{2}{\hbar}\int_0^t \|V\|\ \|F(s)\|\ \big\|\mathcal{F}_m(s)\big\|_1 ds. \tag{34}$$

Now $\|\mathcal{F}_m(s)\|_1 \leq \|F(s)\|_1^m$ by Proposition 2.3 since $\mathcal{F}_m = F^{\otimes m} m! A_m$. Furthermore, $\|F(s)\|_1 = \|F(0)\|_1$ and $\|F(s)\| = \|F(0)\|$ for all $s > 0$ by Proposition 4.3 of [6]. Substituting into (34) produces the bound

$$m(m-1)\frac{2\|V\|}{\hbar}\|F(0)\| \, \big\|F(0)\big\|_1^m t \tag{35}$$

on the trace norm of the last term in (33).

5 Accuracy of the TDHF approximation

We have shown that if D is a density operator on $\mathbb{F}_{\mathbb{H}}$ of the form $D = \oplus D_n$ such that $\sum n^2 \mathrm{Tr}(D_n) < \infty$, then

$$\mathcal{N}_1(t) \;=\; \mathcal{U}_t^{(1)} \mathcal{N}_1(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(1)} \left[V_{j,2}, \mathcal{N}_2(s)\right]_{:1} ds \;,$$

where $\mathcal{N}_1$ and $\mathcal{N}_2$ are the one-particle and two-particle reduced number density operators for a system which evolved under the dynamics (1) from the initial state D. In this section we will compare $\mathcal{N}_1(t)$ to the solution of the TDHF equation

$$\begin{aligned} F(t) \;&=\; \mathcal{U}_t^{(1)} F(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(1)} \left[V,\; F(s)^{\otimes 2} 2A_2\right]_{:1} ds \\ F(0) \;&=\; \mathcal{N}_1(0) \end{aligned} \tag{36}$$

whose initial condition is $\mathcal{N}_1(0)$. When the initial state D is a Gibbs density for noninteracting fermions, then the distance between $\mathcal{N}_1(t)$ and $F(t)$ in trace norm can be controlled at short times (Theorem 5.1). In the mean field limit, $F(t)$ is an asymptotically accurate approximation to $\mathcal{N}_1(t)$ at all times t, provided that the initial states are Gibbs states for noninteracting fermions (Theorem 5.2).

We state and discuss Theorems 5.1 and 5.2 before going on to prove them:

Theorem 5.1 *Let G be the density operator on $\mathbb{F}_{\mathbb{H}}$ of a Gibbs equilibrium state for noninteracting fermions, as in Proposition 2.2. Let $\mathcal{N}_1(t)$ and $\mathcal{N}_2(t)$ denote $\mathcal{N}_1(\mathcal{W}_t(G))$ and $\mathcal{N}_2(\mathcal{W}_t(G))$, respectively, where $\mathcal{W}_t$ is the dynamics with two-particle interactions defined in (25).*

Let $F(t)$ be the solution of the TDHF equation (36).

Let τ denote $(2\|V\|\|\mathcal{N}_1\|_1)^{-1}\hbar$. Then

$$\big\|\mathcal{N}_1(t) - F(t)\big\|_1 \;\leq\; \frac{3}{2}\left(\frac{t}{\tau - t}\right)^2 \tag{37}$$

for $t < \tau$.

This theorem implies, for instance, that

$$\left\|\mathcal{N}_1(\tau/2) - F(\tau/2)\right\|_1 \;\leq\; \frac{3}{2}.$$

This is remarkable because there are about $\frac{1}{2}\|\mathcal{N}_1\|_1^2$ interactions driving the dynamics (1) and the error could be much larger *prima facie*: it could be proportional to $\|\mathcal{N}_1\|_1$. Unfortunately, the bound (37) on the error of the TDHF approximation is valid only when $t < \tau$, which is inversely proportional to $\|\mathcal{N}_1\|_1$, and we have no explicit bounds for larger t. If $\|\mathcal{N}_1\|_1$ is too large, the bound (37) is useless, for then it is valid for too short a time.

The time-of-validity of (37) ends up being inversely proportional to $\|\mathcal{N}_1\|_1$ because the number of two-particle interactions is proportional to $\|\mathcal{N}_1\|_1^2$, and none of these interactions is weaker than any other *a priori*. In the thermodynamic limit, where $\|\mathcal{N}_1\|_1 \longrightarrow \infty$ with constant spatial density, the total interaction energy grows like $\|\mathcal{N}_1\|_1$ (rather than the square of $\|\mathcal{N}_1\|_1$) if the interaction potential is short-ranged. We do not know how to derive the TDHF equation in the thermodynamic limit, but we can derive it in the *mean field* limit, where the strength of the interaction is scaled in inverse proportion to $\|\mathcal{N}_1\|_1$.

For each value of the parameter $\lambda > 0$, consider the Hamiltonian

$$H^\lambda \;=\; \sum_{i,j}\langle j|L|i\rangle a_j^\dagger a_i \;+\; \lambda \sum_{i,j,k,l}\langle kl|V|ij\rangle a_k^\dagger a_l^\dagger a_j a_i. \tag{38}$$

If the initial state is given by a Gibbs equilibrium density D^λ for noninteracting fermions, then Proposition 3.1 implies that all reduced number density operators exist and satisfy

$$\begin{aligned}
\mathcal{N}_m^\lambda(t) \;&=\; \mathcal{U}_t^{(m)}\mathcal{N}_m^\lambda(0) \;-\; \lambda\frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{1\leq i<j\leq m}\left[V_{ij}, \mathcal{N}_m^\lambda(s)\right] \\
&-\; \lambda\frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(m)} \sum_{j=1}^{m}\left[V_{j,m+1}, \mathcal{N}_{m+1}^\lambda(s)\right]_{:m} ds \\
\mathcal{N}_m^\lambda(0) \;&=\; \mathcal{N}_m(D^\lambda(0)).
\end{aligned} \tag{39}$$

We will show that if λ is inversely proportional to $\|\mathcal{N}_1^\lambda\|_1$, then $\mathcal{N}_m^\lambda(t)$ is close to $F^\lambda(t)^{\otimes m} m! A_m$ in trace norm, where

$$\begin{aligned}
F^\lambda(t) \;&=\; \mathcal{U}_t^{(1)}F^\lambda(0) \;-\; \frac{i}{\hbar}\int_0^t \mathcal{U}_{t-s}^{(1)}\left[V,\; F^\lambda(s)^{\otimes 2} 2A_2\right]_{:1} ds \\
F^\lambda(0) \;&=\; \mathcal{N}_1^\lambda(0).
\end{aligned} \tag{40}$$

Theorem 5.2 *Let $\left\{D^\lambda\right\}_{\lambda>0}$ be a family of Gibbs equilibrium densities for noninteracting fermions, as in Proposition 2.2, with*

$$\limsup_{\lambda\to 0} \lambda\|\mathcal{N}_1(D^\lambda)\|_1 \;<\; \infty.$$

Let $\mathcal{N}_m^\lambda(t)$ be the solution of (39), let $F^\lambda(t)$ be the solution of the TDHF equation (40) and let $\mathcal{F}_m^\lambda(t) = F^\lambda(t)^{\otimes m} m! A_m$. Then

$$\lim_{\lambda\to 0} \left\|\mathcal{N}_m^\lambda(t) - \mathcal{F}_m^\lambda(t)\right\|_1 / \|\mathcal{N}_1^\lambda\|_1^m \;=\; 0$$

for all $t > 0$ and all $m \in \mathbb{N}$.

We will derive Theorems 5.1 and 5.2 from Lemma 5.1 below.

Let $\mathcal{N}_m(t)$ be as in Proposition 3.1, and let $\mathcal{F}_m(t)$ satisfy the TDHF hierarchy. In the hypotheses Theorems 5.1 and 5.2 we suppose that $F(0) = \mathcal{N}_1(D(0))$, but for now let us only assume that

$$\|F(0)\| \leq 1 \qquad \text{and} \qquad \|F(0)\|_1 = \|\mathcal{N}_1(D(0))\|_1. \tag{41}$$

The trace norm of $\mathcal{N}_1(t)$ is independent of t, and we shall denote it simply by $\|\mathcal{N}_1\|_1$. Assuming (41), the bound (35) is itself bounded by

$$m(m-1)\frac{2\|V\|}{\hbar}\, \left\|\mathcal{N}_1\right\|_1^m t. \tag{42}$$

Subtracting equations (33) from equations (30) and using (42) leads to the estimates

$$\begin{aligned}
\left\|\mathcal{N}_m(t) - \mathcal{F}_m(t)\right\|_1 \;&\leq\; \left\|\mathcal{N}_m(0) - \mathcal{F}_m(0)\right\|_1 \\
&+\; \frac{m(m-1)}{\hbar}\|V\|\Big(2\|\mathcal{N}_1\|_1^m + \|\mathcal{N}_m\|_1\Big)t \\
&+\; m\frac{2\|V\|}{\hbar}\int_0^t \left\|\mathcal{N}_{m+1}(s) - \mathcal{F}_{m+1}(s)\right\|_1 ds\ .
\end{aligned}$$

Iterating this estimate n times, one obtains

$$\begin{aligned}
\left\|\mathcal{N}_m(t) - \mathcal{F}_m(t)\right\|_1 \;&\leq\; \sum_{j=0}^{n} a_{m+j}\binom{m+j-1}{j} C^j t^j \;+\; \sum_{j=0}^{n}\frac{b_{m+j}}{j+1}\binom{m+j-1}{j} C^j t^{j+1} \\
&+\; C^n \frac{(m+n)!}{(m-1)!}\int_0^t\int_0^{t_1}\cdots\int_0^{t_n} \left\|\mathcal{N}_{m+n+1}(s) - \mathcal{F}_{m+n+1}(s)\right\|_1 ds dt_n \cdots dt_1
\end{aligned} \tag{43}$$

with $C = 2\|V\|/\hbar$ and

$$\begin{aligned}
a_m \;&=\; \left\|\mathcal{N}_m(0) - \mathcal{F}_m(0)\right\|_1 \\
b_m \;&=\; \frac{m(m-1)}{\hbar}\|V\|\Big(2\|\mathcal{N}_1\|_1^m + \|\mathcal{N}_m\|_1\Big).
\end{aligned} \tag{44}$$

To make use of these estimates we need some control over the size of the integrand in (43). We will assume that there exists a constant B such that

$$\|\mathcal{N}_m\|_1 \;\leq\; B\|\mathcal{N}_1\|_1^m \tag{45}$$

for all m. Then the last term on the right hand side of (43) is bounded by

$$\binom{m+n-1}{m-1}(B+1)\big\|\mathcal{N}_1\big\|_1^{m+n}C^n t^n,$$

which tends to 0 as n tends to infinity if m fixed and $C\|\mathcal{N}_1\|_1 t < 1$. Furthermore, assuming (45), we can bound b_m of (44) by $C(B/2+1)\|\mathcal{N}_1\|_1^m m(m-1)$ and we find the following:

Lemma 5.1 *Suppose that D is a density operator on $\mathbb{F}_{\mathbb{H}}$ of the form $D = \oplus D_n$, such that (45) holds for all $m \in \mathbb{N}$. Let $\mathcal{W}_t$ be as defined in (25) and let $\mathcal{N}_m(t)$ denote $\mathcal{N}_m(\mathcal{W}_t(D))$. Let $F(t)$ be the solution of a TDHF equation (31) whose initial condition $F(0)$ satisfies (41), and let $\mathcal{F}_m(t)$ be as in (32). Then, with $C = 2\|V\|/\hbar$,*

$$\begin{aligned}\frac{\big\|\mathcal{N}_m(t)-\mathcal{F}_m(t)\big\|_1}{\big\|\mathcal{N}_1\big\|_1^m} \;&\leq\; \sum_{j=0}^{\infty}\frac{\big\|\mathcal{N}_{m+j}(0)-\mathcal{F}_{m+j}(0)\big\|_1}{\big\|\mathcal{N}_1\big\|_1^{m+j}}\binom{m+j-1}{m-1}\big(C\|\mathcal{N}_1\|_1 t\big)^j \\ &+\; \big\|\mathcal{N}_1\big\|_1^{-1}\frac{B+2}{2}\sum_{j=0}^{\infty}(m+j-1)\binom{m+j}{m-1}\big(C\|\mathcal{N}_1\|_1 t\big)^{j+1}\end{aligned}$$

when $C\|\mathcal{N}_1\|_1 t < 1$.

Proof of Theorem 5.1 When G is the density operator of a Gibbs equilibrium state and $F(0) = \mathcal{N}_1(G)$, then $\|F(0)\| = \|\mathcal{N}_1(G)\| \leq 1$ by Proposition 2.1, $\big\|\mathcal{N}_m(0)-\mathcal{F}_m(0)\big\|_1 = 0$ for all m by Proposition 2.2, and $B = 1$ in (45) by Proposition 2.4. Upon simplifying the inequality in Lemma 5.1, one obtains Theorem 5.1. □

Proof of Theorem 5.2 For $\mathcal{N}_m^\lambda(t)$ and $\mathcal{F}_m^\lambda(t)$ as in the hypothesis of Theorem 5.2, observe that

$$\|F(t)\| \leq 1 \qquad \text{and} \qquad \|F(t)\|_1 = \|\mathcal{N}_1(D(t))\|_1$$

at all times t, and $\mathcal{N}_m^\lambda$ satisfies (45) with $B = 1$. Thus, we may apply Lemma 5.1, with a few changes: t and $t+\Delta t$ may be substituted for 0 and t, and C should be replaced by $C\lambda$. These substitutions yield

$$\begin{aligned}\frac{\big\|\mathcal{N}_m^\lambda(t+\Delta t)-\mathcal{F}_m^\lambda(t+\Delta t)\big\|_1}{\|\mathcal{N}_1^\lambda\|_1^m} \;&\leq\; \sum_{j=0}^{\infty}\frac{\big\|\mathcal{N}_{m+j}^\lambda(t)-\mathcal{F}_{m+j}^\lambda(t)\big\|_1}{\|\mathcal{N}_1^\lambda\|_1^{m+j}}\binom{m+j-1}{m-1}\big(C\lambda\|\mathcal{N}_1^\lambda\|_1\Delta t\big)^j \\ &+\; \frac{1}{\|\mathcal{N}_1^\lambda\|_1}\frac{3}{2}\sum_{j=0}^{\infty}(m+j-1)\binom{m+j}{m-1}\big(C\lambda\|\mathcal{N}_1^\lambda\|_1\Delta t\big)^{j+1}\end{aligned} \tag{46}$$

for $\Delta t < (C\lambda\|\mathcal{N}_1^\lambda\|_1)^{-1}$. Since $u = \limsup_{\lambda\to 0} C\lambda\|\mathcal{N}_1^\lambda\|_1$ is finite by hypothesis, taking the lim sup of both sides of (46) implies that

$$\lim_{\lambda\to 0} \left\|\mathcal{N}_m^\lambda(s) - \mathcal{F}_m^\lambda(s)\right\|_1 / \|\mathcal{N}_1^\lambda\|_1^m \;=\; 0 \qquad \forall\; m \in \mathbb{N} \tag{47}$$

holds at time $s = t + \Delta t$ if it holds at time $s = t$ and $\Delta t < 1/u$. Since (47) holds at $s = 0$, an inductive argument proves that it holds at all times $s > 0$. □

6 Appendix: the proofs of Propositions 2.1, 2.2, and 2.3

6.1 Proof of Proposition 2.1

We begin by proving

Proposition 6.1 *If D_n is an n-particle fermionic density operator then*

$$\left\|D_{n:1}\right\| \;\leq\; 1/n. \tag{48}$$

Proof: Thanks to the convexity of the norm and the linearity of the partial trace, it suffices to prove (48) for fermionic pure states. Let $\mathbb{H}$ denote the single-particle Hilbert space, and let Ψ be a unit vector in $\mathbb{H}^{(n)}$. Since $P_{\Psi:1}$ is a compact Hermitian operator, there exists a unit vector $u \in \mathbb{H}$ such that

$$\left\|P_{\Psi:1}\right\| \;=\; \langle u, P_{\Psi:1}(u)\rangle. \tag{49}$$

Let $\{\phi_j\}_{j\in J}$ be a basis of $\mathbb{H}$ containing u. For each subset $\mathbf{s} = \{j_1, \ldots, j_n\}$ of J, let $\Psi(\mathbf{s})$ denote one of the two Slater determinants that may be formed from the vectors $\phi_{j_1}, \ldots, \phi_{j_n}$ (the two choices differ only in sign). The set of vectors $\Psi(\mathbf{s})$ is an orthonormal basis of $\mathbb{H}^{(n)}$ and so $\Psi = \sum_{\mathbf{s}} \langle \Psi(\mathbf{s}), \Psi\rangle \Psi(\mathbf{s})$. By definition of the partial trace, $\langle u, P_{\Psi:1}(u)\rangle$ equals

$$\sum_{j_1,\ldots,j_{n-1}\in J} \Big\langle u\otimes\phi_{j_1}\otimes\cdots\otimes\phi_{j_{n-1}},\; P_\Psi(u\otimes\phi_{j_1}\otimes\cdots\otimes\phi_{j_{n-1}})\Big\rangle. \tag{50}$$

From (49) and (50)

$$\begin{aligned}
\|P_{\Psi:1}\| \;&=\; \sum_{j_1,\ldots,j_{n-1}\in J} \left\|P_\Psi(u\otimes\phi_{j_1}\otimes\cdots\otimes\phi_{j_{n-1}})\right\|^2 \\
&=\; \sum_{j_1,\ldots,j_{n-1}\in J} \sum_{\mathbf{s}} \left|\langle\Psi(\mathbf{s}),\Psi\rangle\right|^2 \left|\left\langle\Psi(\mathbf{s}), u\otimes\phi_{j_1}\otimes\cdots\otimes\phi_{j_{n-1}}\right\rangle\right|^2.
\end{aligned} \tag{51}$$

But

$$\left|\left\langle \Psi(\mathbf{s}), u \otimes \phi_{j_1} \otimes \cdots \otimes \phi_{j_{n-1}} \right\rangle\right|^2 \;=\; \begin{cases} 1/n! & \text{if} \quad \mathbf{s} = \{u, \phi_{j_1}, \ldots, \phi_{j_{n-1}}\} \\ 0 & \text{otherwise} \end{cases}$$

whence

$$\|P_{\Psi:1}\| \;\leq\; \sum_{\mathbf{s}} |\langle \Psi(\mathbf{s}), \Psi \rangle|^2 \, \frac{(n-1)!}{n!} \;=\; \frac{1}{n} \sum_{\mathbf{s}} |\langle \Psi(\mathbf{s}), \Psi \rangle|^2 \;=\; \frac{1}{n}$$

by (51). □

Now we return to the proof of Proposition 2.1.

Let D be a density operator on $\mathbb{F}_{\mathbb{H}}$ that commutes with the number operator N and satisfies $\mathrm{Tr}(ND) < \infty$. Then D has the form (14) and $\sum n\mathrm{Tr}(D_n) < \infty$, so that $\mathcal{N}_1(D) = \sum nD_{n:1}$ is defined and

$$\big\|\mathcal{N}_1(D)\big\| \;\leq\; \sum_{n=1}^{\infty} n \big\|D_{n:1}\big\|.$$

By Proposition 6.1 the operator norm of D_n is less than or equal to $\mathrm{Tr}(D_n)/n$, whence

$$\big\|\mathcal{N}_1(D)\big\| \;\leq\; \sum_{n=1}^{\infty} n\mathrm{Tr}(D_n)/n \;=\; \mathrm{Tr}(D) \;=\; 1.$$

6.2 Proof of Proposition 2.2

From (22), with $\mathbb{P}(\mathbf{s})$ as in (20),

$$\mathcal{N}_n(G) \;=\; \sum_{\mathbf{s}: N(\mathbf{s}) \geq n} \frac{N(\mathbf{s})!}{(N(\mathbf{s})-n)!} \mathbb{P}(\mathbf{s}) \left(P_{\Psi(\mathbf{s})}\right)_{:n}.$$

Substituting the expressions (13) and (12) for $\left(P_{\Psi(\mathbf{s})}\right)_{:n}$ and collecting terms, we find that

$$\begin{aligned} \mathcal{N}_n(G) \;&=\; \sum_{\mathbf{s}: N(\mathbf{s}) \geq n} \mathbb{P}(\mathbf{s}) \left(N(\mathbf{s}) P_{\Psi(\mathbf{s})}\right)_{:1}^{\otimes n} n! A_n \\ &=\; \sum_{\substack{distinct \\ j_1, \ldots, j_n \in J}} \Big[\sum_{\mathbf{s} \ni j_1, \ldots, j_n} \mathbb{P}(\mathbf{s}) \Big] \big(P_{\phi_{j_1}} \otimes \cdots \otimes P_{\phi_{j_n}}\big) n! A_n \;, \end{aligned} \tag{52}$$

where $N(\mathbf{s})$ is the size of $\mathbf{s}$. The sum in (52) is made over *distinct* $j_1, \ldots, j_n$ since $(P_{\phi_{j_1}} \otimes \cdots \otimes P_{\phi_{j_n}})A_n$ equals the zero operator if $j_r = j_s$ for any $r \neq s$. In particular,

$$\mathcal{N}_1(G) \;=\; \sum_{j \in J} \Big[\sum_{\mathbf{s} \ni j} \mathbb{P}(\mathbf{s}) \Big] P_{\phi_j} \;=\; \sum_{j \in J} p(j) P_{\phi_j}. \tag{53}$$

Thus,

$$\mathcal{N}_1(G)^{\otimes n} n! A_n \;=\; \sum_{\substack{distinct \\ j_1,\ldots,j_n \in J}} \prod_{i=1}^{n} p(j_i) \big(P_{\phi_{j_1}} \otimes \cdots \otimes P_{\phi_{j_n}}\big) n! A_n. \tag{54}$$

The sum in (54) is again restricted to distinct $j_1, \ldots, j_n$ because of the presence of the anti-symmetrizer A_n. Substituting (21) into (54) yields (52), proving the proposition.

6.3 Proof of Proposition 2.3

Suppose T is a Hermitian trace class operator. There exists an orthonormal basis $\{e_j\}_{j\in J}$ of $\mathbb{H}$ such that

$$T \;=\; \sum_j \lambda_j P_{e_j} \qquad \text{with} \qquad \sum_j |\lambda_j| \;=\; \big\|T\big\|_1.$$

The operator $T^{\otimes n} n! A_n$ is diagonalizable with respect to the basis of Slater determinants formed from n distinct members of $\{e_j\}$. Indeed,

$$\begin{aligned} T^{\otimes n} n! A_n \big(\sqrt{n!}\; A_n(e_{j_1} \otimes \cdots \otimes e_{j_n})\big) \;&=\; \sqrt{n!} A_n n! T^{\otimes n} (e_{j_1} \otimes \cdots \otimes e_{j_n}) \\ &=\; \Big(n! \prod_{s=1}^{n} \lambda_{j_s}\Big) \sqrt{n!}\; A_n(e_{j_1} \otimes \cdots \otimes e_{j_n}). \end{aligned}$$

The trace norm of $T^{\otimes n} n! A_n$ is the sum of the absolute values of its eigenvalues, whence

$$\big\|T^{\otimes n} n! A_n\big\|_1 \;=\; \sum_{\substack{distinct \\ j_1,\ldots,j_n \in J}} \prod_{s=1}^{n} |\lambda_{j_s}|\,. \tag{55}$$

Note that the sum in (55) is over ordered sequences $j_1, j_2, \ldots, j_n$ rather than subsets $\{j_1, \ldots, j_n\}$. But

$$\sum_{\substack{distinct \\ j_1,\ldots,j_n \in J}} \prod_{s=1}^{n} |\lambda_{j_s}| \;\le\; \sum_{j_1,\ldots,j_n \in J} \prod_{s=1}^{n} |\lambda_{j_s}| \;=\; \prod_{s=1}^{n} \sum_{j\in J} |\lambda_j| \;=\; \|T\|_1^n$$

proving (24).

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